

# **ISSUE RESOLUTION STATUS REPORT**

## **KEY TECHNICAL ISSUE: EVOLUTION OF THE NEAR-FIELD ENVIRONMENT**

**Division of Waste Management  
Office of Nuclear Material Safety & Safeguards  
U.S. Nuclear Regulatory Commission**

**Revision 3**

**August 2000**



History of Major Changes to “Issue Resolution Status Report (IRSR), Key Technical Issue: Evolution of the Near-Field Environment (ENFE)”

<u>Revision #</u>	<u>Section</u>	<u>Date</u>	<u>Modification</u>
Rev0	all	September 1997	None. Initial issue.
Rev1	2.0	August 1998	Redefinition/clarification of subissues; addition of new subissue on near-field criticality
Rev1	3.0	August 1998	Expanded discussion of relationship between ENFE IRSR and other IRSR and repository performance. Added new sections on potential nuclear criticality in the near field and on TSPA-VA Methods and Assumptions.
Rev1	4.0	August 1998	Expanded discussion of relationships between Acceptance Criteria (AC) and repository design and concept of operation, Total System Performance Assessments (TSPA), 10 CFR 63, and the TSPA IRSR. Added Review Methods (RM). Added sections on microbial effects on seepage and flow, waste package chemical environment, chemical environment for radionuclide release, and radionuclide transport through engineered and natural barriers. New subissue on potential near-field criticality with RM, AC, and technical bases identified
Rev1	5.0	August 1998	Revised site characterization analysis open items' status and their description to reflect performance assessment approach. New sections summarizing subissue resolution achieved in the revision, and those coupled thermal-hydrologic-chemical processes potentially affecting repository performance Revised to reflect progress in issue resolution, and to reflect changes in the NRC and DOE programs. Revised to reflect the proposed 10 CFR Part 63

History of Major Changes to “Issue Resolution Status Report (IRSR), Key Technical Issue: Evolution of the Near-Field Environment (ENFE) (cont’d)”

<u>Revision #</u>	<u>Section</u>	<u>Date</u>	<u>Modification</u>
Rev2	5.0	July 1999	Updated discussion of status of resolution and site characterization and study plan comments to reflect review of TSPA-VA. Revised Rev1 to reflect application of acceptance criteria to review of TSPA-VA
Rev3	Executive Summary	August 2000	Added per NRC Plain English directive
Rev3	1.0	August 2000	Address migration of ACs and RMs to Yucca Mountain Review Plan, nature of documents reviewed, and clarify resolution definitions.
Rev3	3.0	August 2000	Updated to reflect DOE’s new repository design, changes in NRC integrated subissues, and new discussion on solubility.
Rev3	4.0	August 2000	ACs and RMs migrated to the Yucca Mountain Review Plan. Updated to reflect DOE’s new repository design (e.g., drip shield).
Rev3	5.0	August 2000	Moved summary to beginning. ACs were revised. Major revisions were made to reflect progress in issue resolution based on application of ACs to review of the preliminary draft DOE Analysis and Model Reports and Process Model Reports

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## ACRONYM LIST

CRWMS M&O	Civilian Radioactive Waste Management System Management and Operating Contractor
DOE	U.S. Department of Energy
ENFE	Evolution of the Near-Field Environment
FEPs	Features, Events, and Processes
IRSR	Issue Resolution Status Report
KTI	Key Technical Issue
NRC	U.S. Nuclear Regulatory Commission
THC	Thermal-Hydrologic-Chemical
THMC	Thermal-Hydrologic-Mechanical-Chemical
TSPA	Total System Performance Assessment
TSPAI	Total System Performance Assessment and Integration
TSPA-SR	Total System Performance Assessment for Site Recommendation
TSPA-VA	Total System Performance Assessment for Viability Assessment
TSPA-93	Total System Performance Assessment conducted in 1993
TSPA-95	Total System Performance Assessment conducted in 1993

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## QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

**DATA:** CNWRA-generated original data contained in this report meets quality assurance requirements described in the CNWRA Quality Assurance Manual. Sources for other data should be consulted for determining the level of quality for those data.

**ANALYSES AND CODES:** The TPA code Versions 3.1 and 3.2 were developed under TOP-018 procedures, which implements the quality assurance guidance contained in the CNWRA quality assurance manual.

## EXECUTIVE SUMMARY

The Evolution of the Near-Field Environment Key Technical Issue relates to all aspects of the evolution of the near-field geochemical environment that have the potential to affect the performance of the proposed repository. The near-field geochemistry will be perturbed from ambient conditions by variations in temperature and pressure associated with heat production of the waste, introduction of foreign materials into the mountain, variations in fluid flow, and consequent chemical reactions. The effects of coupled thermal-hydrologic-chemical processes influence parameter values and conceptual models used in performance assessment calculations. This report is focused on the adequacy of the methodology, testing, and modeling efforts conducted by the U.S. Department of Energy (DOE) to evaluate the effects of the evolution of the near-field geochemical environment on repository performance. The following five subissues are addressed in this report: (i) effects of coupled thermal-hydrologic-chemical processes on seepage and flow, (ii) effects of coupled thermal-hydrologic-chemical processes on the waste package and drip shield chemical environment, (iii) effects of coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release, (iv) effects of coupled thermal-hydrologic-chemical processes on radionuclide transport through engineered and natural barriers, and (v) effects of coupled thermal-hydrologic-chemical processes on potential nuclear criticality in the near field.

With this revision (Revision 3) of this report, the status of issue resolution was updated based on information available to the staff prior to May 15, 2000. DOE models, abstractions, and analyses were assessed based on information provided in the DOE Total System Performance Assessment-Site Recommendation Methods and Assumptions report, Revision 3 of the DOE Repository Safety Strategy, and the preliminary, draft, DOE process model reports and analysis and model reports that were available.

The purpose of issue resolution is to assure that sufficient information is available on an issue to enable the NRC to docket the license application. Resolution at the staff level does not preclude an issue being raised and considered during the licensing proceedings, nor does it prejudice what the NRC staff evaluation of that issue will be after its licensing review. Issue resolution at the staff level during prelicensing is achieved when the staff has no further questions or comments at a point in time regarding how the DOE is addressing an issue. Pertinent additional information could raise new questions or comments regarding a previously resolved issue.

Issues are "closed" if the DOE approach and available information acceptably address staff questions such that no information beyond what is currently available will likely be required for regulatory decision making at the time of initial license application. Issues are "closed-pending" if the NRC staff has confidence that the DOE proposed approach, together with the DOE agreement to provide the NRC with additional information (through specified testing, analysis, etc.) acceptably addresses the NRC's questions such that no information beyond that provided, or agreed to, will likely be required at time of initial license application. Issues are "open" if the NRC has identified questions regarding the DOE approach or information, and the DOE has not yet acceptably addressed the questions or agreed to provide the necessary additional information in the license application. The status of the five subissues and comments regarding their resolution are summarized in the following table.

Subissue	Status of Resolution	Path to Resolution
Effects of coupled thermal-hydrologic-chemical processes on seepage and flow	Open	Information is needed on potential thermal alteration of zeolites and volcanic glasses. DOE needs to evaluate the effect of cementitious materials on flow. Neglect of mineral precipitation on fracture surfaces must be justified. DOE must provide adequate technical bases for neglecting several thermal-hydrologic-chemical processes in its model abstractions.
Effects of coupled thermal-hydrologic-chemical processes on the waste package chemical environment	Open	DOE must improve: technical bases for microbially influenced corrosion of Alloy 22; drip shield degradation; excluded features, events and processes; and major assumptions in Total System Performance Assessment-Site Recommendation Methods and Assumptions report.
Effects of coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release	Open	DOE must provide adequate technical bases for neglecting several thermal-hydrologic-chemical processes in its model abstractions.
Effects of coupled thermal-hydrologic-chemical processes on radionuclide transport through engineered and natural barriers	Open	Technical bases for exclusion of features, events, and processes screening needs to be consistent and sufficient justification must be provided. DOE must demonstrate that effects of coupled thermal-hydrologic-chemical processes will not adversely affect matrix diffusion and sorptive minerals, or include the effects in abstractions.
Effects of coupled thermal-hydrologic-chemical processes on potential nuclear criticality in the near field	Closed, pending confirmation	Open items concerning the DOE criticality analysis methodology raised in the NRC safety evaluation report need to be closed. Document technical basis for exclusion of features, events, and processes.

## 1.0 INTRODUCTION

Site characterization activities are specified in the U.S. Nuclear Regulatory Commission (NRC) geologic repository regulations and in the proposed Commission rule (U.S. Nuclear Regulatory Commission, 1999a). The Commission has noted that ongoing review of information from site investigation and characterization activities, particularly those activities with long completion schedules, allows for the early identification and resolution of potential licensing issues. Moreover, the NRC strategic planning assumptions call for the early identification and resolution of issues at the staff level. The principal means for achieving this goal is through formal, prelicensing consultation with the U.S. Department of Energy (DOE). These consultations are required by law and occur in an open manner that permits observation by the State of Nevada, Tribal Nations, affected units of local government, and interested members of the public. Obtaining input and striving for consensus from the technical community and interested parties helps the issue resolution process. The issue resolution approach attempts to reduce the number of, and to better define, issues that may be in dispute during any potential NRC licensing review.

Consistent with NRC regulations on prelicensing consultations and a 1992 agreement with DOE, staff-level resolution can be achieved during prelicensing consultation. The purpose of issue resolution is to assure that sufficient information is available on an issue to enable the NRC to docket the license application. Resolution at the staff level does not preclude an issue being raised and considered during the licensing proceedings, nor does it prejudice what the NRC staff evaluation of that issue will be after its licensing review. Issue resolution at the staff level during prelicensing is achieved when the staff has no further questions or comments at a point in time regarding how the DOE is addressing an issue. Pertinent additional information could raise new questions or comments regarding a previously resolved issue.

Issues are “closed” if the DOE approach and available information acceptably address staff questions such that no information beyond what is currently available will likely be required for regulatory decision making at the time of initial license application. Issues are “closed-pending” if the NRC staff has confidence that the DOE proposed approach, together with the DOE agreement to provide the NRC with additional information (through specified testing, analysis, etc.) acceptably addresses the NRC's questions such that no information beyond that provided, or agreed to, will likely be required at time of initial license application. Issues are “open” if the NRC has identified questions regarding the DOE approach or information, and the DOE has not yet acceptably addressed the questions or agreed to provide the necessary additional information in the license application.

The NRC high-level radioactive waste program was realigned during fiscal year 1996-1997. The realignment was in response to: (i) a reduction in Congressional budget appropriations for NRC in fiscal year 1996; (ii) the reorganization of DOE's geologic repository program at Yucca Mountain, Nevada; and (iii) a 1995 report issued by the National Academy of Sciences to advise the U.S. Environmental Protection Agency regarding the technical bases for new geologic disposal standards for Yucca Mountain. In response to these developments, the NRC high-level radioactive waste program was realigned to focus prelicensing work on those topics most critical to the postclosure performance of the proposed geologic repository; these topics are called Key Technical Issues (KTIs). [This approach is summarized in Chapter 1 of the staff's fiscal year 1996 Annual Progress Report (see Sagar, 1997).]

The current Division of Waste Management approach is to focus most activities on issue resolution of the respective KTIs, at the staff level. The division's activities have been re-prioritized to streamline and improve the integration of the technical work necessary to achieve staff-level resolution. Regulatory attention is focused where technical uncertainties will have the greatest affect on the assessment of repository safety, and is achieved by identifying KTIs, integrating their activities into a risk-informed approach, and evaluating their significance for postclosure repository performance. Early feedback among all parties is essential to define what is known, what is not known, and where additional information is likely to make a significant difference in the understanding of future repository safety.

An important step in our approach to issue resolution is to provide DOE with feedback regarding issue resolution. Issue Resolution Status Reports (IRSRs) are the primary mechanism that we use to provide DOE with feedback on KTI subissues. IRSRs focus on: (i) acceptance criteria for issue resolution; and (ii) the status of resolution, including areas of agreement or when we have comments or questions. Feedback is also contained in the staff's Fiscal Year 1996 Annual Progress Report (Sagar, 1997), which summarized the significant technical work toward resolution of all KTIs during the fiscal year. Finally, open meetings and technical exchanges with DOE provide additional opportunities to discuss issue resolution, identify areas of agreement and disagreement, and develop plans to resolve such disagreements. In addition, we are currently using the IRSRs to develop the Yucca Mountain Review Plan for any potential repository license application.

Each IRSR contains five sections. This Introduction is Section 1.0. Section 2.0 defines the KTI, all the related subissues, and the scope of the particular subissue that is the subject of the IRSR. Section 3.0 discusses the importance of the subissue to repository performance including: (i) qualitative descriptions; (ii) relationship to total system performance; (iii) results of available sensitivity analyses; and (iv) relationship to the DOE Repository Safety Strategy [see Civilian Radioactive Waste Management System Management and Operating Contractor (CRWMS M&O), 2000a; U.S. Department of Energy, 1998a]. Section 4.0 provides the technical bases for our review methods and acceptance criteria that the staff will use in subsequent reviews of DOE submittals. These acceptance criteria are guidance for the staff and, indirectly, for DOE as well. Note that the review methods and acceptance criteria previously listed in Section 4.0 of the Evolution of the Near-Field Environment (ENFE) IRSR Revision 2 are being used to develop the Yucca Mountain Review Plan. In the process, they have undergone some modifications. The acceptance criteria listed in Section 5.0 are consistent with and reflect, to the extent practicable, those modifications. Our technical bases for the acceptance criteria are explained in detail in Section 4.0 to document the rationale for our decisions regarding issue resolution. Section 5.0 summarizes and provides an analysis of the DOE models, abstractions, and analyses, and concludes the IRSR with the status of resolution, indicating those items resolved at the staff level and those items remaining open. The staff will track these open items, and a resolution will be documented in future reports.

The technical bases contained in the IRSR was the basis for our review of information in the DOE Viability Assessment (U.S. Department of Energy, 1998b), and will be the basis for our review of DOE's Site Recommendation Considerations Report and supporting Analysis and Model Reports and Process Model Reports. Our comments on these documents are intended to facilitate DOE's efforts to focus its program and develop a high-quality license application. For Revision 2 of the ENFE IRSR, we reviewed the preliminary design concept, the total system performance assessment, the license application plan, and supporting documents (U.S.

Department of Energy, 1998b,c,d). Through these reviews, we identified a set of technical comments regarding the supporting data and models within the Total System Performance Assessment-Viability Assessment (U.S. Nuclear Regulatory Commission, 1999b). Detailed comments on the Viability Assessment, based on the application of review methods and acceptance criteria, were provided in Section 5.0 of the ENFE IRSR, Revision 2. With this revision (Revision 3) of the IRSR, the status of issue resolution was updated based on information on the DOE models, abstractions, and analyses taken from the DOE Total System Performance Assessment-Site Recommendation methods and assumptions report (CRWMS M&O, 1999a), Revision 3 of the DOE Repository Safety Strategy (CRWMS M&O, 2000a), and the DOE Process Model Reports and Analysis and Model Reports that were available for review in preliminary draft, or final form by May 15, 2000.

The NRC staff recognizes the preliminary nature of the draft Analysis and Model Reports and Process Model Reports; specifically, they have not been accepted by DOE. Thus, the staff has not used the information contained in those draft documents to resolve any open subissues in this report. To aid the issue resolution process, however, the staff has reviewed and provided comments on the sufficiency of the information in the preliminary documents to address staff concerns. After a review of the final Process Model Reports or other documents that indicate DOE's acceptance of the information in the preliminary documents, the staff will consider whether it is appropriate to close the subissues or any portion the subissues.

## 2.0 KEY TECHNICAL ISSUE AND SUBISSUES

Revision 3 of the Issue Resolution Status Report (IRSR) addresses all the Evolution of the Near-Field Environment (ENFE) subissues. This report provides the technical bases for the acceptance criteria that will be used to judge the U.S. Department of Energy (DOE) evaluations regarding the effects of the ENFE on repository performance at Yucca Mountain. DOE evaluations will be presented in the Site Recommendation Consideration Report and in a potential license application. The staff evaluates in this version of the IRSR whether the DOE assessment of the effects of ENFE on repository performance includes (i) important design features, physical phenomena and processes, and consistent and appropriate assumptions; (ii) sufficient data to adequately define relevant parameters and conceptual models; (iii) parameter values used in the performance assessment abstractions that are consistent with site characterization data, design data, laboratory experiments, field measurements, and natural analog data; (iv) consideration of alternative models; and (v) performance assessment abstractions that are justified by comparison with process-level models and empirical observations.

Both the U.S. Nuclear Regulatory Commission (NRC) and the DOE are evaluating the potential Yucca Mountain repository using a performance assessment approach. The NRC site-specific regulations for the proposed Yucca Mountain repository have been issued as the proposed 10 CFR Part 63 (U.S. Nuclear Regulatory Commission, 1999a). The proposed standards are risk-informed and performance-based. The near-field environment is defined from NRC's perspective in terms of potential impact on the performance of the proposed geologic repository. The near field is considered to be the portion of the site where changes in the physical and chemical properties, resulting from the construction of the underground facility or from the heat generated by the emplaced radioactive waste, affect performance of the repository. The extent of the near field may vary substantially depending on the specific processes of concern. With respect to repository performance, large portions of the mountain may be affected significantly by some thermal-hydrologic-chemical (THC) coupled processes. Other coupled THC processes may have effects only close to or within the engineered barrier system. Coupled processes considered for this IRSR are THC interrelations associated with the near field of the proposed repository at Yucca Mountain. Thermal-hydrologic couplings and thermal-mechanical couplings are addressed primarily in the IRSRs on Thermal Effects on Flow (U.S. Nuclear Regulatory Commission, 1998a,1999c) and Repository Design and Thermal Mechanical Effects (U.S. Nuclear Regulatory Commission, 1998b), respectively (see also; <http://www.nrc.gov/NMSS/DWM/irsr.htm>).

The objective of the ENFE KTI is to assess all aspects of the evolution of the near-field geochemical environment that may affect the performance of the potential repository. The near-field geochemistry will be perturbed from ambient conditions by variations in temperature and pressure associated with the heat production of the waste, introduction of foreign materials into the mountain, variations in fluid flow, and consequent chemical reactions. Coupled THC and thermal-chemical processes can cause changes in parameter values and conceptual models used in various modules of performance assessment computer codes. The consequent effects on performance from the ENFE are expressed as the results of coupled processes. Acceptance criteria are established to assist in judging DOE's evaluations of the effects of the ENFE on repository performance. The scope of ENFE KTI work includes review of the various DOE documents as well as applicable documents in the open literature. We participate in

meetings with DOE to discuss issues related to the KTI and observe Quality Assurance audits of DOE. Independent technical investigations and sensitivity studies related to the effects of coupled THC processes on total system performance are conducted.

Four system attributes have been identified by the DOE in Revision 2 of their Repository Safety Strategy (U.S. Department of Energy, 1998a) as being the most important for predicting the performance of the engineered and natural barriers of the proposed repository. These system attributes include: limited water contacting the waste packages, long waste package lifetime, slow rate of radionuclide release, and reduction in radionuclide concentration during transport. These system attributes serve as one way to classify the effects of coupled processes on performance that result from the ENFE. For instance, the safety strategy (U.S. Department of Energy, 1998a) notes hydrothermal reactions may irreversibly change the hydraulic properties of the rock that could change the flow system in the near field and affect the quantity of water contacting waste packages. In addition, nuclear criticality has been identified as a potential disruptive process to the repository system [U.S. Department of Energy, 1998a; Civilian Radioactive Waste Management System Management and Operating Contractor (CRWMS M&O), 2000a].

The five subissues of the ENFE KTI have been constructed to address these system attributes. Because consequences on performance from the ENFE are expressed as the results of coupled processes, the subissues of the ENFE KTI are:

- Effects of coupled THC processes on seepage and flow
- Effects of coupled THC processes on waste package chemical environment
- Effects of coupled THC processes on chemical environment for radionuclide release
- Effects of coupled THC processes on radionuclide transport through engineered and natural barriers
- Effects of coupled THC processes on potential nuclear criticality in the near field

The scope of Revision 3 encompasses all five subissues. To adequately evaluate the impact of the ENFE on the performance of the repository requires addressing four aspects of coupled processes for each subissue. These aspects are: (i) identification of the coupled processes that could affect performance, (ii) characterization of the natural system, (iii) characterization of engineered materials and repository design, and (iv) abstraction of the effects of coupled processes into a performance assessment. The systematic approach adopted in this IRSR will provide a framework to determine the potential importance to performance of coupled THC processes. Resolution of the subissues will require that each of the four aspects be adequately addressed.

The potential effects of coupled processes on performance for each subissue must be identified. This first aspect has been completed for each subissue in Revision 1 of the IRSR. The second aspect that needs to be addressed is characterization of the natural system (minerals, groundwater and gaseous species, microbiological organisms; their masses and fluxes) and how it will influence and be influenced by coupled processes. The site geochemistry offers a large buffering capacity that will moderate chemical disturbances. Controls on the ambient geochemistry would be expected ultimately to govern many properties of the near-field environment. Understanding these controls provides a basis for predictions of near-field effects. Furthermore, the site geochemistry poses initial and boundary conditions for

modeling the induced evolution of the near field. Extensive data on ambient site mineralogy and rock chemistry are mainly based on studies conducted prior to construction of the Exploratory Studies Facility (Bish, et al., 1996). In addition, an increasing amount of data is becoming available from the underground (e.g., Paces, et al., 1996), from thermal testing, and additional data are anticipated from exploration of the east-west drift at Yucca Mountain (CRWMS M&O, 1997a). The characterization of the natural system and how it will influence and be influenced by coupled processes has been addressed for each subissue.

The third aspect required for resolution of the subissues is to evaluate how engineered materials and repository design will influence coupled processes. The effects of engineered materials on the near-field environment have been partially evaluated in performance assessment studies (TRW Environmental Safety Systems, Inc., 1996). These effects also are studied as part of the ongoing DOE thermal testing program. However, the evaluation of how engineered materials and repository design will influence coupled processes will remain unresolved until specification of a final design and an analysis of its consequences on performance is completed.

Each of these first three aspects bears on the fourth, the adequacy of any representation of the effects of coupled processes in a performance assessment. The adequacy of the DOE treatment of coupled processes in the performance assessment will be evaluated, guided by the constraint that the performance assessment should not provide over-optimistic estimates of repository performance. The performance assessments, or other analyses, however, provide information on the importance of particular coupled processes and the level of detail required to address particular subissues.

### 3.0 IMPORTANCE TO REPOSITORY PERFORMANCE

The consequences of coupled geochemical processes can affect several aspects of the proposed Yucca Mountain high-level waste repository performance. Ambient near-field geochemical conditions will be perturbed by variations in temperature and pressure associated with the heat production of the waste and introduction of foreign materials into the mountain. The changes in gas, water, and solid phase compositions and masses in the near field can affect hydrologic and mass transport characteristics, alteration of the waste package and waste form materials, and waste element speciation and solubility. The capability of the repository system to isolate waste will depend strongly on the near-field geochemistry. Thus, the performance of the repository will also depend on the effects of coupled geochemical processes. Coupled geochemical processes that could significantly impair the ability of the repository to isolate waste should be included in evaluations of repository performance.

Repository performance is evaluated through numerical models. Coupled processes might be included in these models directly, or they might be represented through modifiers to attributes, boundary conditions, or aspects of the numerical model. Uncertainty in how the coupled processes might change repository behavior may be treated by evaluating the range of postulated effects, possibly through alternate conceptual models.

For instance, precipitation or dissolution of minerals as a result of coupled THC processes will affect porosity and permeability. Flow attributes, such as porosity and permeability, are generally treated as variables (parameters) within performance assessment code modules. Thus, the values of these parameters in performance assessment modules would change as a result of the coupled processes. Likewise, the effects of coupled processes will modify values of parameters used in waste package, waste form (radionuclide release), and flow and transport modules of performance assessment codes. In addition, if the modifications to the system resulting from coupled processes are large enough, then alternative conceptual models of physical processes embodied in existing performance assessment modules or scenarios not considered (e.g., nuclear criticality) would need to be assessed for their impact on total system performance. The importance of ENFE investigations to overall repository performance is currently uncertain because the effects of coupled process have not yet been sufficiently evaluated in DOE performance assessments. To the extent that coupled processes have the potential to significantly impair repository performance, ENFE investigations remain important pending the results of further evaluations.

The subissues of the ENFE KTI concern the effects of coupled processes on the rate of seepage and flow, chemical environments for the waste package and radionuclide release, radionuclide transport through engineered and natural barriers, and potential nuclear criticality in the near field. Each of the subissues is directly related to a major system attribute or to a principal factor of the DOE Repository Safety Strategy (U.S. Department of Energy, 1998a; CRWMS M&O, 2000a). This relationship between the subissues and the safety strategy is discussed in more detail in Section 3.1. A discussion of the importance to performance of each subissue and how the subissues and the effects of coupled processes are addressed within the staff performance assessment framework is presented in Section 3.2. Evaluation of the effects of coupled processes in the near-field environment in performance assessment studies completed prior to DOE's Viability Assessment is presented in Section 3.3. Finally, NRC sensitivity analyses of the effects of coupled processes on repository performance are outlined in Section 3.4.

### 3.1

## U.S. DEPARTMENT OF ENERGY REPOSITORY SAFETY STRATEGY

The original DOE strategy for waste containment and isolation at the Yucca Mountain site was presented in its 1988 Site Characterization Plan (U.S. Department of Energy, 1988). DOE updated their strategy in the Repository Safety Strategy as a result of additional site characterization data, advances in the engineered system design, and a changing regulatory framework (U.S. Department of Energy, 1998a; CRWMS M&O, 2000a). The updated safety strategy reflects recent site characterization information, new waste package and repository designs, more realistic performance calculations, and the assumption of a dose- or risk-based standard. The primary goals of the strategy are near-complete containment of radionuclides within the waste packages for several thousand years and acceptably low annual doses to a member of the public living near the site (U.S. Department of Energy, 1998a; CRWMS M&O, 2000a). The updated strategy continues to rely on engineered and natural barriers to contain and isolate the waste from the public. In Revision 2 of the DOE Repository Safety Strategy (U.S. Department of Energy, 1998a), four system attributes are identified as the most important for predicting the performance of engineered and natural barriers. These system attributes are: (i) limited water contacting waste packages, (ii) long waste package lifetime, (iii) slow rate of release of radionuclides from the waste form, and (iv) concentration reduction of radionuclides during transport through engineered and natural barriers. The four system attributes are examined in detail as part of this IRSR, as they are affected by coupled THC processes in the near field. These attributes also correspond to four of the five subissues addressed in the ENFE KTI. The fifth subissue on near-field criticality relates to a potential disruptive scenario noted in the Repository Safety Strategy (U.S. Department of Energy, 1998a; CRWMS M&O, 2000a).

A number of working hypotheses have been developed by DOE to guide testing of the most important postclosure safety issues that relate to each of the attributes. The hypotheses provide a basis that DOE can use to explain analyses related to total system performance. These hypotheses can be used to organize, manage, and explain the rationale for DOE testing. For the first attribute, limited water contacting the waste packages, DOE developed four hypotheses. The four seepage-related hypotheses that are affected by coupled THC processes and addressed in part in this IRSR, are: (i) percolation flux at the repository depth can be bounded, (ii) seepage into drifts will be a fraction of percolation flux, (iii) thermally induced seepage can be bounded, and (iv) seepage that contacts a waste package can be limited. For the second system attribute, long waste-package lifetime, two testable hypotheses are addressed in part in this IRSR. The hypotheses affected by the ENFE are that corrosion rates are very low at low relative humidity, and that corrosion of the corrosion-resistant barrier is slow. The safety strategy notes that environmental characteristics of the waste package affect its corrosion rate. However, no testable hypotheses have been formulated to address this issue. For the third system attribute, slow rate of release of radionuclides from the waste form, four testable hypotheses are addressed in this IRSR. The hypotheses affected by the ENFE are that: (i) containment time is sufficient to prevent oxidation of spent fuel, (ii) water contacting the waste can be limited, (iii) release rate of soluble radionuclides is controlled by slow waste form dissolution, and (iv) the release rate of poorly-soluble radionuclides is controlled by solubility rather than colloid stability. Finally, for the fourth system attribute, concentration reduction during radionuclide transport through engineered and natural barriers, two testable hypotheses are also addressed in this IRSR. The DOE hypothesizes that physical properties of engineered and natural barriers reduce concentrations during transport. They also hypothesize

that chemical properties of engineered and natural barriers reduce concentrations during transport. Both the physical and chemical properties of the engineered and natural barriers are affected by the ENFE. Although unspecified among DOE hypotheses, criticality is noted in the safety strategy as a potential disruptive scenario.

### **3.2 IMPORTANCE OF SUBISSUES TO TOTAL REPOSITORY SYSTEM PERFORMANCE**

The near-field environment is currently considered to be an important factor in repository performance. The consequences of coupled THC processes may affect many aspects of repository performance. For instance, dissolution and precipitation of quartz or other minerals may occur both above and below the repository horizon as a result of the changing thermal regime (Sagar, 1997), and could, thus, impact both the seepage into the drifts and transport of radionuclides away from the drifts. DOE will need to adequately demonstrate and quantify the consequences of coupled processes on repository performance. This demonstration will require that DOE consider the interactions of coupled processes both within and among key elements of the natural and engineered subsystems of the repository.

Our strategy for reviewing the performance of the potential high-level waste repository at Yucca Mountain is described in the Total System Performance Assessment and Integration (TSPAI) IRSR (U.S. Nuclear Regulatory Commission, 1998c, 2000a). The performance assessment IRSR provides the framework and context for other KTI IRSRs, and integrates the results of those IRSRs. Its overall goal is to delineate a systematic approach for determining compliance with an overall system performance objective. The ENFE KTI supports the resolution of the TSPAI KTI by describing the information needed in key performance areas and by pursuing issue resolution in those areas. Those elements that are important to a post-closure performance assessment of a facility at the Yucca Mountain site are defined as integrated subissues.<sup>1</sup> Therefore, the approach that we will use to independently evaluate DOE's post-closure performance assessment will focus on integrated subissues. The integrated subissues are illustrated in Figure A-1 in Appendix A.

As highlighted in Figure A-1, the ENFE is an important factor that needs to be considered in the abstraction of five key elements of the engineered and natural subsystems. The five integrated subissues that the ENFE influences are: (i) engineered barrier degradation, (ii) quantity and chemistry of water contacting waste packages and waste forms, (iii) radionuclide release rates and solubility limits, (iv) flow paths in the unsaturated zone, and (v) radionuclide transport in the unsaturated zone. The acceptance criteria are designed to ensure that information necessary to describe the effects of coupled near-field THC processes on the integrated subissues is acceptable. The importance of coupled THC effects on performance for each ENFE KTI subissue and the relationship between the integrated subissue and each subissue are described more fully in the following sections.

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<sup>1</sup>In Revision 2 of the ENFE IRSR, integrated subissues were referred to as Key Elements of System Abstraction (KESA).

### 3.2.1 Importance to Performance of Coupled Thermal-Hydrologic-Chemical Effects on Seepage and Flow

The effects of coupled THC processes on seepage into the drift and flow in the unsaturated zone is the first subissue of the ENFE KTI. The integrated subissue (see Appendix A) that is influenced by the ENFE within the scope of the seepage and flow subissue is the flow paths in the unsaturated zone. There are three main near-field environmental factors that have the potential to affect performance. Each of these factors needs to be considered in the evaluation of the integrated subissue. The processes are dehydration of zeolitic horizons, coupled THC processes that affect the porosity and permeability of the natural system, and coupled THC processes at the interface of the natural system and the engineered components.

The first factor that may affect performance of the repository is the potential dehydration of zeolitic minerals. Major geochemical changes in the near field are likely to depend primarily on the availability of water. Although unsaturated, the rocks at Yucca Mountain contain significant water, commonly 10 percent of the rock volume (e.g., data from Flint, 1997). A large amount of zeolitic water is also potentially available in certain horizons, primarily beneath the repository, that could be released at elevated temperatures. The potential importance of zeolite dehydration to performance has been recognized by DOE (Bish, et al., 1996). The spatially varying distribution of the zeolitic horizons in Yucca Mountain, and the thermal-loading strategy will cause spatially and temporally variable dehydration of zeolites. Water released from the dehydration of zeolites could affect both the spatial and temporal distribution of flow, which is also addressed in the Thermal Effects on Flow IRSR (U.S. Nuclear Regulatory Commission, 1999c). Flow through these thermally-affected zeolitic horizons will also be influenced by the loss of host rock volume associated with the dehydration process, creating new fractures and widening existing fractures, thereby, leading to possible increases in fracture flow.

The second factor that may affect performance of the repository is the effect of coupled THC processes on the porosity and permeability structure of the natural system. Given the temperature-dependent solubility of different minerals, it is possible that fluids (both liquid and gas phase) moving by thermally driven convection will redistribute chemical components such as  $H^+$ ,  $Cl^-$ ,  $O_2$ ,  $CO_2$ ,  $SiO_2$ ,  $Ca^{2+}$ . Most extensive and rapid chemical reactions will occur where water evaporates, depositing solutes, and where water vapor transported by distillation condenses. Because water is drawn by capillarity into the finest pores of the rock, evaporation and precipitation may have the greatest effects in the rock matrix. However, gaseous transport of water vapor to cooler zones of condensation is likely to occur dominantly in fractures. Therefore, condensation of initially-dilute mildly-acidic water, from the dissolution of  $CO_2$  into the condensate, and mineral dissolution are likely to occur on fracture surfaces. The thermal effects on the natural system are both temporally and spatially variable as a result of repository design (edge effects) and the radioactive decay of the waste. Extensive development of heat pipe effects and refluxing at elevated temperatures, which is described in the Thermal Effects on Flow IRSR (U.S. Nuclear Regulatory Commission, 1999c), could cause changes in porosity over regulatory time frames of thousands of years. Small changes in porosity can produce orders of magnitude changes in permeability (Lichtner and Walton, 1994). Thus, the dissolution and transport of mineral constituents such as silica and calcium, followed by precipitation during evaporation, could modify the permeability distribution in the natural system surrounding the repository horizon.

The final set of coupled process within the subissue of seepage and flow that may affect performance of the repository is likely to be spatially limited to near the drifts of the repository. In the Total System Performance Assessment conducted for the Viability Assessment (TSPA-VA), the reference design description of the ground control system indicated the use of both pre-cast and cast-in-place concrete liners for emplacement drifts (TRW Environmental Safety Systems, Inc., 1997a). Interaction of cement with the tuffaceous host rock and ambient pore fluids and gas could have an important effect on seepage and flow. The chemistry of pore fluids in contact with hydrated cement phases is characterized by a persistent alkaline pH (>10). Hyperalkaline cement pore water increases silica solubility. Silica is a major component of the proposed Yucca Mountain repository host rock unit, the Topopah Spring Tuff. This unit is composed predominantly of alkali-feldspar, quartz, cristobalite, and tridymite (Bish, et al., 1996). Thus, migration of the high-pH cement pore water into the host rock is likely to result in strong alteration of the tuff by changing its porosity and permeability (Lichtner, et al., 1997). The strong interaction of the concrete with the tuff is also likely to influence the quantity of water that can be transmitted into the drifts by both the matrix and fractures. For example, model calculations by Lichtner, et al. (1998) indicated that cement-tuff interaction could reduce porosity within the tuff matrix and also cause calcification of cement, resulting in zero porosity almost right at the contact between the cement and the tuff. The concrete may also react with and remove both dissolved carbonate and bicarbonate and gaseous CO<sub>2</sub>. The carbonation of the concrete will affect mechanical properties (Parrott, 1994) of the ground control system. In addition, the concrete drift liners will have mechanical stability that likely will depend on the spatially variable mechanical stability of the geologic medium [this topic is addressed in the Repository Design and Thermal Mechanical Effects IRSR (U.S. Nuclear Regulatory Commission, 1998b)]. Thus, the stability of the drift liners and their interactions with the surrounding geologic medium will influence both the spatial and temporal distribution of flow. It should be noted that the Enhanced Design Alternative-II, the design selected for the Site Recommendation Consideration Report, eliminated the use of concrete liners. Thus, the effect of coupled processes involving cementitious materials is expected to be significantly less compared to the TSPA-VA reference design.

The flow paths in the unsaturated zone will be affected as a result of the evolution of the near-field geochemical environment. To adequately describe this integrated subissue in a post-closure performance assessment (see TSPA IRSR; U.S. Nuclear Regulatory Commission, 2000a), the effects of coupled THC processes on seepage and flow will need to be considered.

### **3.2.2 Importance to Performance of the Effects of Coupled Thermal-Hydrologic-Chemical Processes on Waste Package Chemical Environment**

The effect of coupled THC processes on waste package chemical environment is the second subissue of the ENFE KTI. With the addition of the drip shield as a design option in the Enhanced Design Alternative II design, this subissue now also addresses the drip shield chemical environment. The two integrated subissues (see Appendix A) that are influenced by the ENFE within the scope of the waste package chemical environment subissue are: (i) quantity and chemistry of water that contacts the waste package and waste form, and (ii) engineered barrier degradation. The three factors that were discussed in the seepage and flow subissue (dehydration of zeolites, dissolution and precipitation of minerals in the natural system, and reactions within and between cementitious materials and the host rock) also will affect the waste package chemical environment subissue and the two integrated subissues, but

will not be discussed in detail in this section. Only those aspects of these coupled processes that differ between the two subissues and their effect on the integrated subissues will be presented here. One additional factor, the generation of natural and spontaneous self potentials, is no longer thought to affect the waste package chemical environment significantly (see Section 4.2.1).

Successfully abstracting the drip shield and waste package corrosion processes within a performance assessment framework is primarily addressed in the Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1998d, 1999d). This abstraction requires incorporating the various potential modes of corrosion and the functional dependence of the corrosion process on environmental factors. Both the modes and rates of corrosion are directly dependent on the pH,  $\text{Cl}^-$ , oxygen content, and total carbonate in the near field of the repository and possibly other components. As a result, coupled THC processes will assert a strong influence on waste package corrosion. For instance, the low air mass fraction that may be generated temporarily within the near field as a result of boiling (Wilder, 1996; Lichtner, 1997; Hardin, 1998) will lower the oxygen concentration and directly influence the possibility of corrosion in the thin film of water that may be present on the waste package surface.

The number, location, and time when waste packages are contacted and affected by dripping water will be influenced by the coupled processes addressed in the seepage and flow subissue. In addition, the chemistry of the water that contacts the waste package will be the result of coupled THC processes with natural and engineered materials. Depending on the temperature of the waste package and the drifts, and the flux of water intercepting the waste package, further concentration of the fluid on the surface of the waste package due to evaporative effects is possible. Thus, to adequately describe the integrated subissues on quantity and chemistry of water and engineered barrier system degradation in a performance assessment, the effects of coupled THC processes on waste package chemical environment will need to be considered.

### **3.2.3 Importance to Performance of the Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Chemical Environment for Radionuclides Release from the Engineered Barrier System**

The effect of coupled THC processes on the chemical environment for radionuclide release is the third subissue of the ENFE KTI. The quantity and chemistry of fluids contacting waste forms and radionuclide release rates and solubility limits will be affected as a result of the evolution of the near-field geochemical environment. These two integrated subissues (see Appendix A) are addressed within the chemical environment for radionuclide release subissue and are addressed in more detail in the Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1998d, 1999d). The effects of coupled processes on the integrated subissues necessary to describe radionuclide release are presented below.

Prior to water contacting the spent fuel, degradation of the cladding must occur. If credit for cladding were to be taken in a performance assessment, the effects of coupled THC processes on the chemical environment of cladding would need to be considered. Both the chloride concentration and the redox potential at the cladding surface control whether localized corrosion of fuel cladding by pitting will occur (Cragolino and Galvele, 1978; Maguire, 1984). Under the environmental and potential conditions leading to pitting, stress corrosion cracking of zirconium and Zircaloy occurs in the presence of an applied stress (Cox, 1990). The presence

of the fluoride anion in the environment, although its concentration is relatively low, may increase the uniform dissolution of zirconium alloy. This is the result of the greater stability of the  $ZrF_6^{2-}$  complexes compared to that of the passive  $ZrO_2$  film.

Corrosion of the spent fuel (predominantly  $UO_2$ ) pellets by contact with the groundwater is the most important process affecting the long-term performance of this waste form. The corrosion of the pellets is modified by chemical and physical interactions in the near field. Several environmental factors are known to affect the dissolution rate of  $UO_2$  in aqueous environments. The nature and concentrations of the anionic species present in the groundwater are extremely important in determining the rate of corrosion of spent fuel. Dissolved  $CO_3^{2-}$ ,  $F^-$ ,  $SO_4^{2-}$ ,  $PO_4^{3-}$ , and  $Cl^-$  can accelerate the corrosion rate (Blesa, Morando, and Grambow, 1994; Grambow, 1989). A major near-field environmental factor affecting  $UO_2$  waste form performance is the redox potential or Eh. The redox potential of the near-field environment will be influenced by radiolysis, the air mass fraction in the waste packages, and reactions with metallic components of the engineered barrier system.

The effect of pH on the rate of dissolution of spent fuel depends on the pH range. Under oxidizing conditions, only a slight dependence of corrosion rate on pH has been observed at pH values lower than four, whereas at pH values between four and eight, the rate decreases linearly with pH (Grambow, 1989). At higher pH values, the rate of dissolution seems to be unaffected by pH changes. The pH of fluid contacting spent fuel will be influenced by hydrolysis of highly charged cations such as  $Cr^{3+}$  from the degradation of the waste package, air mass fraction in the waste package, reaction with metallic components of the engineered barrier system, and radiolysis.

In addition, the modification of the pH of the leachate attributed to the formation of  $HNO_3$  by alpha-radiolysis of humid air, as well as the generation of formate and oxalate from inorganic carbon, may raise the solubility of actinides (Finn, et al., 1994a). Through interactions with oxidizing components, including radiolytic products, spent fuel will eventually oxidize, forming a large quantity of uranyl ( $UO_2^{2+}$ )-bearing solids. Other species, such as  $SiO_2(aq)$ ,  $H_3SiO_4^-$ ,  $H_2SiO_4^{2-}$ , and products from waste package degradation, can react with  $U^{6+}$  to precipitate complex uranyl silicates or other secondary minerals, which may tend to reduce the corrosion rates and exposure of fresh surface by forming a protective layer over the fuel pellets. Therefore, secondary oxidation products will accumulate and uranyl minerals will have a large effect on near-field physical and chemical conditions. The importance of the chemistry of the fluid interacting with spent fuel is captured in the release models within performance assessment codes (Mohanty, et al., 1997; TRW Environmental Safety Systems, Inc., 1997b).

Secondary uranium phases are likely to have several important effects on the near-field environment. First, physical disruption of structural components (e.g., cladding or degraded containers), due to the large volume increase accompanying oxidation and hydration of  $UO_2$ , may occur. Second, both porosity and permeability could be reduced due to the volume expansion. Third, neptunium, plutonium, and other radioactive waste species may be incorporated into secondary uranium phases by coprecipitation. Fourth, the secondary uranium phases may limit ingress of water and oxidants to unaltered wastes. Finally, the secondary uranium phases may control the solubility or dissolution rate of spent fuel. Thus, secondary uranium phases may control the source term for radionuclide (not just uranium) releases from the breached waste packages. With regard to long-term performance of the potential repository, secondary alteration products resulting from interactions of spent fuel with the near-

field environment, rather than unaltered spent fuel, will likely control releases of many radionuclides from the waste packages.

The second main waste form planned for the proposed repository at Yucca Mountain is high-level waste borosilicate glass. Environmental factors affecting the general or localized dissolution rate of borosilicate glasses are pH,  $F^-$ , and  $Fe^{2+}$ . Ultimate glass waste form alteration products are likely to be clay or zeolite minerals. These alteration products are analogous to alteration products of the natural volcanic glasses existing at Yucca Mountain (Murphy and Pabalan, 1994). However, they are likely to incorporate augmented quantities of components of the engineered barrier system, such as iron and calcium. Clay minerals generally have low solubilities. Some quantity of radioactive waste species are likely to be incorporated through solid solution in mineral alteration products of glass waste forms. Contribution of the high-level waste glass to the source term could be significant if the rate at which radionuclides can be released and transported in a colloidal form is higher than the rate of release of radionuclides from spent fuel.

Interactions between cementitious materials and the near-field system can be potentially beneficial for mitigating release of radionuclides. The persistent alkaline pH (>10) characteristic of pore fluids in contact with hydrated cement phases favors precipitation of a wide variety of radionuclides, including transuranics (Glasser, et al., 1985; Atkins, et al., 1990). On the other hand, alkaline conditions can be detrimental to the stability of nuclear waste glass. For instance, experiments by Heimann (1988) indicated that cement and glass interaction leads to accelerated dissolution and alteration of the nuclear waste glass compared to a system without cement present.

Thus, the effects of coupled THC processes on the chemical environment for radionuclide releases from the engineered barrier system appear likely to be important to the performance of the potential Yucca Mountain repository and will need to be considered in the abstraction of release of radionuclides.

### **3.2.4 Importance to Performance of the Effects of Coupled Thermal-Hydrologic-Chemical Processes on Radionuclide Transport Through Engineered and Natural Barriers**

The effect of coupled THC processes on the transport of radionuclides through engineered and natural barriers is the fourth subissue of the ENFE KTI. The two integrated subissues (see Appendix A) that are influenced by the ENFE within the scope of the radionuclide transport subissue are: (i) flow paths in the unsaturated zone, and (ii) radionuclide transport in the unsaturated zone. The three coupled processes that were discussed in the seepage and flow issue are equally important for this subissue. Each of these processes needs to be considered in the evaluation of each of the integrated subissues.

Radionuclides transported through the engineered barrier system and the geologic setting can be gaseous species, associated with colloids, or as dissolved species in aqueous solution. Each mode of transport is influenced by several geochemical parameters; thus, an assessment of the relative importance of each type of transport will depend on the specific geochemical and hydrologic characteristics of the near-field environment. One mechanism for removing

radionuclides from solution is the precipitation of stoichiometric radioelement compounds or coprecipitation as impurities in other minerals as fluids travel through and react with both the engineered barrier system materials and natural system minerals. Coupled THC processes will cause changes in system chemistry parameters such as Eh, pH, and component concentrations that will influence the solubilities of radionuclide-bearing minerals.

Another retardation mechanism, which is strongly controlled by geochemical parameters, such as solution pH, is sorption. For example, sorption of actinide species, such as  $\text{UO}_2^{2+}$ ,  $\text{NpO}_2^+$ , and  $\text{Am}^{3+}$ , on oxides and oxyhydroxides through surface complexation mechanisms is characterized by a sharp sorption "edge," where sorption increases sharply with increasing pH from essentially zero over a relatively narrow, low pH range (Sagar, 1997). The pH of the sorption edge differs for different actinides. The amount of radionuclide sorbed also depends on radionuclide concentration and the number of available sorption sites. For clay and zeolite minerals, sorption of radionuclides, such as  $\text{Cs}^+$ ,  $\text{Sr}^{2+}$ , and  $\text{UO}_2^{2+}$ , can also occur through an ion exchange mechanism, which depends on the nature and concentration of competing cations present in solution (Sagar, 1997). The stability of the sorbent minerals, the amount of sorbent minerals, and the pH will be influenced by coupled THC processes resulting from the changing thermal conditions.

The oxidation state in the near field may affect sorption behavior. For example, under oxidizing conditions, technetium is principally present as pertechnetate ( $\text{TcO}_4^-$ ) and does not sorb strongly, whereas, under reducing conditions,  $\text{Tc}^{4+}$  is predominant and sorbs more strongly (Lieser and Bauscher, 1988). Elevated temperatures expected for the near-field environment may also affect sorption, but there are few data for evaluating the magnitude of the effect. In the near field, boiling in response to thermal loading would tend to partition  $^{14}\text{CO}_2$  into the gas phase, enhancing gas transport. Local fluctuations of reducing and oxidizing conditions in the near field due to an unstable hydrologic regime could also induce secondary chemical effects, such as the formation of colloids (Buddemeier and Hunt, 1988; McCarthy and Zachara, 1989). Thus, the redox conditions, which also affect sorption behavior, will be influenced by coupled THC processes.

The potential importance of colloids in the transport of radionuclides is also of concern in performance assessments. The stability of the colloidal suspension of charged particles varies as a function of ionic strength, solution chemistry, and pH. Each of these parameters will change as the near field evolves in response to coupled THC processes. Colloid transport through the near field may be retarded in several ways, including through interaction with cementitious materials (Savage, 1997).

Thus, the effects of coupled THC processes on transport of radionuclides in the near field will be important to the performance of the potential Yucca Mountain repository and will need to be considered in the abstraction of radionuclide transport.

### **3.2.5 Coupled Thermal-Hydrologic-Chemical Processes Affecting Potential Nuclear Criticality in the Near Field**

Coupled THC processes that could affect potential nuclear criticality in the near-field environment is the fifth subissue of the ENFE KTI. The presence of fissile radionuclides, such as U-235 and Pu-239, in spent nuclear fuel creates a potential for a sustained neutron chain

reaction (criticality event). Such an event could arise if there is failure of the waste canister, dissolution of the fissile material, and redeposition outside the canister in the near-field environment. For example, Bowman and Venneri (1995) claimed that autocriticality and explosive conditions could be reached based on a conceptual model where neutron absorbers (e.g., boron and lithium) and subcritical concentrations of U-235 and Pu-239 and other fissile materials are mobilized from waste forms in the potential Yucca Mountain repository and deposited in a concentration and geometry sufficient to reach criticality. However, several reviews rejected Bowman and Venneri's conclusion as implausible because of the low probabilities that could lead to critical configurations. Sanchez, et al. (1995) reported possible supercritical conditions in systems of plutonium-SiO<sub>2</sub>-H<sub>2</sub>O and plutonium-tuff-H<sub>2</sub>O, but concluded that the probability of forming such conditions is extremely low. Choi and Pigford (1997) concluded from their technical analysis of the potential for autocatalytic criticality that, based on simplified geometries, there is a theoretical potential for appreciable energy release from autocatalytic (or self-enhancing) criticality, but that additional analysis is required to determine the extent and consequences of such an event. Potential effects on repository performance of criticality in the near field include an increase in the fission product inventory, a decrease in the fissile radionuclide inventory, and an increase in thermal output.

### **3.3 CONSIDERATION OF COUPLED NEAR-FIELD PROCESSES IN PREVIOUS PERFORMANCE ASSESSMENTS**

Some limited consideration of the effects of coupled processes on the performance of the proposed Yucca Mountain repository have been included in performance assessment studies conducted prior to the Viability Assessment. Those recent studies that have addressed the effects of coupled geochemical processes include: (i) total system performance assessments performed by DOE, the 1993 Total System Performance Assessment (TSPA-93) (Wilson, et al., 1994), and the 1995 Total System Performance Assessment (TSPA-95) (TRW Environmental Safety Systems, Inc., 1995); (ii) one performance assessment prepared by the Electric Power Research Institute—Yucca Mountain Total System Performance Assessment, Phase 3 (Kessler and McGuire, 1996); (iii) one sensitivity study by DOE—Status/Summary Report for Fiscal Year 1996 Activities Within the Performance Assessment Overview Study on the Consequences of Cementitious Materials (TRW Environmental Safety Systems, Inc., 1996); (iv) one assessment prepared by NRC—Iterative Performance Assessment, Phase 2 (Wescott, et al., 1995); and (v) sensitivity studies conducted by the NRC and the Center for Nuclear Waste Regulatory Analyses (CNWRA) staffs using the NRC TPA code (Manteufel, et al., 1997). The manner in which these studies incorporate effects of coupled processes is summarized in the following sections. DOE proposed a much expanded consideration of near-field chemical effects in their TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b).

#### **3.3.1 U.S. Department of Energy Total System Performance Assessment 1993**

The source-term model included a near-field geochemistry module containing geochemistry parameters for use in the container failure, cladding failure, and waste form dissolution modules. The input parameters in the near-field geochemistry module are pH, Eh, Cl<sup>-</sup>, F<sup>-</sup>, and total carbonate concentrations. However, in TSPA-93, Eh and Cl<sup>-</sup> concentration were not used. The temperature, fractional time the waste packages are wet, the pH, F<sup>-</sup>, and total carbonate concentrations were used in estimating the container corrosion rate, cladding failure rates, and alteration rates of the waste form. Although these chemical parameters could have been

allowed to vary as a function of time, in TSPA-93, they were held constant for each simulation. The pH,  $F^-$ , and total carbonate concentrations used were from wells J-13 and Ue25p#1. It is recognized that these values are appropriate for far field, saturated zone conditions, but may misrepresent the conditions in the near-field environment. Consequently, although the coding architecture was available for simulating some limited near-field conditions, only the temperature-time history can be considered a reasonable approximation for the near field.

TSPA-93 reported that “increased temperature from the repository may cause more aggressive groundwater chemistries and increased solubilities for radionuclides in the near field; however, when the solute is transported out of the near field, the potentially lower solubilities in the far field would cause precipitation and thus would be the limiting factor. The experts have made this assumption primarily because the dearth of information about the near-field water chemistry makes accurate predictions of solubility impossible for this region. A potential concern must be mentioned with regard to this assumption. The high thermal loads being considered for the potential repository (e.g., 114 kilowatt per acre) may cause near-field conditions to extend throughout the unsaturated zone” (Wilson, et al., 1994; p. 9-3).

### **3.3.2 U.S. Department of Energy Total System Performance Assessment 1995**

Possible geochemical variations in near-field environmental conditions were considered in TSPA-95. However, minimal effects of changes in the geochemical environment were employed in the performance calculations. For example, with regard to solubilities, it was stated, “Because the actual changes to the near-field environment are not yet well-defined, incorporation of such effects either into [solubility] distributions, such as those discussed above, or into models for predicting the solubility-controlling phases for each radionuclide is not currently possible” (TRW Environmental Safety Systems, Inc., 1995; p. 6-11). Although some pH-dependent solubility relations were derived, it was concluded that, “Although the derived functions incorporate pH-dependence explicitly, the near-field pH evolution is uncertain to the extent that adequate constraints do not exist for making a pH choice other than a random selection from a distribution” (TRW Environmental Safety Systems, Inc., 1995; p. 9-25). For alternate solubility models, which use functional dependencies on pH and temperature, only pH 7 was considered.

In general, solubilities used in TSPA-95 were highly uncertain, which is partly represented by distributions spanning many orders of magnitudes (TRW Environmental Safety Systems, Inc., 1995). Comparisons to solubilities for selected elements, determined in independent computations using EQ3 (Wolery, 1992) for ranges of possible geochemical conditions, revealed that most TSPA-95 solubilities were comparable or higher (more conservative). Two exceptions are radium and tin. Calculated solubilities of  $RaSO_4$  and cassiterite ( $SnO_2$ ) for a range of possible water chemistries and temperatures were near  $10^{-6}$  molal. This value is near the upper limit of the TSPA-95 range for radium and 10 times the upper limit of the TSPA-95 range for tin. However, considering  $RaSO_4$  or  $SnO_2$  to limit solubility is perhaps unnecessarily conservative because these trace metals will likely be incorporated as minor components of other phases.

Although not addressed in terms of effects of coupled processes, fracture and matrix interactions were analyzed as part of the performance assessment (TRW Environmental Safety Systems, Inc., 1995). DOE incorporated the effect of matrix diffusion in its conceptual model

for flow and transport in the unsaturated zone. Support for that conceptual model from experimental or field data was not provided (U.S. Nuclear Regulatory Commission, 1996). Hydrological and geochemical data from the vicinity of the Yucca Mountain site and from analog sites suggest that matrix diffusion type processes may have limited effects on the rate of radionuclide migration (U.S. Nuclear Regulatory Commission, 1996; Baca and Jarzempa, 1997).

Gas phase transport of radionuclides was not evaluated in terms of performance (individual dose) because DOE judged it to be insignificant to performance. This may be a reasonable conclusion because of mixing of gaseous components in the atmosphere; however, no calculations were provided to support it.

### **3.3.3 Total System Performance Assessment—Viability Assessment Methods and Assumptions**

The Viability Assessment methods and assumptions report (TRW Environmental Safety Systems, Inc., 1997b) provides a good description of the near-field geochemical environment evolution as a consequence of coupled THC processes. TRW Environmental Safety Systems, Inc. (1997b) repeatedly recognizes the significance of near-field chemistry on repository performance. “A key aspect of the degradation of the engineered barriers (in particular, the waste package, cladding, and waste form) is the environment in which these engineered components exist” (p. 4-9). Table 4.3-1 of TRW Environmental Safety Systems, Inc. (1997b) notes that the drift-scale thermo-chemical model will provide information including pH,  $p\text{CO}_2$ ,  $\text{CO}_3^{2-}$ ,  $\text{Cl}^-$ ,  $\text{F}^-$ , silica,  $\text{PO}_4^{3-}$ , and  $\text{SO}_4^{2-}$ . Section 6.3 of TRW Environmental Safety Systems, Inc. (1997b) provides a fairly comprehensive description of coupled THC effects on the drift scale and outlines a fairly ambitious program to model drift-scale chemical characteristics as a function of time. “For the TSPA-VA, explicit consideration of the near-field geochemical environment evolution constitutes a major step forward for directly including the potential chemical variations affecting source-term performance; however, this initial effort is only a relatively simplified representation of the complex interaction of this heterogeneous system” (p. 6-38).

A key assumption in the TSPA-VA methods and assumptions report is that mechanical and chemical changes do not alter hydrologic properties (TRW Environmental Safety Systems, Inc., 1997b, p. 6-22). This assumption is rather severe, and is recognized in TRW Environmental Safety Systems, Inc. (1997b). “Chemical or mechanical changes to the fracture properties influence the resulting gas-phase and liquid-phase flow fields predicted by the models (drift- and mountain-scale), and, thereby, potentially affect heat and radionuclide transport as well. Although the response of the mountain to these effects will not be fully coupled in the TSPA-VA analyses, simplifications that patch thermal-mechanical and/or thermal-chemical influences into a unsaturated zone thermal-hydrologic simulation have been proposed as a series of sensitivity studies” (p. 6-20).

Additional near-field aspects are addressed within the engineered barrier system transport discussion in Section 6.6 of TRW Environmental Safety Systems, Inc. (1997b). Mobilization of radionuclides from the waste form will be combined with the flux and chemistry of water moving through the system and with other transport parameters, including retardation and permeability of the engineered barrier system materials (e.g., waste package, corrosion products, and invert

materials). The aqueous and colloidal transport of radionuclides through the engineered barrier system components and radionuclide sorption along the engineered barrier system transport pathways as a function of pH and temperature will be abstracted. The output from the engineered barrier system transport model will be a release of radionuclides from the engineered barrier system to the geosphere.

### **3.3.4 Electric Power Research Institute Yucca Mountain Total System Performance Assessment**

The Electric Power Research Institute has evaluated some of the processes that the ENFE KTI has responsibility to address. They used a code called Integrated Multiple Assumptions and Release Calculations to assess the performance of the individual components that contribute to the performance of the repository system (Kessler and McGuire, 1996). IMARC is a deterministic code in which an event tree approach is used. Advection and diffusion between the source term compartment and the following compartments could be modeled using the code. The waste form (source term), corrosion products found in the corroded section of the container, gravel backfill below and sometimes above the container, concrete invert (both concrete matrix and fracture), and rock matrix and fractures immediately surrounding the drift were each modeled as compartments. The study evaluated the effect of microbial processes on waste package containment and the effect of near-field transport processes (diffusion and advection) on performance.

### **3.3.5 U.S. Department of Energy Performance Assessment Overview Study on the Consequences of Cementitious Materials**

An overview study to address potential postclosure performance issues concerning the use of large quantities of cementitious materials within the potential waste emplacement drifts was conducted by DOE (TRW Environmental Safety Systems, Inc., 1996). Preliminary consequence sensitivity analyses were conducted using a performance assessment model (the RIP code) by modifying the TSPA-95 basecase (parameter set as 83 MTU per acre, backfill, high infiltration rate, climatic variation, drips on the waste package and fluid pH of 7) to reflect the potential interaction of alkaline fluids from cementitious materials. Two scenarios were analyzed, one in which enhanced aqueous complexing of dissolved radionuclides at high pH results in negatively charged species. This was accomplished by setting distribution coefficients,  $K_d$ s, for all radionuclides to zero for distances of 10 and 100 meters into the unsaturated zone and for the entire unsaturated zone. The second scenario simulates the first scenario with the added effect of pH-dependent waste form dissolution rates and pH-dependent solubility limited concentrations of neptunium, plutonium, and Am. The dissolution rate and solubility limits were evaluated at a fluid pH of 11. The results of these simulations were compared to the TSPA-95 basecase results. A single realization, expected-value calculated dose at the accessible environment, at  $10^4$ ,  $10^5$ , and  $10^6$  years was compared to the basecase. In addition, the complementary cumulative distribution function of 100 realizations of calculated dose at the accessible environment at times of  $10^4$  and  $10^5$  years were compared to the basecase.

For the  $10^4$  year time frame, the results indicate essentially no change from the basecase for both scenarios where there is no retardation up to 100 meters into the unsaturated zone. For unretarded transport through the entire unsaturated zone, only a minor contribution to peak

dose from Np-237 (~1 percent ) is calculated for the first scenario, but Np-237 becomes the primary contributor to peak dose at about 8,700 years for the second scenario.

For the  $10^5$  year time frame and 10 meters of unretarded unsaturated zone transport, there is essentially no difference for the first scenario compared to the basecase. In the second scenario, the peak dose is increased by about a factor of 10 and dominated by Np-237 at greater than 60,000 years. For unretarded transport through 100 meters and the entire unsaturated zone, the peak dose results from Np-237 for both scenarios, and dose contributions from Th-229 and U-233 are also elevated. For the wholly unretarded unsaturated zone pathway, Np-237 contributes the highest doses after ~20,000 and ~10,000 years in the first and second scenarios, respectively. In the latter case, the peak dose is about 300 times the basecase value, and Pu-239 is the second highest dose contributor at ~60,000 to 100,000 years, comparable to the dose contribution from Th-229.

For the  $10^6$  year time frame, the results for the first scenario indicate that the long-term Np-237 peak dose is: (i) relatively unchanged for 10 meters of unretarded transport, (ii) about twice the basecase for 100 meters unretarded transport, and (iii) about 4 times the basecase for unretarded transport throughout the unsaturated zone. In the latter case, dose contributions from Ac-227 and Pa-231 essentially equal the contribution from Np-237. For the second scenario, the long-term Np-237 peak dose is higher and shifted to earlier times for both 10 and 100 meters of unretarded unsaturated zone pathway. For unretarded transport throughout the entire unsaturated zone, the major dose contributors are Pu-239 and Pu-242 after about 150,000 years.

In all the cases simulated, except for the second scenario in which transport is unretarded for the entire unsaturated zone, there is little change to peak doses at 10,000 years because peak doses are dominated by I-129 and Tc-99, which are assumed to be unretarded. If transport is unretarded for only 10 m, Np-237 peak doses in the first scenario are relatively unaffected at all time frames, but in the second scenario the peak dose is increased by a factor of 10 and 5 in the  $10^5$  and  $10^6$  year time frames, respectively. For unretarded transport through 100 meters and the entire unsaturated zone, Np-237 peak doses are about 0.5 and 1.5 orders of magnitude, respectively, higher than the basecase for the first scenario, and about 1.0 and 2.5 orders of magnitude, respectively, higher for the second scenario.

For the  $10^6$  year time frame, the doses from Pu-239 and Pu-242 are about five orders of magnitude higher for the first scenario with unretarded transport through the entire unsaturated zone compared to the basecase, although they do not reach 10 millirem levels and do not become major contributors to peak dose within that time frame. However, for the second scenario, the plutonium isotope doses are about seven orders of magnitude higher than the basecase and are major contributors to peak dose after about 150,000 years. The peak dose from either plutonium isotope is only about half that of Np-237 for that case.

### **3.3.6 U.S. Nuclear Regulatory Commission Iterative Performance Assessment Phase 2**

An auxiliary analysis was included in Wescott, et al. (1995, Appendix K) that addressed coupled near-field processes and their effects on the carbon geochemical system. A simple one-dimensional, uniform, time varying gas flow field was coupled to equilibrium aqueous

speciation, CO<sub>2</sub> gas evolution and transport, and calcite precipitation in a transient thermal field. The model was used to explore possible mechanisms for <sup>14</sup>C retardation and gas phase release. An initial pulse of <sup>14</sup>CO<sub>2</sub> was modeled to exit the mountain after less than 2,000 years of transport. Subsequently, <sup>14</sup>C releases to the atmosphere were small and some <sup>14</sup>C remained trapped in precipitated calcite. However, these results were not incorporated in analyses of complementary cumulative distribution functions.

### **3.4 U.S. NUCLEAR REGULATORY COMMISSION/CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES SENSITIVITY ANALYSES**

In sensitivity analyses conducted by the NRC and the Center, the effects of some coupled processes on repository performance were assessed in terms of effects on dose. These effects and the importance of parameter values assigned to physical properties in the analysis were determined by systematically performing sensitivity analyses. Both process-level models and abstracted models in the performance assessment code were used to assess effects of coupled processes expected to be active in the near-field environment. Process-level models used by the ENFE KTI are detailed models formulated on basic principles that govern heat and mass transfer and chemical reaction and transport for the range of expected conditions at the repository. Abstracted models within the NRC TPA code are designed to represent the physical processes by extracting only higher order effects identified in process-level models (Manteufel, et al., 1997).

Process-level models contained within the MULTIFLO code (Lichtner and Seth, 1996a,b; Lichtner, 1997) were used by the ENFE KTI to guide the input values chosen for parameters in the NRC KTIs sensitivity analyses. In particular, some of the input values used for the Container Life and Source Term KTI sensitivity analyses were derived from MULTIFLO process-level modeling. Both process-level and abstracted models were used in the ENFE KTI to assess the effects of coupled processes in terms of sensitivity of dose to variations in model assumptions and parameter values.

The ENFE KTI sensitivity analyses primarily focused on evaluating the potential effects engineered materials (cementitious materials, and steel used in waste packages) may have on the performance of the repository. Conceptual models of waste package degradation and radionuclide release were also evaluated as part of the ENFE KTI sensitivity analyses. The results of the preliminary efforts using the TPA Version 3.1 code are summarized in the following sections and also discussed by Jarzemba, et al. (1999).

#### **3.4.1 Cement-Affected Near-Field Environment**

Changes in water chemistry may result from interactions between cementitious materials and groundwater. In particular, hyperalkaline fluids (pH >10) may result from these interactions. These fluids are capable of precipitating radionuclides, including transuranics, thus, altering the source term and transport behavior of radionuclides. Cement phases provide a multitude of sorption sites that could aid in retarding radionuclide migration. In addition, dissolution of the geologic barrier (e.g., tuff) by a hyperalkaline fluid could lead to a widening of fractures and enhanced groundwater flow. On the other hand, migration of a hyperalkaline fluid could result in precipitation of calcite and calcium-silica-hydrate phases along fractures and reduce fracture

porosity and hydraulic conductivity. In addition, mineral precipitation could seal fracture surfaces and enhance transport of radionuclides by preventing diffusion into the matrix.

The potential importance of the above processes to Yucca Mountain repository performance is not known. Few studies have been conducted regarding the effect of hyperalkaline plume migration in the near-field environment of a geologic repository similar to Yucca Mountain. Process-level model calculations by Lichtner, Pabalan, and Steefel (1998) suggest that strong alteration of the tuff host rock at Yucca Mountain and of cement in contact with the tuff host rock could result from diffusion-controlled interaction of cement and tuff pore waters and the respective minerals. Results from Lichtner, Pabalan, and Steefel (1998) show that porosity reduction within the tuff matrix could isolate it from fracture pore water, a process that would reduce the importance of matrix diffusion as a retardation mechanism. The model calculations also predict calcification of the cement, a reaction that is predicted to be more pronounced in a partially saturated system, compared to a fully saturated system.

To provide preliminary information regarding the potential effects of hyperalkaline plume migration on repository performance, sensitivity analyses were carried out using the TPA Version 3.1 code. Four scenarios simulating the effects of a hyperalkaline plume were studied:

- Scenario 1: Enhanced fracture flow due to dissolution and widening of fractures
- Scenario 2: Increased matrix flow and matrix diffusion due to dissolution of the tuff host rock
- Scenario 3: Reduced diffusion from the fractures into the matrix caused by mineral precipitation along and coating of fracture walls
- Scenario 4: Reduced fracture flow caused by mineral precipitation and plugging of the fractures

The above scenarios are focused on the potential effect of a hyperalkaline plume on hydraulic properties and the consequent effect on repository performance. The possible effect of hyperalkaline pH on radionuclide retardation and solubility and, consequently, on repository performance was not evaluated. Literature data indicate that sorption  $K_d$ s of radionuclides, particularly actinides, in cementitious environments are high (Bradbury and Sarott, 1995; Campbell and Krupka, 1997). For example, a comparison of  $K_d$ s of different elements specified in the TPA code input file with  $K_d$ s of those elements in cement environments taken from Bradbury and Sarott (1995) indicate that, except for technetium, radium, cesium, and selenium, radionuclide  $K_d$ s are higher, typically orders of magnitude higher, in cement-affected environments compared to a Yucca Mountain environment unaffected by the presence of cementitious materials. Thus, the lower radionuclide sorption in hyperalkaline environments assumed in DOE sensitivity analyses (TRW Environmental Safety Systems, Inc., 1996) may be incorrect, although conservative.

### **3.4.1.1 Assumptions and Modeling Approach**

It is not possible to explicitly model Scenarios 1, 2, 3, and 4 with TPA Version 3.1. Thus, an indirect approach was attempted in which parameters available in the TPA code input file were

varied to approximate the effects postulated in each scenario. Four types of hydraulic parameters that can be specified in the input file are of specific interest, namely: (i) matrix permeability, (ii) matrix porosity, (iii) fracture permeability, and (iv) fracture porosity. Widening of fractures or enhanced matrix flow was simulated by increasing the permeability and porosity of the fracture or matrix, whereas, reduction in fracture flow or matrix flow was simulated by reducing the permeability and porosity of the fracture or matrix. Reduced matrix diffusion due to mineral precipitation along fracture walls was simulated by decreasing the matrix porosity and permeability.

The potential spatial extent of the zone affected by a hyperalkaline plume is unknown. Sensitivity analyses with respect to the spatial extent of this zone cannot be done with TPA Version 3.1 code because the thickness of the zone cannot be specified in the input file. Thus, for the sensitivity calculations, the full thickness of the Topopah Spring Tuff unit was used to represent the altered zone. For Scenarios 1 and 3, calculations using the Topopah Spring Tuff unit would be more conservative relative to calculations using a discrete (e.g., 10 meters) hyperalkaline plume-affected repository layer. On the other hand, calculations for Scenario 4 using the Topopah Spring Tuff unit would be less conservative compared to those using a discrete layer for the altered zone. For Scenario 3, using the Topopah Spring Tuff unit would be more conservative if fracture flow was the dominant groundwater flow mechanism, otherwise, it would be less conservative.

The sensitivity of TPA results to the matrix permeability, matrix porosity, fracture permeability, and fracture porosity of the Topopah Spring Tuff unit was determined by allowing these parameters to be sampled in the TPA runs. The range of values and the probability distribution function of the parameter of interest specified in the input file were modified for each particular scenario. Because only four parameters are of specific interest, it was possible to study cases in which only one of these four parameters was sampled. In other cases, several parameters were sampled.

#### **3.4.1.2 Results**

It was determined that the lack of sensitivity of repository performance to variations in the parameter of interest was largely due to the approximation adopted in the TPA Version 3.1 code that if travel time in the unsaturated zone is less than 30 years, the travel time for transport of radionuclides in the unsaturated zone is assumed to be zero. In that case, performance is independent of the flow and transport properties of the unsaturated zone below the emplacement horizon. Because the parameters resulted in short calculated travel times, that were then set to zero, the sensitivity of repository performance to these parameters was not thoroughly tested. Revisions to the TPA code are planned that will include the capability to model transport in the near-field environment so that the effects of changes in near-field hydraulic properties on repository performance can be evaluated. Additional discussion of our sensitivity and uncertainty analyses using the TPA Version 3.1 code are presented in Jarzempa, et al. (1999).

### **3.4.2 Effects of Corrosion Products from Waste Packages on the Near-Field Environment**

Corrosion of waste containers could result in alteration products, such as iron oxyhydroxides, which could either sorb and retard radionuclide migration or form pseudo-colloids that adsorb and could enhance transport of radionuclides. The effectiveness of the near-field environment in physical and chemical filtering of iron oxyhydroxides from suspension will determine whether steel alteration products will retard or enhance transport of radionuclides. If the flow is predominantly through the matrix, it is likely that iron oxyhydroxides will be effectively filtered from suspension. Thus filtration will act as an additional barrier to transport of radionuclides. Fracture flow may lead to unretarded or enhanced flow, if the fracture apertures are large enough for iron oxyhydroxide transport, and if the chemical conditions of the solution (e.g., low ionic strength) lead to more stable suspensions. If fracture apertures are too small, or if the chemical conditions favor flocculation and settling, then iron oxyhydroxide transport will not be effective.

#### **3.4.2.1 Assumptions and Modeling Approach**

To investigate the potential effect of iron oxyhydroxide formation on dose, two end-member cases were considered in TPA calculations relevant to the effects of waste package corrosion products. In one case, radionuclides were assumed to be retarded by the iron oxyhydroxides. In the second case, radionuclide migration was assumed to be enhanced by transport of iron oxyhydroxides. An indirect approach was used in which parameters available in the TPA code were varied to approximate the effects of these two end-member cases. Potential effects on radionuclide transport were studied by using radionuclide distribution coefficients,  $K_d$ s, applicable to the cases. As an approximation of the spatial extent of iron oxyhydroxide transport, and to be consistent with the physical situation considered in the cement sensitivity analyses, the zone of transport was assumed to extend vertically downward from the emplacement horizon to the bottom of the Topopah Spring Tuff. Because of large uncertainties in estimating the amount of colloid-size iron oxyhydroxide generated by waste package degradation, possible effects on solubility or increased loading to levels in excess of solubility limits was not considered. In addition, the possibility of plugging fractures and matrix porosity by particulates was acknowledged, but not specifically considered. It seems likely that effects on hydrologic parameters in the near field due to iron oxyhydroxides will be overshadowed by those induced by cement-water interaction.

Of the elements currently considered in the TPA database, not all are likely to sorb strongly onto iron oxyhydroxides to form pseudocolloids. Those not considered in this analysis are Tc-99, I-129, C-14, Cl-36, and Cs-135. It is possible that the anions  $\text{Cl}^-$  and  $\text{I}^-$  will sorb on iron, but there are no available data. In addition, anion sorption may be greatly reduced at high pH values possible in the near field. For technetium, the effect of iron will be more significant in helping to reduce  $\text{Tc}^{7+}$  to  $\text{Tc}^{4+}$ , with lower solubility and increased sorption, but there is a lack of data to estimate technetium sorption behavior. Similar to sodium, cesium is a weakly hydrolyzable cation and is not likely to interact strongly with iron. Carbon has been shown to sorb onto iron oxyhydroxide, but the effect is probably small relative to the effects of carbonate precipitation.

Two conceptual models were considered simultaneously:

- Colloid transport through fractures. Fracture apertures in the Topopah Spring are on the order of 100 microns and larger (Wilson, et al., 1994), and may be large enough to transport colloids (<1 micron) and lead to early breakthrough due to hydrodynamic forces. For this scenario, the fracture retardation factor ( $R_d$ ) was modified.
- Because the TPA code already assumes no retardation in fractures (i.e.,  $R_d = 1.0$ ), the only consideration is enhanced transport ( $R_d < 1$ ). In the absence of any definitive estimates of colloid transport, a maximum of 10 percent enhancement of fracture colloid transport was assumed ( $0.9 \leq R_d \leq 1.0$ ). Because only iron oxyhydroxide pseudocolloids were considered here, the same distribution (uniform) was assumed for all radioelements. This conceptual approach is conservative because it neglects any colloid filtration.
- Colloid filtration by pores of the matrix and enhanced retardation by iron oxyhydroxides relative to the tuff mineralogy. A specific filtration mechanism (chemical attachment, straining) is not implied. Two populations of pore sizes cluster around 5 and 200 microns (Peters, et al., 1984). The larger pore size may be sufficient to transport colloids, but the likelihood of constricted pore throats and slow matrix flow suggest that colloid transport through the matrix is limited. This conclusion is consistent with field observations at the Peña Blanca site where higher uranium concentrations are associated with hematite in fractures, but uranium migration into the matrix was limited (Pearcy, Prikyrl, and Leslie, 1995). Matrix  $K_d$  values were modified for each of the radioelements for the Topopah Spring Tuff to reflect the enhanced sorption by iron oxyhydroxides in the tuff. For the purposes of the sensitivity analysis, the  $K_d$  distributions developed for iron oxyhydroxides through expert elicitation for TSPA-93 (Wilson, et al., 1994) were used to identify the minimum, maximum, and mean (if appropriate)  $K_d$ . Curium was not considered in TSPA-93 due to its 3+ valence; however, curium was assigned the same  $K_d$  distribution as americium and niobium. This approach is non-conservative but was used to determine the potential importance of this retardation mechanism.

In the simulations, 220 realizations (equal to  $10n$  for  $n = 22$  variables) were run for 10,000 years at five kilometers. Both  $R_d$  and  $K_d$  were allowed to vary during the simulation.

### 3.4.2.2 Results

The reason for the lack of sensitivity of repository performance to variations in the parameter of interest is the same as that discussed for the cement scenario (i.e., the parameters used in the calculations resulted in short travel time in the Topopah Spring Tuff). Thus, the sensitivity of repository performance to those parameters was not thoroughly tested. Revisions to the TPA code are planned that will include the capability to model transport in the near-field environment so that the effects of colloid transport in the near field on repository performance can be evaluated. Additional discussion of our sensitivity and uncertainty analyses using the TPA Version 3.1 code is presented in Jarzemba, et al. (1999).

### 3.4.3 Conceptual Model of Waste Package Degradation—Brine Formation on Container Surface

Aqueous corrosion is one of the degradation processes that may lead to waste package failure. Water films on metal surfaces will contain a variety of components, including chloride and other soluble anionic species. Chloride ions are known to promote localized corrosion and enhance general corrosion of container materials. Evaporation of water contacting the waste container could lead to increasing chloride concentrations and eventually to brine formation on the outer overpack surface, enhancing the susceptibility of both the outer and inner containers to localized corrosion.

To determine the possible effect of high chloride concentrations and oxygen concentration on dose, calculations have been completed using the EBSFAIL and EBSREL modules of the TPA code. Waste package corrosion depends on the corrosion potential ( $E_{\text{corr}}$ ) and the critical potential required to initiate localized corrosion. The value of  $E_{\text{corr}}$  is strongly dependent on oxygen concentration. In EBSFAIL, the critical potential, conservatively represented by the repassivation potential ( $E_{\text{rp}}$ ), is assumed to depend only on container material, chloride concentration, and temperature. For a given temperature, localized corrosion only occurs above a critical chloride concentration. At a given temperature,  $E_{\text{rp}}$  decreases with increasing chloride concentration while  $E_{\text{corr}}$  increases with increasing oxygen concentration. High chloride and oxygen concentrations are expected to induce earlier failures of waste packages after their surfaces become wet because conditions for the initiation of localized corrosion will be established earlier.

#### 3.4.3.1 Assumptions and Modeling Approach

The conceptual model for the evolution of the concentration of chloride and oxygen in the near-field environment is part of an integrated process model that provides the chemical composition of the aqueous and gaseous environment as a function of time in the immediate vicinity of the waste packages. The model takes into consideration the evaporative effects produced in a partially saturated environment by the heat released from the waste package due to radioactive decay. When evaporation takes place, an increase in salinity occurs, and pH increases as  $\text{CO}_2$  is degassed from solution. At the same time, oxygen concentration decreases following the decrease in the oxygen partial pressure due to purging of the air by evaporation of ambient groundwater in the partially saturated tuff. The computer code, MULTIFLO (Lichtner and Seth, 1996b), is used to calculate concentrations of dissolved aqueous and gaseous species in the matrix pore space (Lichtner, 1997). This information is provided in tabular form to the TPA Version 3.1 code (Manteufel, et al., 1997).

To address uncertainties in the MULTIFLO calculations, the results of MULTIFLO are multiplied by a chloride multiplication factor. The uncertainties include the potential for evaporative concentration of solutes that could accumulate as fluid drips onto the waste packages, and spatial variability in the chloride concentration between the various subareas. The TPA code does not explicitly take into account the potential for dripping water during the thermal period when the relative humidity in the drift is below the critical relative humidity necessary for a water film to develop on the surface of the waste package. Evaporative concentration of solutes could occur, leading to saturated salt solutions on the waste package (Walton, 1993). A uniform distribution was adopted for chloride concentration factor, ranging from 1 to 30. The

upper value selected, when multiplied by the maximum concentration from the MULTIFLO calculations, is close to the solubility of NaCl at temperatures ranging from 25 °C to the boiling point of water at the repository horizon.

The values of dissolved oxygen concentration calculated by MULTIFLO are not inputs in EBSFAIL. A constant value equal to the oxygen partial pressure in air is used, and the concentration of oxygen in solution is calculated in EBSFAIL by using Henry's law. The partial pressure of oxygen is an input parameter for both EBSFAIL and EBSREL. A fixed, constant value of 0.21 atmosphere is used in the TPA Version 3.1 code input file assuming the maximum value expected in the near-field environment when the temperature of the waste package decreases below the boiling point of water. For some of the sensitivity runs, a triangular distribution was selected for the oxygen partial pressure. A minimum value for the oxygen partial pressure of  $2.1 \times 10^{-5}$  atmosphere, reflecting carefully de-aerated water, and maximum and peak values of 0.21 atmosphere were adopted, corresponding to a mean value of 0.14 atmosphere.

Twenty different sensitivity studies were performed. Each sensitivity studied differed from the others by the values assigned to chloride multiplication factor, oxygen partial pressure, distance to the receptor group, simulation period, or the distribution assumed for selected UZFLOW parameters. These studies examined the sensitivity of maximum dose, time of the peak dose, and the cumulative engineered barrier system release to chloride multiplication factor and oxygen partial pressure. The receptor group was assumed to be located either 5 or 20 kilometers away from the repository. The simulation period ranged from 10,000 to 30,000 years. One hundred realizations were performed for each simulation. Scatter plots were used to evaluate the influence of parameters on the dependent variables.

### **3.4.3.2 Results**

Cumulative engineered barrier system release was found to be highly sensitive to chloride multiplication factor within a very narrow range of variation, exhibiting a step function response near value of 10.0, where cumulative releases increase by approximately one order of magnitude. A similar step function is also apparent for oxygen partial pressure, with higher releases occurring for partial pressures greater than about  $3.0 \times 10^{-2}$  atmosphere. The chloride concentration needs to exceed a threshold value to initiate localized corrosion for Alloy 625. Therefore, even in the presence of sufficient oxygen, low chloride concentrations can result in low releases of radionuclides. Different alloys are expected to have different chloride threshold concentrations for the initiation of localized corrosion.

Use of a chloride multiplication factor to address the uncertainties in chloride concentration, as evaluated by MULTIFLO (Lichtner and Seth, 1996b) calculations, appears to be appropriate. The decrease in oxygen partial pressure to values almost an order of magnitude lower than those prevailing in the atmosphere does not appear to significantly influence repository performance as measured by cumulative release from the engineered barrier system or maximum dose for a 20,000-year simulation period. This decrease in oxygen partial pressure is considered to accommodate the expected spatial variability, unless the oxidation reactions with man-made materials results in a nearly complete consumption of oxygen. Additional discussion of our sensitivity and uncertainty analyses using the TPA Version 3.1 code is presented in Jarzempa, et al. (1999).

### 3.4.4 Conceptual Model of Oxidation Rate Controlled Limits on Radionuclide Release

Radionuclide release rates in the TPA code basecase scenario are based on extrapolations of the results of short-term laboratory experiments. Uncertainties in long-term extrapolations and in the dependence of release rates on environmental characteristics introduce considerable uncertainty in determination of the source term for the potential repository at Yucca Mountain. An alternate approach is to use data from natural analog systems to constrain release rates on a geologic time scale. The Nopal I uranium deposit at Peña Blanca, Chihuahua, Mexico, has been studied in detail as an analog of the proposed repository at Yucca Mountain. Using data from that site, conservative estimates of the maximum average rate of oxidative alteration of primary uraninite (an analog of spent fuel) have been calculated (Murphy and Percy, 1992; Murphy, Percy, and Pickett, 1997). The rate is a maximum because a maximum limit was used for the infiltration that removed uranium, and a minimum limit was used for the period of oxidation. The rate is an average because it is integrated over the entire period of oxidation.

#### 3.4.4.1 Assumptions and Modeling Approach

This sensitivity analysis was based on the premise that the oxidation rate of spent fuel in the potential Yucca Mountain repository will be comparable to the oxidation rate of uraninite at Peña Blanca. Similarities between these sites in geology, geochemistry, climate, and hydrology support this premise. Calculation of the bounding rate of oxidation depends on the amount of uranium removed from the site. Peña Blanca site-specific characteristics were used to determine this quantity, based on conservative estimates of the uranium solubility and the groundwater flux through the system. The maximum average uranium oxidation rate for the Yucca Mountain repository was determined by scaling for the masses of uranium at each site. For radionuclides incorporated in the matrix of spent fuel, the oxidation rate was assumed to limit their release rate.

Murphy, Percy, and Pickett (1997) calculated a maximum bounding oxidation rate (i.e., the maximum average oxidation rate) for uraninite at Peña Blanca based on the approach of Murphy and Percy (1992) and a revised minimum time for oxidation based on radiometric ages of late-forming uranophane (an oxidized uranium mineral) of 3 million years (Pickett and Murphy, 1997). Calculation of the bounding rate depends on the amount of uranium that has been removed from the site, which is given by the product of the water flux and uranium concentration yielding the relation (Murphy and Percy, 1992)

$$R_0 = (U_e / t) + FC \quad (3-1)$$

where  $R_0$  stands for the oxidation rate (e.g., grams per year),  $U_e$  represents the amount of oxidized uranium remaining at the deposit,  $t$  represents the period of time of oxidation,  $F$  stands for the water infiltration rate (e.g., liter per year), and  $C$  denotes the concentration of uranium in solution exiting the system. Peña Blanca site-specific characteristics were used to determine this quantity based on estimates of the uranium solubility and conservative limits on the groundwater flux through the system. The calculated maximum average rate is 140 grams per year for 320 metric tons of oxidized uranium remaining at the site and an estimated upper limit of 86 metric tons of uranium removed from the site over the 3-million-year period of leaching

subsequent to onset of oxidation. Because the oxidation rate of uraninite was rapid relative to transport out of the system, the limit on oxidation rate should be applicable to analyses for Yucca Mountain, although hydrologic transport rates may differ between the sites.

Scaling the potential Yucca Mountain repository to Peña Blanca, 70,000 tons of uranium at a hypothetical Yucca Mountain repository are  $70,000/408 = 172$  times the initial estimated (maximum) quantity of uranium at Peña Blanca. Supposing the whole of the repository inventory of spent fuel oxidizes at a comparable rate that at Peña Blanca, the total rate is given by  $172 \times 140$  grams per year = 24 kilograms per year. This value represents a maximum average uranium oxidation rate for the potential Yucca Mountain repository, based on the maximum average rate of oxidation estimated for the Peña Blanca deposit, and is used to estimate the release rate for radionuclides in the spent fuel matrix.

Uranium solubilities [see Equation (1)] were used in calculation of the maximum average oxidation rate at the Peña Blanca deposit to estimate the maximum amount of uranium that has been removed from the deposit. Determination of the uranium solubility was based on the premise that secondary, oxidized alteration products control uranium solubility, which is indicated by the Peña Blanca data. This approach differs from that adopted in the DOE TSPA-95 to determine uranium solubility. TSPA-95 adopted a log  $\beta$  distribution of solubilities for uranium centered on values selected by the expert elicitation that supported TSPA-93 (Wilson, et al., 1994). It is stated in TSPA-95 that "it was not expected that solubility-limits for uranium would be a factor in release of uranium in Yucca Mountain even though the ground water contains dissolved silica which could cause uranium silicates to precipitate." In this sensitivity analysis, the alternate hypotheses were taken that secondary uranyl minerals control uranium solubility and that oxidation rates are fast relative to transport of uranium out of the waste emplacement horizon. These alternate hypotheses are supported by the Peña Blanca natural analog data.

For the maximum average oxidative alteration rate analysis for the Peña Blanca site, Murphy and Percy (1992) adopted a uranium solubility of  $10^{-7}$  molar. The solubility was calculated by estimating the water chemistry and using uncertain thermodynamic data for the uranophane dissolution equilibrium constant. To examine possible ranges of solubilities based on secondary mineral formation, a suite of model aqueous solutions were computed for the Yucca Mountain site. A variety of possible water chemistries and solubility controlling minerals was examined. The mean of the base 10 logarithm of the uranium solubility (in moles per liter) in these analyses was  $-7$ , corresponding to the value calculated previously by Murphy and Percy (1992). The standard deviation of the 32 computed values of the logarithm of uranium solubility was 1.8 units.

#### **3.4.4.2 Results and Discussion**

One hundred runs of the code were performed, each using one perfectly correlated pair of sampled uranium solubility and corresponding leach rate. Dose was calculated in sets of analyses for a 5-kilometer distance from the repository for various times up to 100,000 years. The dose for release rates based on the Peña Blanca natural analog were lower than those calculated using other release models contained within the EBSRELEASE module. The mean dose is about an order of magnitude lower than the basecase release model. The results from the natural analog study are not necessarily conservative, because we assume the release of

radionuclides in the matrix is directly related to release of solubilized uranium. This direct relationship between uranium and all other radionuclides is only possible if radionuclides initially in the spent fuel matrix are quantitatively captured in secondary uranium minerals that form as a result of the oxidation of uranium.

Additional discussion of system level sensitivity studies of the natural analog alternative source term model is presented by Jarzempa et al. (1999). Furthermore, Murphy and Codell (1999) present Yucca Mountain performance assessment results calculated using the TPA 3.2 code to test the sensitivity of results to two alternate source term models. One model is based on the assumption that the release rate of matrix components from spent nuclear fuel is limited by the maximum average oxidation rate of uraninite at the Nopal I uranium deposit scaled to the proposed repository inventory. The second model is based on the assumption that matrix components of spent fuel are incorporated in secondary schoepite, and that their releases are governed by the equilibrium solubility of schoepite, which is computed as a function of temperature, pH, and aqueous carbonate and uranyl speciation. Probabilistic doses are calculated for a critical group located 20 kilometers from the waste emplacement at 10,000 and 50,000 years after repository closure. Both alternate models yield lower doses than the basecase model in TPA 3.2 analyses.

## **4.0 REVIEW METHODS AND ACCEPTANCE CRITERIA**

This IRSR is focused on providing a path to resolution. The discussion will be revised to become more specific once performance assessment studies are completed, other analyses and testing progressed, the license application design of the repository is specified, and the NRC regulations for the potential Yucca Mountain repository (10 CFR 63; U.S. Nuclear Regulatory Commission, 1999a) are finalized. A systematic approach to resolving subissues is necessary. The extent and nature of the coupled THC processes that may affect repository performance are strongly dependent on both the design (e.g., thermal-loading strategy, ventilation, materials, etc.) and concept of operation of the repository (e.g., length of preclosure period). As a result, this IRSR is formulated in a manner that will lead to resolution of the subissues by allowing progress in determining which effects of coupled THC processes need to be included in any performance assessment of the proposed repository.

DOE needs to consider coupled THC interactions both within and between key elements of the engineered and natural subsystems of the repository, as discussed in Section 3, to adequately demonstrate and quantify the consequences that ENFE might have on repository performance. The acceptance criteria and review methods being developed for the Yucca Mountain Review Plan focus on 14 model abstractions, which are equivalent to the integrated subissues in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 2000a).

Several relatively comprehensive review documents have been published that are relevant to coupled THC processes in the near field of the proposed repository at Yucca Mountain, Nevada (Manteufel, et al., 1993; Wilder, 1996; Bish, et al., 1996; Angell, et al., 1996; Hardin and Chestnut, 1997; Hardin, 1998). Although material in these reviews supports much of the technical basis for this IRSR, references to primary sources of data are generally provided in the text.

### **4.1 THE EFFECTS OF COUPLED THERMAL-HYDROLOGIC-CHEMICAL PROCESSES ON SEEPAGE AND FLOW**

In Revision 3 of the Repository Safety Strategy (CRWMS M&O, 2000a) the DOE identified seven principal factors of the postclosure safety case. Three of these principal factors are potentially affected by persistent THC alterations of the flow properties: (i) limited seepage of water into the emplacement drifts, (ii) retardation of radionuclide migration in the unsaturated zone, and (iii) dilution of radionuclide concentrations during migration. Other factors identified by the DOE for the postclosure safety case, though given lower importance, include (i) coupled processes—effects on unsaturated zone flow, (ii) coupled processes—effects on seepage, and (iii) advective transport in the unsaturated zone. One integrated subissues (U.S. Nuclear Regulatory Commission, 2000a). requires input from ENFE Subissue 1: flow paths in the unsaturated zone.

Drift seepage enters into a performance assessment framework through the integrated subissue on flow paths in the unsaturated zone. Drift seepage depends on flow properties at the tunnel crown, and is thus potentially affected by THC-induced changes in these properties. Radionuclide dilution enters into a performance assessment framework through several integrated subissues. One of these integrated subissues—advective flow paths in the unsaturated zone—is potentially affected by THC-induced changes. In particular, dilution in the

unsaturated zone depends on the advective flow paths through this zone and on the distribution of flow between matrix and fractures, which are potentially altered by THC processes. The principal factor, retardation of radionuclide migration, is also covered by several integrated subissues, one of which is relevant to this integrated subissue on flow paths in the unsaturated zone. THC-induced changes in flow paths can affect retardation indirectly by altering the fraction of contaminants passing through highly sorbing zones. For example, THC-induced changes in rock flow properties may alter the partitioning between the fractures and the more sorbing matrix or cause highly sorbing zeolitic zones to be bypassed.

Contributions from the Unsaturated and Saturated Flow Under Isothermal Conditions (USFIC) KTI (U.S. Nuclear Regulatory Commission, 1999e) and the Thermal Effects on Flow KTI (U.S. Nuclear Regulatory Commission, 1999c) will be required, in addition to those of the ENFE KTI, to successfully address the issue of THC effects on seepage and flow. The DOE must adequately address the potential for coupled THC processes to alter seepage and flow in its assessments of repository performance.

#### **4.1.1 Technical Bases for Reviewing the Effects of Coupled Thermal-Hydrologic-Chemical Processes on Seepage and Flow**

The technical bases are primarily focused on explaining why the results of coupled THC processes may be important to seepage and flow. Past performance assessment analyses of the effects of coupled THC processes on the potential repository seepage behavior and groundwater flow have been limited. In addition, the relevance to performance of a particular coupled THC process is highly dependent on repository design (e.g., thermal-loading strategy, materials used, etc.). Thus, the focus is on ensuring that analysis of coupled THC effects is completed. The sophistication of the analysis of the effects of coupled THC processes on seepage and flow that could be found acceptable by staff depends on the information available at the present, any plans to obtain the additional information as part of the long-term testing program, and the ability of codes to model coupled processes and determine their impact on repository performance.

For instance, results of coupled THC modeling (see Chapter 4 of Sagar, 1997, for an example) might be used to infer the potential amounts of primary minerals that are either dissolved or the mass of secondary minerals that may precipitate due to the coupling. This information might be combined with existing site distribution of primary and secondary minerals (abundance, and location in the matrix or fracture or both) and the molar volume of the minerals to calculate the increase or decrease in porosity caused by the coupled THC process. If the resulting changes in porosity and other hydrologic properties are conservatively bounded by the range of values sampled for a particular modeled hydrologic unit in the basecase performance assessment, the effect of that coupled process on seepage and flow need not be considered further.

##### **4.1.1.1 Coupled Thermal-Hydrologic-Chemical Processes Affecting Flow of Water**

At Yucca Mountain, host rocks are silicic tuffs [70 to 80 percent SiO<sub>2</sub>; (Byers, 1985)]. These rocks are variably vitric (glassy), devitrified, or altered. Devitrification of the glass results in an assemblage of silica minerals and alkali feldspar. The tuffs can be altered, primarily to the silica-rich zeolites, clinoptilolite and mordenite, or to analcime at depth (Broxton, Bish, and Warren, 1987; Bish and Chipera, 1989). The saturated zone groundwater is predominantly a

dilute, oxidizing, sodium bicarbonate solution rich in dissolved silica. However, there is evidence from one well (UE-25 WT #17) that redox conditions can locally be mildly reducing ( $-100$  millivolts  $\leq E_h < 0$  millivolt). The unsaturated zone groundwaters differ substantially from saturated zone water, being more concentrated and dominated by calcium chloride or calcium sulfate in rocks nearer the ground surface (Yang, 1992; Yang, Peters, and Thorstenson, 1993; Yang et al., 1996; Yang, Rattray, and Yu, 1996). Aqueous silica concentrations in excess of cristobalite saturation are observed in tuffaceous aquifers at Yucca Mountain (Kerrisk, 1987), and higher concentrations are observed in the unsaturated zone (Yang, et al., 1996). The high silica contents are generated by reaction of infiltrating meteoric water with siliceous volcanic glass (White, Claassen, and Benson, 1980). Glass alteration in the Calico Hills formation below the repository horizon is accompanied by incongruent precipitation of mordenite and clinoptilolite. These minerals are zeolites with important sorptive characteristics (Ames, 1964; Pabalan, et al., 1998; Bertetti, Pabalan, and Almendarez, 1998). In the proposed repository horizon and the near-field environment at Yucca Mountain, the tuffs are devitrified to alkali feldspars and silica mineral polymorphs (cristobalite, quartz, and tridymite). Here, smectite is the dominant aluminosilicate alteration phase, occurring in abundances up to 5 percent (Bish, 1988). In lower volcanic units at Yucca Mountain, primary glass has been altered to analcime  $\pm$  kaolinite rather than clinoptilolite. This mineralogic change, which is generally associated with the disappearance of cristobalite with depth, is consistent with a decrease in the activity of aqueous silica (Kerrisk, 1983). Fracture and cavity mineralogy is characterized by calcite, smectite, silica minerals, zeolites, and manganese oxides (Carlos, 1989; Carlos, Bish, and Chipera, 1991; Paces, et al., 1996).

The gas phase in the vadose zone at Yucca Mountain is primarily air, approximately saturated with liquid water, and enriched in CO<sub>2</sub> relative to the atmosphere. Gas chemistry analyses show limited variability in the CO<sub>2</sub> content in space and time (Thorstenson, et al., 1990). The partial pressure of CO<sub>2</sub> in the gas phase has a strong effect on the pH of the coexisting groundwater, which, in turn, affects aqueous speciation, solubilities, and mineral stabilities (Arthur and Murphy, 1989; Murphy, 1993).

Under elevated temperatures, the rates of alkali feldspar dissolution and growth of secondary phases, such as smectite, clinoptilolite, silica minerals, and calcite, would be accelerated. Thermodynamic analyses for smectites (Ransom and Helgeson, 1994) and clinoptilolites (Bowers and Burns, 1990) have quantified their decreasing stability with increasing temperature and decreasing aqueous silica content. In addition, the swelling capacity of uncompact smectite has been shown to be irreversibly decreased by alteration in a water vapor environment at temperatures above 150 °C (Couture, 1985). Field evidence for temperature-induced changes can be obtained by regarding the natural environment at depth as an analog of the near-field environment (Apted, 1990). Observations at Yucca Mountain include the transitions with increasing depth from clinoptilolite to analcime to albite and from smectite to ordered illite/smectite to illite. The clay mineral data have been interpreted to give thermal profiles with temperatures ranging up to 300 °C for an extinct hydrothermal system at depth at the north end of Yucca Mountain (Bish, 1989; Bish and Aronson, 1993). Reduction in permeability of tuff adjacent to fractures has been documented in rocks near the intrusion of a basaltic sill (Matyskiela, 1997).

Experimental studies of hydrothermal reaction of tuffs and natural waters from Yucca Mountain at 90 to 250 °C have been conducted (Knauss, et al., 1984; Knauss, Beiriger, and Peifer, 1987; Knauss, 1987). These studies demonstrate dissolution of primary minerals, precipitation of

secondary phases, and variations in water chemistry. Secondary precipitation of clay minerals, zeolites, cristobalite, and calcite have been observed. Experiments in which CO<sub>2</sub> loss occurred showed more extensive secondary mineralization and particularly more calcite precipitation than in pressurized closed-system experiments. Water chemistry variations were generally small, and achieved approximately steady-state conditions in long-term experiments. The aqueous silica concentration was observed to increase substantially at elevated temperatures corresponding to the increased solubility of silica minerals. The water chemistry in selected experiments was reasonably represented as a function of time with partial equilibrium and kinetic reaction path models of the water-rock interactions (Delany, 1985). Application of the results of these experiments to the near-field environment at Yucca Mountain must be judicious, because the high temperatures, high pressures, saturated conditions, and short time scales of the experiments are unrepresentative of expected conditions at Yucca Mountain.

Mass transfer calculations that account for partial equilibrium and reaction kinetics in gas-water-rock interactions have provided geochemical models related to the Yucca Mountain site and near-field environment (Kerrisk, 1983; Ogard and Kerrisk, 1984; Delany, 1985; Arthur and Murphy, 1989; Murphy, 1993; Murphy and Pabalan, 1994; Lichtner and Seth, 1996a). The aqueous silica concentration and the CO<sub>2</sub> pressure have been shown to be particularly important in defining the solid-phase assemblage and the aqueous-solution composition. At present, these models are limited principally by the lack of thermodynamic and kinetic data. However, there have been several recent attempts to obtain such data by experimental and estimation techniques (e.g., Bowers and Burns, 1990; Johnson, et al., 1991; Ransom and Helgeson, 1994; Murphy, et al., 1996; Chipera and Bish, 1997) which, in some cases, appear to yield inconsistent results. Calculations of time-dependent processes are further hampered by the difficulty in realistically characterizing reactive surfaces in geologic environments.

Coupled THC effects on seepage and flow may be affected by changes in porosity and permeability of the host rock. The porosity and permeability may be enhanced by dissolution of primary minerals that make up the matrix of the medium. Conversely, precipitation of secondary minerals may serve to plug available porosity, reducing permeability. Mineral solubilities depend on the pressure and temperature of the system of interest, as well as solution pH, p(CO<sub>2</sub>), p(O<sub>2</sub>), and salinity. Many common minerals, such as quartz and metal sulfides, exhibit a prograde solubility, whereby precipitation is favored with decreasing temperature. Silica scale in geothermal wells is due, in large part, to the cooling of fluids that are supersaturated, with respect to silica, as they rise to the surface (Thomas and Gudmundsson, 1989). In contrast, carbonates, such as calcite and dolomite, exhibit retrograde solubility and precipitate from solution with increasing temperature. Precipitation and dissolution may also be controlled by kinetic processes. For example, although calcite precipitates readily in geothermal systems, quartz precipitation is kinetically controlled at temperatures below 200 °C and may not occur under supersaturated conditions (Thomas and Gudmundsson, 1989).

Gas flow from the near field, driven by vaporization of water, is predicted to be away from the near field in all directions (Pruess, Wang, and Tsang, 1990; Tsang and Pruess, 1987). This process would tend to purge air containing O<sub>2</sub> from the near-field environment. The vapor pressure of water at temperatures above 95 °C exceeds the hydrostatic pressure of less than 0.1 megapascal at Yucca Mountain. Thus, the gas phase in the near field would tend to be dominated by H<sub>2</sub>O. The different minerals have different temperature-dependent solubilities.

As a result, solutions (both liquid and gas phase) moving by thermally driven convection will redistribute chemical components such as pH, chloride, oxygen, CO<sub>2</sub>, silica, and calcium.

Silica redistribution in the Yucca Mountain near-field environment is likely to be controlled by the dissolution of glass, feldspar, and cristobalite, and amorphous silica precipitation. The rate of this redistribution will depend on the aqueous silica activity and the relative rates of reaction. Silica redistribution has been observed in laboratory heater experiments with Yucca Mountain tuff under unsaturated conditions (Rimstidt, Newcomb, and Shettel, 1989). Silica and iron, dissolved near the heater, were transported in solution and precipitated as amorphous silica, iron hydroxides, clay, and zeolite at the cooled end of the system. In the nonisothermal, transient experiments of Lin and Daily (1990) on samples of the Topopah Spring Tuff, permeability was progressively reduced by three orders of magnitude from  $1.3 \times 10^{-14}$  to about  $10^{-17}$  square meter due to narrowing of fracture apertures by silica deposition. Experiments of Vaughan (1987) using granite cores indicated that, although porosity was reduced by a relatively small amount, permeability was reduced by over 95 percent. Chigira and Watanabe (1994) also observed narrowing of pore throats by silica precipitation in flow experiments using powdered granite and amorphous silica powder. They calculated that at 90 °C porosity would be halved in 135 years.

Bish (1993) developed a premise that the thermal regime will induce a progressive Ostwald ripening effect in which a sequence of zeolites will form and, if enough time is available, culminate in the most stable assemblage of albite + quartz. This alteration will lead to a net volume reduction, potentially increasing porosity and permeability.

The effects of chemistry on flow are commonly neglected in thermal-hydrologic simulations. For the Viability Assessment repository design and thermal-loading pattern, development of an extensive region of counter circulating water and vapor above the emplacement drifts was predicted to cause changes in porosity, permeability, and solution composition over regulatory time frames of thousands of years (e.g., Hardin, 1998). Small changes in porosity can affect orders of magnitude changes in permeability (Lichtner and Walton, 1994). Thus, the dissolution and transport of silica, followed by precipitation during evaporation, could modify the permeability distribution around the repository horizon. Some numerical simulations have been performed in an attempt to predict the redistribution of pH, chloride, silica, and calcium in the near-field environment and the effect on permeability (Lichtner and Seth, 1996a; Lichtner and Turner, 1997). The increased drift spacing and lower areal heat loading in the Enhanced Design Alternative-II repository design may allow water to drain through the interdrift regions and may prevent the formation of a zone of perched condensate and the associated region of counter-circulating water and vapor above the repository. This potential lack of a vigorous heat pipe and refluxing decreases the likelihood of large changes in porosity and permeability above emplacement drifts.

Major geochemical changes in the near field are likely to depend primarily on the availability of water. Although unsaturated, the rocks at Yucca Mountain contain abundant water, commonly 10 percent of the rock volume. A large amount of zeolitic water is also available in certain horizons that could be released at elevated temperatures. Most extensive and rapid chemical reactions will occur where water evaporates, depositing solutes, and where water vapor condenses. Evaporation and mineral precipitation may occur in the rock matrix or in the fractures, depending on the local hydrologic conditions. However, gaseous transport of water vapor to cooler zones of condensation is likely to occur dominantly in fractures. Therefore,

condensation of initially dilute acidic water, due to dissolution of gaseous CO<sub>2</sub> into the condensate, and mineral dissolution are likely to occur on fracture surfaces. Together, these processes could lead to changes in average permeability and in permeability distributions for matrix and fractures. However, if water condensed on fracture surfaces dissolves minerals there and precipitates secondary phases with larger volumes before the water is imbibed, then fracture permeability could decrease as well. The locus of dissolution and precipitation reactions, with respect to fractures and matrix, could affect the hydrologic behavior of the near-field system, including seepage into drifts, and is presently poorly constrained in coupled hydrochemical modeling.

#### **4.1.1.2 Effects of Engineered Materials on Seepage and Flow**

The TSPA-VA design assumed cementitious materials, in the form of concrete inverts and linings, would be used for the estimated 179 kilometers of emplacement drifts of the potential Yucca Mountain high-level waste repository in addition to the planned use of cement in roadways for construction, and emplacement ramps and service mains. In the Enhanced Design Alternative-II design, cementitious materials are not planned to be used for ground support, except in the form of grout to anchor the rock bolts of the ground support system. The potential effect of cementitious materials on seepage and flow in the Enhanced Design Alternative-II design is expected to be substantially reduced compared to the TSPA-VA design. However, the current design does call for large amounts of cementitious materials in ventilation shafts and in ventilation tunnels below the emplacement drifts.

Interaction of cement with the tuffaceous host rock and ambient groundwater could have an important effect on seepage. The chemistry of pore fluids in contact with hydrated cement phases is characterized by persistent alkaline pH (>10). Hyperalkaline cement pore water is thermodynamically incompatible with silica, a major component of the proposed Yucca Mountain repository host rock unit. Thus, migration of the high-pH cement pore water into the host rock is likely to result in strong alteration of the tuff. The Maqarin natural analog site for studying cement interaction in northern Jordan suggests that rapid interaction between cement-equilibrated, pore waters, and silicate-bearing rocks can be expected (Lichtner, Pabalan, and Steefel, 1998). This may result in sealing of fractures and alteration of the host rock minerals. Calculations by Lichtner, Pabalan, and Steefel (1997, 1998) suggest that strong alteration of the tuff host rock at Yucca Mountain and of cement in contact with the tuff could result from interaction of cement and tuff pore waters and the respective minerals.

Because of the low silica concentration of the cement pore water, the host rock would begin to dissolve on contact with the hyperalkaline fluid (Lichtner and Eikenberg, 1995). Dissolution of tuff could lead to widening of the fractures and enhancement of seepage and groundwater flow through the repository. As the host rock dissolved and the silica concentration increased, calcium silicate hydrate phases would precipitate and clog the pore spaces. Alternatively, precipitation of calcite and calcium silicate hydrate phases along the interface of the fracture and matrix could seal the fractures from the matrix, producing isolated channels through which groundwater could flow. However, if sufficient amounts of calcite and CSH phases are precipitated along fracture walls, reduction in fracture porosity and permeability, or fracture plugging, could result in changes in seepage and groundwater flow through the repository. Reduction in horizontal permeability that might result from fracture plugging tends to increase seepage by limiting the capillary diversion effect, whereas the associated increase in capillary

pressures tends to have the opposite effect. Results of Lichtner, Pabalan, and Steefel (1997, 1998) show that porosity reduction within the tuff matrix could isolate it from fracture pore water and could affect seepage. Precipitation of calcite would also occur, as the low CO<sub>2</sub>-high calcium cement pore fluid mixes with the ambient groundwater containing high CO<sub>2</sub> concentrations (Lichtner and Eikenberg, 1995; Steefel and Lichtner, 1994).

The results of Lichtner, Pabalan, and Steefel (1998) indicate that calcification of the cement could occur as would be expected under ambient conditions. In the case of counter-diffusive transport, calcification is more pronounced in the case of a partially saturated system compared to a fully saturated one. Both the large-block test and drift-scale test thermal tests could provide important insights that could constrain the potential effects on seepage resulting from cementitious materials interacting with the tuff.

#### **4.1.1.3 Microbial Effects on Seepage and Flow**

Within the last decade data have been collected showing existence of diverse communities of microorganisms living in the deep subsurface environment (Amy and Haldeman, 1997). The role of microorganisms in many important geochemical phenomena has long been recognized (Ehrlich, 1996). Subsurface bacteria may mediate many processes that may impact the performance of a potential repository. These processes include canister corrosion and leaching of the glass waste form. Other potentially important microbial processes are supplying organic byproducts as potential complexants and direct uptake of radionuclides and either immobilization or mobilization depending on whether the microorganism is attached to an immobile surface. Finally microbial processes can alter pore-water and gas chemistry and microbes can produce exopolymeric materials that may block pores and fractures in the rock matrix impeding the flow of water. Only the latter two processes are discussed in this subissue. The conditions necessary for microbial activity in relation to all of these deep subsurface processes will be substantially the same and will be discussed below.

The potential importance of microbial processes to nuclear waste repositories has been recognized for over 10 years in international radioactive waste disposal programs (West, 1995). Extensive research and modeling programs are ongoing in other countries (Christofi and Philp, 1997), particularly in Switzerland (Brown and Sherrif, 1998), Canada (Stroes-Gascoyne, 1996), United Kingdom (West, 1995), and Sweden (Pedersen, 1996; Pedersen and Karlsson, 1995). While more work has been focused on low-level and intermediate-level waste repositories, as the quantity of organic carbon is greater than that expected for high-level waste repositories, several countries have assessed the potential importance of microbial processes in proposed high-level waste repositories (Christofi and Philp, 1997).

In contrast, microbiological study in the United States high-level waste program has only recently begun to determine the importance of microbial processes to repository performance. The efforts have focused on identifying microbial processes relevant to the Yucca Mountain site, determining parameters critical to evaluation of the disturbed environment (temperature and nutrients added), and identifying the most effective means of evaluating these two factors (Horn and Meike, 1995; Wilder, 1996). Microbes could compromise the integrity of the waste package, modify water and gas chemistry outside the bounds predicted by abiotic chemical calculations, and alter the rate of radionuclide transport from breached waste packages (Horn and Meike, 1995). Research efforts on the effect of microorganisms on radionuclide transport

at Yucca Mountain were summarized and evaluated (Hersman, 1996). One conclusion from this study was that the elevated subsurface concentrations of CO<sub>2</sub>, relative to the atmospheric concentration, is the result of biogenic activity. Finally, an initial attempt to model the potential effects on repository performance of microbial processes in the near field was been proposed as part of the TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). This effort attempted to address potential changes in gas chemistry (CO<sub>2</sub>) from microbial processes. The effort relied on models that have been developed in Europe to assess the potential importance of microbial processes on high-level waste repositories (TRW Environmental Safety Systems, Inc., 1997b; Grogan and McKinley, 1990).

Horn and Meike (1995) reviewed the conditions necessary for microbial activity in relation to deep subsurface repositories, such as Yucca Mountain. For metabolic activity, microorganisms require an energy source (usually organic carbon); water; a source of nitrogen, sulfur, and phosphorus; and suitable environmental conditions, such as temperature and pH (Horn and Meike, 1995). Most microorganisms are heterotrophic, using organic carbon compounds as their energy source. Oligotrophic bacteria (those that can grow in nutrient-poor water) require a minimum organic carbon content 1 to 15 milligrams per liter of water (Ehrlich, 1996). The organic carbon content of the groundwater in the vicinity of the repository (J-13 Well water) is in the range of 0.15 to 0.55 milligram per liter, comprising principally humic acids and low-molecular weight fatty acids (Means, Maest, and Crerar, 1983). This is well below the level suggested for oligotrophic bacterial growth. Nevertheless, microbial activity in the soil zone of the unsaturated zone is attributed as the reason for elevated CO<sub>2</sub> concentration in the gas phase (Hersman, 1996; Thorstenson, et al., 1998).

Unlike other international programs for high-level waste disposal where potential disposal sites are located in the saturated zone, the Yucca Mountain site is located in the oxidizing unsaturated zone. This has a profound consequence on the potential importance of microbial processes at Yucca Mountain. The activity of microbes is limited in the unsaturated zone by the low water potential (Kieft, et al., 1993,1997). Other limits on the potential for microbial activity in the subsurface of a waste repository include the available nutrients (nitrogen and phosphorus) and the increased temperature from radioactive decay (Pedersen, Motamedi, and Karnland, 1995).

Using samples collected from the Exploratory Studies Facility, Kieft, et al. (1997) conclusively demonstrated that water is the major limiting factor to growth and microbial activity at Yucca Mountain. Microbial abundance was low, and, in general, indicated microbes were most abundant in locations that were from less consolidated, nonwelded, or bedded tuffs, such as the Paintbrush tuff (Kieft, et al., 1997). Additions of N and P to crushed tuff samples resulted in little further stimulation of microbial activity. Addition of organic carbon to the crushed tuff samples stimulated growth more than just added water. Thus, there is a potential for increased microbial activity at Yucca Mountain with the addition of water and carbon.

The predominant mode of growth of bacteria in nature is as a biofilm attached to a surface, particularly in oligotrophic environments (Amy and Haldeman, 1997). Bacteria attach to solid surfaces, where nutrients concentrate. These surfaces provide a more abundant food source than the bulk fluid. For saturated conditions, provided sufficient nutrients and carbon are present, the growth of microbes in biofilms and the production of exopolymeric materials by microbes can be sufficient to substantially reduce the permeability of rock units and soil (Amy and Haldeman, 1997).

The tuff at Yucca Mountain is unsaturated, pore fluids are oligotrophic, and microbial activity is limited under ambient conditions. Microbial activity in the near field during the postclosure period, when elevated temperatures prevail as a result of localized heating by the waste, will be even further inhibited by the drying out of the tuff. Even if the tuff completely saturates as the repository cools, microbial activity will be limited due to the lack of nutrients. Thus, changes in porosity and permeability that could affect seepage and flow and the repository's performance, due to the growth of microbes and the production of exopolymeric material as the near field evolves, are unimportant and need not be considered.

The potential importance of changes of pore-water and gas chemistry to repository performance is that mineral precipitation and dissolution is controlled by the composition of fluids. Calcite precipitation and dissolution are directly controlled by the partial pressure of CO<sub>2</sub> in equilibrium with pore fluids in the unsaturated zone. Volatilization of CO<sub>2</sub> as water evaporates is expected to increase gas phase CO<sub>2</sub> concentration to many times that of the ambient conditions (Lichtner, 1997). This has been observed in the drift-scale test at Yucca Mountain (unpublished results of Lawrence Berkeley Laboratory and Lawrence Livermore National Laboratory). Ambient gas chemistry appears to be controlled by near surface microbial processes (Thorstenson, et al., 1998). The CO<sub>2</sub> concentration in the gas phase as the near-field environment evolves will be primarily controlled by THC processes as a result of the fast kinetics of these processes and the oligotrophic nature of the tuffs at Yucca Mountain.

Based on the groundwater studies, organic carbon is expected to be limiting for microbial growth and activity. Nevertheless, it needs to be recognized that unknown, but limited, quantities of xenobiotic carbon will be introduced into the emplacement drifts (Haldeman, et al., 1996; Horn and Meike, 1995) during the preclosure operation of the repository. These xenobiotic compounds will include diesel fuel, lubricating oil, and other organic compounds associated with the operation of machinery (Haldeman, et al., 1996; Hardin and Chestnut, 1997). Current site characterization operations within the Exploratory Studies Facility (Wilder, 1996) and the Enhanced Characterization of the Repository Block Cross Drift include a spill program (CRWMS M&O, 1998a,b) that requires spill minimization and clean-up activities for spillage of organic fluids. While there is a tremendous taxonomic diversity of bacteria capable of utilizing petroleum products as a carbon source, the limited quantity of water in the drifts (due to ventilation during the preclosure and radioactive decay heating in the postclosure) and the limited quantities of organic carbon added to the drifts should limit the potential importance of microbial activity to repository performance.

Additional insights on the potential importance of microbial processes in the near field can be derived from experiments designed to study the gross effects of microbial activity on repository geochemistry, radionuclide sorption, and integrity of repository and host rock materials (West, et al., 1998). Fermentation cells were loaded with carbon steel coupons, ion-exchange resins embedded in cement paste, cement-pumice composite blocks, and marl. The cells were then filled with fluids likely to be encountered in a potential Swiss repository and inoculated with a diverse mixture of microbes. The presence of microbes in these experiments did not appear to influence the overall geochemistry in any of the experiments, which was dominated by cement dissolution (West, et al., 1998). Geochemical modeling of the experiments could reproduce many of the observed features without explicitly invoking microbial activity, while discrepancies between the experiments and models were mostly explained by data or model limitations (West, et al., 1998).

Thus, changes in pore-water and gas chemistry due to microbial activity that could affect seepage and flow and repository performance will be insignificant relative to chemical changes expected as the result of THC processes and interaction of engineered materials with the surrounding tuff. Therefore, the effects of microbial processes on seepage and flow need not be considered in a performance assessment. Should the design of the repository change to one in which large quantities of organic carbon would be left in the emplacement drifts, the potential for microbial activity that could affect seepage and flow would need to be reevaluated.

#### **4.2 THE EFFECTS OF COUPLED THERMAL-HYDROLOGIC-CHEMICAL PROCESSES ON THE WASTE PACKAGE CHEMICAL ENVIRONMENT**

In Revision 2 of its Repository Safety Strategy (U.S. Department of Energy, 1998a), the DOE identified long waste package lifetime as a key factor in its safety strategy. In Revision 3 of the Repository Safety Strategy (CRWMS M&O, 2000a), DOE considered performance of the drip shield and performance of the waste package as two of the seven principal factors of the postclosure safety case. Other factors identified by DOE for the postclosure safety case, though given lower importance, include “environments on the drip shield” and “environments on the waste package.”

The model abstraction section of the Yucca Mountain Review Plan is divided into 14 different abstractions. These abstractions are equivalent to the integrated subissues in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a). Waste package lifetime is embodied in a performance assessment framework by the integrated subissues on mechanical disruption of engineered barriers, degradation of engineered barriers, and quantity and chemistry of water contacting waste packages and waste forms. The effects of coupled THC processes only influence the abstraction of the latter two integrated subissues. Degradation of the engineered barriers, which is addressed explicitly in the Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1998d, 1999d), depends strongly on the near-field chemical environment. Performance of the waste packages may also be affected by alternate engineered barrier subsystem design features. The ENFE KTI will help to evaluate the quantity and chemistry of water contacting the drip shields and waste packages. Contributions from the Container Life and Source Term, Repository Design and Thermal Mechanical Effects, Unsaturated and Saturated Flow under Isothermal Conditions, and Thermal Effects on Flow KTIs will be required, in addition to contributions from the ENFE KTI, to successfully abstract the two integrated subissues that are necessary to assess the corrosion of the drip shields and waste packages. DOE must adequately consider coupled THC processes affecting the drip shield and waste package chemical environment in its assessments of repository performance.

##### **4.2.1 Technical Bases for Reviewing the Effects of Thermal-Hydrologic-Chemical Processes on Waste Package Chemical Environment**

The technical bases for reviewing coupled THC effects on waste package chemical environment are given in this section. These bases are primarily focused on explaining why the results of coupled THC processes may be important to the drip shield and waste package chemical environment. Limited analyses in past performance assessments on the effects of coupled geochemical processes on the potential repository waste package environment have been completed. In addition, the relevance to performance of a particular coupled THC process is highly dependent on repository design (e.g., thermal-loading strategy and waste

package material). This dependence has resulted in focusing on ensuring that analyses of the effects are completed. The sophistication of the analysis of the effect of a coupled process on the waste package chemical and drip shield environment that could be conducted and found acceptable by staff depends on the extent to which DOE will rely on the barrier performance, information available at present, plans to obtain the additional information as part of the long-term testing program, and ability of codes to model coupled processes and determine their impact on repository performance.

Previous versions of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1998e, 1999f) reviewed the technical bases for earlier versions of the DOE performance assessments of Yucca Mountain. With this revision of the ENFE IRSR, the focus is on updating the technical bases to reflect the effects of the revised repository design features as they effect the waste package chemical environment through coupled THC processes. The Enhanced Design Alternative-II selected as the reference design by the DOE during the License Application Design Selection process (CRWMS M&O, 1999b) for TSPA-SR calls for a waste package with a corrosion resistant (Alloy 22) outer barrier and stainless steel inner barrier, optional backfill, a titanium drip shield, a steel invert with granular ballast, and steel ground support. Since the selection of the Enhanced Design Alternative II design, DOE has removed backfill from the reference design (CRWMS M&O, 2000b).

#### **4.2.1.1 Coupled Thermal-Hydrologic-Chemical Processes Affecting Waste Package Chemical Environment**

The modes and rates of corrosion of container materials are determined by their corrosion potentials in the near-field environment contacting them (Sagar, 1997). The corrosion potential is the potential difference between the container material and a nonpolarizable, reference electrode that is in contact through an electrolyte. The corrosion potential is established by the combination of oxidative and reductive reactions at the container material-solution interface. This potential is an electrochemical parameter that depends on environmental variables, such as temperature, pH, concentration of oxygen, and other reducible species. The potential also is dependent on the composition and surface conditions of the container material. Evaluation of the effects of corrosion potentials on waste package lifetime are treated in detail in the Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1999d). In the geophysical literature, another potential is noted, called the self-potential or Earth potential. It has been suggested that natural and spontaneous electrical potentials, known as self-potentials, may generate electrical currents that may affect the performance of the waste containers (Wilder, 1996). The components of the self-potential are (see Chapter 8 in Wilder, 1996):

- (1) **Streaming potential**: This is the potential difference observed due to the pressure differential between two points in the solution/electrolyte phase in continuous contact with a nonconducting solid phase (e.g., rock). The magnitude of the streaming potential decreases with an increase in electrolyte concentration (Newman, 1991) and a decrease in pore radius. In the presence of a conductive solid, the streaming potential decreases through rearrangement of electronic structure within the solid, and a streaming current results.

- (2) Thermoelectric potential: In metallic corrosion literature, thermoelectric potentials are referred to as thermogalvanic potentials. The thermogalvanic potentials consist of thermal diffusion potential (Soret effect) and Nernstian potential due to activity differences at different temperatures.

In contrast to the corrosion potential, the self-potential is established in the electrolyte phase only.

Large self-potentials (greater than 500 millivolts) have been measured during the single-heater test within the Exploratory Studies Facility at Yucca Mountain. The measurements were reported to have been made using the same electrodes that were used for conductivity measurements and were spaced approximately 1 meter apart. Details regarding the input impedance of the voltmeter and the electrode materials are not available at this time. The potential difference measured above the heater element was about 100 millivolts and below the heater was about 800 millivolts. The conductivity measurements indicated that saturation may have increased below the heater. The measured values of self-potentials may be subject to large errors due to (i) the use of polarizable electrodes designed for conductivity measurements, not for self-potential measurements, (ii) the possible use of low-impedance voltmeters that will introduce significant polarization of the electrodes, (iii) the presence of conductive materials, (iv) the discontinuity in the electrolyte layer between measuring points, especially in a variably saturated medium, (v) the contact resistances, and (vi) the lack of filtering of telluric and other sources of noise introduced by the presence of metallic heater elements. These sources of errors have been discussed in geophysical literature (e.g., Corwin and Hoover, 1979) as well as electrochemical literature (e.g., Bard and Faulkner, 1980). For example, assuming the electrode materials were made of platinum typically used in conductivity measurements, the polarization of the electrodes for the oxygen reduction reaction can, by itself, cause a potential difference of over 500 millivolts (Hoare, 1967). Further information is needed regarding the details of the measurement to assess the accuracy of self-potential measurements made at the Exploratory Studies Facility. An accurate measurement of self-potentials would entail using nonpolarizable electrodes (e.g., silver/silver chloride or tungsten/tungsten oxide electrodes), high impedance voltmeter (with an input impedance of at least 1 gigaohm), and signal processing to reduce other sources of noise.

Self-potential is a potential gradient along the solution path, whereas, the corrosion potential is the potential difference across the metal-solution interface. Furthermore, corrosion potentials calculated for container performance explicitly consider the relevant charge transfer reaction equilibrium and kinetics at each spatial location (Mohanty, et al., 1997). Therefore, measured self-potentials should have no effect on waste package performance. If during the performance confirmation period, however, the corrosion potential of the containers is monitored using reference electrodes located at various distances from the container surface, the measured value may be affected by self-potentials. In such a case, an accurate survey of self-potentials at spatial locations relevant to monitoring electrodes should be made. The experimental precautions for accurate measurement of self-potentials mentioned previously apply to this survey.

The ability to calculate moisture redistribution at the drift scale is essential to determine how fast the waste package will corrode. The time period at which the relative humidity exceeds a critical value,  $RH_c$ , is an important factor in determining the container performance. When the relative humidity is less than the  $RH_c$ , corrosion of the titanium drip shield and the Alloy 22

waste package will be negligible. Dry oxidation of the engineered barriers may occur, but, the rate of oxidation is expected to be slow for the temperatures specified in the Enhanced Design Alternative-II design. At relative humidity  $> RH_c$ , corrosion of the titanium drip shield and the Alloy 22 barrier will occur. The type of corrosion and the corrosion rate will depend on the condition of the material (i.e., residual stresses) and the chemistry of the water contacting these engineered barriers. The time at which rewetting of the containers occurs depends on the near-field environment. At this scale, the geometry of the individual waste package becomes important, unlike the repository-scale model in which the waste is assumed to be distributed uniformly. There have been several attempts to model drift-scale moisture distribution (Nitao, 1988; Pruess, Wang, and Tsang, 1990). The problem is difficult because of the variation in scale within the computation domain. Symmetric boundary conditions are usually imposed, implying an infinite array of evenly spaced waste packages. Thermohydrologic calculation of moisture redistribution is treated specifically in the Thermal Effects of Flow IRSR (U.S. Nuclear Regulatory Commission, 1998a, 1999c). Coupled THC effects on moisture redistribution have been noted to induce large effects, such as changes in permeability and porosity (e.g., Hardin, 1998). In general, these effects have been neglected in the thermohydrologic assessments used for Yucca Mountain performance calculations. Also, THC processes will affect the chemistry of gas and water that would interact with the waste package.

Elevated temperatures in the near field at Yucca Mountain are expected to lead to important geochemical changes. For the Viability Assessment waste package design, oxidation of the carbon steel outer overpack under dry conditions would be controlled by the waste package temperature and the composition of the gaseous phase. In particular, the oxygen partial pressure would be important for the oxidation of the carbon steel outer barrier. Localized reducing conditions could be promoted by near-field hydrologic effects and phase variations. For the Enhanced Design Alternative-II design, oxidation of the titanium drip shield and the Alloy 22 waste package would be expected to occur at a very slow rate and not result in significant consumption of gaseous species by these engineered barriers. Initial gas flow from the near field, driven by vaporization of water, was predicted to be away from the near field in all directions (Pruess, Wang, and Tsang, 1990; Tsang and Pruess, 1987). This process would tend to purge air containing  $O_2$  and  $CO_2$  from the near-field environment. The vapor pressure of water at temperatures above  $95\text{ }^\circ\text{C}$  exceeds the hydrostatic pressure of less than 0.1 megapascal at Yucca Mountain. Thus, the gas phase in the near field would tend to be dominated by  $H_2O$  under these conditions. Diffusion of air toward zones of relatively high water vapor pressure could reintroduce oxygen to the near field (Tsang and Pruess, 1987). Estimates of the temporal extent of this period of reduced air mass fraction, based on thermal-hydrologic modeling, range from hundreds of years to a few thousand years, depending on thermal loading of the repository (Wilder, 1996; Lichtner, Pabalan, and Steefel, 1997).

Aqueous corrosion of steel sets used as ground support in the repository near field can create a local decrease in Eh and in the corrosion potential. Provided the air mass fraction in the near field remains high, the extent of the reduced zone may be small for various reasons. First, electrochemical reduction of oxygen on iron surfaces is irreversible (far from equilibrium) and is diffusion-limited in the aqueous phase. Second, corrosion of iron in an oxidizing environment leads to the formation of iron oxides and oxyhydroxides that can further decrease the rate of electrochemical reduction of  $O_2$ . Finally, the initial formation of  $\eta\text{-FeOOH}$  can lead to a secondary reduction reaction with  $Fe_3O_4$ . This last process should occur for alternating wet and dry environments (Nishikata, et al., 1994). During the dry period,  $Fe^{2+}$  oxides or oxyhydroxides

are oxidized by air to  $\eta$ -FeOOH, and the cyclic process proceeds because of the electronic conductivity of the inner layer of  $\text{Fe}_3\text{O}_4$ .

#### 4.2.1.2 Effects of Waste Package Corrosion Processes on Waste Package Chemical Environment

Corrosion products could have a significant effect on the near-field geochemical environment, which in turn, may affect the nature of ongoing corrosion processes. For example, corrosion of the outer steel overpack of the waste package considered in the TSPA-VA (U.S. Department of Energy, 1998b) might have led to locally reducing conditions in the near field and strong gradients in oxidation potential, despite the prevailing oxidizing nature of the geologic setting (Murphy and Pabalan, 1994). In this example, the corrosion rate of the outer steel overpack in the Viability Assessment waste package design would then adjust to these changes.

Recent changes to the proposed repository design may reduce the uncertainties associated with the performance of the waste package. These changes include the elimination of both the carbon steel outer overpack and the concrete lining in the drift. The Enhanced Design Alternative-II repository design specifies steel sets as ground support, a titanium-alloy drip shield, a corrosion resistant Alloy 22 waste package outer barrier, and a type 316 NG (nuclear grade) stainless steel inner barrier. Geochemical parameters that affect the performance of the steel sets used as ground support, via uniform passive or localized corrosion, stress corrosion cracking, and hydrogen embrittlement, include pH, Eh, temperature,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{HCO}_3^-/\text{CO}_3^{2-}$  ratio. For the steel sets used as ground support, alkaline conditions (pH ranging from 8 to 11) provide an environment that results in the formation of a tightly-adhering, passive film, thought to be  $\eta$ - $\text{Fe}_2\text{O}_3$  on carbon steel. This passive film slows down uniform corrosion rates by several orders of magnitude, but makes the metal more prone to the localized breakdown of passivity leading to the occurrence of pitting or crevice corrosion. Should the pH of the water contacting steel sets be neutral (pH = 7) or acidic, then active uniform corrosion will occur. Stress corrosion cracking can occur in a  $\text{HCO}_3^-/\text{CO}_3^{2-}$  environment at a pH of about 8 when the corrosion potential (related to the Eh of the environment) reaches a critical value and when the total carbonate concentration is above a certain minimum value.

Corrosion of the drip shields and the waste packages will occur after the formation of water on the surfaces of these engineered barriers as a result of either condensation or dripping. The type of corrosion will be dependent on the chemistry of the water in contact with the metal surfaces. Materials selected for the drip shield (titanium grade 7) and the waste package outer barrier (Alloy 22) are designed to be resistant to localized corrosion. In the absence of localized attack, the degradation of these engineered barriers will occur at a slow rate determined by the passive current density. The localized corrosion of the titanium drip shield and the Alloy 22 waste package outer barrier may result in rapid penetration of the engineered barriers and the possible release of radionuclides. Breakdown of the passive film on titanium grade 7 has been shown dependent on the concentration of fluoride (Brossia and Cragolino, 2000). Localized corrosion of Alloy 22 is possible in oxidizing environments with high chloride concentrations (Dunn, Pan, and Cragolino, 2000). Elevated chloride and fluoride concentrations have been reported in laboratory tests designed to simulate the effect of evaporation on the chemistry of the environments on the drip shields and waste packages (TRW Environmental Safety Systems, Inc., 1999).

If penetration of the Alloy 22 waste package outer barrier occurs by localized dissolution, further local acidification may result from hydrolysis of  $\text{Cr}^{3+}$ . Experimental evidence for acidification within crevices of nickel-chromium-molybdenum alloys has been documented (Cavanaugh, et al., 1983; Sridhar and Dunn, 1994). At higher temperatures, crevice pH lower than 1 may be found in cracks or pits. It has been shown that the presence of molybdenum, which is added to increase the corrosion resistance of these alloys, can decrease the pH further, depending on the potential inside the crevice. Increased local acidification and chloride concentration may result in the rapid initiation and penetration of the Type 316 NG stainless steel waste package inner barrier.

Thus, depending on the rate of movement and dripping of water into the container, the water in immediate contact with the drip shield and the waste package outer barrier may become acidic. Several factors may decrease the corrosion potential established at the drip shield and the waste package outer barrier surface below the repassivation potential for localized corrosion, precluding the occurrence of this phenomenon and, therefore, the generation of acidic conditions.

#### **4.2.1.3 Effects of Cementitious Materials on Waste Package Chemical Environment**

Cement is used primarily for its structural (e.g., high compressive strength) and physical (e.g., low permeability) properties. Cements are extremely fine-grained, high-surface area materials containing somewhat soluble and thermodynamically metastable phases that are unstable with respect to crystalline cement phases. These properties and the partially interconnected pore network of the solids make these materials potentially reactive with the near-field environment and the engineered barrier system.

Cementitious materials may cause changes in near-field chemistry through the time period of regulatory interest, particularly if seepage occurs preferentially along cement-grouted rock bolts. Interactions with cementitious materials will tend to keep the aqueous carbonate concentrations low. A study by Atkinson, Everitt, and Guppy (1989) indicated that interaction of groundwater typical of a clay environment with cement could maintain a pH above 10.5 for a few hundred thousand years, using the low flow rates assumed in that study. Results of these types of studies are highly dependent on the assumptions used in the calculations, such as groundwater flow rates, amount of cementitious materials present in the repository, and stability of the calcium silicate hydrate gel. Simple extrapolation of results from experiments using laboratory-aged cement pastes is likely to be invalid because the solid and aqueous chemistry of cements will change considerably within the relevant time frame (10 to 10,000 years), even in a closed system (Atkins, Glasser, and Kindness, 1991). For example, Atkinson, Everitt, and Guppy (1989) indicated that, if recrystallization of the calcium silicate hydrate gel occurred in the long term, lower pH could result because of the lower solubility of the crystalline calcium silicate hydrate phases. Formation of crystalline calcium silicate hydrate phases by recrystallization of preexisting calcium silicate hydrate gel is likely in a high-level waste repository, because of the long time frame involved and the elevated temperatures imposed by radioactive decay heat from emplaced nuclear wastes. Even a modest temperature excursion to 55 °C for 6 to 12 months can result in partial transformation of the calcium silicate hydrate gel to more stable, though poorly crystallized, phases, such as jennite and tobermorite (Atkins, Damidot, and Glasser, 1994). Thus, modeling of cement interactions with the near-field

environment and its potential effect on waste package lifetimes must consider the likelihood that cement chemistry is dominated by phases other than those present in the initial material because the dominant phases control the long-term evolution of solution pH, a key parameter for container corrosion. Although a number of simulations of the evolution of cement-pore fluid and some simulations of groundwater-cement interactions have been conducted using estimated data (Glasser, Macphee, and Lachowski, 1987; Atkinson, Everitt, and Guppy, 1989; Reardon, 1992; Lichtner and Eikenberg, 1995; Neall, 1996), most of these simulations were conducted for 25 °C and assumed the presence of amorphous CSH gel. Thus, the results may not be relevant to cement-water interactions in a high-level waste repository.

In the TSPA-VA (U.S. Department of Energy, 1998b) design, the use of cementitious materials, in the form of concrete inverts and linings, was considered for the estimated 179 kilometers of emplacement drifts of the proposed Yucca Mountain high-level waste repository, in addition to the planned use of cement in roadways for construction and emplacement ramps and service mains. In the TSPA-SR design, cementitious materials are not planned for ground support, except in the form of grout to anchor the rock bolts of the ground support system. The potential effect of cementitious materials on the chemical environment for waste package and drip shield corrosion in the TSPA-SR design is expected to be substantially reduced compared to the TSPA-VA (U.S. Department of Energy, 1998b) design. Nevertheless, standard spacing of rock bolts could result in grouted rock bolts occurring above each waste package and drip shield in certain rock units (CRWMS M&O, 2000d). Thus, the effects of cementitious materials on the waste package and drip shield environment still needs to be addressed in a performance assessment of these barriers. The degree to which cementitious materials can affect the chemical environment for waste package and drip shield corrosion will depend to a large part on materials inventory, their location, and the reactivity with groundwater of the specific cement material formulation used.

#### **4.2.1.4 Microbial Effects on Waste Package Chemical Environment**

The potential importance of microbial processes to nuclear waste repositories has been recognized internationally for more than 10 years, and microbial influenced corrosion of the waste package has been one of the primary concerns (Pedersen and Karlsson, 1995; Stroes-Gascoyne, 1996; Christofi and Philp, 1997). One experiment was explicitly designed to study the gross effects of microbial activity on repository geochemistry, radionuclide sorption, and integrity of repository and host rock materials (West, et al., 1998). This study demonstrated that only localized corrosion of carbon steel could be ascribed to microbial processes. The high-level waste program has also realized the potential importance of microbial processes on corrosion of waste packages (Geesey, 1993; Horn and Meike, 1995; CRWMS M&O, 1997b; TRW Environmental Safety Systems, Inc., 1997b). Microbial influenced corrosion is also addressed in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d, 1999d).

The conditions necessary for microbial activity in the near field were discussed in Section 4.1.1.3. Only those aspects that differ and are pertinent to the chemical environment of the waste package and drip shield are presented here. The amount of nutrients introduced into the repository drifts, which will be potentially available for microbial growth, will be a strong function of both the repository design and the concepts of operation (Wilder, 1996). The potential for microbial growth may be increased greatly if backfill is used. Increased nutrient loading would occur by introduction of the backfill (Stroes-Gascoyne and Gascoyne, 1998; Wilder, 1996).

Microbial activity in the tuff system has been demonstrated to be water and organic carbon limited (Kieft, et al., 1997). Thus, if spills of organic fluids are not minimized, the potential for microbial activity would increase. One additional control on the potential for microbial activity within the emplacement drifts will be temperature and, consequently, the relative humidity (Stroes-Gascoyne, et al., 1996; CRWMS M&O, 1997b). For instance, as long as the temperature is above 100 °C or the relative humidity is less than about 60 to 70 percent, then microbial activity will not occur (Stroes-Gascoyne, 1996; Geesey, 1993). It is likely that the probability of growth of microbes will vary as a function of temperature (CRWMS M&O, 1997b).

While microbial activity in the volcanic tuffs will be limited by a lack of nutrients, additional nutrients may be available within the engineered barrier system. An analysis of the supply nutrients and energy-producing reactions, which would consume the nutrients, is warranted. A mass balance type of model could be used to determine the potential for microbial activity (Grogan and McKinley, 1990). Without a mass balance for the repository, it is unknown whether organisms isolated and grown under laboratory conditions will even grow under repository conditions. A mass balance inventory approach will establish the limiting conditions for microbial growth. It will also establish the potential for microbial growth and the potential for microbial induced corrosion (CRWMS M&O, 1997b). An evaluation of energy-producing reactions combined with the material inventory will provide a conservative estimate of the maximum microbial growth that can be sustained in the repository (McKinley, et al., 1997). This mass balance approach has been tested by comparing predictions from natural analogues with field observations (West, 1995; West, et al., 1995). Three countries (Switzerland, Canada, and United Kingdom) have applied this extremely conservative (West, 1995; McKinley, et al., 1997) approach to their high-level waste repositories. Each country has demonstrated that microbial activity is not important to repository performance (McKinley, West, and Grogan, 1985; McKinley and Hagenlocher, 1993; West, 1995; McKinley, et al., 1997).

If the mass balance results indicated that microbial activity was likely for Yucca Mountain, then the potential for microbial induced corrosion would need to be assessed. An assessment of microbial induced corrosion was attempted in the DOE waste package expert elicitation (CRWMS M&O, 1997b). Microbial effects on the in-drift geochemical environment will be considered in the TSPA-SR.

#### **4.3 THE EFFECTS OF COUPLED THERMAL-HYDROLOGIC-CHEMICAL PROCESSES ON THE CHEMICAL ENVIRONMENT FOR RADIONUCLIDE RELEASE**

In Revision 2 of its Repository Safety Strategy (U.S. Department of Energy, 1998a), DOE has identified slow rate of release of radionuclides from the waste form as a key attribute of the repository system that would help its objective of acceptably low annual doses to a person living near the site (U.S. Department of Energy, 1998a). In Revision 3 of the Repository Safety Strategy (CRWMS M&O, 2000a), the DOE considered solubility limits of dissolved radionuclides as one of the seven principal factors of the postclosure safety case. Other factors identified by DOE for the postclosure safety case, though given lower importance, include (i) performance of commercial spent nuclear fuel waste form, (ii) performance of DOE-owned spent nuclear fuel, Navy fuel, and plutonium disposition waste form, and (iii) performance of defense high-level waste form. Two of the fourteen integrated subissues NRC has determined that need to be abstracted into a performance assessment are (i) quantity and chemistry of

water contacting waste packages and waste forms, and (ii) radionuclide release rates and solubility limits. Radionuclide release from waste forms and from the engineered barrier system will depend on the chemical environment, which will be conditioned by coupled THC processes. The abstraction of the quantity and chemistry of water contacting waste packages and waste form incorporates THC processes that will determine the chemical environment for radionuclide release. Contributions from the Container Life and Source Term, Thermal Effects on Flow, and Unsaturated and Saturated Flow under Isothermal Conditions KTIs also will be required to resolve this integrated subissue. DOE must adequately evaluate the chemical environment for radionuclide release and appropriately consider coupled THC processes affecting this chemical environment in its assessment of repository performance.

#### **4.3.1 Technical Bases for Reviewing the Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Chemical Environment for Radionuclide Release**

The bases are primarily focused on explaining why the effects of coupled THC processes may be important to the chemical environment controlling radionuclide release from the waste form and engineered barrier system. As mentioned previously in Section 3, limited analyses in past performance assessments on the effects of coupled THC processes on radionuclide release have been completed. In addition, the relevance to performance of a particular coupled THC process is highly dependent on repository design (e.g., thermal-loading strategy, waste package material, etc.). This has resulted in acceptance criteria that primarily focus on ensuring that analyses of THC effects are completed. The sophistication of the analysis of the effect of coupled processes on the chemical environment for release that could be conducted and found acceptable by staff depends on the credit to which DOE will ascribe to this barrier, the information available at the present, any plans to obtain additional information as part of the long-term testing program, and the ability of codes to model coupled processes and determine their impact on repository performance.

Previous versions of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1998e, 1999f) have addressed the technical bases for earlier versions of the DOE performance assessments of Yucca Mountain. This revision of the ENFE IRSR focuses on preserving the previous evaluations and updating the technical bases for the review methods and acceptance criteria. This revision also describes how the current repository design features will effect coupled THC processes and their consequences on the chemical environment for radionuclide release. For example, the Enhanced Design Alternative-II selected by the DOE during the License Application Design Selection process (CRWMS M&O, 1999b) for use in the TSPA-SR (CRWMS M&O, 1999a). This new design calls for the use of backfill—later removed from consideration, a titanium drip shield, a steel invert with granular ballast, steel ground support, and a waste package with an outer corrosion-resistant material of Alloy 22 and an inner stainless steel structural material. Interaction of fluids with these engineered materials or their degradation products could substantially affect the chemical environment for radionuclide release. For instance, acidic pH solutions may result from oxidation and hydrolysis of chromium contained in stainless steel, or high pH solutions may be generated due to interaction of water with cementitious grout material used to anchor the rock bolts of the ground support system. The altered pH may significantly increase the solubility and the rate of release of radionuclides. The DOE assessment of this impact would be acceptable if it demonstrated using geochemical equilibrium modeling codes, such as EQ3 (Wolery, 1992) or others, that the solubility range

chosen for its reference case performance assessment was conservative relative to the results of the equilibrium calculations.

#### 4.3.1.1 Coupled Thermal-Hydrologic-Chemical Processes Affecting Spent Fuel, Cladding, and Borosilicate Glass Degradation

Near-field environmental factors, including Eh, pH, temperature,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{HCO}_3^-$ , and  $\text{F}^-$ , are governed by near-field THC processes. These factors affect degradation modes of spent fuel and its Zircaloy cladding. Zirconium alloys are susceptible to a form of hydrogen embrittlement called delayed hydride cracking. This phenomenon is promoted by the precipitation of brittle zirconium hydrides ( $\text{ZrH}_{2-x}$ ) in areas of stress concentrations upon cooling from high temperature (Cox, 1990). Slow cooling may induce reorientation of plate-like hydrides into an axial rather than circumferential distribution, facilitating failure (Chan, 1996). Although cladding creep at moderate temperatures is not dependent on environmental factors, it is considered a plausible mode of failure (Santanan, Raghavan, and Chin, 1992). Above a certain critical potential, Zircaloy is susceptible to pitting corrosion in chloride-containing environments (Cragolino and Galvele, 1978). Such a potential can be attained naturally under slightly oxidizing conditions (i.e., in the presence of  $\text{Fe}^{3+}$ ). Under the environmental and potential conditions leading to pitting, stress-corrosion cracking of zirconium and Zircaloy occurs in the presence of an applied stress (Cox, 1990). Whereas a decrease in Eh protects the fuel cladding from localized corrosion and stress-corrosion cracking, it can promote failure by delayed hydride cracking. Despite its relatively low concentration, the presence of the fluoride anion in the environment may increase the uniform dissolution of zirconium alloy. The increase in dissolution is the result of the greater stability of the  $\text{ZrF}_6^{2-}$  complexes compared to that of the passive  $\text{ZrO}_2$  film.

Corrosion of spent fuel (predominantly  $\text{UO}_2$ ) by contact with the groundwater is the most important process affecting the long-term performance of this waste form. The groundwater will be modified by chemical and physical interactions in the near field. A major factor determined by the near-field environment is the redox potential or Eh. Eh generally increases by gamma- or alpha-radiolysis. The corrosion rate of spent fuel is determined by the corrosion potential  $E_{\text{corr}}$ .  $\text{UO}_{2+x}$  is a relatively good electronic conductor because of its deviation from stoichiometry. As a result,  $E_{\text{corr}}$  is a well-defined electrochemical parameter for spent fuel immersed in an aqueous environment. The rates of reduction of species, such as  $\text{O}_2$  and  $\text{H}_2\text{O}_2$ , are coupled to the rate of oxidation of  $\text{UO}_{2+x}$ , establishing  $E_{\text{corr}}$  as a mixed potential on the interface between the oxide and solution (Shoesmith, et al., 1989). The effect of the potential is important due to the oxidative nature of the dissolution of  $\text{UO}_2$ .

The pH has an effect on the rate of dissolution of spent fuel that depends on the pH range. Under oxidizing conditions, only a slight dependence of corrosion rate on pH has been observed at pH values lower than 4. At pH values between 4 and 8, the rate of dissolution decreases linearly with pH (Grambow, 1989). At higher pH values, the rate of dissolution seems to be unaffected by pH changes. As in the case of other metals, valuable information can be compiled in terms of Eh-pH diagrams for the uranium- $\text{H}_2\text{O}$  system in the presence of certain anions (Paquette and Lemire, 1981). Specific domains for the dominant degradation modes can be superimposed onto to the Eh-pH diagrams. Temperature increases the rate of dissolution of  $\text{UO}_2$ , although the functional dependence is not well established over a wide range of temperatures.

The nature of the anionic species present in the groundwater and their concentrations are extremely important in determining the rate of corrosion of spent fuel. Anions such as  $\text{CO}_3^{2-}$ , that form stable soluble complexes with  $\text{U}^{6+}$  cations, substantially increase the rate of oxidative dissolution (Blesa, Morando, and Regazzoni, 1994). At low  $\text{CO}_3^{2-}$  concentrations (0.001 molar), the rate of dissolution is proportional to the total concentration (Blesa, Morando, and Regazzoni, 1994). At intermediate concentrations of  $\text{CO}_3^{2-}$  (0.5 molar), the dissolution rate depends on the square root of the total concentration (Grambow, 1989). At a higher  $\text{CO}_3^{2-}$  concentration (1.0 molar at 100 °C), the corrosion rate reaches a constant value. Finally, at even higher concentrations, the corrosion rate decreases, probably due to the formation of surface films (Needes, Nicol, and Finkelstein, 1975). These concentrations, while high for the nominal water composition, may occur due to evaporative processes in the near field. Corrosion is accelerated by anions in the sequence  $\text{Cl}^- < \text{PO}_4^{3-} < \text{SO}_4^{2-} < \text{F}^- < \text{CO}_3^{2-}$ . In the case of  $\text{PO}_4^{3-}$  and  $\text{SO}_4^{2-}$ , a maximum in the corrosion rate is observed at intermediate concentrations (about  $1.5 \times 10^{-2}$  molar) (Blesa, Morando, and Regazzoni, 1994).

Other species, such as  $\text{SiO}_2(\text{aq})$ ,  $\text{H}_3\text{SiO}_4^-$ , and  $\text{H}_2\text{SiO}_4^{2-}$ , can react with  $\text{U}^{6+}$  to precipitate complex uranyl silicates. These secondary minerals may tend to reduce the corrosion rates and exposure of fresh surface by forming a protective layer over the spent fuel. Under certain circumstances, acceleration of spent fuel dissolution can occur as a result of spallation of the alteration layers.

Rapid increases in the concentration of spent fuel dissolution products may lead to saturation of the medium with secondary alteration products. This accelerated precipitation of secondary phases could eventually preferentially release certain radionuclides. Bates, et al. (1995) found that intermittent additions of controlled amounts of groundwater to spent fuel led to precipitation of most of the transuranic elements (Am, Cm, and Pu). Neptunium was not precipitated and remained in solution. Wilson (1990) conducted semi-static experiments (i.e., involving periodic removal of a leachant aliquot and replacement with fresh solution). Wilson (1990) observed that actinide (U, Pu, Am, Cm, and Np) concentrations reached constant values rapidly. This observation suggests that steady-state conditions between spent fuel dissolution and secondary-phase formation are established. Formation of  $\text{U}^{6+}$  secondary phases, such as uranophane, was confirmed. Actinide concentrations, with the exception of neptunium, measured at 85 °C were lower than at 25 °C. This suggests that the solubility limiting phases are formed more rapidly at the higher temperature. Alternatively, this effect could be the consequence of the retrograde solubility of secondary products. The presence of plutonium, americium, and curium as colloids in the leachates was reported, but the formation of precipitated secondary phases predominated at 85 °C.

Under oxidizing conditions and in the presence of carbonate anions, there is a large driving force for the dissolution of the  $\text{UO}_2$  matrix. Soluble radionuclides, such as Cs-137, Sr-90, and Sb-125, exhibited congruent dissolution from spent fuel in flow-through tests. The release rate of these fission products decreased with time to a steady-state value similar to the release rate of uranium from the  $\text{UO}_2$  matrix (Gray, Leider, and Steward, 1992). In semi-static tests, the fractional release of Sr-90, Cs-137, I-129, and Tc-99 increased with temperature, and almost linearly with time. Species such as  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{H}_3\text{SiO}_4^-$ ,  $\text{H}_2\text{SiO}_4^{2-}$ , and  $\text{CO}_3^{2-}$  precipitated from solution in tests conducted at higher temperatures. Bates, et al. (1995) suggested that the corrosion rate of the matrix and release of radionuclides are accelerated in unsaturated tests compared to those under semi-static conditions. This situation leads to incongruent release of individual fission products and actinides, probably controlled by the formation of particulates in

solution. In addition, the corrosion rate, and especially the rate of radionuclide release, depend on the characteristics of the spent fuel (e.g., composition, degree of burnup). Pre-oxidation of the fuel was not considered to be a factor in the acceleration of the dissolution rate (Bates, et al., 1995). However, the modification of the pH of the leachate, attributed to the formation of HNO<sub>3</sub> by alpha-radiolysis of humid air as well as the generation of formate and oxalate from inorganic carbon, may raise the solubility of actinides (Finn, et al., 1994a). All these effects are postulated to become even more important at relatively large surface area-to-groundwater volume ratios such as may be expected in the unsaturated zone at Yucca Mountain. However, acid generating processes may be counteracted by alkalinity deriving from cement-water interactions.

Through interactions with oxidizing components, including radiolytic products, spent fuel will eventually oxidize and form a large quantity of UO<sub>2</sub><sup>2+</sup>-bearing solids. Natural analog (Pearcy, et al., 1994) and experimental (Wronkiewicz, et al., 1992) studies indicate that schoepite, soddyite, and uranophane are among the secondary minerals likely to form from spent fuel oxidation. Furthermore, these studies indicate that rates of oxidation of reduced uraninite and unirradiated fuel (both analogs of spent fuel) are rapid relative to transport of uranium away from the natural geologic setting or the experimentally simulated waste package, respectively. Therefore, secondary oxidation products will accumulate and uranyl minerals will have a large effect on near-field physical and chemical conditions.

Secondary uranium phases are likely to have several important effects on the near-field environment. First, physical disruption of structural components (e.g., cladding or degraded containers), due to the large volume increase accompanying oxidation and hydration of UO<sub>2</sub>, may occur. Second, both porosity and permeability could be reduced because of the volume expansion. Third, neptunium, plutonium, and other radioactive waste species may be incorporated into secondary uranium phases by coprecipitation. Fourth, the secondary uranium phases may limit ingress of water and oxidants to unaltered wastes. Finally, the secondary uranium phases may control the solubility or dissolution rate of spent fuel. Thus, secondary uranium phases may control the source term for radionuclide (not just uranium) releases from the breached waste packages. With regard to long-term performance of the proposed repository, secondary alteration products resulting from interactions of spent fuel with the near-field environment, rather than unaltered spent fuel, will likely control releases of many radionuclides from the engineered barrier system.

Experimental (Holland and Brush, 1980) and theoretical (Murphy, 1997) studies indicate that the solubilities of uranyl minerals, such as schoepite and uranophane, are retrograde with temperature. The emplacement horizon will reach its maximum temperature shortly after waste emplacement (e.g., within tens or hundreds of years). Thereafter, it will experience an environment of continuously decreasing temperature. Consequently, the solubilities of alteration products of spent fuel will tend to increase with time. In contrast, through a process of Ostwald ripening, increasingly stable secondary phases, with lower solubilities, will crystallize.

In DOE and NRC performance assessments, solubility is invoked as a conservative upper limit to the radionuclide concentration in aqueous solution. In principle, mineral solubilities and radionuclide concentrations can be calculated based on thermodynamic data and the chemical characteristics of the system, i.e., temperature, pressure, and composition. In practice, maximum radionuclide concentrations are further complicated by metastability of solid phases,

metastable supersaturation, solid solution (coprecipitation), colloid stability, oxidation-reduction disequilibria, disequilibrium dissolution of source material, sorption, etc. The gross disequilibrium between spent nuclear fuel and the Yucca Mountain geochemical environment creates a strong potential to generate metastable conditions including solution concentrations in excess of minimum equilibrium solubilities. Consequently, radionuclide solubilities used in performance assessments have tended to be uncertain (i.e., sampled over a large range) and conservative (i.e., high relative to the most thermodynamically stable possibilities). A comparison of solubilities used in the DOE TSPA-VA to calculated solubilities based on uncertain thermodynamic data for several radionuclide bearing solids and a variety of possible solution compositions shows that the TSPA-VA solubility values tend to meet the criterion of conservatism.

One approach in experimental determination of radionuclide solubilities for performance assessment that will not underestimate solubilities is to approach steady-state concentrations from a state of supersaturation (as well as from undersaturation). This approach was advocated in the NRC position paper on solubilities (U.S. Nuclear Regulatory Commission, 1984) and followed in experimental studies conducted by Nitsche, et al. (1993, 1994) for neptunium solubilities. Resulting neptunium solubilities are high and contribute to high neptunium contributions in estimated doses in performance assessments.

Reducing the values of neptunium solubility in performance assessments from the empirical values used in previous performance assessment iterations requires substantial justification. The solid phase proposed to control lower solubilities should be shown by empirical and theoretical means to form and to remain stable, e.g., by reversal of the stability of this solid phase from undersaturated and supersaturated conditions and by thermodynamic calculations of its stability for expected environmental conditions. Also, measurements of the distribution of the radionuclide between the solid phase and the aqueous phase should be made and used together with an evaluation of anticipated aqueous speciation to determine radionuclide concentrations. Empirical measurements of concentrations under a particular set of experimental conditions provide weak justification for maximum concentration limits in performance assessments because relevant repository conditions will be different and will vary.

Coprecipitation of a minor species in a phase composed predominantly of other species does not lead to maximum concentration limits in the same sense that solubility of a stoichiometric phase limits the concentrations of species in solution. A strong potential exists for a small amount of substitution of a minor or trace species. This potential may be reduced as the amount of substitution increases for thermodynamic reasons (entropy and enthalpy of mixing) related to structural (crystallographic) effects. Nevertheless, concentrations of trace elements may be severely limited by coprecipitation. Concentrations of many trace constituents in natural environmental waters are limited by sorption and coprecipitation (solid solution) phenomena.

The likelihood of limitation of radionuclide concentrations by coprecipitation with uranyl phases in a Yucca Mountain repository is indicated by several lines of evidence: (i) the abundance of uranium in the spent nuclear fuel waste form, (ii) the wealth of experimental, theoretical, and natural analog data indicating that relatively stable uranyl minerals will form on oxidation of uranium in the Yucca Mountain environment, and (iii) experimental and theoretical crystallographic indications of coprecipitation of minor radionuclides in secondary uranyl minerals.

However, data for the equilibrium solubilities (thermodynamic properties) of uranyl phases are uncertain, and experimental data for the distribution of radionuclides between aqueous solution and the structures of these phases are sparse.

The second main waste form planned for the proposed repository at Yucca Mountain is borosilicate glass. Environmental factors affecting the general or localized dissolution rate of borosilicate glasses include Eh, pH, temperature,  $\text{Cl}^-$ ,  $\text{HCO}_3^-$ ,  $\text{H}_3\text{SiO}_4^-$ ,  $\text{F}^-$ , and  $\text{Fe}^{2+}$ . As in the case of metals, the interrelationship of Eh and pH on the dissolution of waste glass can be displayed in a potential-versus-pH diagram (Jantzen, 1992). In general, Eh has practically no effect on the dissolution of the glass matrix. Silicon, boron, and aluminum, which are the principal network formers of borosilicate glasses, do not undergo changes in oxidation state within the range of Eh values expected under repository conditions. The effect of pH is far more important. The rate of dissolution is strongly accelerated at alkaline pH due to matrix dissolution. At pH lower than 4, the rate is accelerated by diffusion-controlled hydrogen ion exchange for alkali ions. Many anions have a minor effect on the solubility and rate of dissolution of borosilicate glasses. However, fluoride accelerates the dissolution substantially through the formation of  $\text{SiF}_6^{2-}$  complexes. The value of relative humidity is important in the durability of glasses in humid air. Glasses are also susceptible to environmentally assisted cracking in aqueous environments (McCauley, 1995). The effect of this phenomenon on radionuclide releases may be far less important than that associated with generalized dissolution.

Alteration of glass depends primarily on the activity of aqueous silica. In the ambient geochemical environment, and for predicted geochemical conditions in the host rock, the aqueous silica concentration is large (Yang, 1992; Yang, et al., 1996). A glass waste form would be expected to be fairly unreactive for these conditions. Other components of the glass (e.g., boron) will also affect the stability of the glass. Alkalinity will be produced by interactions of water with cementitious materials. Also lower silica activity, as a consequence of precipitation of silicates by interactions of groundwater and unstable engineered materials, could enhance the alteration of glass waste forms. Ultimate glass waste form alteration products are likely to be clay or zeolite minerals. These are analogous to alteration products of the natural volcanic glasses existing at Yucca Mountain. Both the clay and zeolite minerals are likely to incorporate augmented quantities of components of the engineered barrier system, such as iron and calcium. Clay minerals generally have low solubilities. Some quantity of radioactive waste species is likely to be incorporated in mineral alteration products of glass waste forms.

#### **4.3.1.2 Effects of Engineered Materials on the Chemical Environment for Radionuclide Release**

The effects of engineered materials are likely to be important to the chemical environment for radionuclide release. A variety of metal alloys that are thermodynamically unstable in contact with oxidizing water are being considered as container materials for the Yucca Mountain repository. In addition, if carbon steel is used as structural support for the drifts, then reactions affecting the containers would also affect the structural supports. Although corrosion of these materials may be slow, it would consume oxidants in the near-field environment and, thus, could affect the continued corrosion of the spent fuel. Therefore, corrosion of container materials may lead to locally reducing conditions in the near field and strong gradients in

oxidation potential, despite the prevailing oxidizing nature of the geologic setting (Murphy and Pabalan, 1994). Reducing conditions will likely have a limited effect on radionuclide release. However, depending on the geometry of the flow path of water through the engineered barrier system, the flux of oxygen, and the mass and rate of oxidation of easily oxidizable metals along the transport path, local conditions could be chemically reducing.

Corrosion products from metallic components, mostly in the form of metal cations, can affect corrosion rates of spent fuel directly through precipitation reactions forming secondary minerals that may slow the rate of dissolution. Conversely, corrosion rates can be increased by indirect action of corrosion products that may change the redox potential and the pH of the environment. The redox potential can increase by the action of reducible cations, such as  $\text{Fe}^{3+}$ . The pH can decrease by oxidation of chromium, present in stainless steel, to  $\text{Cr}^{3+}$ , or by hydrolysis of highly-charged cations, such as  $\text{Fe}^{2+}$  and  $\text{Cr}^{3+}$ , among others. Low molecular weight organic compounds, including carboxylic acids, can be produced by degradation of fuel, lubricants, or other organic materials, either by chemical or biochemical mediated processes. These compounds may accelerate the rate of corrosion of spent fuel due to the formation of complexing species and chelating species. In addition, published literature (McVay and Buckwalter, 1983; Barkatt, et al., 1989; Bourcier, et al., 1992) and ongoing CNWRA experiments indicate, under some conditions, dissolution rates of silicate glasses are strongly enhanced by dissolved iron. Thus, waste package corrosion products such as  $\text{Fe}^{3+}$  could be important in determining the dissolution rate of the glass waste form.

In the TSPA-VA design, the use of cementitious materials, in the form of concrete inverts and linings, was considered for the estimated 179 kilometers of emplacement drifts of the proposed Yucca Mountain high-level waste repository, in addition to the planned use of cement in roadways for construction, and emplacement ramps and service mains. Cement is used primarily for its structural (e.g., high compressive strength) and physical (e.g., low permeability) properties. However, its effect on the near-field chemical environment of a repository could be pronounced. Cements are extremely fine-grained, high-surface area materials containing somewhat soluble and thermodynamically metastable phases (e.g., a gel-like phase designated calcium silicate hydrate because it contains Ca, Si, and  $\text{H}_2\text{O}$ ) that are unstable with respect to crystalline cement phases. These properties and the partially interconnected pore network of the solids make these materials potentially reactive with the near-field environment and the engineered barrier system.

Interactions between cementitious materials and the near-field system can be potentially beneficial for mitigating release of radionuclides. The persistent alkaline pH (>10) characteristic of pore fluids in contact with hydrated cement phases favor precipitation of a wide variety of radionuclides, including transuranics (Glasser, et al., 1985; Atkins, et al., 1990). For example, interaction of cement with aqueous  $\text{U}^{6+}$  can result in the formation of Ca-bearing phases uranophane or becquerelite, a poorly-crystallized Ca-uranyl hydrate (Atkins, Beckley, and Glasser, 1988; Atkins et al., 1990). On the other hand, alkaline conditions can be detrimental to the stability of nuclear waste glass. For example, experiments by Heimann (1988) indicated that cement-glass interaction leads to accelerated dissolution or alteration of the nuclear waste glass compared to a system without cement present. The degree to which cementitious materials can affect the chemical environment for radionuclide release will depend to a large part on its inventory and on the reactivity with groundwater of the specific cement material formulation used.

In the TSPA-SR design, cementitious materials are not planned to be used for ground support, except in the form of grout to anchor the rock bolts of the ground support system. The potential effect of cementitious materials on the chemical environment for radionuclide release in the TSPA-SR design is expected to be substantially reduced compared to the TSPA-VA design.

#### 4.3.1.3 Radiolysis Effects on Radionuclide Release

Radiolysis has complex effects on aqueous oxidation-reduction conditions. According to Dubessy, et al. (1988), the dose of absorbed gamma rays is only 0.02 times the dose of absorbed alpha particles in a given time. Also, according to Spinks and Woods (1976) (cited in Dubessy, et al., 1988), a single 1 mega-electron-volt alpha particle can ionize  $10^5$  molecules as it loses energy. Therefore, the primary cause of water radiolysis is alpha-particle radiation. Radiolysis occurs close to the site of radioactive decay and can affect wetted surfaces of radioactive waste forms.

Radiolytic oxidizing species, such as  $\text{OH}^\bullet$ , where “ $\bullet$ ” denotes a free radical,  $\text{H}_2\text{O}_2$ ,  $\text{HO}_2^\bullet$ , and  $\text{O}^{2-}$  (Spinks and Woods, 1976), could oxidize reduced species [e.g.,  $\text{Fe}^0$  in the waste package to  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ,  $\text{N}_2(\text{aq})$  to  $\text{NO}_2^-$  or  $\text{NO}_3^-$ , and  $\text{U}^{4+}$  to  $\text{U}^{6+}$ ]. Molecular hydrogen ( $\text{H}_2$ ) produced as the result of the combination of two  $\text{H}^\bullet$ , in contrast, is relatively non-reactive.  $\text{H}_2$  is likely to diffuse away from the site of radiolysis. Various experimental studies using gamma radiation suggest that radiolysis will promote waste form (both spent fuel and glass) instability and radionuclide mobility through both enhancement of oxidative processes and lowering of pH (Wronkiewicz, Young, and Bates, 1991; Wronkiewicz et al., 1993; Sunder, et al., 1992; Sunder and Christensen, 1993). As pointed out by Van Konynenburg (1986), such processes are enhanced by unsaturated conditions expected in the proposed Yucca Mountain repository. On the other hand, bicarbonate could limit the radiolytic pH lowering (Van Konynenburg, 1986), as could cement-water interactions.

#### 4.3.1.4 Microbial Effects on the Chemical Environment for Radionuclide Release

The potential importance of microbial processes on the chemical environment for radionuclide release from nuclear waste repositories has been recognized in Europe (Pedersen and Karlsson, 1995; Lessart, et al., 1997; Christofi and Philp, 1997). Metabolic by-products of microorganisms may lead to solubilization and increased mobility of radionuclides (Christofi and Philp, 1997). Complexation of actinides (e.g., U and Np) by organic acids could increase their solubilities by about two orders of magnitude (McKinley, West, and Grogan, 1985).

The potential for enhanced high-level waste glass dissolution and the production of chelating and complexing ligands as a result of microbial activity has also been recognized by DOE as being potentially important to radionuclide release. These topics are addressed in the Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1998d, 1999d). Horn and Meike (1995) concluded that microbial activity may promote dissolution of glass by producing acidic conditions, and may increase the solubility of metals via chelation. Investigations of Yucca Mountain indigenous microbes, documented in Hersman (1996), suggested that they are capable of producing enough chelating agents (e.g., siderophores) to chelate 0.2 gram per liter of Pu-239 in the subsurface pore water. Corrosion of natural basaltic glass (Thorseth, Furnes, and Tumyr, 1995) and simulated nuclear-waste glass (Staudigel, et al., 1995) by biologically mediated dissolution has been demonstrated. Despite these studies,

DOE did not consider microbial effects on radionuclide release in its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b).

It will not be necessary to address microbial effects on radionuclide release in its performance assessments if DOE demonstrates, through the use of a mass balance of nutrients and energy-producing reactions approach (McKinley, et al., 1997), or another approach, that microbial activity within the engineered barrier system is unlikely to be of significance. This approach has been discussed in detail in Section 4.2.1.4 and a detailed discussion of the limitations for microbial activity has been presented in Section 4.1.1.3. While microbial activity in the volcanic tuffs will be limited by a lack of nutrients, additional nutrients may be available within the engineered barrier system. Thus, an analysis of the supply nutrients and energy-producing reactions, which would consume the nutrients, could be used to demonstrate that microbial effects on radionuclide release could be neglected. DOE stated that it would use this type of approach to evaluate the potential for microbiological processes in the near-field geochemical environment (TRW Environmental Safety Systems, Inc., 1997b).

#### **4.4 THE EFFECTS OF COUPLED THERMAL-HYDROLOGIC-CHEMICAL PROCESSES ON RADIONUCLIDE TRANSPORT THROUGH ENGINEERED AND NATURAL BARRIERS**

In Revision 2 of its Repository Safety Strategy (U.S. Department of Energy, 1998a), the DOE considered radionuclide concentration reduction during transport through engineered and natural barriers as a key performance attribute of the proposed repository. In Revision 3 of the Repository Safety Strategy (CRWMS M&O, 2000a), DOE considered retardation of radionuclide migration in the unsaturated zone as one of the seven principal factors of the postclosure safety case. Two of the fourteen integrated subissues NRC has determined that need to be abstracted into a performance assessment are (i) radionuclide release rates and solubility limits, and (ii) radionuclide transport in the unsaturated zone. Each of these integrated subissues (see Appendix A) will be affected by coupled THC processes and is necessary to appropriately describe transport of radionuclides through engineered and natural barriers. Contributions from Unsaturated and Saturated Flow under Isothermal Conditions and Radionuclide Transport KTIs will also be required to resolve radionuclide transport on the unsaturated zone integrated subissue. DOE must adequately evaluate the effects of coupled THC processes on the transport of radionuclides through engineered and natural barriers in its assessments of repository performance.

##### **4.4.1 Technical Bases for Reviewing the Effects of Coupled Thermal-Hydrologic-Chemical Processes on Radionuclide Transport Through Engineered and Natural Barriers**

The bases are primarily focused on explaining why the results of different coupled processes may be important to radionuclide transport. As mentioned in Section 3, the dearth of analysis in past performance assessments on the effects of coupled geochemical processes on the potential radionuclide transport behavior has resulted in an approach that primarily focus on ensuring that some type of analysis of the effects be completed. The sophistication of the analysis of the effect of a coupled process on transport of radionuclides that could be conducted and found acceptable by the staff is dependent on the credit to which DOE ascribes to the barrier, the information available at present, any plans to obtain the additional information

as part of the long-term testing program, and the ability of codes to model coupled processes and determine their impact on repository performance.

Previous versions of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1998e, 1999f) have documented the evaluation of the technical bases for earlier versions of DOE performance assessments of Yucca Mountain. This revision of the ENFE IRSR focuses on preserving the previous evaluations and updating the technical bases to reflect the effects of the current repository design features on radionuclide transport through engineered and natural barriers. For example, the Enhanced Design Alternative-II was selected by the DOE during the License Application Design Selection process (CRWMS M&O, 1999b) for TSPA-SR (CRWMS M&O, 1999a). This new design calls for the use of backfill—later removed from consideration, a titanium drip shield, a steel invert with granular ballast, steel ground support, an Alloy 22 outer barrier, and a stainless steel inner barrier. Interaction of infiltrating fluid with corrosion products could substantially affect the values assumed for retardation in the unsaturated zone. One acceptable approach to evaluate the effect of this design would be to conduct sensitivity studies using sorption coefficient ( $K_d$ ) values associated with iron corrosion products. If substantial degradation in the performance of the repository resulted from the use of these alternative  $K_d$ s, it would be expected that these alternative values be used to assess performance of the repository, or methods be employed to estimate realistic value of  $K_d$ s in the presence of corrosion products.

#### **4.4.1.1 Coupled Thermal-Hydrologic-Chemical Processes Affecting Radionuclide Transport Through Engineered and Natural Barriers**

A number of processes may operate in the near field to control the migration of radionuclides from the waste forms through the engineered barriers and into the geologic setting. The following is a brief summary of those processes that may be significant within the near-field environment.

A major concern in performance assessments is the transport of radionuclides through the engineered barrier system and the geologic setting. Transport of radionuclides can occur as gaseous species, as species in colloidal form, or dissolved in aqueous solution. Each mechanism for radionuclide transport is influenced by several geochemical parameters. Thus, an assessment of the relative importance of each mechanism will depend on the specific geochemical and hydrologic characteristics of the near-field environment. However, it is useful to qualitatively describe the effects of changing geochemical parameters on near-field radionuclide retardation and transport processes.

#### **Precipitation and Coprecipitation**

One mechanism for removing radionuclides from solution is precipitation of stoichiometric radioelement compounds. Coprecipitation as an impurity in other minerals can also remove radionuclides from solution. Changes in system chemistry parameters, such as Eh, pH, and component concentration, influence the solubilities of radionuclide-bearing minerals. For example, reduction of  $UO_2^{2+}$  to  $U^{4+}$  greatly reduces uranium in solution through precipitation of reduced uranium minerals, such as uraninite (e.g., Langmuir, 1987). Under oxidizing conditions, increases in dissolved silica and other species can stabilize minerals, such as

soddyite and uranophane. These minerals will sequester not only uranium, but also other actinides through coprecipitation (Murphy and Prikryl, 1996).

Porosity and permeability could be enhanced by dissolution of the primary minerals and reduced by precipitation of secondary minerals. Given the temperature-dependent solubilities of different minerals, it is possible that thermally convecting solutions will dissolve and redistribute minerals such as opal and calcite. This process could affect transport of radionuclides in the unsaturated zone and in the saturated zone beneath the repository. It is also possible that precipitation and dissolution of minerals in response to temperature increases will affect the porosity and permeability of backfill, if it is placed above the titanium drip shield as originally considered in the Enhanced Design Alternative-II design. In addition to its effect on radionuclide transport, changes in porosity and permeability of the uppermost portion of the saturated zone could affect the extent of vertical mixing of fluids leaving the unsaturated zone. Simulations by Travis and Nuttall (1987) suggest that reduced permeability due to quartz precipitation may enhance waste isolation. In contrast, Verma and Pruess (1988) determined that silica redistribution in a saturated fractured medium did not have a significant effect on near-field temperatures, pore pressures, or fluid flow. Matyskiela (1997) reports large changes in porosity and permeability in Paintbrush Tuff, where it has been intruded by a basaltic sill. He argued that the tuff was altered under conditions analogous to the proposed repository near field. The alteration is reported to have sealed the matrix from the fractures. If this type of alteration occurs at Yucca Mountain, it could lead to enhanced fracture flow and minimal matrix diffusion.

The TSPA-SR design proposed by DOE is anticipated to lead to lower temperatures in the wall rock than previous repository designs (CRWMS M&O, 1999a,b, 2000b). Temperatures at the drift wall are not anticipated to exceed 150 °C, and while temperatures may exceed boiling (96 °C) in the host rock, the boiling isotherms are not anticipated to coalesce in the drift pillars, enhancing the shedding of refluxed water between drifts (CRWMS M&O, 1999a). The effects of the new design temperatures on porosity and permeability have not been evaluated, but DOE proposes that the lower temperatures will reduce uncertainty in simulating the effects of temperature.

## **Sorption**

The principal concern of radionuclide transport in the near field is the advective transport of radionuclides dissolved in aqueous solution through the engineered barrier system to the geologic setting. Minerals in different components of the near-field environment may act to sorb radionuclides, removing them from solution and retarding the transport of radionuclides.

Oxides and oxyhydroxides of metals, such as iron, manganese, and silicon, are common fracture lining minerals in the Yucca Mountain system (Carlos, et al., 1993). These minerals may also be created by oxidation of materials introduced during the construction and operation of the repository (e.g., steel containers and rock bolts). The oxidation state in the near field may affect sorption behavior. For example, under oxidizing conditions, technetium is principally present as pertechnetate ( $\text{TcO}_4^-$ ) and does not sorb strongly. Under reducing conditions,  $\text{Tc}^{4+}$  is the predominant species of technetium and sorbs more strongly (Lieser and Bauscher, 1988). Electrostatic sorption is a function of surface charge. Titration experiments with oxyhydroxides indicate that surface charge is a function of system chemistry, particularly pH

(Davis and Kent, 1990). Sorption of cations, such as  $\text{UO}_2^{2+}$ ,  $\text{NpO}_2^+$ , and  $\text{Am}^{3+}$ , on oxyhydroxides exhibit a sharp sorption edge. Depending on radionuclide concentration and the number of available sites, sorption of cations increases from zero to nearly 100 percent over a relatively narrow pH range. In the presence of complexing ligands, such as  $\text{CO}_3^{2-}$ , cation sorption typically decreases to zero with further increases in pH (e.g., Kohler, Wieland, and Leckie, 1992; Pabalan and Turner, 1997). For anions and oxyanions, such as  $\text{TcO}_4^-$  and  $\text{SeO}_4^{2-}$ , the reverse is true. For these ions sorption typically decreases in a gradual fashion with increasing pH (Davis and Kent, 1990). Reactive surface areas can be high for the amorphous forms of oxyhydroxides. Thus these minerals are potentially important as a sorbent phase. In addition, the potential for forming a sorptive oxide coating on less sorptive particles, such as quartz or feldspar, suggests an additional role for these minerals in radionuclide sorption (Robert and Terce, 1989).

Other sorptive phases, such as clays, occur at Yucca Mountain as secondary replacement products. In addition, clays may also develop in the near field as alteration products of vitrified waste and spent fuel. Clays have interlayer exchange sites and the large surface area resulting from their layered structure. These minerals can have a high cation exchange capacity. Smectite, vermiculite, and some kaolinite group clays expand upon interaction with water or organic fluids. This process can change the interlayer spacing and affect the degree to which radionuclides can penetrate the interlayer ion exchange sites and sorb onto clays (Goldberg, Forster, and Heick, 1991). Increasing ionic strength can reduce interlayer spacing. Under these conditions, ion exchange on planar sites is likely to be less. The edge sites (perpendicular to the silicate layers) also exhibit a surface charge that varies as a function of pH. The pH variation is similar to that described previously for oxides and oxyhydroxides. Actinide sorption on clays is pH-dependent (Zachara and McKinley, 1993; Pabalan and Turner, 1997; Turner, Pabalan, and Bertetti, 1998).

Zeolites, such as clinoptilolite, heulandite, and analcime, may also be important for retarding transport of radionuclides in the near field at Yucca Mountain. Zeolites exhibit a fixed charge developed by substitution of  $\text{Al}^{3+}$  for  $\text{Si}^{4+}$  in the zeolite structure. The charge imbalance is compensated by  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  in the intracrystalline exchange sites. Sorption is typically by way of ion exchange in the intracrystalline sites (Davis and Kent, 1990). This behavior occurs for the alkaline and alkaline earth elements, such as the short-lived radioisotopes of  $\text{Cs}^+$  and  $\text{Sr}^{2+}$ . There also appears to be a component of pH-dependent surface charge involved in sorption of these elements as well (Pabalan, et al., 1993; Pabalan and Turner, 1993).

At increasing temperature and pH, calcite may be stable in the near-field environment (Murphy and Pabalan, 1994). For radionuclide sorption, the surface charge of carbonate minerals is dominated by the balance between the dominant cation ( $\text{Ca}^{2+}$  or  $\text{Mg}^{2+}$ ) and the carbonate anion ( $\text{CO}_3^{2-}$ ). For this reason, sorption on carbonates is a complex function of pH, solution chemistry, and  $p(\text{CO}_2)$ . Recent modeling efforts have focused on adapting surface complexation models to describe sorption at the interface between water and carbonate minerals (van Cappellen, et al., 1993).

Mineral precipitation and dissolution can also affect the retardation of radionuclide migration due to introduction or removal of sorptive minerals. Minerals such as zeolites, clays, and oxides can be dissolved and reprecipitated, depending on temperature and fluid chemistry (Bish, 1993; Murphy, et al., 1996). In addition, removal of radionuclides from solution by precipitation or

coprecipitation is also affected by the temperature and chemistry of the solution (Murphy and Prikryl, 1996). Walton, Ross, and Juhnke (1985) demonstrated this reaction experimentally. They machined circular flow channels into granite blocks and constructed thermal-convection loops to study the effects of heat and mass transport on radionuclide migration. A 40 °C temperature difference was applied across the system. Several radionuclides (Sb-125, Co-60, and Mn-54) were concentrated at the hot side of the experiment. This was probably due to sorption on iron oxyhydroxides. Ce-144 and Tc-99 were present in elevated concentrations on the cold side of the apparatus.

Most sorption experiments are run at room temperature (20 to 30 °C). The effects of elevated temperature on sorption are poorly understood. Machesky, Palmer, and Wesolowski (1994) indicate that the zero-point-of-charge ( $\text{pH}_{\text{ZPC}}$ ) of rutile decreases with increasing temperature. This change suggests that negative charge development is enhanced for oxyhydroxides with increasing temperature. This observation also suggests that the pH edge for cation sorption would move to lower pH values at higher temperatures. Limited batch data for temperatures up to 85 °C suggest that sorption coefficients for americium, barium, cerium, cesium, europium, plutonium, strontium, and uranium on crushed tuff materials either remain constant or increase with increasing temperature (Meijer, 1990). This assumption has been made in DOE performance assessment transport models (TRW Environmental Safety Systems, Inc., 1995). However, there is a lack of sample characterization before and after sorption, and large experimental uncertainties persist. These uncertainties and the limited sorption data make it difficult to extrapolate over ranges in physical and chemical conditions that are likely in the near field. The effects of temperature are likely to be greater for mineral precipitation and dissolution than sorption. Additional effort is necessary to constrain temperature effects on radionuclide transport through the near field. One of the criteria used in selecting the reference design for TSPA-SR is to reduce uncertainty in performance assessment calculations by maintaining lower temperatures than previous repository designs. By selection of blending, waste package spacing, drift spacing, and in-drift ventilation, temperatures during 10,000 years following permanent closure are estimated not to exceed 150 °C at the drift wall, and the boiling isotherm (96 °C) is predicted to not coalesce in the pillars between drifts (CRWMS M&O, 1999b).

## Diffusion

Diffusion is a retardation mechanism that is potentially important in the near field at Yucca Mountain. One example of diffusion is the migration of chemical constituents in water in fractures, where transport may be relatively rapid, into the matrix where flow and transport are slow. Field studies at the Nopal I uranium deposit in the Peña Blanca mining district, Chihuahua, Mexico, suggest that diffusion into the matrix is of limited importance in uranium retardation (Percy, Prikryl, and Leslie, 1995). Instead, uranium transport in fractured tuff appears to be dominated by fracture flow. Uranium retardation appears to be limited to precipitation of a suite of secondary uranyl minerals. The suite of precipitated minerals progress with time from hydrated uranyl oxides to uranyl silicates. Finally, uranium appears to be coprecipitated with iron oxyhydroxides and clays. A similar paragenesis has been observed in long-term drip experiments using water, related to that from the J-13 Well at Yucca Mountain, and unirradiated  $\text{UO}_2$  (Wronkiewicz, et al., 1992).

In TSPA-SR reference design (CRWMS M&O, 1999a,b, 2000b), diffusion of radionuclides through the invert beneath the waste package is the primary mechanism of release. As noted

in CRWMS M&O (1999b), testing is still necessary to determine the diffusion coefficients for the invert material, and the compatibility among the different parts of the waste package support system (waste package, invert, supports, ballast).

### **Gas Transport**

Vaporization would partition  $^{14}\text{CO}_2$  into the gas phase, enhancing gaseous radionuclide transport in the near field. Increased pH, perhaps through interaction with human-introduced materials in the near field, could result in increased partitioning of  $^{14}\text{CO}_2$  into the liquid phase. Codell and Murphy (1992) performed 1-D simulations of C-14 transport in unsaturated rock. The results indicated an early initial release of C-14 to the gas phase.  $\text{CO}_2$  was predicted then to dissolve into the aqueous phase and calcite precipitation served to sequester C-14 at longer times. The amount of gas transport is also sensitive to the thermal load imposed by the repository. Higher thermal loads cause venting of gas at the surface in numerical simulations (Light, et al., 1989). Releases also depend on the travel time to the surface, which depends on the Darcy velocity and the partitioning coefficient between the gaseous and aqueous phases.

DOE performance assessment models do not explicitly include  $^{14}\text{CO}_2$  gas transport (TRW Environmental Safety Systems, Inc., 1995; U.S. Department of Energy, 1998b). The decision by DOE not to include this mode of transport is consistent with recent recommendations of the National Academy of Sciences (National Research Council, 1995). The Academy considered that  $^{14}\text{CO}_2$  release at the accessible environment will be sufficiently diluted through mixing in the atmosphere to pose negligible individual risk. Other potential gas phase species, such as I-129 and Cl-36, are assumed in some TSPA-95 scenarios to be transported as gases without any retardation through the engineered barrier system, and then to be dissolved in the aqueous phase (TRW Environmental Safety Systems, Inc., 1995).

### **Colloid Transport**

Colloids involving radionuclides typically are called radiocolloids and have been divided into two types (Maiti, Smith, and Laul, 1989; Manaktala, et al., 1995). "True" or "real" colloids generally are formed from hydrolysis, polymerization, condensation, or precipitation of radionuclide compounds in solution. True colloid stabilization is favored under alkaline conditions, such as might persist in the near field. This is especially true for highly charged, redox-sensitive species such as actinides (Maiti, Smith, and Laul, 1989; Choppin and Mathur, 1991). Olofsson, et al. (1982a,b) indicated that the formation of colloids is favored for the actinides in lower (+3, +4) valence states. In the near field, where radionuclide concentrations can be relatively high, there is the potential for locally reducing conditions, and the formation of true colloids could be favored.

Pseudocolloids are formed when the radioelements sorb on small particles already present in the groundwater. In the near field, these particles may be either natural or introduced by human activity. The particles could include organic carbon, calcite, silica, clay particles, and oxyhydroxide compounds of metals such as iron, manganese, and aluminum. The presence, stability, composition, and sorptive capacity of these particles depend on the chemistry of the groundwater system. Parameters including pH, Eh, ionic strength, and  $p(\text{CO}_2)$  will affect colloid behavior. Further complicating the behavior of pseudocolloids is the possibility of non-sorptive particles being coated with sorptive materials (Robert and Terce, 1989). Experimental evidence

also has demonstrated that colloids can be formed as secondary precipitates and clay alteration products. These mineral phases can be released from high-level waste forms (Bates, et al., 1992; Ebert and Bates, 1992; Finn, et al., 1994a).

### **Colloid Stability**

The stability of the particles in suspension is of critical importance in colloid-mediated transport of radionuclides in the near field. Human activity associated with a high-level waste repository is the most likely source of colloidal materials in the near field. Dissolution of vitrified waste forms; secondary alteration products from spent fuel, glass, waste packages, and concrete; and organic matter used in drilling, construction, and repository operations (Travis and Nuttall, 1985) are all potential sources for colloids.

For metal oxyhydroxides, particle stability is a function of pH, Eh, particle size, and the total concentration of the metal (e.g., Fe, Mn, Al, Ti, Si) in solution. The presence of other ligands, such as  $\text{HCO}_3^-$  and  $\text{SO}_4^{2-}$ , can also affect the formation of oxides by consuming metal ions in the precipitation of carbonate and sulfate solids. Changes in solution chemistry can result in desorption of radioelements from the colloids and resorption onto the immobile medium. In this case, colloid transport becomes less of an issue in performance assessments.

The stability of the colloidal suspension of charged particles varies as a function of ionic strength, solution chemistry, and pH. Higher ionic strengths solutions may occur in the near field. The electrostatic double layer will collapse under these conditions. As a result, the charged particles will begin to flocculate (agglomerate) and come out of suspension due to gravity settling and filtration. Variations in solution chemistry and moisture content of the near field will influence the magnitude of the ionic strength effect. For example, at low pH, the positive surface charge of variably charged surfaces, such as clay edge sites and oxyhydroxides, is high. This results in increased bonding of positively charged crystallite edges to negatively charged planar sites. Positively charged oxides will also bond to negatively charged clay surfaces and organic macromolecules (Ryan and Gschwend, 1990). Under these conditions, dispersion is low, flocculation and agglomeration occurs, and the suspension is destabilized. As the pH increases towards the  $\text{pH}_{\text{ZPC}}$ , the positive surface charge of the oxides decreases and bonding to clays diminishes. At high pH, edge sites and oxyhydroxides exhibit a negative surface charge and actively repel the negatively charged clays. Thus, dispersion is enhanced, and the colloids are kept in suspension (Suarez, et al., 1984). Localized reducing conditions could be promoted by near-field hydrologic effects and phase variations. Local fluctuations of reducing and oxidizing conditions in the near field, due to an unstable hydrologic regime, could also induce secondary chemical effects such as the formation of colloids (Buddemeier and Hunt, 1988; McCarthy and Zachara, 1989).

### **Colloid Filtration**

The effectiveness of colloids in enhancing or retarding radionuclide migration depends on the efficiency of particle transport through the groundwater system. Colloid migration may be enhanced relative to fluid flow due to volume exclusion effects and reduced interaction between the particle and medium. The presence of a gas phase may influence particle transport by particle attachment to the bubble surface (Wan and Wilson, 1994). Conversely, colloids may

be retarded through various physical and chemical filtration mechanisms resulting from interaction between the different phases of the colloid-rock-water system.

McDowell-Boyer, Hunt, and Sitar (1986) divided filtration processes into three basic classes: (i) surface (cake) filtration, (ii) straining, and (iii) physical-chemical filtration. Surface filtration involves building a barrier at the interface between the water and pore. This type of filtration occurs when the particles are too large to enter the pores of the medium. As the particles are stopped at the surface, they are held in place by the fluid flow. A mat or cake of colloids is gradually formed. With time, the filter cake thickens and its porosity and permeability decrease through compression. Fluid flow through the mat decreases, and there is a pressure drop across the cake. Filter-cake permeability is also a function of particle aggregation. Destabilized colloidal suspensions (e.g., high ionic strength) tend to form a more porous arrangement than those cakes formed from highly dispersed stable suspensions (McDowell-Boyer, Hunt, and Sitar, 1986). If the particles are small enough to enter the porous medium, the tortuous path they must follow may eventually lead to a constriction that is too small for them to pass through. This leads to a straining of the colloids from solution.

Particles may also be removed from suspension by interaction with the pore walls. The interactions may be through physical processes or chemical processes. Once particles have been deposited, there is the possibility that they may be resuspended. The distances calculated for the energy attachment well (0.3 to 1 nanometer) are generally smaller than the diameter of the particle. London-van der Waals forces generally predominate at these distances. However, energy provided from Born repulsive forces, or thermal and hydrodynamic energy, can overcome the attraction energy well. This will lead to particle erosion and re-entrainment of colloids into solution. An additional possibility is that a decrease in the solution ionic strength may extend the electrostatic double layer. This would lead to particle release into solution (Kallay, Barouch, and Matijevic, 1987). Kallay, Barouch, and Matijevic (1987) also indicate that sweeping the resuspended particle away from the surface is necessary to prevent reattachment.

The size of colloids makes them vulnerable to several different filtration mechanisms. However, it is also possible that particle size (Bales, et al., 1989) will lead to volume exclusion and a less tortuous, more rapid path through the near field (Hunter, 1987). In pores and fractures, the water velocity distribution is such that the maximum velocity is along the centerline of the fracture. The minimum velocity occurs at the fracture wall. Because of their size, colloids can never “experience” the minimum water velocity. Thus, the average colloid velocity will be larger than that of the water. In general, this effect, called hydrodynamic chromatography (de Marsily, 1986), becomes more pronounced with increasing particle diameter. In addition, electrostatic repulsion associated with charged particles will tend to keep the particles away from the surfaces. This behavior further enhances the increased velocity effect. Hydrodynamic chromatography in a natural environment varies as a function of solution chemistry (de Marsily, 1986) because the particle charge is a function of pH and ionic strength.

#### **4.4.1.2 Effects of Engineered Materials on Radionuclide Transport Through Engineered and Natural Barriers**

Earlier analyses of the effects of engineered and man-introduced materials on transport of radionuclides in the near field assumed the TSPA-VA reference case design for the proposed

repository. This design assumed pre-cast concrete liners, rather than carbon steel ribbing, for drift support. The current TSPA-SR reference design eliminates the concrete liner and calls for a titanium drip shield. The reference design also replaces the concrete invert of earlier TSPA designs with a steel invert with granular ballast and steel ground support. For these reasons, the effects of engineered and man-introduced materials other than cementitious material are likely to be important to radionuclide release in the current design.

The principal organic components of natural soils and waters are humic materials (Choppin, 1988). Other organics may be introduced into the near field during repository construction and operation (e.g., solvents, fuels, etc.). The anionic charge of organic molecules allows them to bind readily to cationic species in solution. Humic substances can complex ions in solution, principally through oxygen donor sites. These substances can bind relatively highly charged cations, such as heavy metals and transuranic radionuclides. This behavior would reduce sorption of radionuclides onto minerals. For example, studies of Kohler, Wieland, and Leckie (1992) indicate that the presence of ethylenediaminetetraacetic acid (EDTA) in millimolar concentrations ( $10^{-4}$  to  $10^{-3}$  molar) can significantly reduce the amount of  $\text{Np}^{5+}$  sorbed on kaolinite. EDTA concentrations of the order  $10^{-6}$  molar, however, have only a slight effect on the sorption behavior. Thus the concentration of organic matter in the near field will determine whether complexation or sorption will control radionuclide transport behavior. Organic molecules may become bound as gels and coatings to the surface of inorganic particles such as clays and oxides. The sorptive behavior of the inorganic particles changes to reflect the organic coating (Robert and Terce, 1989).

The TSPA-VA design called for extensive use of concrete in constructing inverts, supports, and drift liners (U.S. Department of Energy, 1998b). The current TSPA-SR reference design significantly reduces the amount of concrete in the proposed repository (CRWMS M&O, 1999a,b). It is likely there still will be cementitious materials in the near-field environment in the form of grout used to cement rock bolts and in ventilation shafts, and there may be additional design changes prior to the submittal of any potential license application. Therefore, the transport of radionuclides potentially still may be affected by the presence of cementitious materials in the near-field environment of the proposed repository. Cement hydration products provide a multitude of sorption sites that could aid in retarding the migration of radionuclides (Atkins, et al., 1990; Atkins, Glasser, and Kindness, 1991) from the engineered barrier system to the host rock. In addition, the persistent alkaline pH ( $>10$ ) of pore fluids in contact with hydrated cement phases favors precipitation of a wide variety of radionuclides. Mineral alteration due to alkaline solutions and precipitation of secondary phases could reduce the sorptive and retardation ability of the geologic barrier. This alteration could also affect the hydraulic properties (porosity and permeability) of the tuff. Lichtner and Eikenberg (1995) used a geochemical transport model (MPATH) to predict that interaction between a hyperalkaline plume released from a cement-based low-level radioactive waste repository and a marl host rock would result in a rapid decrease in porosity of the host rock several meters from the repository. This decrease in porosity was due to precipitation of secondary phases. Their model predicted a porosity increased at the interface of the marl host rock and the cement due to mineral dissolution.

Flow of a hyperalkaline fluid along fractures in the tuffaceous host rock of the proposed repository is a potentially adverse scenario. Dissolution of the tuff could lead to widening of the fractures and enhancement of groundwater flow and transport of radionuclides. Alternatively, precipitation of calcite and calcium silicate hydrate phases along the fracture and matrix

interface could seal the fractures from the matrix. This would produce isolated channels through which transport of radionuclides could occur relatively unimpeded by matrix diffusion. However, if sufficient amounts of calcite and calcium silicate hydrate phases are precipitated along fracture walls, reduction in fracture porosity and permeability, or fracture plugging, could result in diminished flow and radionuclide transport. Preliminary calculations by Lichtner, Pabalan and Steefel (1997) suggest that strong alteration of the Yucca Mountain tuff host rock and of cement in contact with the tuff could result from interaction of cement and tuff pore waters and the respective minerals.

Engineered materials affect the potential transport of colloidal radionuclides. Oxides and oxyhydroxides of metals, such as iron, manganese, and silicon, may be created by oxidation of materials introduced during the construction and operation of the repository (e.g., metal containers and rock bolts). These highly sorbent minerals may form pseudocolloids and facilitate radionuclide transport within the near field. Colloids that could be formed within the waste package or from the spent fuel would most likely be agglomerated as a result of interaction with alkaline fluids associated with cementitious materials (Savage, 1997). If a concrete invert is used for the drifts, any colloids generated hydrologically up-gradient (i.e., in the waste package) would be subject to alkaline pore fluids associated with the invert.

#### **4.4.1.3 Radiolysis Effects on Radionuclide Transport Through Engineered and Natural Barriers**

Experiments on spent fuel leaching without imposed irradiation (Finn, et al., 1994a,b) are representative of potential autoradiolytic effects from spent fuel alpha radiation. These experiments imply that nascent hydrogen,  $H^\bullet$ , plays a role in reducing carbonate in solution to formate and oxalate. During transport of radionuclides, the coexisting reduced and oxidized species could become separated, leading either to a net reduction or net oxidation of the environment where radionuclides are concentrated. Furthermore, it is possible that reduced  $U^{4+}$  may form mobile complexes with the formate and oxalate radiolysis products (Finn, et al., 1994a,b).

Organic material may be introduced into the near field during construction and operation of the repository. Naturally occurring  $U^{4+}$  in sedimentary rocks is commonly correlated with organic matter (Pierce, Mytton, and Gott, 1955; Pierce, Gott, and Mytton, 1964; Nash, Granger, and Adams, 1981). The process by which the association arises is not fully understood. Uranium is readily transported in the uranyl ( $UO_2^{2+}$ ) state as carbonate complexes. Uranyl adsorption on organic material containing oxygen-bearing functional groups and as  $-COOH$ ,  $-COO$ , and  $-OH$  is favored and probably represents the first step of uranium mineralization. Subsequent reduction of the adsorbed uranyl ion by the organic matter or other reducing species and eventual precipitation of a  $U^{4+}$  mineral (e.g., uraninite or coffinite) follow. It appears, however, that the organic material hosting the uranium sometimes accumulates from solution in the form of asphaltite- or thucholite-type nodules (e.g., Pierce, Mytton, and Gott, 1955). The growth of these nodules could be an indication of autogenous radiolysis, during which water-miscible hydrocarbons are scissioned by radiation and condense. Similar processes may occur in the near field with human-introduced organic matter.

Alternatively, this process might be inhibited by the reduction of bicarbonate to formate or acetate. The reactive  $H^\bullet$  could reduce adsorbed uranyl complexes to uraninite. As more  $UO_2$

precipitates, a chemical potential gradient in  $\text{UO}_2^{2+}$  carbonate complexes would be set up that would diffuse toward the precipitated  $\text{UO}_2$ , thereby increasing the probability of uranium adsorption, reduction, and precipitation. Further study of uranium coprecipitation with organic material is required to establish the validity of these hypotheses.

Both oxidizing and reducing radiolytic effects on waste forms can be hypothesized. However, the preponderance of evidence suggests that oxidation (and possibly acidification) will be dominant over reduction. Notably, the DOE TSPA-95 (TRW Environmental Safety Systems, Inc., 1995) does not consider potential radiolytic effects on the source term or radionuclide transport. The preceding discussion demonstrates that quantification of radiolytic effects is fraught with uncertainty. Nevertheless, it should be possible to calculate, quantitatively, ranges of possible states. Once this is accomplished, the effects of radiolysis could be incorporated into performance assessment models by ensuring that probability distribution functions for parameters such as solubility, sorption coefficient, and release rate cover the ranges of possible effects.

#### **4.4.1.4 Microbial Effects on Radionuclide Transport Through Engineered and Natural Barriers**

Microbial effects on the transport of radionuclides have been examined in foreign nuclear waste repository programs (Christofi and Philp, 1997; Brown and Sherrif, 1998; Lessart, et al., 1997). These effects also have been studied in crushed volcanic tuff systems (Brown, Bowman, and Kieft, 1994). Potential impacts on radionuclide transport caused by microbial processes involve sorption (Brown, Bowman, and Kieft, 1994; Stroes-Gascoyne, 1996; Christofi and Philp, 1997), cellular uptake and potential transport as colloids (Pedersen, 1996; Christofi and Philp, 1997), and chelation and complexation of radionuclides (Brown and Sherif, 1998).

Biomass, such as biofilms that grow under oligotrophic conditions expected at Yucca Mountain, can increase sorption of some radionuclides (Brown and Sheriff, 1998; Stroes-Gascoyne, 1996). However, microbes can also decrease sorption of other radionuclides by changing the pH of the pore fluids (Brown, Bowman, and Kieft, 1994). Irreversible cellular uptake of radionuclides by microorganisms can be treated as colloidal transport (McKinley, West, and Grogan, 1985; Pedersen and Karlsson, 1995). This process has been demonstrated to be negligible for the Swedish program. The ability of microorganisms to be transported as colloids in an unsaturated zone is strongly affected by irreversible sorption of the microbes onto the gas-water interface (Wan, Wilson, and Kieft, 1994). A series of experiments designed to study the gross effects of microbial activity on repository geochemistry, radionuclide sorption, and integrity of repository and host rock materials indicated that microbial effects were not important (West, et al., 1998). Thus, it appears that microbial effects on radionuclide transport through engineered and natural barriers appears to be unimportant.

However, DOE has argued that potentially deleterious impacts of microbial activity on the radioactive waste environment at Yucca Mountain include the increased rate of transport of radionuclides from breached waste packages (Horn and Meike, 1995; Hersman, 1996). Both chelation (Hersman, 1996) and colloidal transport of microbially sorbed radionuclides (Horn and Meike, 1995) are postulated to be potentially important to repository performance. Nevertheless, DOE did not address microbial effects on radionuclide transport in its TSPA-VA (U.S. Department of Energy, 1998b).

It will not be necessary for DOE to address microbial effects on radionuclide transport in their performance assessments if they demonstrate that microbial activity in the repository is unlikely to be of significance. This could be demonstrated through the use of a mass balance of nutrients and energy-producing reactions approach (McKinley, et al., 1997). This approach has been discussed in detail in Section 4.2.1.4 and a detailed discussion of the limitations for microbial activity has been presented in Section 4.1.1.3 of this IRSR.

#### **4.5 COUPLED THERMAL-HYDROLOGIC-CHEMICAL PROCESSES AFFECTING POTENTIAL NUCLEAR CRITICALITY IN THE NEAR FIELD**

The presence of fissile radionuclides, such as U-235 and Pu-239, in the spent nuclear fuel creates a potential for sustained neutron chain reaction (criticality event). Such an event could arise if there is failure of the waste package, dissolution of the fissile material, and redeposition outside the waste package in the near-field environment. The acceptance criteria considered relevant to this ENFE subissue are limited to consideration of criticality resulting from coupled THC processes in the near-field environment of the proposed Yucca Mountain repository. Criticality issues related to canister and waste form design issues are considered in the Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1998d, 1999d). Criticality issues in the far field of the proposed repository are addressed in the Radionuclide Transport IRSR (U.S. Nuclear Regulatory Commission, 1998f, 1999g). The review process will determine whether nuclear criticality in the near-field environment due to coupled THC processes has been adequately considered by the DOE. Potential effects on repository performance of criticality in the near field include an increase in the fission product inventory, a decrease in the fissile radionuclide inventory, and an increase in thermal output.

##### **4.5.1 Technical Bases for Reviewing the Effects of Coupled Thermal-Hydrologic-Chemical Processes Affecting Potential Nuclear Criticality in the Near Field**

The technical bases for the acceptance criteria for nuclear criticality as a result of coupled THC processes are given in this section. The sophistication of the analysis of nuclear criticality as a result of coupled THC processes that could be conducted and found acceptable by the NRC staff is dependent on the information available at present and is a function of whether DOE screens out from the performance assessment the criticality scenario, and the ability of computer codes to model nuclear criticality as a result of coupled THC processes.

As noted by Choi and Pigford (1997), fissile materials scheduled for long-term geologic disposal in the proposed high-level waste repository at Yucca Mountain theoretically can reach criticality in a geologic medium. These materials may include weapons-grade plutonium, highly enriched uranium from naval and research reactors, and small amounts of plutonium and enriched uranium from commercial and DOE-owned spent fuel.

##### **4.5.1.1 Principles of Criticality Safety and Factors That Affect Criticality**

Several parameters affect the potential for nuclear criticality in a given system. A brief summary, taken from a report by Cragnolino, et al. (1997) on the Hanford Tank Waste Remediation System, is provided here. Before a self-sustained neutron chain reaction, or critical state, can be achieved, a number of physical conditions must exist. One required condition is the presence of a sufficient amount of fissile material to absorb neutrons and

undergo fission. Each fission event generates several high-energy neutrons. These neutrons undergo interactions in which they either lose energy, are absorbed, or are lost from the system by leakage. The critical state of a system is determined by the number of neutrons lost by absorption or leakage relative to the number of neutrons from fission events that are available to produce subsequent fissions. If more neutrons are absorbed or lost by leakage than are produced by fission, the system is subcritical. If an equal number of neutrons are produced as are lost or absorbed, the system is critical. If more neutrons are produced than are lost or absorbed, the system is supercritical.

The critical state is mathematically represented by a parameter called  $k_{\text{eff}}$ . This parameter is defined as the number of neutrons in one generation divided by the number of neutrons in the preceding generation. A critical state has a neutron population that remains constant, with  $k_{\text{eff}}$  equal to one. A subcritical system has a neutron population that decreases in time,  $k_{\text{eff}}$  less than one. A supercritical system has a neutron population that increases in time,  $k_{\text{eff}}$  greater than one. If the size of the system is effectively infinite and has no neutron leakage, the parameter of interest is called  $k_{\infty}$ . The value of  $k_{\text{eff}}$  is always less than or equal to the value of  $k_{\infty}$ . Typically, an upper bound for subcritical conditions is to have a calculated  $k_{\text{eff}}$  value of less than 0.95, with a 95-percent confidence level.

Fission occurs more readily after neutrons have undergone several scattering reactions. As a result of the scattering reactions, the energy of the neutrons has decreased such that the neutrons are in thermal equilibrium with the scattering medium. The process of reducing the neutron energy is known as “slowing down” or “moderation.” Moderation is most effectively accomplished by materials of low atomic mass and high ratios of neutron scattering to absorption coefficients. Hydrogen (e.g., in water) is one example of the materials that are called moderators. At optimum moderation, a minimum amount of fissile material is required to sustain a chain reaction. At other than optimum moderation, more fissile material is required to reach criticality. The amount of fissile material required for criticality is also affected by the concentration of the fissile material. The geometry of the system containing the fissile material also affects the potential for criticality. Finally, the presence (or absence) of other materials that compete with fissile material for absorption of neutrons also affects the potential for criticality.

To prevent inadvertent criticality in a system, specific controls and limitations are placed on the factors that affect criticality. For high-level waste disposal operations, the factors most important to criticality include: (i) concentration of fissile material, principally Pu-239 and U-235, (ii) the amount and properties of neutron absorbers or moderators present with the fissile material, (iii) the geometry of the system containing the fissile material, and (iv) the presence or absence of neutron reflectors adjacent to the system.

### **Fissile Material Concentration**

In order to achieve criticality, the fissile material must be present in certain concentrations, regardless of the size of the system. The critical concentration depends on the solids, water, and mixture characteristics present in the repository environment. In pure water, a Pu-239 concentration of 7.2 grams per liter is generally reported as the minimum critical concentration of plutonium (Knief, 1992).

## Neutron Absorbers

Neutron absorbers, such as boron or gadolinium, reduce the reactivity of a fissile mixture by reducing the thermal neutron flux. These materials generally absorb neutrons and release gamma or alpha particles. The alpha particles and gamma rays do not contribute to further fission events. There is a unique minimum absorber to fissile material mass ratio for all absorbers. Above this ratio, the system will remain subcritical, independent of any other influences. Neutron absorbers are likely to be engineered into waste forms (e.g., gadolinium in glass logs containing weapons-excess Pu) or waste packages (e.g., stainless steel in basket materials) for disposal. Exclusion of neutron absorbers, such as B, in model calculations leads to conservative estimates of the subcritical margin of safety.

## Geometry

Geometry plays a role in determining subcritical limits because of its influences on neutron leakage. Neutrons that leak out of the system will not contribute to any further fissions. Therefore, reducing the number of neutrons that escape the system will increase the reactivity of the system. If the geometries for high-level waste disposal can be shown to be subcritical for a homogeneous material containing fissile and absorber atoms, or for an infinite medium, then any finite slice of the infinite medium will be even more subcritical.

The effects of geometry are typically discussed in terms of a sphere. This is because a sphere is the most reactive geometry and, thus, constitutes the bounding case. For plutonium in relatively low concentrations, the critical sphere size is very large. Other geometries considered in models include infinite slabs and infinite-length cylinders.

## Neutron Reflectors

Neutron reflectors surrounding fissile material may increase the reactivity of the system. The reflectors would return neutrons that have leaked out of the system back to the fissile material where they would be able to contribute to further fissions. Reflectors will reduce losses from geometry effects. Thus for conservative calculations that assume infinite dimensions, reflectors have no effect. It is unknown whether reflectors will be present around the fissile material. However, calculations that take credit for neutron leakage losses from the system must take into account the effects of neutron reflectors that surround the system on any side, if they are present.

### 4.5.1.2 Theoretical Autocatalytic Criticality in the Near Field

While the waste canisters and waste forms are intact, design features to prevent criticality are expected to function as planned. Thus criticality should not be an issue (Bowman and Venneri, 1995; Choi and Pigford, 1997). Following canister failure, however, Bowman and Venneri (1995) presented a conceptual model where added neutron absorbers (e.g., boron and lithium) and subcritical concentrations of Pu-239, U-235 and other fissile materials are mobilized from waste forms in the Yucca Mountain repository environment. They postulated that fissile material could be deposited in a concentration and geometry sufficient to reach criticality. Bowman and Venneri (1995) also proposed a series of feedback mechanisms, with the rock itself acting as a moderator in a low-water environment. This means that, as the system

reaches criticality, water would boil off and disperse plutonium into a greater volume of rubbleized rock. This was postulated to eventually create a geometry that is autocatalytic (or self-enhancing).

In general, Bowman and Venneri (1995) considered a spherical geometry of homogeneous mixtures of Pu-239 with water and SiO<sub>2</sub> as a proxy for rock. For the idealized spherical plutonium-H<sub>2</sub>O-SiO<sub>2</sub> geometry considered, Bowman and Venneri (1995) noted that a sphere of about 25 centimeters radius is the smallest geometry that can sustain an autocatalytic reaction. However the radius of the sphere depends on the mole fraction of silica. Bowman and Venneri (1995) proposed that a spherical mass of as little as 2 kilograms of Pu-239 may be enough for autocatalytic criticality. Canavan, et al. (1995) provide some qualitative discussion of the validity and probability of the Bowman and Venneri (1995) hypothesis. A similar approach for U-235 would require an even larger critical mass than Pu-239. Bowman and Venneri (1995) propose the natural reactors at Oklo, Gabon (Cowan, 1975), as evidence that such reactions have occurred in nature.

A number of internal reviews of the hypothesis presented by Bowman and Venneri (1995) were conducted at Los Alamos National Laboratory (LANL). Several critiques of the hypothesis proposed by Bowman and Venneri (1995) have been prepared (Murphy, Jarzempa, and Lichtner, 1995; Parks, Williamson, and Hyder, 1995; Van Konynenburg, 1995). Most of these critiques focus on several key aspects of the Bowman-Venneri hypothesis that limit its applicability in the Yucca Mountain environment, including:

- The lack of specificity on plausible radionuclide transport mechanisms that could lead to assembling a spherical geometry. One proposed mechanism is colloid transport into fractures surrounding the waste form. It is still not clear if colloid transport through the unsaturated zone can move and concentrate the amounts of plutonium that are necessary. At present, experiments suggest that glass does not alter to form pure plutonium phases, but instead alters to a suite of clays and secondary minerals (Bates, et al., 1992). In addition, if plutonium is bound to existing particles and transported as pseudocolloids, the mass of plutonium required for criticality will be larger than that required for a pure plutonium phase.
- Low plutonium solubility limits ( $10^{-6}$  to  $10^{-12}$  molar) that potentially require large volumes of water to provide the mass of Pu-239 necessary for autocatalytic criticality. For the lower solubility, it is estimated that tens of cubic kilometers of water would have to transport through a 100-centimeter radius sphere to transport the potentially critical 15 kilograms of Pu-239 by means of dissolution and redeposition (Murphy, Jarzempa, and Lichtner, 1995).
- The poor analogy between SiO<sub>2</sub> and the host rock at Yucca Mountain. Other constituents in the waste (e.g., U-238), waste canister (e.g., iron), rock (e.g., K, Al, and Na), and in the groundwater (Cl) would serve as neutron absorbers. This would require significantly larger amounts of fissile material for criticality, changing the dynamic behavior of the critical system, and possibly eliminating the potential for self-enhancing autocriticality. Also, dissolution of poisons, such as boron and lithium, is dependent on system chemistry, and preferential

leaching scenarios relative to silica are likely to be much more complex than the simple model proposed by Bowman and Venneri (1995).

- Consideration of realistic porosity and hydrologic saturation would greatly limit the mole fraction of water in the system, eliminating all but the largest geometries (and greatest masses of Pu-239) from consideration in the Yucca Mountain system. Also, the heterogeneity of the Yucca Mountain system makes it difficult to picture the idealized spherical geometry and homogeneous mixture considered by Bowman and Venneri (1995) (Van Konynenburg, 1995).

Other, less efficient geometries might be more feasible, but site-specific calculations for the Yucca Mountain system would be necessary to evaluate the masses of Pu-239 or U-235 required to achieve criticality. We do not consider that the scenarios and conditions postulated in the Bowman and Venneri paper are shown to be credible for any realistic repository situation (U.S. Nuclear Regulatory Commission, 1995). Nevertheless, we expect a rigorous, site-specific technical analysis of repository criticality safety (U.S. Nuclear Regulatory Commission, 1995). DOE should provide adequate technical justification or a conservative basis for neglecting criticality in a performance assessment (see further discussion in Section 5.4.5).

#### **4.5.1.3 Coupled Thermal-Hydrologic-Chemical Processes Affecting Potential Criticality in the Near Field**

Criticality in the near field would require transport of fissile components from the primary location of the waste form(s) and reconcentration at a location in the near field. The reconcentration of fissile materials would have to be in sufficient quantity and in an adequate configuration with respect to neutron reflection and escape. In addition, sufficient neutron moderators and sufficiently few poisons would be required for criticality. In general, entropy will tend to drive dispersion of initially concentrated fissile nuclides. Concentration of species may occur at interfaces (e.g., redox fronts, interfaces between strata or hydrologic systems). Precipitation may occur in zones of fluid mixing. Precipitation from a solution may also occur due to its movement along a thermal gradient.

A variety of hypothetical concentration environments may be possible in the near field of the proposed repository due to processes such as the following:

- Mixing of an oxidizing solution carrying uranyl ions with a solution rendered reducing by interactions with easily corroded iron construction materials
- Filtration of plutonium colloids where energetic fracture flow is terminated (e.g., at a perched water body)
- Precipitation at or near the water table where the temperature (or other chemical) gradient may change rapidly

The Disposal Criticality Analysis Methodology Topical Report (U.S. Department of Energy, 1998e) describes the methods DOE will use to estimate the probabilities and consequences of criticality events in the proposed repository. Included in the report is a list of configuration classes representing sites and processes, such as those listed above, that may lead to near-

field criticality. Geochemical aspects of the configurations and their probabilities and consequences will be modeled consistent with other near-field geochemical modeling to be conducted in support of a potential license application. The NRC has accepted the methodology contingent on closure of 25 open items (U.S. Nuclear Regulatory Commission, 2000b) and, as part of any potential license application review process, will evaluate the criticality analysis and its implications for performance assessment (see Section 5.4.5).

## 5.0 STATUS OF ISSUE RESOLUTION AT THE STAFF LEVEL

We have reviewed and commented on the DOE site characterization and performance assessment programs in areas related to the ENFE. The site characterization concerns were documented in the Staff Site Characterization Analysis of the DOE Site Characterization Plan, Yucca Mountain Site, Nevada (U.S. Nuclear Regulatory Commission, 1989). Additionally, a letter from the NRC to DOE with results of our review of the DOE thermohydrology testing and modeling program (U.S. Nuclear Regulatory Commission, 1997a) described concerns on the DOE coupled THC modeling efforts. Our concerns on near-field topics within the DOE performance assessment program have been documented (U.S. Nuclear Regulatory Commission, 1996, 1998g, 1999b). These concerns address the DOE program prior to the publication of the DOE TSPA-VA (U.S. Department of Energy, 1998b). The site characterization, thermohydrology, and performance assessment comments, and their status of resolution, are described in the following sections. Section 5.4 documents in detail our application of acceptance criteria to the preliminary draft DOE Process Model Reports, and preliminary draft Analysis and Model Reports available for review prior to May 15, 2000. In addition other sources of recent DOE information, such as the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) and Revision 3 of the DOE Repository Safety Strategy (CRWMS M&O, 2000a) were reviewed.

Sections 5.0.1 – 5.0.6 summarize our review and list items we believe DOE must address in order to close the subissues. Table 5-1 provides an updated summary of the status of subissue resolution.

**Table 5-1. Summary of issue resolution for ENFE KTI subissues**

Subissue	Status of Resolution
Effects of coupled thermal-hydrologic-chemical processes on seepage and flow	Open
Effects of coupled thermal-hydrologic-chemical processes on waste package chemical environment	Open
Effects of coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release	Open
Effects of coupled thermal-hydrologic-chemical processes on radionuclide transport through engineered and natural barriers	Open
Effects of coupled thermal-hydrologic-chemical processes affecting potential nuclear criticality in the near field	Closed, pending confirmation

Issues are “closed” if the DOE approach and available information acceptably address staff questions such that no information beyond what is currently available will likely be required for regulatory decision making at the time of initial license application. Issues are “closed-pending” if the NRC staff has confidence that the DOE proposed approach, together with the DOE agreement to provide the NRC with additional information (through specified testing, analysis, etc.) acceptably addresses the NRC's questions such that no information beyond that provided,

or agreed to, will likely be required at time of initial license application. Issues are “open” if the NRC has identified questions regarding the DOE approach or information, and the DOE has not yet acceptably addressed the questions or agreed to provide the necessary additional information in the license application.

#### **5.0.1 Resolution Status of U.S. Nuclear Regulatory Commission Staff Comments on the U.S. Department of Energy Site Characterization Plan**

As noted in Section 5.1 of this revision, all items relating to the ENFE KTI resulting from our review of the DOE Site Characterization Plan and study plans are considered resolved.

#### **5.0.2 Resolution Status of Evolution of the Near-Field Environment Key Technical Issue Subissue 1**

The main Analysis and Model Report relevant to this subissue, N0120—Drift Scale Coupled Processes, was not available prior to May 15, 2000 and was not reviewed. The following status summary is based primarily on the acceptance criterion on integration. Staff will update this summary when the Analysis and Model Report becomes available for review.

Based on DOE documents available at the time of this writing, the ENFE subissue on coupled THC effects on seepage and flow is “Open.” To close this subissue DOE should accomplish the following:

- (1) Provide a technical basis for neglecting thermal alteration of zeolites in the Calico Hills unit, demonstrate that thermal alteration of this unit is unimportant to performance, or include the effects in abstracted models
- (2) Provide a technical basis for neglecting thermal alteration of the Paintbrush Tuff, demonstrate that thermal alteration of this unit is unimportant to performance, or include the effects in abstracted models
- (3) Provide a description of the tests used to verify that TOUGHREACT Version 2.2 is producing correct solutions to the underlying mathematical models
- (4) Provide an improved technical basis for neglecting mineral precipitation in a highly localized zone at the interface between fractures and matrix
- (5) Provide adequate screening arguments for those FEPs identified by Pickett and Leslie (1999) or in Section 5.4.1.2 as having inadequate or missing screening arguments. The two FEPs on dehydration reactions identified as missing from the preliminary DOE FEP database should also be addressed
- (6) Evaluate the potential for cementitious materials (in ventilation shafts and tunnels) to interact with tuff host rock, thereby altering hydrological properties and flow paths below the repository

### **5.0.3 Resolution Status of Evolution of the Near-Field Environment Key Technical Issue Subissue 2**

DOE has made significant advances in its total system performance assessment abstraction of the waste package chemical environment. Efforts by DOE to improve the in-drift geochemical model abstractions by using the new linkage software GRIM represent a constructive response to NRC staff reviews (U.S. Nuclear Regulatory Commission, 1999f). The current DOE approach was designed to provide key advantages over the approach used in the TSPA-VA (U.S. Department of Energy, 1998b), including improved data traceability, explicit coupling between in-drift seepage flux and reaction processes, and the ability to track the time-dependent output of key variables. Based on specific review comments presented in the subsequent sections and the more general comments provided below, however, the status of this ENFE Subissue 2 remains open.

After completing the review of all DOE Analysis and Model Reports and supporting documents relevant to ENFE Subissue 2, staff will evaluate the overarching impact of DOE's approach to in-drift geochemical submodel integration on repository performance. A key aspect of this review will be an analysis of the reliability of DOE's major assumption (CRWMS M&O, 1999a) that coupled THC processes can be decoupled, evaluated separately, and then recoupled without adversely affecting predictions of repository performance. Several implications of this assumption are described in conjunction with various acceptance criteria in subsequent subsections within Section 5.4.2. DOE must provide sufficient technical bases to support their treatment of coupled THC processes. Another important aspect of the staff review on submodel integration will be the effectiveness of DOE's approach in tracking the evolution of water composition. The individual in-drift submodels predict changes in water composition due to chemical interactions with a limited number of repository materials, and it is not yet clear how DOE will weigh the output from several different submodels and integrate it into a single RIP cell. Integration must also be demonstrated between the output from different submodels. It is not appropriate, for example, to assume that the initial water compositions for all submodels is similar to that of J-13 Well water, when each of the in-drift geochemical submodels describes changes in the infiltrating water composition due to chemical interactions with different types of engineered barrier materials. All DOE assumptions must be explicitly stated, and adequate technical bases must be provided.

There is a second major assumption in DOE's approach (CRWMS M&O, 1999a) relevant to ENFE Subissue 2. It is also lacking a sufficient technical basis. This assumption, that all reactions proceed to equilibrium, must be supported by calculations or sensitivity studies demonstrating a low consequence on predicted repository performance.

DOE should evaluate uncertainties associated with the PT4 database for modeling the behavior of electrolyte solutions and provide a more representative list of FEPs associated with the Salts/Precipitates Analysis and Model Report.

DOE should conduct further studies to identify and characterize the conditions that could lead to enhanced drip shield degradation, in particular, take the potential impact of elevated fluoride concentrations into account, place reasonable constraints on the aqueous compositions at Yucca Mountain that might lead to drip shield degradation, and use this information to define

appropriate abstractions of engineered barrier degradation for performance assessment calculations.

The TSPA-SR design has changed from the design proposed after the License Application Design Selection process (CRWMS M&O, 1999b). The most significant change is the removal of backfill from the reference design. In future revisions of the Analysis and Model Reports and Process Model Reports, DOE will either need to provide additional analyses to evaluate the effect of no-backfill on the engineered barrier system or provide a technical basis for the applicability of the earlier calculations. Possible effects include temperature, flow rates, and water chemistry. Resolution of ENFE Subissue 2 requires that current repository design features are considered in all relevant DOE abstractions, technical bases, and supporting calculations.

DOE identified a number of excluded FEPs. As necessary, DOE will need to provide a more thorough technical basis or supporting calculations to justify a number of these exclusions, especially those based on low consequence. DOE also needs to revise the FEP database so that all the FEPs included in the in-drift geochemical model abstraction are also included in the FEP database. Several exclusions such as liner degradation and backfill erosion are acceptable based on the current TSPA-SR reference design. However, should the design change, continued exclusion of these FEPs will require additional technical bases consistent with the revised design features.

Several DOE model abstraction approaches outlined in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) are appropriate, but the technical basis and supporting calculations (sensitivity analyses and bounding values) necessary to support the parameter selections or characterize the uncertainty in the models have not yet been reviewed.

To expedite resolution of ENFE Subissue 2, DOE should communicate the results of their work in the framework provided by the acceptance criteria listed in Section 5.4.2.

#### **5.0.4 Resolution Status of Evolution of the Near-Field Environment Key Technical Issue Subissue 3**

DOE abstractions relevant to coupled THC effects on the chemical environment for radionuclide release represent significant improvements relative to DOE's approach used in the TSPA-VA and represent a constructive response to our review of the TSPA-VA. There is improved data traceability, explicit coupling between in-package chemistry and waste form and waste package degradation processes, and abstraction of time-dependent output of key variables. However, based on specific review comments presented in the subsequent sections, the status of this ENFE Subissue 3 remains open. DOE's abstraction approaches outlined in the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j) are appropriate, but the potential effects of some THC processes were neglected without sufficient technical basis. For example, there was no consideration of early-stage, high-temperature phenomena such as evaporation and condensation on the composition of water entering the waste package; the effect of corrosion products on high-level waste glass degradation was not considered; radiolysis effects on waste form degradation was neglected; and enhancement of cladding corrosion in the presence of high chloride concentrations was excluded. Also at issue is the spatial variation in chemistry that is likely to occur in the waste package, which could

result in local pH values more acidic than values calculated using a volume-averaged mass. For example, pH in crevices and other tight spaces differ from bulk pH values due to dissolution reactions that become spatially separated from oxidation/reduction reactions. These locally more acidic pH solutions could enhance the dissolution rate of the waste form and the release of radionuclides. DOE will need to provide more adequate technical bases, for example, using sensitivity analyses, for neglecting these processes in their abstractions of the chemical environment for radionuclide release.

DOE identified a number of excluded FEPs from its abstractions relevant to coupled THC effects on the chemical environment for radionuclide release. As necessary, DOE will need to provide more adequate technical bases or supporting calculations to justify a number of these exclusions. Staff will review the pertinent Analysis and Model Reports and Process Model Reports to determine which FEPs have been included in the DOE abstraction of chemical environment for radionuclide release, if those FEPs have been incorporated adequately, and determine whether excluded FEPs have an adequate technical basis to support their exclusion.

DOE predictions of in-package chemistry based on the EQ3/6 code have not been verified by empirical observations. In particular, staff has concerns regarding the use of the EQ3/6 code because it does not incorporate electrochemical reactions. Experiments to simulate certain aspects of waste package geometry and materials may aid in gaining confidence in the EQ3/6 results.

To address our comments on colloid-assisted radionuclide release, DOE could either conduct additional analyses or incorporate critical assessments of the effects on colloidal radionuclide release that are beyond the scope of available experimental work. Parameters that may need to be addressed include (i) temperature, (ii) oxidation potential, (iii) major cation and anion concentrations, and (iv) in-package hydrologic conditions.

As planned, DOE should incorporate the results from ongoing spent fuel corrosion tests in deciding whether to include irreversible radionuclide attachment in the colloid abstraction for spent fuel. These tests should employ methods similar to, or more rigorous than, those that helped to identify radionuclide-bearing, waste form-generated colloids in high-level waste glass tests. If DOE does not alter the abstraction, it should provide a technical argument that exclusion of irreversible attachment will not neglect a potential means for increasing radionuclide concentrations.

In addition, DOE should more explicitly evaluate whether the experimental results are adopted in a manner that is conservative with respect to ionic strength and pH effects (e.g., on colloidal plutonium concentration). Finally, DOE should extend the colloidal release abstraction to other radionuclides or provide a technical basis for exclusion of important radionuclides.

#### **5.0.5 Resolution Status of Evolution of the Near-Field Environment Key Technical Issue Subissue 4**

DOE made significant advances in its total system performance assessment abstraction of radionuclide transport through engineered and natural barriers. Based on the evaluation described in Section 5.4.4, however, this subissue (ENFE Subissue 4) remains open.

The TSPA-SR design is different from the design used in developing the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b). The most significant change is the removal of backfill from the reference design. In future revisions of the Analysis and Model Reports and Process Model Reports, DOE will either need to provide additional analyses to evaluate the effects of no-backfill on the engineered barrier system or provide a technical basis for the applicability of the earlier calculations. Possible effects include temperature, flow rates, and water chemistry.

DOE identified a number of excluded FEPs. As necessary, DOE will need to provide a more thorough technical basis or supporting calculations to justify a number of these exclusions, especially those based on low consequence. In addition, apparent discrepancies between the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) and the supporting preliminary draft FEPs Analysis and Model Report (CRWMS M&O, 2000q) will need to be reconciled. Particular attention should be paid to the exclusion of colloid transport FEPs, either using detailed process models or sensitivity/bounding analyses, to demonstrate low consequence. Several exclusions such as liner degradation and backfill erosion are acceptable based on the current TSPA-SR reference design. However, if the design changes, continued exclusion of these FEPs will require additional technical bases consistent with the revised design features.

Several DOE model abstraction approaches outlined in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) and summarized in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) are appropriate, but the technical bases and supporting calculations (sensitivity analyses and bounding values) necessary to support the parameters selected for the models are not yet available and will need to be provided.

In several cases, DOE used bounding analyses. A good example is the assumption of no retardation for transport through the invert. Given the short flow path through the invert (< 1 meter) relative to the flow path to the critical group (~20 kilometers), this assumption is not likely to underestimate dose to the critical group and is acceptable. However, the analysis in DOE's Repository Safety Strategy, Revision 3 (CRWMS M&O, 2000a) suggests that diffusive transport through the invert may have a significant effect on dose, and additional bounding analyses may be necessary.

DOE (CRWMS M&O, 2000a) has proposed to rely on radionuclide retardation due to sorptive minerals in the unsaturated zone. Currently, DOE has screened out potential adverse effects on the sorptive minerals due to from coupled THC process. DOE must demonstrate that the effects of THC processes from the repository will not adversely affect the stability of the sorptive minerals found in the Calico Hills vitric units beneath the repository, provide a technical basis for neglecting THC effects on zeolites in the Calico Hills unit, demonstrate that thermal alteration of this unit is unimportant to performance, or include the effects in abstracted models.

DOE relies on the Integrated Site Model to provide input to compare sorptive zeolites and vitric minerals at Busted Butte to minerals beneath the repository horizon. This derived information provides input to process models and the total system performance assessment. A statistical basis is needed to support the derived input for performance assessment models involving near field and unsaturated zone flow and radionuclide transport.

## **5.0.6 Resolution Status of Evolution of the Near-Field Environment Key Technical Issue Subissue 5**

The criticality subissue is resolved at the staff level, pending (i) closure of open items concerning the DOE criticality analysis methodology raised in the NRC safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) and (ii) DOE documentation of the technical basis for screening of criticality FEPs in the performance assessment.

### **5.1 U.S. NUCLEAR REGULATORY COMMISSION REVIEW OF U.S. DEPARTMENT OF ENERGY SITE CHARACTERIZATION PLAN AND STUDY PLANS**

The NRC review of the DOE Site Characterization Plan (U.S. Nuclear Regulatory Commission, 1989) and Study Plan 8.3.1.3.2.1 resulted in nine comments and four questions related to subject matter within the ENFE KTI. Since the time of the Site Characterization Analysis, DOE has adopted a revised program plan (U.S. Department of Energy, 1996a) and the Repository Safety Strategy, Revision 2 (U.S. Department of Energy, 1998a). The safety strategy included a number of hypotheses concerning safety attributes of the proposed high-level waste repository (U.S. Department of Energy, 1998a). DOE's refocused program, a result of Congressional direction (U.S. Department of Energy, 1996a; p. 11), reflects the predecessor to the Repository Safety Strategy (U.S. Department of Energy, 1996b). Now the DOE program focuses on the remaining technical questions that have been demonstrated, through total system performance assessment analyses, to be important to waste containment and isolation. As a result of the refocused program, many of the study plans proposed in the Site Characterization Plan have changed in scope, or been deferred or canceled (U.S. Department of Energy, 1997a; Appendices A and G). NRC has refocused its prelicensing program to address those issues most significant to repository performance (Sagar, 1997). In addition, the proposed NRC site-specific regulations for the proposed repository are performance based (U.S. Nuclear Regulatory Commission, 1999a). Some of the Site Characterization Analysis and Study Plan comments are no longer valid as the result of the changes in the overall DOE program and the NRC refocused program. Additional information from both the DOE and ongoing work by the NRC and Center for Nuclear Waste Regulatory Analyses staff has become available to close open items. Two of the Site Characterization Analysis comments were closed in Revision 0 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1997b). The rest of these open items are closed with respect to the Site Characterization Analysis and Study Plan. However, the technical concerns of some of these issues remain with the staff. These concerns have been incorporated into the discussions in Sections 5.4.1 - 5.4.5. The NRC disposition of the Site Characterization Analysis and Study Plan comments and questions is provided below and is summarized in Table 5-2.

#### **Site Characterization Analysis Comment 25:**

The Site Characterization Plan does not provide the rationale for additional testing to obtain information on the effects of package degradation products and interactions between and among radionuclides on sorption (U.S. Nuclear Regulatory Commission, 1989, p. 4-29).

NRC DISPOSITION OF COMMENT: CLOSED

**Table 5-2. Summary of Evolution of the Near-Field Environment (ENFE) Key Technical Issue (KTI) open item status**

Item ID	Status	Comments on Site Characterization and Study Plans	Reference
OSC0000001347C025	Closed	No rationale for additional testing to determine the influence of package degradation products and radionuclide interactions on sorption	This report
OSC0000001347C029	Closed	Lack of study of the effects of radioactive decay heat, the nuclear radiation field, and the effect of certain microorganisms introduced during site construction	U.S. Nuclear Regulatory Commission (1997b)
OSC0000001347C079	Closed	No demonstration that waste package corrosion test environment is representative of actual repository environment	U.S. Nuclear Regulatory Commission (1998d)
OSC0000001347C081	Closed	Assumptions regarding waste container surface wetness for cracking behavior studies inconsistent with information presented for corrosion model	U.S. Nuclear Regulatory Commission (1998d)
OSC0000001347C084	Closed	Incomplete consideration of potential conditions and events which could impact waste package and engineered barrier system issue resolution strategies and testing programs	U.S. Nuclear Regulatory Commission (1998d)
OSC0000001347C089	Closed	Use of grouts, cements, and organic materials in the repository can alter pH and affect corrosion and leach rates	U.S. Nuclear Regulatory Commission (1998d)
OSC0000001347C090	Closed	Effects of varying oxygen concentrations on waste package corrosion are not considered in the waste package environment model	U.S. Nuclear Regulatory Commission (1998d)
OSC0000001347C092	Closed	Delineation of the boundary of the disturbed zone does not include changes in physical or chemical properties as a result of waste-generated heat	U.S. Nuclear Regulatory Commission (1997b)

**Table 5-2. Summary of Evolution of the Near-Field Environment (ENFE) Key Technical Issue (KTI) open item status (cont'd)**

Item ID	Status	Comments on Site Characterization and Study Plans	Reference
OSC0000001347Q030	Closed	Expected quality of water that may contact waste packages and changes in quality over time	This report
OSP0000831321C001	Closed	Inadequate information collected to determine textural relationships of minerals along groundwater pathways	U.S. Nuclear Regulatory Commission (1999f)
OSP0000831321Q002	Closed	Potential for bias in retardation calculations from use of vertical core thin sections to obtain groundwater pathway mineral data	U.S. Nuclear Regulatory Commission (1999f)
OSP0000831321Q004	Closed	Methods for determining changes in lithology for mineral studies	U.S. Nuclear Regulatory Commission (1999f)
OSP0000831321Q005	Closed	The difference between software and model verification/validation for mineralogy studies	U.S. Nuclear Regulatory Commission (1999f)

Comment 25 relates to Subissue 3 (chemical environment for radionuclide release from the engineered barrier system) and Subissue 4 (effects of coupled THC processes on transport of radionuclides through engineered and natural barriers). Although the Container Life and Source Term KTI has lead responsibility for this comment, the ENFE KTI also considered the comment important to issue resolution. We considered this comment open, based on the decision that the DOE commitment to study the effects of microbial activity in the near-field environment, in itself, was not sufficient reason to resolve the comment (U.S. Nuclear Regulatory Commission, 1991). Comment 25 raised two issues: effects of waste package degradation products on sorption, and interaction among and between radionuclides on sorption. We consider interaction between and among radionuclides insignificant in sorption processes because of the low solubilities of radionuclides. NRC technical concerns are embodied in the DOE Repository Safety Strategy, Revision 2, Hypotheses 13 and 14.

DOE proposed to evaluate the potential impact to performance of waste package degradation products within its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). We still consider the effects of degradation products on repository performance important. Our review of the TSPA-VA confirms that the technical concern of this comment remains with the staff. For instance, the effects of the alteration of cladding and basket materials on the chemical within the waste packages were omitted (also addressed in Section 5.4.3). Sorption in the engineered barrier system was only evaluated for the invert of the drift. The TSPA-VA approach did not address the effect of waste package degradation products on radionuclide transport. These concerns are also addressed in Section 5.4.4.

**Site Characterization Analysis Comment 29:**

This comment concerns activities to evaluate the effects of radioactive decay heat, the nuclear radiation field, and the effect of non-site specific microorganisms introduced during site construction (U.S. Nuclear Regulatory Commission, 1989; p. 4-32).

**NRC DISPOSITION OF COMMENT: CLOSED**

Comment 29 relates to Subissue 2 (waste package lifetime), Subissue 3 (rate of release of radionuclides from breached waste packages), and Subissue 4 (transport of radionuclides through engineered barriers and natural barriers). We considered this comment resolved based on the DOE commitment to study the effects of microbial activity in the near-field environment (U.S. Nuclear Regulatory Commission, 1991). DOE stated that this work would be covered in Study Plan 8.3.4.2.4.1 (Characterization of Chemical and Mineralogical Changes in the Post-Emplacement Environment). The original comment is considered resolved, however the staff, through its focused review of the evolving DOE program, will track progress in the DOE characterization of microbial effects as part of the issue resolution process for the ENFE KTI.

**Site Characterization Analysis Comment 79:**

It has not been demonstrated that the test environment in waste package corrosion tests is fully representative of the repository environment (U.S. Nuclear Regulatory Commission, 1989; p. 4-66).

## NRC DISPOSITION OF COMMENT: CLOSED

Comment 79 relates to Subissue 2 (chemical environment of waste package). The ENFE KTI has lead responsibility for this comment. We considered this comment open because DOE indicated that the test environments for waste package corrosion tests will evolve as site data and detailed designs become available (U.S. Nuclear Regulatory Commission, 1991). The NRC technical concerns are embodied in Hypothesis 7 of the DOE Repository Safety Strategy, Revision 2.

DOE proposed to evaluate the potential impact to performance of the waste package chemical environment within the waste package degradation module of its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). The DOE model was expected to include the interaction between inner corrosion resistant alloy and outer corrosion allowance material. Crevice formation between the outer barrier or outer barrier corrosion products and the inner barrier could occur. This would cause pH reduction in the crevice. This reduction in pH is due to hydrolysis of dissolved metal ions from corrosion of both barriers. Accumulation of corrosion products inside the crevice was proposed to be considered in the TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b, p. 6-60).

The chemistry within pits and crevices may vary substantially from the bulk fluid chemistry in contact with the waste package surface (Sridhar, Lichtner, and Dunn, 1996). DOE has chosen Alloy 22 as the corrosion resistant alloy. The most plausible chemical environment under which this alloy can undergo localized corrosion (Brossia, Dunn, and Sridhar, 1998) is associated with the reduction of pH and generation of concentrated  $\text{FeCl}_3$  solutions that may occur in crevices (Sridhar, Lichtner, and Dunn, 1996). We still consider the test environment in waste package corrosion tests important and address this item further in Section 5.4.2.

The Container Life and Source Term KTI resolved this comment within their IRSR as a result of an Appendix 7 meeting on DOE's waste package testing program that occurred in February 1998 (U.S. Nuclear Regulatory Commission, 1998d). The testing program is discussed in Wang, et al. (1998). The ENFE staff concurs with the conclusion of the Container Life and Source Term KTI. This comment is resolved.

### **Site Characterization Analysis Comment 81:**

Investigation into the stress corrosion behavior of the container alloys assumes that the container surface will be either homogeneously dry or homogeneously wet, but in the corrosion model (7.4.5.4.6), it stated that "the waste package will most likely not be uniformly wet."

## NRC DISPOSITION OF COMMENT: CLOSED

The Container Life and Source Term KTI has lead responsibility for Comment 81. This comment was open because DOE indicated that the test environments for waste package corrosion tests will evolve as site data and detailed designs become available (U.S. Nuclear Regulatory Commission, 1991). Discussions with the Container Life and Source Term KTI team suggest that this comment does not require consideration of the chemistry of the waste package environment. Thus, Comment 81 is judged to be outside the scope of the ENFE KTI.

The Container Life and Source Term KTI resolved this comment within their IRSR as a result of an Appendix 7 meeting on DOE's waste package testing program that occurred in February 1998 (U.S. Nuclear Regulatory Commission, 1998d). The testing program is discussed in Wang, et al. (1998). The ENFE staff concurs with the conclusion of the Container Life and Source Term KTI. This comment is resolved.

**Site Characterization Analysis Comment 84:**

The issue resolution strategies and testing programs for design of the waste package and the engineered barrier system do not take into account the full range of reasonable likely natural conditions ("anticipated processes and events") that, with current understanding of the site, might be expected to affect performance of these barriers (U.S. Nuclear Regulatory Commission, 1989; p. 4-68).

NRC DISPOSITION OF COMMENT: CLOSED

The Container Life and Source Term KTI has lead responsibility for Comment 84. We considered Comment 84 open because the tests and analyses did not reflect the full range of potential anticipated processes and events and, as need be, unanticipated processes and events (U.S. Nuclear Regulatory Commission, 1991). Discussions with the Container Life and Source Term KTI team suggest that this comment does not require consideration by the ENFE KTI. Thus, Comment 84 is judged to be outside the scope of the ENFE KTI.

The Container Life and Source Term KTI resolved this comment within their IRSR as a result of an Appendix 7 meeting on DOE's waste package testing program that occurred in February 1998 (U.S. Nuclear Regulatory Commission, 1998d). The testing program is discussed in Wang, et al. (1998). The ENFE staff concurs with the conclusion of the Container Life and Source Term KTI. This comment is resolved.

**Site Characterization Analysis Comment 89:**

Grouts, cements, and organic materials used in the repository may change the local pH of the repository and affect corrosion of the metal waste containers and the local leach rates of radionuclides from the glass (U.S. Nuclear Regulatory Commission, 1989; p. 4-71).

NRC DISPOSITION OF COMMENT: CLOSED

Comment 89 relates to Subissue 2 (chemical environment of the waste package), Subissue 3 (chemical environment for radionuclide release from the engineered barrier system), and Subissue 4 (transport of radionuclides through engineered and natural barriers). The ENFE KTI has lead responsibility for this comment, and the Container Life and Source Term KTI also considers the comment important to issue resolution. We have considered Comment 89 open. DOE indicated that testing programs will investigate how water chemistry is changed by the waste package and other repository materials. DOE also indicated that testing programs would investigate how such changes affect the corrosion of the containers and the leaching of radionuclides. However, no details were provided (U.S. Nuclear Regulatory Commission, 1991). DOE intends to submit a study plan (8.3.4.2.4.5 - Effects of man-made materials on water chemistry) to the NRC to address this issue and to submit a supplemental response to this comment to the NRC (U.S. Department of Energy, 1997a).

In addition, DOE has proposed to evaluate the potential importance of variation in the pH on the performance of the waste package and glass waste form in its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). We still consider the introduction of large quantities of cements potentially important to repository performance. We address this issue further in Sections 5.4.2 and 5.4.3.

The Container Life and Source Term KTI partially resolved this comment within their IRSR as a result of an Appendix 7 meeting on DOE's waste package testing program that occurred in February 1998 (U.S. Nuclear Regulatory Commission, 1998d). The testing program is discussed in Wang, et al. (1998). The Container Life and Source Term IRSR indicated that a full range of the evolving chemical environments at the proposed Yucca Mountain repository should be considered in the long-term glass corrosion studies (U.S. Nuclear Regulatory Commission, 1998d, p. 55). The ENFE staff concurs with the conclusion of the Container Life and Source Term KTI. Thus, the technical concerns of this comment remain with the staff and are discussed in Section 5.4.3.

**Site Characterization Analysis Comment 90:**

The effects of varying oxygen concentrations on corrosion of the metal canisters are not considered (U.S. Nuclear Regulatory Commission, 1989; p. 4-71).

NRC DISPOSITION OF COMMENT: CLOSED

Comment 90 is related to Subissue 2 (chemical environment of waste package). The Container Life and Source Term KTI has lead responsibility for this comment, and the ENFE KTI also considers the comment important to issue resolution. This comment was considered open based on DOE's response. DOE indicated that the effects of varying oxygen concentration on the corrosion of the metal container will be considered when site data, detailed designs, and performance scenarios are available (U.S. Nuclear Regulatory Commission, 1991).

Ongoing long-term corrosion tests using corrosion tanks and relative humidity chambers are being performed at Lawrence Livermore National Laboratory in support of DOE's waste package program (Wang, et al., 1998). These tests are designed to evaluate the behavior of container materials under air-saturated conditions. This test environment is conservative and is consistent with the quick return of ambient oxygen levels as the repository cools that has been predicted in coupled THC processes modeling of the near field (Lichtner, 1997). Thus, this comment is considered to be closed by the ENFE KTI, based on the testing program.

The Container Life and Source Term KTI also resolved this comment within their IRSR as a result of an Appendix 7 meeting on DOE's waste package testing program that occurred in February 1998 (U.S. Nuclear Regulatory Commission, 1998d). The testing program is discussed in Wang, et al. (1998). The ENFE staff concurs with the conclusion of the Container Life and Source Term KTI. This comment is resolved.

**Site Characterization Analysis Comment 92:**

The approach for delineation of the boundary of the disturbed zone does not include all physical or chemical properties which will have changed as a result of heat generated by the emplaced radioactive wastes such that the resultant change of properties may have a significant effect on

the performance of the geologic repository (U.S. Nuclear Regulatory Commission, 1989; p. 4-72).

NRC DISPOSITION OF COMMENT: CLOSED

Comment 92 relates to all of the subissues of the ENFE KTI. We considered this comment resolved based on DOE's response which indicates Activity 1.6.5.2 (Definition of the Disturbed Zone) will reevaluate and if necessary refine the boundary of the disturbed zone. This ongoing activity may be a result of changes in the NRC guidance and in DOE's understanding of repository property effects (U.S. Nuclear Regulatory Commission, 1991). The original comment is considered resolved. However, the staff, through its focused review of the evolving DOE program, will track progress in DOE's characterization of the ENFE and its impact on performance as part of the issue resolution process within the ENFE KTI.

**Site Characterization Analysis Question 30:**

It is stated that the expected quality of the water is such that it will have little impact on the long-term integrity of the waste packages. What is the expected quality of the water and how might this quality vary over the lifetime of the repository (U.S. Nuclear Regulatory Commission, 1989; p. 4-115)?

NRC DISPOSITION OF COMMENT: CLOSED

Question 30 addresses Subissue 2 (chemical environment of waste package) and is related to concerns in Subissue 3 (chemical environment for radionuclide release from the engineered barrier system) and Subissue 4 (transport of radionuclides through engineered and natural barriers). The ENFE KTI has lead responsibility for this question, and the Container Life and Source Term KTI also considers the question important to issue resolution. We considered this question open because DOE indicated that the expected water quality will be unknown until the results of activities in Study Plan 8.3.4.2.4.1 are completed. The DOE is addressing resolution of this comment in a site characterization framework (Wang, et al., 1998).

In addition, DOE has proposed to evaluate the potential impact to performance of the waste package chemical environment within the waste package degradation module in its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). We still consider the chemical environment (water quality) of the waste package to be important to repository performance. The technical concerns of this comment remains with the staff and are addressed in Section 5.4.2.

**STUDY PLAN 8.3.1.3.2.1**

**Comment 1:** Although the Study Plan calls for gathering data on "textural relationships of minerals along potential groundwater pathways," which are important to establish stratigraphic location of core samples and to determine the accessibility of potentially sorbing phases to radionuclides, only the candidate host rock will be analyzed petrographically. Thus, it appears that inadequate information will be collected on rock outside the repository. The NRC staff recommended that the study include petrographic analysis for determining textural relationships of minerals along the transport pathways between Topopah Spring Tuff and the accessible environment.

NRC DISPOSITION OF COMMENT: CLOSED

The comment was incorporated from the Open Item Tracking System. The tracking system indicated that the comment was still open and was not being addressed in an IRSR. Based on a review of this comment (U.S. Nuclear Regulatory Commission, 1992), this comment is closed. The basis for closure of the comment was previously documented (U.S. Nuclear Regulatory Commission, 1992).

**STUDY PLAN 8.3.1.3.2.1**

**Question 2:** Could the effect of characterizing thin sections of core primarily cut in a vertical orientation significantly bias the estimations of types, abundances, distributions, compositions, and textural relationships of minerals along potential groundwater pathways such that calculated radionuclide retardation would be overestimated?

NRC DISPOSITION OF COMMENT: CLOSED

The question was incorporated from the open item tracking system. This database indicated that the question was still open and was not being addressed in an IRSR. Based on a review of this comment (U.S. Nuclear Regulatory Commission, 1992), this comment is closed. The basis for closure of the comment was previously documented (U.S. Nuclear Regulatory Commission, 1992).

**STUDY PLAN 8.3.1.3.2.1**

**Question 4:** What is the method for determining changes in lithology?

NRC DISPOSITION OF COMMENT: CLOSED

The question was incorporated from the open item tracking system. This database indicated that the question was still open and was not being addressed in an IRSR. Based on a review of this comment (U.S. Nuclear Regulatory Commission, 1992), this comment is closed. The basis for closure of the comment was previously documented (U.S. Nuclear Regulatory Commission, 1992).

**STUDY PLAN 8.3.1.3.2.1**

**Question 5:** What is the difference between software verification and validation and model verification and validation?

NRC DISPOSITION OF COMMENT: CLOSED

The question was incorporated from the open item tracking system. This database indicated that the question was still open and was not being addressed in an IRSR. Based on a review of this comment (U.S. Nuclear Regulatory Commission, 1992), this comment is closed. The basis for closure of the comment was previously documented (U.S. Nuclear Regulatory Commission, 1992).

## **5.2 U.S. NUCLEAR REGULATORY COMMISSION REVIEW OF U.S. DEPARTMENT OF ENERGY THERMAL MODELING AND TESTING PROGRAM**

We commented on the DOE Thermohydrology Testing and Modeling Program (U.S. Nuclear Regulatory Commission, 1997a). The three comments addressed the effects of the thermal perturbation on flow in the near field and DOE's efforts to conservatively bound the effects of THC coupled processes on repository performance. Our comment on conservatively bounding the effects of coupled THC on repository performance is applicable to each of the subissues in the ENFE IRSR. Only Comment 3 in the letter (U.S. Nuclear Regulatory Commission, 1997a) will be addressed in this IRSR and the other two comments were addressed in the TEF IRSR (U.S. Nuclear Regulatory Commission, 1998a). Comment 3 addressed whether the DOE testing and modeling strategy included means for bounding the effects of THC coupled processes on repository performance.

The DOE responded (U.S. Department of Energy, 1997b) to the comment on THC coupling by indicating significant progress has been achieved in modeling THC coupled effects. They cited several recent publications that document this progress. These documents were reviewed and additional information on the thermal testing program has been gained by our participation in the thermal testing workshops and near-field/altered-zone expert elicitation. It is clear that major progress has been made in the experiments (laboratory and field thermal tests) and modeling efforts to examine and bound coupled THC processes (CRWMS M&O, 1998c). Progress has been made in process-level modeling and results have been obtained from the experiments and modeling efforts. However, DOE has not fully evaluated the potential impacts on repository performance. A key assumption in the TSPA-VA methods and assumptions report was that mechanical and chemical changes do not alter hydrologic properties (TRW Environmental Safety Systems, Inc., 1997b). The DOE has stated that "simplifications that patch thermal-mechanical and/or thermal-chemical influences into an unsaturated zone thermal-hydrologic simulation have been proposed as a series of sensitivity studies" (TRW Environmental Safety Systems, Inc., 1997b, p. 6-20). We consider the proposed sensitivity studies important in determining whether THC processes significantly impact repository performance. Our concern on the bounds of coupled THC processes contained in Comment 3 of Comments on the Department of Energy Thermohydrology Testing and Modeling Program (U.S. Nuclear Regulatory Commission, 1997a) is considered resolved. We will review the TSPA-SR to ensure that the effects of coupled THC processes on repository performance have been adequately considered assessed in the repository performance calculations. The other two comments were addressed in the TEF IRSR (U.S. Nuclear Regulatory Commission, 1998a) and were resolved. Thus, all comments associated with our letter to the DOE on their Thermohydrology Testing and Modeling (U.S. Nuclear Regulatory Commission, 1997a) are resolved.

## **5.3 EVOLUTION OF THE NEAR-FIELD GEOCHEMICAL ENVIRONMENT CONCERNS WITHIN U.S. DEPARTMENT OF ENERGY'S PERFORMANCE ASSESSMENTS**

We have provided comments to DOE on its performance assessment activities conducted prior to the publication of the Viability Assessment (U.S. Nuclear Regulatory Commission, 1996, 1998g, 1999b). Many conservatisms were considered in TSPA-95. However, several potential

waste package failure modes were not considered and, as a result, the calculations may be non-conservative (U.S. Nuclear Regulatory Commission, 1996). We raised concerns related to DOE's humid-air corrosion model. The model used by DOE did not account for the effect of aggressive groundwater. Also, modifications of the environment by the presence of corrosion products and the evolution of the chemical composition of the environment with time were not considered. We also questioned if episodic wetting and drying might increase the corrosion rate (U.S. Nuclear Regulatory Commission, 1996). These issues remain a concern with regard to performance of the outer layer of the waste package. We will evaluate how DOE accounts for these effects in its evaluation of waste package and total system performance in TSPA-SR.

It was noted that possible geochemical variations in the near-field environmental conditions were considered reasonably well in the TSPA-95 (U.S. Nuclear Regulatory Commission, 1996). Sensitivity analyses using codes capable of handling coupled processes can be implemented to address the effect of near-field chemistry on the solubility of radionuclides. There were empirical relationships for solubilities of neptunium, plutonium, and americium as a function of pH and temperature in TSPA-95. Sampling on pH would result in concentrations of these radionuclides responding to changes in near-field chemistry in a reasonable manner. DOE has proposed to explicitly account for variations in near-field chemistry in its waste form abstraction in TSPA-SR (CRWMS M&O, 1999a).

NRC has stated its continuing concerns with some aspects of the DOE performance assessment as they relate to an acceptable license application (U.S. Nuclear Regulatory Commission, 1998g, 1999b). Some of the concerns address issues contained in this IRSR. We will review the TSPA-SR information to ensure that all significant features and processes associated with the evolution of the near-field environment were considered by DOE in its performance assessment.

Uncertainty in near-field environmental conditions, as it impacts waste package performance, does not appear to be fully captured by the performance assessment modeling (U.S. Nuclear Regulatory Commission, 1998g, 1999b). There are two concerns arising from the expert elicitations that the DOE has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c). These concerns are that the propagation of uncertainty in near-field environmental conditions is impeded by the use of point values for the corrosion potential. In addition, the expected use of the elicited expert judgements needs to be understood prior to convening the expert panel. For instance, the results from the near-field/altered-zone expert elicitation could not be used to develop the initial and boundary conditions for the waste package corrosion panel. This situation arose because the waste package elicitation was completed prior to the initiation of the near field elicitation. Our concerns on the DOE performance assessments will be evaluated by applying the acceptance criteria to the TSPA-SR.

#### **5.4 STATUS OF SUBISSUE RESOLUTION AT THE STAFF LEVEL**

Revision 1 of the ENFE IRSR focused on developing somewhat general acceptance criteria for all subissues, including the subissue of criticality with the near-field environment. This systematic approach was taken to issue resolution within the ENFE IRSR, as limited performance assessment calculations were available to refine the acceptance criteria to be repository- and design-specific, and performance-based. A technical approach has been presented to evaluate the potential for microbial activity in the near-field environment.

Revision 2 of the ENFE IRSR described the status of issue resolution as a result of the application of the acceptance criteria to DOE's TSPA-VA. In Revision 2, the NRC provided DOE with its evaluation of the Viability Assessment (U.S. Nuclear Regulatory Commission, 1999b). The staff identified several challenges for the DOE to assemble a complete and high-quality license application within the time frame envisioned in the license application plan. Staff reviewed the preliminary design concept, the total system performance assessment, and the license application plan, as well as supporting documents such as the TSPA-VA technical basis document and the near-field/altered-zone models report. Our comments on TSPA-VA were provided to the DOE and addressed key elements of the DOE performance assessment (U.S. Nuclear Regulatory Commission, 1999b). The review approach used to comment on the TSPA-VA is described in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 1998c, 2000a). Review of each of the performance assessment abstractions is based on five technical acceptance criteria (U.S. Nuclear Regulatory Commission, 1998c): (i) integration, (ii) data and model justification, (iii) data uncertainty, (iv) model uncertainty, and (v) model support.

Revision 3 updates the status of issue resolution as a result of the application of acceptance criteria to the preliminary draft DOE Process Model Reports, and preliminary draft Analysis and Model Reports and other recent information that was available prior to May 15, 2000. The preliminary reports reflect the repository concept and design selected by DOE for its TSPA-SR. Each ENFE subissue is pertinent to one or more model abstractions or integrated subissues within the performance assessment. The model abstractions which require input from the ENFE subissue are reiterated in introductory comments for each subissue. For each ENFE subissue, a summary of DOE's approach for the relevant integrated subissues is provided, followed by our evaluation of DOE's approach. The staff evaluation was based on the application of acceptance criteria that may be included in the Yucca Mountain Review Plan. These acceptance criteria are guidance for the staff and, indirectly, for DOE as well. Note that the acceptance criteria previously listed in Section 4.0 of the Evolution of the Near-Field Environment (ENFE) IRSR Revision 2 are being used to develop the Yucca Mountain Review Plan. In the process, they have undergone some modifications. The acceptance criteria listed in Section 5.0 are consistent with and reflect, to the extent practicable, those modifications.

Our review of DOE's approach presented in this revision of the ENFE IRSR is considered preliminary in nature. Only some of the DOE Process Model Reports, and Analysis and Model Reports considered relevant to the ENFE subissues were available for review at the time this revision was prepared (those received prior to May 15, 2000, were reviewed), and most of them were preliminary draft versions. The NRC staff recognizes the preliminary nature of the draft Analysis and Model Reports and Process Model Reports; specifically, they have not been accepted by the DOE. Thus, the NRC staff has not used the information contained in those draft documents to resolve any open subissues in this report. To aid the issue resolution process, however, the staff has reviewed and provided comments on the sufficiency of the information in the preliminary documents to address staff concerns. After a review of the final Process Model Reports or other documents that indicate DOE's acceptance of the information in the preliminary documents, the staff will consider whether it is appropriate to close the subissues or any portion the subissues. Nevertheless, most of those preliminary draft Process Model Reports and preliminary draft Analysis and Model Reports that were reviewed were responsive to the subissues and concerns identified in Revision 2 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1999f). This responsiveness helped to facilitate our review of those reports. Our review will continue as other Process Model Reports and Analysis and Model Reports become available, and will be documented.

#### **5.4.1 Subissue 1: Effects of Coupled Thermal-Hydrologic-Chemical Processes on Seepage and Flow**

DOE's approach to assess the effects of coupled THC processes on seepage and flow must meet the following model abstraction acceptance criteria for each relevant integrated subissue: (i) integration, (ii) data and model justification, (iii) data uncertainty, (iv) model uncertainty, and (v) model support. Quality assurance is handled in a separate section of the Yucca Mountain Review Plan.

One integrated subissue that requires input from the ENFE within the scope of the seepage and flow subissue is Flow Paths in the Unsaturated Zone (U.S. Nuclear Regulatory Commission, 2000a). The features and processes listed in Section 4.1 need to be considered in the evaluation of this abstraction. As part of this evaluation, the staff will use the acceptance criteria to review the preliminary draft DOE Process Model Reports and the supporting preliminary draft Analysis and Model Reports as they become available. This evaluation will also include a review of DOE's treatment of the relevant features, events, and processes (FEPs). Our evaluation will focus on those FEPs that have been excluded from the performance assessment to ensure that an adequate technical basis for their exclusion has been provided by DOE. In addition, the model abstraction acceptance criteria will be applied to those FEPs included in the performance assessment.

The review documented in this revision of the ENFE IRSR focused on a subset of the preliminary draft Analysis and Model Reports and preliminary draft Process Model Reports that reflects the importance of the abstractions to DOE's safety case and relevance to ENFE Subissue 1. The following Analysis and Model Reports are relevant to the ENFE subissue on seepage and flow and are a subset of those in DOE's Analysis and Model Reports/Process Model Reports schedule: Process Model Report on Unsaturated Zone Flow and Transport, U0075—Analysis and Model Report on Seepage Model for Performance Assessment, N0120—Analysis and Model Report on Drift Scale Coupled Processes, U0120—Analysis and Model Report on Drift Seepage Including Coupled Processes, U0105—Analysis and Model Report on Mountain Scale Coupled Processes, and N0080—FEPs for the Near-Field Environment.

Of the relevant Analysis and Model Reports and Process Model Reports, the preliminary draft Analysis and Model Report on Seepage Model for Performance Assessment [U0075 (CRWMS M&O, 2000c)], the preliminary draft Analysis and Model Report on Drift Seepage Including Coupled Processes [U0120 (CRWMS M&O, 2000d)], and the preliminary draft Process Model Report on Unsaturated Zone Flow and Transport (CRWMS M&O, 2000e) were available for review. In the following sections, the resolution status is based on a review of these three documents and the License Application Design S report (CRWMS M&O, 1999b), the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), and Revision 3 of the Repository Safety Strategy (CRWMS M&O, 2000a). The resolution status of this subissue will be further updated as more Analysis and Model Reports and Process Model Reports become available for review.

#### **5.4.1.1 U.S. Department of Energy Approach**

According to Revision 3 of the Repository Safety Strategy, “principal factors” are central to determining and demonstrating long-term safety of the repository system, and will be the focus of the postclosure safety case for the site recommendation and license application. Three of the principal factors that were identified by DOE for the enhanced repository system are potentially affected directly by THC-induced changes to flow properties: (i) limited seepage of water into the emplacement drifts, (ii) retardation of radionuclide migration in the unsaturated zone, and (iii) dilution of radionuclide concentrations during migration. The TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) outlines how these factors will be addressed by abstractions and process-level models in the TSPA-SR to provide the technical basis for the postclosure safety case.

A major assumption embedded in the TSPA-VA was that coupled THC effects were of a transient nature and would not result in long-term changes to rock flow properties. DOE has taken significant steps to develop technical support for this assumption in the reports supporting the TSPA-SR. In particular, abstractions and process-level models are being refined to take into account the potential effects of the new repository design and to improve THC models. THC effects on flow are treated in a series of sensitivity analyses. The sensitivity studies considered only a limited number of parameter states (combinations of important variables). Where appropriate, the results of these sensitivity analyses will be used to update hydrological properties in abstractions of relevant processes.

In Section 5.4.1.1.1, DOE’s screening of FEPs is discussed. In Section 5.4.1.1.2, additional information is provided on DOE’s treatment of individual physical processes that form components of the DOE principal factors. Those components are also potentially affected by THC-induced changes to flow properties. The information is based on Revision 3 of the Repository Safety Strategy, the TSPA-SR methods and assumptions report, and the preliminary draft Unsaturated Zone Flow and Transport Process Model Report. Supporting Analysis and Model Reports were not available for review prior to May 15, 2000. The focus of our review is on those concerns relevant to the ENFE Subissue 1 on effects of coupled THC processes on seepage and flow. Additional information about unsaturated zone flow concerns is provided in the Unsaturated and Saturated Flow under Isothermal Conditions IRSR (U.S. Nuclear Regulatory Commission, 1999e).

##### **5.4.1.1.1 Features, Events, and Processes Screening**

DOE did not include a formal screening process for FEPs in the Viability Assessment. As a result, many important design features, physical phenomena, and couplings were not evaluated in a performance assessment framework. Recently, DOE has developed a more formal documentation of the FEPs identification and screening process for the TSPA-SR. The contents of the preliminary draft DOE FEP database (U.S. Department of Energy, 1999) have been reviewed and those related to the integrated subissue on flow paths in the unsaturated zone and the ENFE subissue on seepage and flow have been identified (Pickett and Leslie, 1999; U.S. Nuclear Regulatory Commission, 2000a). Those FEPs and the screening arguments, as reflected in the preliminary draft DOE FEP database (U.S. Department of Energy, 1999), are presented in Table 5-3. Our evaluation of DOE’s screening of FEPs is presented in Section 5.4.1.2.

**Table 5-3. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 1 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissue on flow paths in the unsaturated zone**

<b>Features, Events, and Processes Number</b>	<b>Features, Events, and Processes Name</b>	<b>Preliminary DOE Screening</b>
1.1.02.00.00	Excavation/construction	Exclude
1.1.02.02.00	Effects of preclosure ventilation	not given
1.1.07.00.00	Repository design	Include (exclude deviations from design)
1.1.08.00.00	Quality control	Include (exclude defects and deviations)
1.1.12.01.00	Accidents and unplanned events during operation	Exclude
1.1.13.00.00	Retrievability	Include
1.2.02.01.00	Fractures	Include (existing, reactivated), Exclude (new, SZ)
1.2.04.02.00	Igneous activity causes changes to rock properties	Include
1.2.06.00.00	Hydrothermal activity	Exclude
1.2.08.00.00	Diagenesis	Exclude
2.1.01.02.00	Codisposal/colocation of waste	Include
2.1.02.08.00	Pyrophoricity	Exclude
2.1.04.03.00	Erosion or dissolution of backfill	Exclude
2.1.04.05.00	Backfill evolution	Include
2.1.05.03.00	Seal degradation	Include
2.1.06.01.00	Degradation of cementitious materials in drift	Include
2.1.06.02.00	Effects of rock reinforcement materials	Include
2.1.06.03.00	Degradation of the liner	Include
2.1.06.04.00	Flow through the liner	Exclude
2.1.06.05.00	Degradation of invert and pedestal	Include
2.1.06.06.00	Effects and degradation of drip shield	unspecified
2.1.06.07.00	Effects at material interfaces	Include
2.1.08.01.00	Increased unsaturated water flux at the repository	Include
2.1.08.02.00	Enhanced influx (Philip's drip)	Include
2.1.08.05.00	Flow through invert	Include

**Table 5-3. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 1 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissue on flow paths in the unsaturated zone (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening
2.1.08.08.00	Induced hydrological changes in the waste and EBS	Include
2.1.08.11.00	Resaturation of repository	Include
2.1.09.12.00	Rind (altered zone) formation in waste, EBS, and adjacent rock	Include
2.1.11.02.00	Nonuniform heat distribution/edge effects in repository	Include
2.2.01.02.00	Thermal and other waste and EBS-related changes in the adjacent host rock	Include
2.2.01.03.00	Changes in fluid saturations in the EDZ	Include
2.2.03.02.00	Rock properties of host rock and other units	Include
2.2.07.05.00	Flow and transport in the UZ from episodic infiltration	Include
2.2.07.06.00	Episodic/pulse release from repository	not given
2.2.07.07.00	Perched water develops	Exclude
2.2.07.15.06	Convection (water transport)	Exclude
2.2.07.15.07	Dispersion (water transport)	Include
2.2.08.01.00	Groundwater chemistry/composition in UZ and SZ	Include
2.2.10.01.00	Repository-induced thermal effects in geosphere	Include
2.2.10.06.00	Thermo-chemical alteration (solubility, speciation, phase changes, precipitation/dissolution)	Include
2.2.10.07.00	Thermo-chemical alteration of the Calico Hills unit	Include
2.2.10.09.00	Thermo-chemical alteration of the Topopah Spring basal vitrophyre	Include
2.2.10.10.00	Two-phase buoyant flow/heat pipes	Include
2.2.10.11.00	Natural airflow in UZ	Include
2.2.10.12.00	Geosphere dryout due to waste heat	Include
2.2.11.01.05	Gas generation and gas sources, far-field	Exclude

**Table 5-3. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 1 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissue on flow paths in the unsaturated zone (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening
2.2.11.02.00	Gas pressure effects	Exclude
3.1.01.01.00	Radioactive decay and ingrowth	Include
SZ = saturated zone      EBS = engineered barrier system UZ = unsaturated zone    EDZ = excavation disturbed zone		

#### 5.4.1.1.2 Flow Paths in the Unsaturated Zone

The integrated subissue on flow paths in the unsaturated zone is concerned with flow paths above and below the repository horizon, as well as seepage into the emplacement drifts. All of these processes are potentially altered by persistent THC-induced changes to rock properties. Detailed discussions of the various processes comprising this integrated subissue can be found in the IRSR for the Unsaturated and Saturated Flow under Isothermal Conditions KTI (U.S. Nuclear Regulatory Commission, 1999e) and in the TSPA I KTI (U.S. Nuclear Regulatory Commission, 2000a).

The approach in the TSPA-SR for flow in the unsaturated zone is similar to that used in the TSPA-VA. A calibrated three-dimensional (3-D) mountain-scale flow model was developed. This mountain-scale model is based on the dual-permeability formulation for fracture/matrix flow with the active-fracture model. The active-fracture model formulation is used to describe the reduced coupling between fractures and matrix expected under unsaturated conditions. Partitioning between the matrix and fracture system is important because flow through the fractures is rapid compared to flow through the matrix. In addition, sorption on minerals is expected to retard radionuclide movement in the matrix but not in the fractures. Hydrologic properties used in the model were obtained by inverse modeling to match measured liquid saturations and other hydrologic variables. These data were obtained in the ambient system, which has not been altered by repository-induced THC effects. THC effects on flow above the repository were not considered in the unsaturated zone flow model; instead, THC models were used in sensitivity studies to demonstrate that bulk changes in hydrologic properties are minor for the ranges of thermal loadings used in the Enhanced Design Alternative-II design.

The supporting THC simulations (CRWMS M&O, 2000e) used the active-fracture dual-continuum formulation for two-phase, nonisothermal flow and chemical transport. The domain of the two-dimensional model extended vertically from the surface to the Calico Hills unit. Horizontally, the model domain spanned one-half of the drift spacing. An unstructured grid with refinements around the drift was used. Equations describing heat, liquid and gas flow, chemical transport, chemical reactions, and porosity and permeability changes were solved sequentially using TOUGHREACT Version 2.2. Two chemical systems were considered, one with a simplified representation of the mineralogy (silica phases, gypsum, and calcite) and a second with a large number of minerals including those representative of the likely secondary phases formed from thermal alteration of zeolitic units. Geochemical data on mineral

abundances and compositions, reaction rates, and water compositions were taken from various sources.

Results of both models show precipitation of calcite in the fracture system in an extended area because of the increase in temperature and the retrograde solubility of calcite. Silica redistribution was confined to regions of limited spatial extent, mostly associated with the position of the boiling front. In all cases, the change in porosity caused by these processes was small compared with the initial porosity. Resulting changes in hydrologic properties are expected to be negligible. These results are in stark contrast to modeling results performed in support of the TSPA-VA, which showed the formation of a robust mineralized cap with reduced permeability above the repository. The reduced THC alteration associated with the Enhanced Design Alternative-II design is a consequence of the increased drift spacing and lower overall heat loading.

The preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e) also contains a discussion on the possibility of matrix sealing by precipitation of silica in a highly localized zone at the fracture-matrix interface. It is argued that the potential for this to occur is small because the kinetic rates for silica dissolution and the silica concentration in the fractures are small. No values are provided for any of these parameters. Theoretical studies by Lichtner, Keating, and Carey (1999) clearly show that the effect is possible and may proceed rapidly relative to the duration of the thermal pulse with some combinations of these parameters. It was also argued that there is little hydrological potential for flow from fractures to matrix (CRWMS M&O, 2000e), a statement that clearly neglects the strong imbibition from fractures to matrix that exists with some saturation conditions.

The potential for thermal alteration of volcanic glasses in the Paintbrush Tuff unit was also addressed in sensitivity studies. The flow fields developed for the TSPA-SR do not allow for possible mineralogical phase changes from volcanic glasses to zeolites and clays. Instead, mountain-scale thermo-hydrologic models were used to calculate temperatures in this unit. The temperatures within the Paintbrush Tuff were calculated to be in the range of 40 to 45 °C with ventilation. Without ventilation, the base of the Paintbrush Tuff reached temperatures of 70 to 75 °C; in areas of low infiltration, the temperatures at the base of the Paintbrush Tuff reached boiling conditions. The preliminary draft Process Model Report provides no technical basis for ignoring possible alterations of glasses at these elevated temperatures (CRWMS M&O, 2000e). In light of the preliminary nature of estimates of the efficiency of ventilation to reduce the heat load of the repository (i.e., no data to support ventilation model), DOE should provide additional clarification of the temperatures expected in the Paintbrush Tuff and additional justification that temperatures in the unit will be too low to affect repository performance.

Drift seepage is an important control on the amount of water contacting the waste package and is potentially altered by THC effects. The approach proposed for drift seepage in the TSPA-SR is similar to that used in the TSPA-VA. Probability distributions are defined for the fraction of waste packages wetted by seepage and for the flow rate in the seeps. These distributions are based on detailed modeling of capillary diversion around the emplacement drifts, as described in preliminary draft U0075—Analysis and Model Report on Drift Seepage Model for Performance Assessment (CRWMS M&O, 2000c).

The drift seepage models used Richard's equation to describe unsaturated flow around the emplacement drifts. The fracture system was treated as an equivalent continuum; flow in the matrix was neglected. The fracture permeability, which is expected to exhibit a high degree of spatial variability, was treated as a random space function. A small number (3 to 5) of realizations was created for this random permeability field using standard geostatistical techniques. Each realization was then used in the solution of Richard's equation to determine a seepage fraction and seep rate. The average permeability for the fracture system was based on air-permeability measurements representative of ambient conditions (i.e., unaltered by coupled THC processes). This average fracture permeability was varied for a range of three orders of magnitude to assess sensitivities to this uncertain parameter. The van Genuchten parameter  $1/\eta$ , which quantifies the strength of moisture retention in the unsaturated fracture system, was treated as spatially constant, except for a small number of sensitivity runs where it was considered related deterministically to the spatially variable fracture permeability. The value of  $1/\eta$  was varied from 30 to 1,000 pascals, which spans the range observed in the ambient system. Other relevant parameters also were considered in the sensitivity studies, including the spatial correlation range, and the variance in  $\ln k_f$ , where  $k_f$  is the fracture permeability. The simulations show that drift seepage decreases with increasing fracture permeability and increasing  $1/\eta$ . Weaker sensitivity was noted to the variance in  $\ln k_f$  and to the spatial correlation range. In general, drift seepage increases with increasing correlation range and increasing  $\ln k_f$  variance.

The potential effects of THC alteration of fracture properties on drift seepage are ignored in the abstractions for the TSPA-SR. This neglect is based on fully coupled THC simulations, which show negligible alteration of the fracture and matrix porosities, as described previously. The reduced degree of mineral precipitation and dissolution associated with the Enhanced Design Alternative-II design is a consequence of the reduced thermal loading.

DOE's preliminary draft abstraction of seepage into drifts (CRWMS M&O, 2000c,d) does take into account one potential THC effect. The seepage flow rate was increased by 55 percent, in part to account for increased seepage along rock bolts. Increased seepage along rock bolts might occur, for example, because of localized dissolution of tuff rock due to chemical interaction with the cementitious grout. The factor of 55 percent is based on results of the drift seepage model with discrete high-permeability features superimposed to represent degraded rock bolts. This factor is considered conservative by the DOE because it is larger than the value calculated from the drift seepage model using the expected number of rock bolts per drift segment (six bolts every 1.5 meters along the drift in non-lithophysal units; CRWMS M&O, 2000d).

Revision 3 of the Repository Safety Strategy also outlines an alternative approach to drift seepage being considered by DOE. This new approach is based on the concept of a seepage threshold—a value of percolation flux below which seepage does not occur. The approach outlined in the Repository Safety Strategy involves determining the seepage threshold from *in situ* measurements. The empirically determined threshold would then be used in performance assessment calculations; that is, drift seepage would not be considered in subareas of the repository where the percolation flux is below the threshold. In practice, this means that seepage will be allowed only in regions of extreme flow. As of May 15, 2000, few details are available on the seepage threshold approach. In particular, it is unclear how DOE plans to justify the neglect of persistent THC-induced changes in rock properties in this approach.

Radionuclide travel time to the water table is strongly controlled by the Calico Hills nonwelded unit. Significant lateral diversion may occur in and above this unit, as indicated by observed regions of perched water. Pathways through the Calico Hills nonwelded unit are controlled by the highly heterogeneous fracture and matrix permeabilities in this unit and by discrete features such as faults. The Calico Hills unit is susceptible to hydrothermal alteration. In particular, the remaining volcanic glass in this unit may be altered to lower permeability zeolites, with important potential consequences for groundwater travel time. This potential alteration was not included in the flow model and abstraction. Expected temperatures at the top of the Calico Hills units were calculated using dual-permeability hydrothermal models. These temperatures reach a maximum of 70 to 75 °C during the period of 2,000 to 7,000 years after closure. A technical basis for neglect of thermal alterations at these elevated temperatures was not provided in the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e).

#### **5.4.1.2 U.S. Nuclear Regulatory Commission Staff Evaluation**

Staff analysis of the DOE approach to abstraction of the effects of coupled THC processes on seepage and flow was conducted based on the acceptance criteria listed in Section 5.4.1.2.1. There are five generic acceptance criteria. Each generic acceptance criteria is followed by specific criteria. The DOE approach to abstract the effects of coupled THC processes on seepage and flow will be acceptable provided that each of the acceptance criteria are met.

The main Analysis and Model Report (N0120—Analysis and Model Report on Drift Scale Coupled Processes) relevant for ENFE Subissue 1 was not available prior to May 15, 2000. The following sections focus on the abstraction architecture described in Revision 3 of the Repository Safety Strategy, the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), and the summary provided in the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e). For this revision of the ENFE IRSR, staff analysis focused on the acceptance criterion on integration. Staff analysis using all five acceptance criteria will be completed as the DOE Analysis and Model Reports become available. An analysis of the FEPs relevant to Subissue 1 of the ENFE IRSR is presented in the first part of this section, followed by discussions specific to the relevant model abstraction integrated subissue.

#### **5.4.1.2.1 Integrated Subissue on Flow paths in the Unsaturated Zone**

##### **5.4.1.2.1.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 1**

Important design features, physical phenomena and couplings, consistent and appropriate assumptions have been incorporated into the abstraction of flow paths in the unsaturated zone in the total system performance assessment and the technical bases are provided. The total system performance assessment abstraction identifies and describes aspects of flow paths in the unsaturated zone and seepage that are important to waste isolation and includes the technical bases for these descriptions. Specifically,

- DOE provides adequate technical bases, including activities such as independent modeling, laboratory or field data, or sensitivity studies, for

exclusion of any thermal-hydrologic-mechanical-chemical (THMC) couplings and FEPs

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

The audit review of Pickett and Leslie (1999) indicated that 58 primary FEPs are relevant to ENFE Subissue 1. In further review of the FEP database, staff identified two additional FEP (2.2.08.03.00—Geochemical interactions in geosphere and 2.2.10.08.00—Thermo-chemical alteration of the saturated zone) relevant to the seepage and flow subissue. This additional review also removed three FEPs from the Pickett and Leslie (1999) list of FEPs relevant to the ENFE Subissue 1. These were 2.1.01.03.00—Heterogeneity of Waste Forms, 2.3.13.03.00—Effects of Repository Heat on Biosphere, and 2.2.07.14.00—Density Effects on Groundwater Flow.

Of the primary FEPs relevant to Subissue 1, 17 are excluded by the DOE screening process, and two have no determination listed. In the audit review of Pickett and Leslie (1999), it was determined that 15 of the excluded 17 FEPs have inadequate or missing screening arguments. Staff reviewed in more detail the other two excluded FEPs (1.2.08.00.00 Diagenesis and 2.2.07.15.06 Convection) and agree with the DOE disposition of these, based on screening arguments presented in the FEP database (U.S. Department of Energy, 1999). No further analysis of the relevance of these FEPs to the ENFE Subissue 1 is anticipated.

Table 5-3 lists primary FEPs relevant to ENFE Subissue 1 and to the integrated subissue on flow paths in the unsaturated zone. This list includes FEPs to be included in the TSPA-SR and those excluded but determined to have inadequate screening arguments. In addition, Pickett and Leslie (1999) identified two FEPs relevant to ENFE Subissue 1 but not included in the FEP database. Both of these FEPs are concerned with dehydration reactions. The first of these missing FEPs is dehydration of zeolites below the repository. Dehydration could result in large-scale volume changes, thereby affecting porosity and, indirectly, flow paths. The second FEP is concerned with direct alteration of flow because of water released from mineralogic dehydration reactions. Both of these FEPs potentially affect the integrated subissue on Flow Paths in the Unsaturated Zone. Technical bases should be provided for the excluded FEPs.

- DOE provides the bases and justification for modeling assumptions and approximations where simplifications for modeling coupled THMC effects on percolation and seepage are used for performance assessment
- DOE temporal abstractions of the flow paths in the unsaturated zone appropriately incorporate the physical couplings or sufficient justification is provided for exclusion of these couplings. The DOE abstraction incorporates or conservatively bounds coupled processes based on, for example, independent models, laboratory and field analyses, literature reviews, natural analog data, and other available information
- DOE estimates of performance are not over-optimistic, given the excluded set of phenomena and the implementation of coupled THMC processes in the total system performance assessment.

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Coupled THC processes that might affect flow paths in the unsaturated zone and seepage into drifts were omitted without technical justification in the TSPA-VA. DOE has taken significant steps toward addressing these potential effects in the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e). Staff agrees that the general approach—to use sensitivity analyses to show that the hydrological parameters are not significantly affected by THC processes—is an appropriate one.

Coupled THC simulations undertaken to support the unsaturated zone flow and transport model show that maximum porosity change caused by mineral precipitation and dissolution is on the order of 1 percent of the initial porosity. Such a small change in porosity is expected to have negligible effects on bulk hydrological properties. The model used in the analysis is near state-of-the-art and appropriate for the task of addressing bulk property modification. Data justification, data uncertainty, model uncertainty, and model support were not reviewed because the supporting Analysis and Model Report was not available prior to May 15, 2000. If future reviews by the staff determine these acceptance criteria are also met, then DOE neglect of the effect of mineral precipitation and dissolution on bulk hydrological parameters used in determining the spatial and temporal distribution of flow would be acceptable.

The DOE preliminary draft thermo-hydrologic calculations (CRWMS M&O, 2000e) show that the base of the Paintbrush Tuff may approach boiling conditions in situations of low infiltration. The Paintbrush Tuff is susceptible to thermal alteration and also plays a potentially important role in the hydrology system by dampening large infiltration events. No technical bases were provided for neglecting the potential alterations to the Paintbrush Tuff. To meet this acceptance criterion, DOE should provide the technical bases for neglect of thermal alteration of the Paintbrush Tuff, demonstrate that the alteration is unimportant to performance, or incorporate the effect of the alterations into future abstractions of spatial and temporal distribution of flow. In light on the preliminary nature of estimates of the efficiency of ventilation to reduce the heat load of the repository (i.e, no data to support ventilation model), DOE should also provide additional clarification of the temperatures expected in the Paintbrush Tuff.

The preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e) contains no discussion of the potential effects of cementitious material on the flow paths below the repository. Staff agrees that the potential for hydrological changes resulting from chemical interactions between tuff rock and cementitious material is reduced greatly because of the limited use of cementitious materials in the Enhanced Design Alternative-II design. There is still a large amount of concrete proposed for the ventilation tunnels and shafts. Much of this concrete will experience elevated temperatures for long periods, with significant potential for chemical interaction with the adjacent tuff rock. An evaluation of the potential for cementitious materials to alter repository performance by altering hydrological properties below the repository is required before the ENFE Subissue 1 can be resolved. DOE indicates (U.S. Department of Energy, 2000) this FEP will be considered in the Site Recommendation. Staff will review this effort when it becomes available.

- For estimates of the amount of seepage flux, the DOE must (i) demonstrate the coupled THC changes in rock mass properties will not focus percolation into drifts, and (ii) rigorously justify estimated diversion of percolation away from the waste package

footprints. Model calculations that account for drift collapse and coupled THMC changes to rock properties could provide such support. If these alteration processes are not accounted for, a technical basis for neglecting them must be provided

#### STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

Coupled THC processes have the potential for producing significant persistent changes to the rock flow properties in the immediate vicinity of the host rock during the 10,000-year compliance period, thereby affecting seepage into the drifts. Coupled THC processes that might affect seepage were not considered explicitly in the TSPA-VA (U.S. Department of Energy, 1998b). The approach for the Site Recommendation is to use auxiliary analyses to demonstrate that THC effects will not alter hydrological properties beyond the range used in the performance assessment abstractions. In broad terms, staff agrees this is an appropriate plan for addressing the subissue.

Coupled THC simulations undertaken to support the drift seepage model show that THC-induced changes in bulk porosity are small. Within a continuum-level framework, the resulting changes in permeability and moisture retention properties would also be small. Data justification, data uncertainty, model uncertainty, and model support were not reviewed because the supporting Analysis and Model Report was not available prior to May 15, 2000. If future review by the staff determines these acceptance criteria are also met, then DOE neglect of bulk hydrological property changes because of mineral precipitation and dissolution would be acceptable. It is acceptable also to neglect the formation of a localized low-permeability layer at the fracture-matrix interface, provided DOE continues to base the seepage abstraction on a single-continuum (fracture only) model for seepage. If, however, future DOE approaches to drift seepage take credit for the beneficial effects of the matrix, the topic of THC-induced changes at the fracture-matrix interface should be addressed more carefully. For example, if an empirically determined drift-seepage threshold, which implicitly takes credit for matrix effects in controlling drift seepage, is used in the TSPA-SR, the topic of matrix sealing due to highly localized mineral precipitation at fracture-matrix interface would need to be revisited.

- DOE peer reviews follow the guidance in NUREG–1297 and NUREG–1298 (Altman, Donnelly, and Kennedy, 1988a,b) or other acceptable approaches

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Staff is unable to make a determination with respect to this acceptance criterion because the supporting Analysis and Model Report was not available prior to May 15, 2000. Staff will review the Analysis and Model Reports and supporting material when they become available.

#### **5.4.1.2.1.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 1**

Sufficient data (field, laboratory, and/or natural analog data) that are consistent with site characteristics are available to define relevant parameters and conceptual models necessary for developing the total system performance assessment abstraction of flow paths in the unsaturated zone, including the distribution of mass flux between fractures and matrix. The

data are also sufficient to assess whether FEPs related to flow paths in the unsaturated zone have been adequately characterized, and whether the technical bases provided for exclusion of those FEPs affecting flow paths in the unsaturated zone are adequate. Specifically,

- If potentially affected flow pathways due to hydrothermally driven geochemical reactions such as zeolitization of volcanic glass are excluded from the total system performance assessment abstraction of flow paths in the unsaturated zone, the reviewer should examine the adequacy of the technical basis for the exclusion and also examine the sufficiency of the transparency and traceability of the data used in defining the technical bases for the exclusion in the abstraction

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

DOE dismisses the possibility for further hydrothermal alteration of the Calico Hills nonwelded unit by calculating temperatures there and arguing that these are below the critical temperature needed for significant alteration. No supporting data for the reaction rates at the calculated temperatures are provided or cited. DOE should provide transparent and traceable data on the reaction rates in support of this exclusion from the abstraction.

Similarly, the DOE preliminary draft thermo-hydrologic calculations show that the base of the Paintbrush Tuff may approach boiling conditions in situations of low infiltration. The Paintbrush Tuff is susceptible to thermal alteration and also plays a potentially important role in the hydrology system by dampening large infiltration events. No technical bases were provided for neglecting the potential alterations to the Paintbrush Tuff. If hydrothermally driven alteration of the Paintbrush Tuff is neglected in abstractions of flow paths in the unsaturated zone, supporting data should be provided for this neglect of thermal alteration of the Paintbrush Tuff, or it should be demonstrated that the alteration is unimportant to performance.

- DOE thermohydrologic tests are designed and conducted:
  - With the explicit objective of testing conceptual and numerical models so that critical thermohydrologic processes can be observed and measured
  - With explicit consideration of thermohydrologic, thermal-chemical, and hydrological-chemical couplings
  - At different scales to discern scale effects on observed phenomena
  - For temperature ranges expected under repository operations conditions
  - To determine if water refluxes back to the heaters during either the heating or cool-down phases of the tests
  - To account for mass and energy losses/gains in the model system
  - Such that the model environment is sufficiently characterized so that the level of uncertainty in property values does not result in unacceptable uncertainty in thermal test interpretation

- Such that accuracy in the measurement of the test environment saturation is sufficient to discern the relative capability of different conceptual models to represent the thermohydrologic processes in heated, partially saturated, fractured porous media

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Staff agrees that the DOE's stated plan to use the results of the Drift Scale Heater Test to test and refine the models and parameters in the THC models is an appropriate approach. The Analysis and Model Reports describing this effort will be reviewed when it becomes available.

- Accepted and well-documented procedures are adopted to construct and calibrate numerical models used

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Based on a review of the preliminary draft unsaturated zone Process Model Report (CRWMS M&O, 2000e), staff agrees that the numerical software TOUGHREACT Version 2.2 used to simulate THC effects on flow and seepage is appropriate and was constructed using accepted and well-documented procedures. Staff will review this in more detail when the supporting Analysis and Model Report becomes available.

- Process-level, conceptual and mathematical models used in the analyses are reasonably complete. In particular:
  - Models are based on well-accepted principles of heat and mass transfer applicable to unsaturated geologic media
  - Models include, at a minimum, an evaluation of important thermohydrological phenomena, the processes of evaporation and condensation, and the effects of discrete geologic features
  - Important thermohydrologic phenomena such as (i) multi-drift dry-out zone coalescence and other multi-drift interactions, (ii) lateral movement of condensate, (iii) repository edge effects, and (iv) condensate drainage through fractures, at a minimum, are addressed
  - Models are capable of accommodating variation in infiltration
  - Mathematical models are consistent with conceptual models, based on consideration of site characteristics
  - Results from different mathematical models are compared to judge robustness of results
  - Adequate technical bases for spatial and temporal variability of parameters and boundary conditions are provided

- Models used to predict shedding around emplacement drifts are shown to contain an adequate level of heterogeneity in media properties
- Thermohydrologic models are demonstrated to be appropriate for the temperature regime expected at the repository
- Media properties of a model contain an adequate level of heterogeneity such that mechanisms such as dripping are not neglected or misrepresented
- Drift wall representations in models contain sufficient physical detail such that processes predicted using a continuum model, such as capillary diversion, are appropriate for the geologic media at the proposed repository horizon
- Physical mechanisms, such as penetration of the boiling isotherm by flow down a fracture, are not omitted from model predictions due to oversimplification of the physical medium or the conceptual model
- Models include changes in boundary conditions (e.g., drift shape and size) and hydrologic properties due to the response of the geomechanical system to thermal loading

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Based on an initial review of the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e), staff agrees that the process and conceptual models used to simulate THC effects on flow and seepage at the continuum level are reasonably complete. Staff will review this in more detail when the supporting Analysis and Model Report becomes available.

- DOE demonstrates that the data on the geology, hydrology, and geochemistry of the unsaturated zone, including the influence of structural features and stratigraphy, used in the total system performance assessment abstraction are based on techniques that may include laboratory experiments, site-specific field measurements, natural analog research, and process-level modeling studies
- Sensitivity or uncertainty analyses are adequate to determine the possible need for additional data
- Some laboratory and field experiments used to support and verify total system performance assessment results may be designed for the postclosure period. In particular, if the testing program for coupled THC processes is not complete at the time of license application submittal, DOE explains why it is not necessary to complete the testing program for the license application and identifies specific plans for completion of the testing program as part of the postclosure program
- Where sufficient data do not exist, the definition of parameters values and conceptual models is based on other appropriate sources such as expert elicitation conducted in accordance with NUREG-1563 (Kotra, et al., 1996)

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Staff is unable to make a determination with respect to these acceptance criteria because the supporting Analysis and Model Report was not available prior to May 15, 2000. Staff will review the Analysis and Model Report and supporting material when they become available.

#### **5.4.1.2.1.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 1**

Parameter values, assumed ranges, probability distributions, and/or bounding assumptions used in the abstraction of flow paths in the unsaturated zone, including the distribution of mass flux between fractures and matrix, are technically defensible and reasonably account for uncertainties and variabilities. The technical basis for the parameter values used in the performance assessment is provided. Specifically,

- DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THMC effects on percolation and seepage

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Based on an initial review of the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e), staff agrees that the numerical software TOUGHREACT Version 2.2 used to simulate THMC effects on flow and seepage is appropriate and was constructed using accepted and well-documented procedures. Staff will review this in more detail when the supporting Analysis and Model Report becomes available. DOE should provide a description of the procedures used to test TOUGHREACT Version 2.2.

- The DOE evaluation of coupled THMC processes properly considered the uncertainties in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, and temporal and spatial variations, in establishing initial and boundary conditions for conceptual models and simulation of THMC coupled processes that affect percolation and seepage
- DOE establishes that reasonable or conservative ranges of parameters or functional relations are used to determine effects of coupled THMC processes on percolation and seepage
- DOE shows that the parameters used to define initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THMC effects on percolation and seepage are consistent with available data
- Coupling of processes has been evaluated using an acceptable methodology. Coupled processes may be uncoupled if it is shown that the uncoupled model results bound the predictions of the fully coupled model results

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Staff is unable to make a determination with respect to these acceptance criteria because the supporting Analysis and Model Report was not available prior to May 15, 2000. Staff will review the Analysis and Model Report and supporting material when they become available.

#### **5.4.1.2.1.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 1**

Alternative modeling approaches consistent with available data and current scientific understanding are investigated and results and limitations are appropriately factored into the abstraction of flow paths in the unsaturated zone, including the distribution of mass flux between matrix and fractures. DOE has provided sufficient evidence that alternative conceptual models of FEPs have been considered, that the models are consistent with available data (e.g., field, laboratory, and natural analog) and current scientific understanding, and that the effect of these alternative conceptual models on total system performance assessment has been evaluated. Specifically,

- DOE provided a reasonable description of the mathematical models included in its analyses of coupled THMC effects on seepage and flow. The description should include a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model

STATUS OF RESOLUTION: OPEN. Further review of the DOE Analysis and Model Reports and Process Model Reports is needed.

The preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e) provides a discussion of an alternative conceptual model in which a highly localized low-permeability zone is formed at the matrix fracture interface due to mineral precipitation. Such sealing of the matrix might lead to significant hydrological changes by decoupling the matrix and fracture systems, even if the overall change in porosity is small. Although the DOE THC model is appropriate for addressing the concern of bulk property modification caused by mineral precipitation and dissolution, it is not an appropriate model for addressing the matrix sealing process. DOE's treatment of this effect in the preliminary draft Process Model Report on Unsaturated Zone Flow and Transport is limited to a nonquantitative discussion and does not represent an adequate technical basis for omission in the total system performance assessment. In particular, the DOE argument hinges on low kinetic rates for silica precipitation in the welded units and on a presumption of low silica concentrations in the fractures, yet no values for these parameters are provided. This approach does not meet the acceptance criterion because the model is not shown to be consistent with available data. Further, the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e) also argues there is little hydrological potential for flow from fractures to matrix under unsaturated conditions, an argument that is clearly incorrect given the strong imbibition from fractures to matrix that can exist under some saturation conditions. If this process is neglected in the TSPA-SR, DOE should either show that the effect is unimportant for performance or provide a sound technical basis for neglecting the effect using data appropriate for the expected conditions.

The clearest path to resolution on this concern may be simply to ignore matrix flow for a limited region below the drifts as a conservative bounding approximation. If the effect of matrix sealing on flow below the repository is neglected in the TSPA-SR, then a more quantitative supporting argument, based on data relevant to the expected repository conditions, should be put forward. If DOE continues to use a conservative single-continuum (fracture only) model for drift seepage, it is acceptable to neglect matrix sealing as it affects drift seepage.

#### **5.4.1.2.1.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 1**

Output from the TSPA abstractions of flow paths in the unsaturated zone and seepage, including the mass flux between matrix and fracture abstraction, is verified through comparison with detailed output from process-level models and/or empirical observations (e.g., laboratory testing, field measurements, and/or natural analogs). Specifically,

- Abstractions of process-level models conservatively bound process-level predictions. In particular, DOE may use an abstracted model to predict seepage flux into an emplacement drift if the abstracted model is shown to conservatively bound process-level model predictions of the influx of water as liquid or vapor into an emplacement drift

STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

In DOE's abstraction of seepage into drifts, the seepage flow rate was increased by 55 percent, in part to account for increased seepage along rock bolts. Increased seepage along rock bolts might occur, for example, because of localized dissolution of tuff rock due to chemical interaction with the cementitious grout. Based on an initial review of the limited information available in the preliminary draft Analysis and Model Reports U0075 (CRWMS M&O, 2000c) and U0120 (CRWMS M&O, 2000d), staff agrees that the 55 percent increase conservatively bounds the results of process-level models that include discrete high-permeability features associated with rock bolts.

- DOE appropriately adopts accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THMC effects on percolation and seepage

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Based on an initial review of the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e), staff agrees that the numerical software TOUGHREACT Version 2.2 used to simulate THC effects on flow and seepage is appropriate and was constructed using accepted and well-documented procedures. Staff will review this in more detail when the supporting Analysis and Model Report becomes available. DOE should provide a description of the procedures used to test TOUGHREACT Version 2.2.

- Results of process-level models have been verified by demonstrating consistency with results/observations from field-scale, thermohydrologic tests. In particular, sufficient

physical evidence should exist to support the conceptual models used to predict thermally driven flow in the near field

- DOE demonstrates that abstracted models for coupled THMC effects on percolation and seepage are based on the same assumptions and approximations demonstrated to be appropriate for closely analogous natural or experimental systems

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

Staff is unable to make a determination with respect to these acceptance criteria because the supporting Analysis and Model Report was not available prior to May 15, 2000.

#### **5.4.2 Subissue 2: Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Waste Package Chemical Environment**

The DOE approach to assess the effects of coupled THC processes on the waste package and drip shield chemical environment must meet the following model abstraction acceptance criteria for each relevant integrated subissue: (i) integration, (ii) data and model justification, (iii) data uncertainty, (iv) model uncertainty, and (v) model support. Quality assurance is handled in a separate section of the Yucca Mountain Review Plan.

Two integrated subissues are influenced by the ENFE within the scope of the waste package chemical environment subissue: (i) Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms, and (ii) Degradation of the Engineered Barriers (U.S. Nuclear Regulatory Commission, 2000a). Both integrated subissues need to be considered in the evaluation of each DOE abstraction. As part of this evaluation, the acceptance criteria listed in Section 5.4.2.2 will be used to review DOE Process Model Reports and the supporting Analysis and Model Reports as they become available. This evaluation will also include a review of the relevant FEPs excluded from the DOE total system performance assessment. Staff evaluation of the degradation of the engineered barriers is addressed in more detail in the Container Life and Source Term IRSR (U. S. Nuclear Regulatory Commission, 1999f).

The following Analysis and Model Reports have been identified in the DOE Analysis and Model Reports/Process Model Reports schedule as relevant to the ENFE subissue on the waste package chemical environment: E0020—Analysis and Model Report on In-Drift Corrosion Products, E0030—Analysis and Model Report on Seepage/Backfill Interaction, E0035—In-Drift Gas Flux and Composition, E0040—Analysis and Model Report on Microbe Communities, E0055—Analysis and Model Report on Seepage/Cement Interaction, E0065—Analysis and Model Report on the In-Drift THC Analysis, E0105—Analysis and Model Report on Precipitates/Salts Analysis, N0120—Analysis and Model Report on Drift Scale Coupled Processes, U0135—Analysis and Model Report on Natural Analogs for Unsaturated Zone, W0035—Analysis and Model Report on General Corrosion and Localized Corrosion of Waste Package Outer Barrier, W0040—Analysis and Model Report on the Pitting and Crevice Corrosion of the Drip Shield in Abstraction Models, W0070—Analysis and Model Report on the Surface Environment of the Waste Package and Drip Shield Outer Barrier, W0085—Analysis and Model Report on General Corrosion and Localized Corrosion of Drip Shield, and W0105—Analysis and Model Report on Hydrogen Induced Cracking of Drip Shield. The relevant Analysis

and Model Reports on FEPs include E0015—FEPs Degradation Modes Analysis, E0110—FEPs Engineered Barrier System Degradation Modes and FEPs Abstraction, N0080—FEPs for the Near-Field Environment, and WP0055—FEPs for the Waste Package. Most of these were not available for review prior to May 15, 2000.

The preliminary draft Analysis and Model Reports relevant to ENFE Subissue 2 that were available and reviewed for this revision of the ENFE IRSR include: E0065—Analysis and Model Report on the In-Drift THC Analysis (CRWMS M&O, 2000f), U0135—Analysis and Model Report on Natural Analogs for Unsaturated Zone (CRWMS M&O, 2000g), E0105—Analysis and Model Report on In-Drift Precipitates/Salts Analysis (CRWMS M&O, 2000h), W0035—General Corrosion and Localized Corrosion of Waste Package Outer Barrier (TRW Environmental Safety Systems, Inc., 2000a), W0070—Analysis and Model Report on the Environment on the Surfaces of the Waste Package and Drip Shield Outer Barrier (TRW Environmental Safety Systems, Inc., 1999), W0085—Analysis and Model Report on General Corrosion and Localized Corrosion of Drip Shield (TRW Environmental Safety Systems, Inc., 2000b), and W0105—Analysis and Model Report on Hydrogen Induced Cracking of Drip Shield (TRW Environmental Safety Systems, Inc., 2000c). Of these preliminary draft Analysis and Model Reports, Revision 0 of the In-Drift THC Analysis and Model Report (CRWMS M&O, 2000f) does not include a discussion of chemical processes, and will not be discussed further. A review of the preliminary draft Process Model Report on Waste Package Degradation is also included.

As appropriate, status based on information contained in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) and in the preliminary draft Process Model Reports and Analysis and Model Reports has been included where the approaches differ from that used in the TSPA-VA. As additional or revised Analysis and Model Reports and Process Model Reports become available for review our evaluation of these reports will be documented.

#### **5.4.2.1 U.S. Department of Energy Approach**

Two of the principal factors identified by DOE for the enhanced repository system are relevant to the ENFE subissue on waste package chemical environment: (i) Performance of the Waste Package Barriers, and (ii) Performance of the Drip Shield. The TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) outlines how these abstractions will be linked to other abstractions or process-level models in the TSPA-SR and a potential license application to provide the technical bases for the postclosure safety case.

The approach used in the TSPA-VA (U.S. Department of Energy, 1998b) will be modified for the TSPA-SR to refine abstractions and process-level models, to take into account the potential effects of the new repository design, and to improve coupling between abstractions. Figure 5-1, taken from the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), shows the basic architecture of the new DOE approach. The TSPA-Site Recommendation conceptual model includes five spatial domains that are linked together by a flow pathway. The spatial domains, sometimes referred to as repository integration program (RIP) cells are (i) the backfill region above the drip shield, (ii) the drip shield environment, defined as the backfill region immediately on or adjacent to the drip shield, (iii) the waste package environment, defined as the region immediately on, or adjacent to, the waste package (this region includes backfill, invert, or corrosion products), (iv) the waste form environment, which includes the interior of the

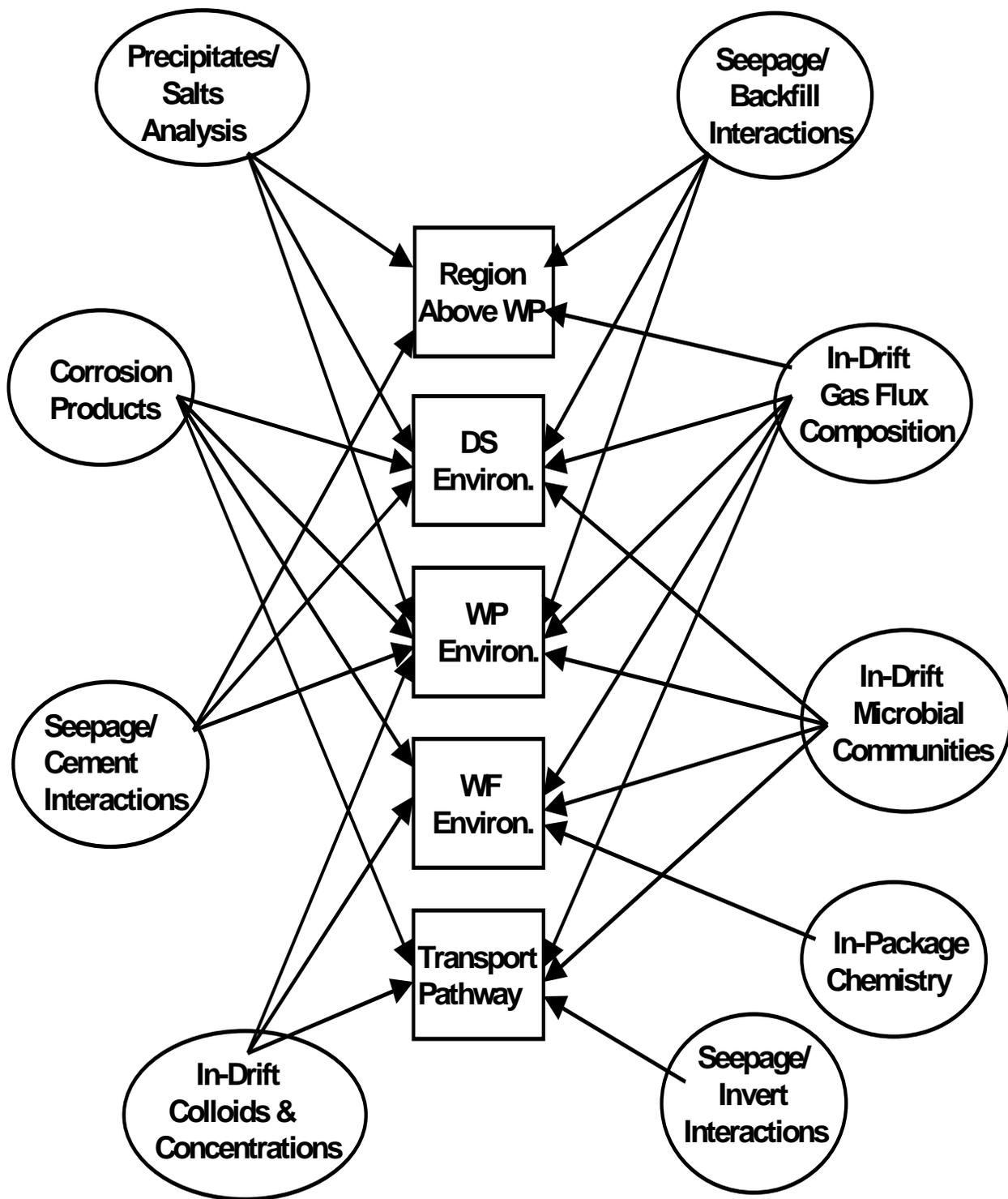


Figure 5-1. The U.S. Department of Energy In-Drift Geochemical Environment Model for TSPA-SR. The evolution of gas and water composition is evaluated by nine different submodels (circles), and the output is fed (see arrows) to relevant mixing cells (rectangles). [after Figure 3-1 and associated text in CRWMS M&O (1999a)].

waste package, and (v) the engineered barrier system transport pathway environment, which includes the region underneath and between the waste package and drift wall.

Analyses of the two principal factors relevant to ENFE subissue on waste package and drip shield chemical environment are embodied in the waste package and drip shield environment cells and in the in-drift geochemical submodels that provide input into this cell. The in-drift geochemical submodels will be linked to the waste package and drip shield spatial domains, which act as mixing cells, using the new Geochemical Repository Integration Model (GRIM) software. GRIM tracks the seepage flow through each cell and transports the cell's effluent to downstream mixing cells according to prescribed flow paths. Altered solids will remain in each cell for reaction at subsequent times. This approach allows only one-way linkage between subsystem models and total system performance assessment components via output/input transfers at the process model level, or within the total system performance assessment analyses. The EQ3/6 software package (Wolery, 1992) with solid-centered flow through capability will be used to evaluate the in-drift geochemistry submodels and to calculate changes to water composition that will be used as input to the waste package cell.

A major assumption of both the TSPA-VA (U.S. Department of Energy, 1998b) and the TSPA-SR is that THC processes can be decoupled, calculated separately, and linked. However, DOE has made significant changes in their approach since the TSPA-VA to address the importance of coupling. The TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) states that the linkage software GRIM is expected to provide the following key advantages over the approach used in the TSPA-VA: (i) it facilitates data traceability, data consistency, and output data reproducibility, (ii) it explicitly couples in-drift seepage flux with some reaction processes, (iii) it allows assessment of a range of conditions, (iv) it provides time-dependent output of key variables that describe the waste package, waste form, and radionuclide transport environments, and (v) it allows assessment of the effects of coupled chemical processes on drip shield/waste package and radionuclide transport environments.

Six submodels of the DOE in-drift geochemical model described in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) are judged relevant to both the integrated subissue on quantity and chemistry of water contacting the drip and waste packages and ENFE subissue on waste package and drip shield chemical environment. These submodels, described in Section 5.4.2.1.2 and reviewed in Section 5.4.2.2.1, are (i) Seepage/Backfill Interactions, (ii) In-Drift Gas Flux and Composition, (iii) In-Drift Microbial Community, (iv) Precipitates/Salts Analysis, (v) Corrosion Products, and (vi) Seepage/Cement Interactions. The following waste package and drip shield environment submodels are expected to be used by DOE to evaluate degradation processes for the TSPA-SR, and are judged to be relevant to both the integrated subissue on degradation of engineered barriers and the ENFE subissue on waste package and drip shield chemical environment: (i) Humid Air Corrosion, (ii) General Aqueous Corrosion, (iii) Crevice Corrosion, (iv) Pitting Corrosion, (v) Stress Corrosion Cracking, and (vi) Hydride Cracking. These submodels are described in Section 5.4.2.1.3 and are reviewed in Section 5.4.2.2.2.

#### **5.4.2.1.1 Features, Events, and Processes Screening**

A formal screening process for FEPs was not developed for the TSPA-VA (U.S. Department of Energy, 1998b), and many important design features, physical phenomena, and couplings were not evaluated in a performance assessment framework. DOE has since developed a formal documentation of the FEPs identification and screening process (Swift, et al., 1999) in

preparation for the TSPA-SR. The DOE identification, screening, and documentation process for FEPs is expected to account for known temporal and spatial variations in conditions affecting coupled THC effects on the waste package and drip shield chemical environment and to relay this information in a thorough and transparent manner. The contents of the preliminary DOE FEP database (U.S. Department of Energy, 1999) have been reviewed, and the FEPs related to the ENFE subissue on waste package and drip shield chemical environment and the integrated subissues on quantity and chemistry contacting the waste packages and drip shield and degradation of engineered barriers have been identified (Pickett and Leslie, 1999; U.S. Nuclear Regulatory Commission, 2000a). The results of the preliminary DOE screening are presented in Table 5-4. Staff evaluation of the preliminary DOE screening is presented in Section 5.4.2.2.

#### **5.4.2.1.2 Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms**

This section describes the individual DOE in-drift geochemical submodels relevant to the ENFE subissue on waste package and drip shield chemical environment and the integrated subissue on quantity and chemistry of water contacting waste packages and waste form. The relevant DOE in-drift geochemical submodels include: (i) Seepage/Backfill Interactions, (ii) In-Drift Gas Flux and Composition, (iii) In-Drift Microbial Community, (iv) Precipitates/Salts Analysis, (v) Corrosion Products, and (vi) Seepage/Cement Interactions. DOE submodels and abstractions related to the integrated subissue on degradation of engineered barriers are described in Section 5.4.2.1.3.

##### **5.4.2.1.2.1 Seepage/Backfill Interactions Submodel**

Backfill was not part of the TSPA-VA (U.S. Department of Energy, 1998b) basecase for the near-field geochemical environment but is included in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a). If the mineralogy of the backfill materials is different from the host rock, the water chemistry may change when fluids percolate through the backfill. These changes could affect the performance of the drip shield and the waste package. According to the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), the seepage/backfill interactions submodel will evaluate chemical reactions between water that enters the drift and backfill materials in the drift to determine the effect on water chemistry. Results will be provided as input to the total system performance assessment drip-shield environment and waste package environment models. However, since the License Application Design Selection process, DOE has removed backfill from the reference design (CRWMS M&O, 2000b). Thus, the potential effects of seepage/backfill interactions on the waste package chemical environment is no longer an issue.

##### **5.4.2.1.2.2 Precipitates/Salts Analysis Submodel**

The effects of precipitates/salts on water chemistry were evaluated in near-field sensitivity analyses in support of the TSPA-VA (U.S. Department of Energy, 1998b), but results were primarily used to assess the need for future work in this area. Bounds were placed on the mass and timing of precipitated minerals that might accumulate on the waste package surface, and the capacity for generating highly concentrated brines during the boiling period was evaluated. The progressive evaporation of J-13 water compositions was modeled in two stages, an early stage with <1 molar concentrations and a late stage with >1 molar concentrations. The EQ3/6 Version 7.2b code package was used to evaluate the early

**Table 5-4. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 2 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissues on quantity and chemistry of water contacting the waste package and waste forms and degradation of engineered barriers**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Degradation of Engineered Barriers
1.1.02.00.00	Excavation/construction	Exclude	X	
1.1.02.01.00	Site flooding (during construction and operation)	Exclude	X	
1.1.02.03.00	Undesirable materials left	Exclude	X	X
1.1.03.01.00	Error in waste or backfill emplacement	Exclude	X	
1.1.07.00.00	Repository design	Include (exclude deviations from design)	X	X
1.1.08.00.00	Quality control	Include (exclude defects and deviations)	X	X
1.1.12.01.00	Accidents and unplanned events during operation	Exclude	X	X
1.1.13.00.00	Retrievability	Include	X	
1.2.04.02.00	Igneous activity causes changes to rock properties	Include	X	
1.2.06.00.00	Hydrothermal activity	Exclude	X	
2.1.01.03.00	Heterogeneity of waste forms	Include	X	
2.1.02.01.00	DSNF degradation, alteration, and dissolution	Include	X	
2.1.02.02.00	CSNF alteration, dissolution, and radionuclide release	Include	X	
2.1.02.03.00	Glass degradation, alteration, and dissolution	Include	X	

**Table 5-4. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 2 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissues on quantity and chemistry of water contacting the waste package and waste forms and degradation of engineered barriers (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Degradation of Engineered Barriers
2.1.02.08.00	Pyrophoricity	Exclude	X	X
2.1.03.01.00	Corrosion of waste containers	Include	X	X
2.1.03.02.00	Stress corrosion cracking of waste containers	Include	X	X
2.1.03.03.00	Pitting of waste containers	Include	X	X
2.1.03.04.00	Hydride cracking of waste containers	Include	X	X
2.1.03.05.00	Microbially mediated corrosion of waste container	Include	X	X
2.1.03.06.00	Internal corrosion of waste container	Include	X	X
2.1.03.07.00	Mechanical impact on waste container	Include	X	X
2.1.03.10.00	Container heating	Include	X	X
2.1.03.11.00	Container form	Include	X	X
2.1.03.12.00	Container failure (long-term)	Include	X	X
2.1.04.01.00	Preferential pathways in the backfill	Include	X	
2.1.04.02.00	Physical and chemical properties of backfill	Include	X	
2.1.04.03.00	Erosion or dissolution of backfill	Exclude	X	
2.1.04.05.00	Backfill evolution	Include	X	
2.1.06.01.00	Degradation of cementitious materials in drift	Include	X	

**Table 5-4. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 2 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissues on quantity and chemistry of water contacting the waste package and waste forms and degradation of engineered barriers (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Degradation of Engineered Barriers
2.1.06.02.00	Effects of rock reinforcement materials	Include	X	
2.1.06.03.00	Degradation of the liner	Include	X	
2.1.06.04.00	Flow through the liner	Exclude	X	
2.1.06.05.00	Degradation of invert and pedestal	Include	X	
2.1.06.06.00	Effects and degradation of drip shield	not given	X	X
2.1.06.07.00	Effects at material interfaces	Include	X	X
2.1.08.04.00	Condensation forms on backs of drifts	Include	X	
2.1.08.07.00	Pathways for unsaturated flow and transport in the waste and EBS	Include?	X	
2.1.08.08.00	Induced hydrological changes in the waste and EBS	Include	X	
2.1.08.11.00	Resaturation of repository	Include	X	
2.1.09.01.00	Properties of the potential carrier plume in the waste and EBS	Include	X	
2.1.09.02.00	Interaction with corrosion products	Exclude?	X	X
2.1.09.03.00	Volume increase of corrosion products	Exclude		X
2.1.09.06.00	Reduction-oxidation potential in waste and EBS	Include	X	X
2.1.09.07.00	Reaction kinetics in waste and EBS	Exclude	X	X

**Table 5-4. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 2 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissues on quantity and chemistry of water contacting the waste package and waste forms and degradation of engineered barriers (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Degradation of Engineered Barriers
2.1.09.08.00	Chemical gradients/enhanced diffusion in waste and EBS	Include	X	X
2.1.09.09.00	Electrochemical effects (electrophoresis, galvanic coupling) in waste and EBS	Exclude	X	X
2.1.09.12.00	Rind (altered zone) formation in waste, EBS, and adjacent rock	Include	X	
2.1.10.01.00	Biological activity in waste and EBS	Include?	X	X
2.1.11.01.00	Heat output/temperature in waste and EBS	Include	X	X
2.1.11.02.00	Nonuniform heat distribution/edge effects in repository	Include	X	
2.1.11.03.00	Exothermic reactions in waste and EBS	Exclude	X	
2.1.11.04.00	Temperature effects/coupled processes in waste and EBS	Include	X	X
2.1.11.06.00	Thermal sensitization of waste containers increases fragility	Include		X
2.1.11.08.00	Thermal effects: chemical and microbiological changes in the waste and EBS	Exclude	X	
2.1.11.09.00	Thermal effects on liquid or two-phase fluid flow in the waste and EBS	Include	X	
2.1.12.01.00	Gas generation	Exclude	X	X

**Table 5-4. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 2 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissues on quantity and chemistry of water contacting the waste package and waste forms and degradation of engineered barriers (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Degradation of Engineered Barriers
2.1.12.03.00	Gas generation (H <sub>2</sub> ) from metal corrosion	Exclude	X	X
2.1.12.04.00	Gas generation (CO <sub>2</sub> , CH <sub>4</sub> , H <sub>2</sub> S) from microbial degradation	Exclude	X	X
2.1.12.05.00	Gas generation from concrete	Exclude	X	
2.1.12.06.00	Gas transport in waste and EBS	Exclude	X	
2.1.13.01.00	Radiolysis	Include	X	X
2.1.13.02.00	Radiation damage in waste and EBS	Include, Exclude (backfill, seals, rock)	X	X
2.2.07.10.00	Condensation zone forms around drifts	Include	X	
2.2.07.11.00	Return flow from condensation cap/resaturation of dry-out zone	Include	X	
2.2.08.01.00	Groundwater chemistry/composition in UZ and SZ	Include	X	
2.2.08.04.00	Redissolution of precipitates directs more corrosive fluids to containers	Exclude	X	
2.2.10.01.00	Repository-induced thermal effects in geosphere	Include	X	
2.2.11.01.05	Gas generation and gas sources, far-field	Exclude	X	
2.3.11.03.00	Infiltration and recharge (hydrologic and chemical effects)	Include	X	

**Table 5-4. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 2 of the Evolution of the Near-Field Environment Key Technical Issues and to the integrated subissues on quantity and chemistry of water contacting the waste package and waste forms and degradation of engineered barriers (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Degradation of Engineered Barriers
3.1.01.01.00	Radioactive decay and ingrowth	Include	X	
SZ = saturated zone                      UZ = unsaturated zone                      DSNF = Department of Energy spent nuclear fuel EBS = engineered barrier system                      CSNF = commercial spent nuclear fuel				

evaporative stages. In these calculations, solid solutions were ignored, and the following 12 minerals were suppressed: quartz, potassium-feldspar, stilbite, mesolite, pyrophyllite, scolecite, muscovite, dolomite, tridymite, calcium-clinoptilolite, albite, and maximum microcline. To simulate late stages of evaporation, simple sets of normative binary salts were constructed to represent the precipitation of the remaining dissolved solids. The types of precipitating salts were chosen based primarily on mass balance and charge balance constraints; however, solubilities reported in the literature also played a role.

According to the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), the precipitates/salts analysis submodel of the TSPA-SR will address more extensive geochemical topics relevant to ENFE Subissue 2. Calculations will be performed to (i) determine the types and amounts of precipitates (including salts) that might form as a result of evaporation driven by temperature gradients within the drift (e.g., from package surface to drift wall), (ii) explore the effects of progressive evaporation on water chemistry, (iii) evaluate changes to water chemistry as a function of the mass of accumulated precipitates, (iv) determine the effect of time and relative humidity on water vapor condensation, (v) account for back reactions involving the dissolution of precipitates previously deposited on the drip shield, waste package, and other engineered barrier system component surfaces, and (vi) predict how much time is required for aqueous species concentrations to return to ambient levels. Results will provide input into the total system performance assessment drip shield environment, waste package environment, and waste form environment models.

In the preliminary draft Analysis and Model Report on precipitates/salts analysis (CRWMS M&O, 2000h), DOE used a geochemical modeling approach to evaluate the chemical effects of evaporation of groundwater and precipitation of salts. The approach focused on variations in solution pH, chloride concentration, ionic strength, and the maximum relative humidity for dry conditions to exist. For calculations at ionic strength less than about 10 [high relative humidity model], a new Pitzer model database was developed that has estimated values for essential aqueous species and estimated values for temperature dependence parameters. The EQ3/6 aqueous speciation and reaction path modeling code was used for the high relative humidity model. The high relative humidity model invoked chemical equilibrium except for selected suppression of precipitation of some minerals. For ionic strengths greater than 10 [low relative humidity model], a mass balance approach was hinged on nitrate salt deliquescence at 50-percent relative humidity and an arbitrary dependence of dissolution of other salts on time with increasing relative humidity. Reactions of groundwaters with rock and engineered materials were not considered in the models. Model results were compared to laboratory results on evaporation of synthetic groundwaters with correspondence deemed to be acceptable within broad ranges of model validity criteria.

#### **5.4.2.1.2.3 Corrosion Products Submodel**

DOE used process-level models to evaluate the effects of corrosion products in the TSPA-VA (U.S. Department of Energy, 1998b). To support the basecase TSPA-VA analyses, the geochemical modeling package EQ3/6 Version 7.0 (Wolery, 1992) was used to calculate the composition of water equilibrated with waste package corrosion products. Corrosion of the iron in steel was represented by the iron-oxyhydroxide phase goethite (Hardin, 1998, Section 6.4.3.1). Calculations were performed for various stages in the thermal-gaseous evolution of the proposed repository. The initial fluid composition was assumed similar to J-13 Well water, except that the total dissolved iron constraint was set to equilibrium with goethite.

In the TSPA-SR, the corrosion products submodel will evaluate changes to water chemistry due to reactions between metallic components, their corrosion products, and water that percolates into the drift. Metallic components may include the drip shield, the waste packages, the waste package structures, the waste package supports, the rail system, the ground support systems, and possibly the structural invert materials. The results of these calculations will be provided as input into the total system performance assessment drip shield environment, waste package environment, waste form environment, and transport pathway environment models.

#### **5.4.2.1.2.4 Seepage/Cement Interactions Submodel**

Concrete is a complex solid containing multiple components. The most reactive component of concrete is cement, which itself contains several mineral phases with variable chemical reactivities. The DOE used process-level models to provide input for the effects of seepage/cement interactions in the TSPA-VA sensitivity analyses (U.S. Department of Energy, 1998b). The capacity of the concrete to compete with the CO<sub>2</sub> system for control of the water composition was evaluated for different temperature-gas composition regimes defined by the thermal-hydrology submodel. Local equilibrium was assumed, and changing physical parameters of the concrete (e.g., porosity and density) were not considered. Bounding calculations were performed to increase confidence in the chosen baseline for variables that are highly uncertain from a conceptual or data standpoint. Sensitivity analyses were performed to evaluate the relative reaction rate values for components within the cement mixtures. These analyses were intended to determine the effect of the magnitude of the rate values on the reacted chemistry and to see if the observed dissolution behavior (Berner, 1990) could be recreated by setting all rates to the same value. The effect of suppressed phases on the resultant fluid chemistry was also evaluated.

Calculations for the TSPA-VA sensitivity analyses (U.S. Department of Energy, 1998b) were performed using the geochemical modeling package EQ3/6 Version 7.0 (Wolery, 1992). Cement was abstracted as a mixture of SiO<sub>2</sub> and CaH<sub>2</sub>SiO<sub>4</sub>, or Ca(OH)<sub>2</sub> and CaH<sub>2</sub>SiO<sub>4</sub> with log K values dependent on the calcium/silicon ratio of the solid. To test how the magnitude of the relative reaction rates influenced the reacted fluid chemistry, baseline reaction rate values for all the concrete components were multiplied and divided by five in two separate suites of simulations. This approach assumes that the relative reaction rates for each phase vary independently of each other. A second sensitivity study was performed to test how well the chosen baseline rates recreated the dissolution behavior of cement as outlined by Berner (1990). Reaction rates (rk1) for all components of the concrete were fixed at 1.0 and 0.1 in two separate runs. Changes in the relative reaction rates required that different sizes of gas reservoirs be available.

At the time of the TSPA-VA calculations, it was assumed there would be a concrete liner around the drifts in the proposed repository. A concrete liner is not currently included in the Enhanced Design Alternative-II repository design (CRWMS M&O, 1999b). Instead, cement is expected to occur only in the form of grout for rock bolts in the emplacement drifts and concrete in ventilation tunnels and shafts. Hence, the role of concrete in totally controlling bulk water compositions in the drift may be substantially reduced. The seepage/cement interactions submodel of the TSPA-SR will evaluate the effects of water/cement reactions on the water chemistry in the drift. The DOE will assume that the cement is initially in equilibrium with the in-drift gaseous phase at the appropriate temperatures. The results of these calculations will be provided as input into the total system performance assessment backfill, drip shield environment, and waste package environment models.

#### **5.4.2.1.2.5 In-Drift Gas Flux and Composition Submodel**

In the TSPA-VA (U.S. Department of Energy, 1998b), calculations were performed to determine the gas flux and composition in the drift as a function of time and temperature. The results of these calculations provided a framework for beginning additional calculations involving the in-drift geochemical conditions following waste emplacement. Ambient pore gas compositions were input into the thermal-hydrology submodel and allowed to migrate in response to heating. Three different sets of conditions were identified in which DOE takes the gas composition to be constant: (i) the ambient regime, (ii) the boiling regime, and (iii) the cool-down regime. The geochemical code EQ3/6 Version 7.2b (Wolery, 1992) was used to calculate gas compositions in the last two regimes by equilibrating the fluids in those regimes with the perturbed gas compositions determined in the thermal-hydrological model.

The in-drift gas flux and composition submodel will evaluate changes in the gaseous phase composition within the repository as a function of time and temperature. Abstractions from the engineered barrier system water distribution and removal model and the engineered barrier system physical/chemical environment model will be used to set cell-to-cell liquid and gas fluxes within the RIP engineered barrier system cell architecture. As a minimum, the in-drift gas flux and composition submodel will consider the following gas constituents: carbon dioxide, oxygen, nitrogen, and steam. Chemical interactions among the water, gas, and materials in the emplacement drift may act as sources or sinks for constituents in the gas phase. The results of these calculations will be provided as input into the total system performance assessment backfill, drip shield environment, waste package environment, waste form environment, and transport pathway environment models.

#### **5.4.2.1.2.6 In-Drift Microbial Communities Submodel**

In the TSPA-VA technical basis document (CRWMS M&O, 1998d), DOE included the potential impacts of in-drift microbial communities on the near-field geochemistry as part of their performance assessment of a potential high-level waste repository at Yucca Mountain. DOE evaluated this topic with the computer code Microbial Impacts to the Near-Field Geochemistry (MING). MING was developed to bound the masses of microorganisms that potentially could be produced in a drift at a potential repository at Yucca Mountain. Following approaches similar to those used in the Canadian high-level waste repository and the Swiss low/intermediate-level waste repository programs, the DOE computer code uses both mass balance and thermodynamics to quantify the impact of microbial populations. Biomass production is based on both nutrient and energy limitations for microorganisms at Yucca Mountain.

DOE work subsequent to the Viability Assessment has focused on bounding the potential abundance of microorganisms in the drift environment based on nutrient and energy limitations. The effects of both thermal loading and introduced materials on microbial abundance will be evaluated as a submodel for the DOE in-drift geochemical model abstraction supporting the TSPA-SR (CRWMS M&O, 1999a).

The microbial communities submodel will evaluate nutrient and energy limitations within the drift environment to place bounds on the ultimate abundance of microbes for the TSPA-SR. The effects of thermal loading and introduced materials will also be evaluated to predict changes in microbe abundance. The results of these calculations will be provided as input into the total system performance assessment drip shield environment, waste package environment, waste form environment, and transport pathway environment models.

#### 5.4.2.1.3 Degradation of Engineered Barriers

The Enhanced Design Alternative-II repository design includes a drip shield made of titanium grade 7 and a waste package with an outer container made of Alloy 22. Although these materials exhibit a high resistance to corrosion, both are susceptible to a variety of localized and general corrosion processes that are influenced by geochemical conditions. The titanium drip shield is expected to develop a protective  $\text{TiO}_2$  film on the surface that confers to the metal an extremely high resistance to corrosion in all natural waters, including those with high  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{2-}$ , silicate,  $\text{NO}_3^-$ ,  $\text{F}^-$ , and  $\text{CO}_3^{2-}$  concentrations with a pH range of 3 to 12. Under certain environmental conditions, titanium alloys can be susceptible to localized corrosion in hot  $\text{Cl}^-$  solutions (Dunn, Pan, and Cragolino, 1999) and when  $\text{F}^-$  concentrations reach a critical value of  $\sim 0.005$  molar (Brossia and Cragolino, 2000). The effects of aggressive environments on the performance of nickel-based alloys, such as Alloy 22, have been studied extensively. Although Alloy 22 is extremely resistant to corrosion as a result of the formation of a chromium-rich oxide film on the metal surface, corrosion rates may increase under certain conditions in solutions with high  $\text{Cl}^-$ ,  $\text{HS}^-$ , and metastable sulfur oxyanion concentrations (Dunn, Pan, and Cragolino, 1999). Localized corrosion of Alloy 22 is favored in oxidizing environments with high  $\text{Cl}^-$  concentrations.

The waste package and drip shield degradation model (WAPDEG) will be integrated with the RIP model in the TSPA-SR to allow time-dependent changes in the environment to be considered directly. Seven degradation submodels will be evaluated within the context of the waste package and drip shield environment RIP cells of the DOE in-drift geochemical model (Figure 5-1). To differentiate between general and localized corrosion processes, the surface of each abstracted drip shield and waste package will be partitioned into discrete regions where degradation processes will be evaluated independently. The following submodels will be used to evaluate waste package and drip shield degradation processes for the TSPA-SR and are judged to be relevant to ENFE Subissue 2: (i) Humid-Air Corrosion, (ii) General Aqueous Corrosion, (iii) Crevice Corrosion, (iv) Pitting Corrosion, (v) Stress Corrosion Cracking, and (vi) Hydride Cracking. Output from these submodels will be in the form of degradation rates and will be used by modules of the WAPDEG (CRWMS M&O, 1999a) computer code to determine longevity and subsequent degradation areas for the waste package and drip shield.

Environmental parameters required to evaluate the degradation submodels will be supplied by the thermal-hydrology abstraction and the in-drift geochemical submodels. The following localized environmental conditions are considered of key importance to the calculation of degradation rates (CRWMS M&O, 1999a): (i) temperature, (ii) in-drift gases (e.g.,  $\text{H}_2\text{O}$ ,  $\text{O}_2$ ,  $\text{CO}_2$ , and  $\text{N}_2$ ), (iii) chemistry of water and mineral films on the waste package (e.g., precipitates, salts, and pH), (iv) presence (or absence) of water dripping on the waste package surface, and (v) relative humidity. Time- and temperature-dependent results from the in-drift geochemical submodels will be abstracted into lookup tables and then used to constrain the degradation calculations.

The following discussion provides a general description of the drip shield and waste package submodels, indicates which geochemical parameters the submodels are sensitive to, identifies which in-drift geochemical submodels will be used to constrain the degradation submodels and indicates how results from the waste package environment submodels will be abstracted into the TSPA-SR. Not all of this information is currently available. Additional information about the degradation submodels may be found in the TSPA-VA (U.S. Department of Energy, 1998b), the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), the Container Life and

Source Term IRSR Revision 3 (in preparation), and the preliminary draft DOE Analysis and Model Reports and Process Model Reports.

#### **5.4.2.1.3.1 Humid Air Corrosion Submodel**

In the TSPA-VA (U.S. Department of Energy, 1998b), in which the outer overpack was designed of carbon steel, humid air was assumed to cause active general corrosion of the steel overpack. Humid-air corrosion was modeled using a parametric equation exhibiting a dependence of the corrosion rate on time, relative humidity, and absolute temperature. The corrosion rate under humid air conditions was based on abstractions of atmospheric corrosion data from tropical, urban, rural, and industrial locations. The DOE model for humid-air corrosion of the Viability Assessment waste package design was based on the concept of a critical relative humidity. The start of corrosion in a humid-air environment was assumed to occur above the critical relative humidity of 65 to 75 percent.

Humid-air corrosion may also occur for the Alloy 22 waste package outer barrier proposed in the TSPA-SR. The corrosion rates would be significantly lower than those of the carbon steel as a result of the passive behavior of the Ni-base alloy. Humid-air corrosion is assumed to occur when the relative humidity is greater than the critical relative humidity (TRW Environmental Safety Systems, Inc., 1999). DOE will evaluate critical relative humidity as a function of the deliquescence point (lowest relative humidity where a saturated solution of the salt can be maintained at a given temperature) for sodium nitrate, and temperature (TRW Environmental Safety Systems, Inc., 1999).

#### **5.4.2.1.3.2 General Aqueous Corrosion Submodel**

The general aqueous corrosion equation used for carbon steel in the TSPA-VA (U.S. Department of Energy, 1998b) did not track geochemical parameters believed to reflect changing environmental conditions in the proposed repository with time. Instead, the dependence of the corrosion rate on time and temperature was obtained from the literature (Boden, 1994; Potter and Mann, 1962) and incorporated into the model development. For example, estimates of the time-dependence of aqueous corrosion were based on long-term corrosion data (up to 16 years) in polluted river water (Coburn, 1978) and in tropical lake water (Southwell and Alexander, 1970). These data include the potential effects of microbial activity and various chemical species dissolved in the waters. The general aqueous corrosion submodel in the TSPA-VA (U.S. Department of Energy, 1998b) is, therefore, strictly applicable to constant exposure conditions. A "corrosion-time" concept was developed to use the model for time-dependent exposure conditions such as in the potential repository (Lee, Atkins, and Dunlap, 1997). A second critical relative humidity (>85 percent) was used to distinguish the regime of general uniform corrosion under aqueous conditions from that of humid-air corrosion.

DOE expects Alloy 22 degradation to occur predominantly by general corrosion in the form of passive dissolution (U.S. Department of Energy, 1998b; CRWMS M&O, 1999a). General corrosion is normally characterized by a relatively uniform thinning of materials without significant localized attack that, in the case of corrosion resistant alloys such as Alloy 22, occurs at a low corrosion rate as a result of passivity. In the TSPA-VA general uniform corrosion of the Alloy 22 inner overpack was based on the results of an expert elicitation where the corrosion rate of Alloy 22 was estimated. Two environmental conditions were considered. The first condition corresponds to relative humidity ranging from 85 to 100 percent in the absence of water dripping. The rates were computed by developing a "composite" distribution using

individual expert assessment. The second condition assumed the presence of water dripping onto the waste packages and, therefore, a wide range of local environments. The environmental conditions selected in the expert elicitation were (i) a moderately oxidizing (340 millivolts versus Standard Hydrogen Electrode) environment with a pH of 3 to 10, (ii) a moderately oxidizing environment with a pH of 2.5, and (iii) a highly oxidizing environment (640 millivolts versus Standard Hydrogen Electrode) environment at a pH of 2.5. These environments were assumed to form at localized corrosion sites even though the corrosion itself was considered uniform. The effect of galvanic coupling was ignored.

In the TSPA-SR, two environmental conditions will be considered for the stabilization of an aqueous film on the waste package surface (TRW Environmental Safety Systems, Inc., 2000a). In one case, the presence of an aqueous environment is expected when the relative humidity in the emplacement drift is greater than the deliquescence point of any salts deposited on the waste package surface. In the other case, when dripping of condensed water occurs on the waste package an aqueous film on the waste package is assumed to form. Below 100 °C, the composition of water that contacts the waste package surface is assumed to be simulated J-13 concentrated water (SCW). Simulated saturated water (SSW) is assumed to be present above 100 °C. Basic saturated water (BSW) has also been identified as another plausible water chemistry that may develop on the waste package surface as a result of dripping and evaporation. Compositions of the water chemistries are provided in Table 5-5. In addition, it is assumed that gamma-radiolysis can increase the corrosion potential of Alloy 22 by 100 millivolts based on short-term exposures of specimens to solutions containing hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>).

General passive corrosion is assumed when the corrosion potential is less than the critical potential for the initiation of localized corrosion. The general corrosion rates are derived from data obtained from the long-term corrosion test facility where numerous test specimens have been exposed to aqueous solutions based on modifications of J-13 Well water (TRW Environmental Safety Systems, Inc., 2000a; McCright, 1998).

**Table 5-5. Concentration of key species in simulated concentrated water (SCW), simulated saturated water (SSW), and basic saturated water (BSW) (TRW Environmental Safety Systems, Inc., 2000a).**

Species	SCW (moles/liter)	SSW (moles/liter)	BSW (moles/liter)
K <sup>+</sup>	0.09	3.62	1.75
Na <sup>+</sup>	1.78	2.12	4.84
F <sup>-</sup>	0.07	0	0.071
Cl <sup>-</sup>	0.19	3.62	3.8
NO <sub>3</sub> <sup>-</sup>	0.10	2.11	2.3
SO <sub>4</sub> <sup>2-</sup>	0.17	0	0.149
HCO <sub>3</sub> <sup>-</sup>	1.15	0	0
pH	—	—	11 to 13

#### 5.4.2.1.3.3 Pitting and Crevice Corrosion Submodels

Pitting and crevice corrosion are types of localized corrosion induced by local variations in electrochemical potential on a microscale. Localized corrosion may be more sensitive to geochemical conditions than general corrosion. The probability that local corrosion will begin is essentially the probability that an aggressive geochemical environment will exist. In the potential repository environment, such aggressive exposure conditions on the drip shield and waste package would be possible with dripping conditions. The probability of localized corrosion also depends on whether drips are accounted for on the top, sides, or bottom of the waste package and drip shield.

In the TSPA-VA (U.S. Department of Energy, 1998b), crevice corrosion of the Alloy 22 inner barrier was assumed to begin at temperatures greater than or equal to 80 °C. The probability of initiating crevice corrosion was equal to zero at 80 °C and increased linearly to one at temperatures of 100 °C. Once started, crevice corrosion was assumed to continue according to a pit growth law. The penetration rate for localized corrosion in the Viability Assessment was expressed as a function of temperature, pH, and the concentrations of NaCl and FeCl<sub>3</sub>. Coefficients in the equation were calculated using data for Alloy 22 obtained over a broad range of environments that lead to both passive corrosion and localized corrosion. At temperatures less than 80 °C, general corrosion was assumed.

Localized corrosion of Alloy 22 is assumed to occur when the corrosion potential is greater than the critical potential for the initiation of localized corrosion. Critical potentials for localized corrosion of Alloy 22, reviewed in the preliminary draft General Corrosion and Localized Corrosion of Waste Package Outer Barrier Analysis and Model Report (TRW Environmental Safety Systems, Inc., 2000a), are limited to pitting repassivation potential data obtained using a lead in pencil geometry. Initiation and repassivation potentials as a function of temperature, were also used as values for the critical potential for the initiation of localized corrosion. These were obtained from anodic polarization curves in cyclic potentiodynamic polarization tests using a variety of electrolytes based on modifications of J-13 Well water. Crevice corrosion stabilization and repassivation potentials were measured in 5-molar LiCl solutions from 60 to 95 °C (Kehler, Ilevbare, and Scully, 2000). These potentials were not used as the critical potential values because the concentrated solutions were deemed to be not representative of any plausible repository environment (TRW Environmental Safety Systems, Inc., 2000d). Although the critical potential for initiation of localized corrosion for a thermally aged specimen was reduced by 100 millivolts, the susceptibility of the thermally aged specimens to localized corrosion was not increased with respect to the base alloy because it is assumed that the corresponding reduction of the critical potential will prevent the initiation of localized corrosion.

The rate of localized penetration of Alloy 22 was estimated using corrosion rates in highly corrosive environments (TRW Environmental Safety Systems, Inc., 2000a) such as 10 percent FeCl<sub>3</sub> at 75 °C; dilute boiling HCl; and a solution containing 7 volume percent H<sub>2</sub>SO<sub>4</sub>, 3 volume percent HCl, 1 weight percent FeCl<sub>3</sub>, and 1 weight percent CuCl<sub>2</sub> at 102 °C. The distribution of localized corrosion rates is centered around the highest passive current density of 10 microamperes per square centimeter that corresponds to a corrosion rate of 100 micrometers per year. The cumulative distribution of penetration rates for localized corrosion are 12.7, 127, and 1,270 micrometers per year for the 0<sup>th</sup>, 50<sup>th</sup>, and 100<sup>th</sup> percentile, respectively (TRW Environmental Safety Systems, Inc., 2000a,d).

#### **5.4.2.1.3.4 Stress Corrosion Cracking Submodel**

Stress corrosion cracking is a failure process characterized by the beginning and propagation of cracks caused by the synergistic interaction of mechanical stress and corrosion reactions. Improper manufacturing can result in pre-existing cracks, and new cracks may start in areas of high-stress concentration, such as at a groove or corrosion pit. Welding residual stress, shrink-fit stress, or weight stress may be sufficient to cause stress corrosion cracking to occur. After a crack begins at specific electrochemical and environmental conditions, the crack will grow when the applied stress intensity factor,  $K$ , is equal to or larger than the threshold value of stress intensity for stress corrosion cracking ( $K_{ISCC}$ ). This process can be represented probabilistically (McCright, 1998). Localized corrosion and stress corrosion cracking are often interrelated because the sites of localized corrosion attack can become the sources of beginning stress corrosion cracking (Farmer and McCright, 1989; Farmer, et al., 1988).

Stress corrosion cracking was not evaluated in the TSPA-VA (U.S. Department of Energy, 1998b). In the TSPA-SR, however, DOE plans to consider stress corrosion cracking of the Alloy 22 outer barrier, particularly in welds. DOE has proposed two models for stress corrosion cracking susceptibility evaluation: the stress corrosion cracking threshold model, and the slip dissolution/film rupture model (TRW Environmental Safety Systems, Inc., 2000d). The stress corrosion cracking threshold model is based on fracture mechanics concepts, and is not directly related to the waste package chemical environment. The slip dissolution/film rupture model relates crack advance to the metal anodic oxidation that occurs when the protective film at the crack tip is ruptured as a result of a tensile stress. Although DOE suggests that the slip dissolution/film rupture equations can be evaluated based on repassivation rate measurements under potentiostatic conditions alone, empirical data obtained for Type 304 and 316 stainless steels in typical boiling water reactor environments and previous crack propagation rates for Alloy 22 are actually used. Crack propagation rates were obtained, in part, from analyses of Alloy 22 exposures in 5 percent NaCl at 90 °C (TRW Environmental Safety Systems, Inc., 2000a). DOE claims that if stress corrosion cracking was to occur, then any cracks that developed would be plugged by corrosion products and therefore would not be available for the transport of water.

#### **5.4.2.1.3.5 Hydrogen Embrittlement Submodel**

Hydrogen embrittlement of Alloy 22 was not evaluated in the TSPA-VA (U.S. Department of Energy, 1998b).

In the preliminary draft Waste Package Degradation Process Model Report (TRW Environmental Safety Systems, Inc., 2000d) and corresponding preliminary draft Analysis and Model Report (TRW Environmental Safety Systems, Inc., 2000c), no distinction is made between the closely related processes hydrogen embrittlement and hydrogen-induced cracking. The approach taken by DOE to evaluate hydrogen-induced cracking is based on the assumption that the dominant cathodic reaction occurring on the metal surface during passive (uniform) dissolution is hydrogen evolution, and the reaction is assigned a reaction rate equal to the passive dissolution rate observed from coupon testing. Of the hydrogen gas produced from this cathodic reaction, a fraction (between 0.02 to 0.10) will enter into the metal as hydrogen atoms, which will then lead to a loss in ductility (i.e., embrittlement). Hydrogen-induced cracking is said to be possible once a critical hydrogen concentration has been exceeded. Based on the uniform corrosion rates observed from coupon testing and the assumptions involved with the fraction of hydrogen that is eventually adsorbed into the metal lattice,

hydrogen-induced cracking was concluded to not have a significant effect on drip shield life expectancy during the 10,000-year performance period.

#### **5.4.2.2 U.S. Nuclear Regulatory Commission Staff Evaluation**

Staff analysis of the DOE approach to abstraction of the effects of coupled THC processes on the waste package and drip shield chemical environment was conducted based on the acceptance criteria listed in Sections 5.4.2.2.1 and 5.4.2.2.2. There are five generic acceptance criteria. Each generic acceptance criterion is followed by specific criteria. The DOE approach to abstract coupled THC effects on the waste package and drip shield chemical environment in a total system performance assessment for the proposed repository at Yucca Mountain will be acceptable from the point of view of the integrated subissue on quantity and chemistry of water contacting waste packages and waste forms provided that each of the acceptance criteria are met.

#### **5.4.2.2.1 Integrated Subissue on Quantity and Chemistry of Water Contacting the Waste Packages and Waste Forms**

##### **5.4.2.2.1.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 2**

Important design features, physical phenomena and couplings, and consistent and appropriate assumptions have been identified and described sufficiently for incorporation into the abstraction of the quantity and chemistry of water contacting waste packages and waste forms in the performance assessment and other related abstractions in the total system performance assessment, and the technical bases are provided. The features, phenomena and couplings, and assumptions used to abstract the quantity and chemistry of water contacting waste packages and waste forms have been provided. The total system performance assessment abstraction is consistent with the identification and description of those aspects of the quantity and chemistry of water contacting waste packages and waste forms that are important to waste isolation. The total system performance assessment abstraction is also consistent with the technical bases for these descriptions of barriers important to waste isolation. Specifically,

- The DOE abstraction is consistent with the detailed information on waste package design and other engineered features
- DOE evaluates the potential for focusing water flow into drifts caused by coupled THMC processes
- DOE reasonably accounts for the chemical composition of the water in the environment surrounding the waste package and drip shield and its evolution with time
- DOE evaluates the effect of gamma-radiolysis of water contacting the waste packages and drip shield
- DOE identifies and adequately considers the effects of the drip shield and backfill on the quantity and chemistry of water contacting waste packages, including the potential for condensate formation and dripping from the underside of the shield

- DOE provides analyses that demonstrate that no deleterious effects are caused by design or site features that DOE does not include in this abstraction
- DOE abstractions, including dimensionality of the abstractions, appropriately account for the various design features, site characteristics, and alternative conceptual approaches
- DOE spatial and temporal abstractions appropriately address the physical couplings (THMC)
- DOE provides the bases and justification for modeling assumptions and approximations where simplifications for modeling coupled THMC effects on seepage and flow and the waste package chemical environment are used for performance assessment
- DOE provides adequate technical bases, including activities such as independent modeling, laboratory or field data, or sensitivity studies, for exclusion of any THMC couplings and FEPs
- DOE identifies and considers likely modes of corrosion for container materials, including dry-air oxidation, humid-air corrosion, and aqueous corrosion processes, such as general corrosion, localized corrosion, microbially induced corrosion, stress corrosion cracking, and hydrogen embrittlement, as well as the effect of galvanic coupling, in determining the quantity and chemistry of water entering the waste packages
- DOE identifies the broad range of environmental conditions within the waste package emplacement drifts that may promote the corrosion processes of engineered barriers, taking into account the possibility of irregular wet and dry cycles and radiolysis that may enhance the rate of degradation
- DOE demonstrates that the conditions and assumptions used to generate look-up tables or regression equations are consistent with all other conditions and assumptions in the total system performance assessment for abstracting the quantity and chemistry of water contacting waste packages
- DOE uses important design features, including waste package design and material selection, backfill, drip shield, ground support, cladding, thermal-loading strategy, and degradation processes, to determine the initial and boundary conditions for calculations of the quantity and chemistry of water contacting waste packages and drip shield
- DOE evaluates in-package criticality or external-to-package criticality within the emplacement drift and provides an adequate technical basis for screening these events. If either event is included in the total system performance assessment, DOE uses acceptable technical bases for selecting the design criteria that mitigate any potential impact of in-package criticality on the repository performance, identifies the FEPs that may increase the reactivity of the system inside the waste package, identifies the configuration classes and configurations that have potential for nuclear criticality, and includes changes in thermal conditions and degradation of engineered barriers in the abstraction of the quantity and chemistry of water contacting the waste packages and waste forms

- The abstraction of the quantity and chemistry of water contacting waste packages and drip shields is consistent with technical bases, data, and models in the flow in unsaturated zone abstraction, degradation of engineered barriers abstraction, mechanical disruption of engineered barriers abstraction, and radionuclide release rates and solubility limits abstraction
- DOE consistently addresses the effect of distribution of flow on the amount of water contacting the waste packages and drip shield in all relevant abstractions
- DOE consistently addresses the effect of waste package corrosion of the quantity and chemistry of water contacting waste packages in all relevant abstractions
- DOE consistently addresses the role of water chemistry parameters such as the pH and carbonate concentration and the effect of released radionuclides on the chemistry of water contacting the waste packages in all relevant abstractions
- DOE consistently addresses the size and distribution of penetrations of the drip shield and waste containers that affect the quantity and chemistry of water that contacts the waste package in all relevant abstractions
- DOE peer reviews follow the guidance in NUREG–1297 and NUREG–1298 (Altman, Donnelly, and Kennedy, 1988a,b), or other acceptable approaches

STATUS OF RESOLUTION: OPEN. Further review of DOE Analysis and Model Reports and Process Model Reports is needed.

The DOE preliminary FEP database (U.S. Department of Energy, 1999) was reviewed to identify FEPs related to the waste package and drip shield chemical environment subissue and the integrated subissues relevant to ENFE (Pickett and Leslie, 1999; U.S. Nuclear Regulatory Commission, 2000a). The audit review of Pickett and Leslie (1999) indicated 67 primary FEPs are relevant to this ENFE subissue and the integrated subissue on the quantity and chemistry of water contacting waste packages and waste forms (Table 5-4). A further review of the DOE FEP database identified two additional FEPs (2.1.09.02.00—Interaction with corrosion products, and 2.1.06.06.00—Effects and degradation of drip shield) relevant to the waste package and drip shield chemical environment subissue. Of the 68 FEPs relevant to ENFE Subissue 2 and the integrated subissue on the quantity and chemistry of water contacting waste packages and waste forms, 23 were either excluded or not assigned an “include” or “exclude” status during the preliminary DOE screening process. In addition, portions of the following three FEPs were excluded: FEP 1.1.07.00.00—Repository design, FEP 1.1.08.00.00—Quality control, and FEP 2.1.13.02.00—Radiation damage in waste and engineered barrier system. Staff reviewed the 26 excluded, partially excluded, or incompletely screened FEPs in the preliminary DOE FEP database (U.S. Department of Energy, 1999). We determined that the technical basis provided was sufficient to support the screening argument, and the staff agrees with, DOE’s preliminary exclusion of two FEPs. The remaining 24 FEPs either should not be excluded based on the current preliminary technical basis, or an insufficient technical basis has been provided to evaluate the screening, chosen by DOE. The bases for these determinations are discussed in the following paragraphs.

Staff judges that DOE exclusion of the following two FEPs is currently acceptable.

#### 2.1.04.03.00—Erosion or dissolution of backfill

Because of recent changes in the reference design (CRWMS M&O, 2000b), the TSPA-SR reference design no longer includes backfill. Based on the information available at this time, the exclusion of this FEP and the technical basis supporting its exclusion is acceptable. Should future design changes call for backfill, the screening of this FEPs will need to be reexamined and the technical basis for screening will need to be updated.

#### 2.1.06.04.00—Flow through the liner

DOE excludes this FEP because the TSPA-SR reference design eliminates the concrete liner that was included in the TSPA-VA design (CRWMS M&O, 2000b). For this reason, DOE does not need to include this design feature in its TSPA abstraction, and the exclusion, and the technical basis, of this primary FEP is acceptable. Should future design changes include a liner, however, DOE would need to provide data on the liner material and laboratory or analog data that can be used to evaluate its effect on repository performance.

Staff either does not support DOE's preliminary screening or concluded that there is an inadequate technical basis for exclusion of the following 24 FEPs.

1.1.02.00.00—Excavation/construction; 1.1.02.01.00—Site flooding (during construction and operation); 1.1.02.03.00 Undesirable materials left; 1.1.03.01.00—Error in waste or backfill emplacement; 1.1.12.01.00—Accidents and unplanned events during operation

These FEPs involve operational and preclosure aspects of the proposed repository and were excluded in DOE's preliminary screening on the basis of low consequence. As explained in the screening argument for FEP 1.1.03.01.00,

The TSPA is based on an assumption that the repository will be constructed, operated, and closed according to design. Deviations from design during the operational period are the subject of an extensive quality control program, and are outside the scope of the long-term performance assessment. Significant deviations that are detected during the operational period will be corrected and, therefore, are excluded from the TSPA on the basis of low probability.

Operational errors cannot be excluded without quality control procedures. Quality control procedures and operational procedures under normal and accident conditions are reviewed in specific sections of the Yucca Mountain Review Plan. Also, DOE references these FEPs with regard to the operational period; preclosure aspects of the proposed repository are reviewed under the preclosure section of the Yucca Mountain Review Plan and not in the postclosure model abstraction section that includes this integrated subissue. Nevertheless, these preclosure activities may impact post-closure repository performance. An inadequate technical basis for screening these FEPs has been provided. In addition, the screening of some of the FEPs is inconsistent with post-closure performance assessment calculations. For instance, the effects of the FEP on undesirable materials is required for the analysis of potential microbial effects on performance (see section 4.1; CRWMS M&O, 1998a). DOE needs to provide a more robust technical basis for exclusion of these FEPs, perhaps by citing project documents where these assumptions and postulated actions (i.e., actions to correct mistakes) are codified. Also DOE needs to screen these FEPs so that the screening is consistent with actual post-closure performance assessment calculations.

#### 1.1.07.00.00—Repository design; 1.1.08.00.00—Quality Control

These two FEPs are included in the DOE screening, but deviations from design and operational error are excluded on the basis of low consequence. As explained in the screening argument for FEP 1.1.03.01.00,

The TSPA is based on an assumption that the repository will be constructed, operated, and closed according to design. Deviations from design during the operational period are the subject of an extensive quality control program. Significant deviations that are detected during the operational period will be corrected and, therefore, are excluded from the TSPA on the basis of low probability.

The excluded portions of these FEPs involve operational and preclosure aspects of the proposed repository. Nevertheless, these pre-closure activities may impact post-closure repository performance. An inadequate technical basis for screening these FEPs has been provided. DOE needs to provide a more robust technical basis for exclusion of these FEPs, perhaps by citing project documents where these assumptions and postulated actions (i.e., actions to correct mistakes) are codified.

#### 1.2.06.00.00—Hydrothermal activity

While this FEP refers to natural hydrothermal activity and is excluded on the basis of low consequence, the preliminary draft Unsaturated Zone Flow Process Model Report (CRWMS M&O, 2000e) states that, “mineralogical data suggest that no hydrothermal alteration has occurred since the waning of Timber Mountain volcanism about 10 Ma.” This statement refers to probability and not consequence. An argument to exclude this FEP based on consequences would demonstrate that if hydrothermal activity occurred, then repository performance would not be adversely affected. In addition, the technical basis does not even address the alternate hypothesis that hydrothermal activity has occurred. This alternative hypothesis is the focus of the ongoing University of Nevada Las Vegas fluid inclusion study. At least some of the fluid inclusion evidence suggest thermal fluids (45 - 80 °C) have passed through Yucca Mountain. Although earlier work has dated the age of one elevated fluid inclusion samples as 160 ka (Dublyansky, 1998), the current fluid inclusion study has found that the fluid inclusions with measured elevated temperatures are not in the very youngest minerals that precipitated (Cline, 2000). Thus DOE should either screen this FEP based on probability, taking into account the observations from the fluid inclusion study, or screen the FEP based on consequences and demonstrate that if hydrothermal fluids did occur then the magnitude and time of the resulting expected annual dose would not be significantly changed by its omission.

Because data and analyses do not currently support the likelihood of hydrothermal activity, the exclusion of this FEP from DOE evaluation of coupled THC effects on the waste package chemical environment is acceptable.

#### 2.1.02.08.00—Pyrophoricity

DOE excluded this FEP on the basis of low consequence. However, an adequate technical bases was not provided to support the exclusion of this FEP. The screening argument for this FEP relies on calculations that have not yet been performed. Results from the proposed calculations would provide an adequate basis for the screening of this FEP. The exclusion of

this FEP is unacceptable because the DOE has not provided a clear technical basis and supporting calculations.

#### 2.1.06.06.00—Effects and degradation of drip shield

DOE provided no exclusion/inclusion statement for this FEP, which evaluates the consequences of drip shield degradation by physical and chemical processes. Staff notes that degradation of the drip shield could significantly affect both the integrated subissue on quantity and chemistry of water contacting waste packages and waste forms, and the integrated subissue on degradation of engineered barriers. Any FEP contained within the DOE in-drift geochemical submodels also must be contained within the FEP database. DOE must commit to a screening stance and provide an adequate technical basis to support their screening decision.

#### 2.1.08.07.00—Pathways for unsaturated flow and transport in the waste and engineered barrier system

This FEP evaluates unsaturated flow and radionuclide transport that may occur along preferential pathways in the waste and engineered barrier system and was given an “include?” status in the screening. The preliminary current screening argument for this FEP explains that “the details of internal pathways providing release from a container are subsumed in an integrated release distribution.” The technical basis for this FEP, however, cannot be subsumed by an integrated release distribution. Screening arguments for this FEP should properly address topics contained within the FEP description. For example, because the FEP description states that “Physical and chemical properties of the engineered barrier system and waste form, in both intact and degraded states, should be considered in evaluating pathways,” staff expects the screening arguments to be based on an evaluation of these topics. DOE must commit to a screening stance and provide an adequate technical basis to support their screening decision.

#### 2.1.09.02.00—Interaction with corrosion products

This FEP is described by the statement “Corrosion products produced during degradation of the metallic portions of the engineered barrier system and waste package may affect the mobility of radionuclides. Sorption/desorption and coprecipitation/dissolution processes may occur”, and was given an “exclude?” status in the preliminary DOE FEP database (U.S. Department of Energy, 1999). DOE must commit to a screening stance and provide an adequate technical basis to support their screening decision.

Staff notes several potential problems associated with this FEP. For example, descriptions of associated secondary FEPs cover a broader range of issues than the primary FEP. For example, secondary FEP 2.1.09.02.03 (Container Corrosion Products) states that, “Container corrosion products could affect the vault environment, causing significant changes to sorption, transport, dissolution, and long-term stability (AECL).” Descriptions of primary FEPs should be sufficiently broad to contain the general aspects of related secondary FEPs. The topics of screening FEPs and classification of FEPs, and the associated acceptance criteria, are contained in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a).

Staff also notes that the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) describes a corrosion products submodel that will evaluate changes to water chemistry from

reactions between metallic components, their corrosion products, and water that percolates into the drift. This submodel is an integral part of the in-drift geochemical model described in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) but does not appear to be adequately addressed anywhere in the preliminary DOE FEP database (U.S. Department of Energy, 1999). Any FEP contained within DOE's in-drift geochemical submodels also must be contained within the FEP database. There should be FEPs in DOE's database, for example, that evaluate how water compositions vary as a result of seepage/corrosion product interactions and how differences in initial water composition affect corrosion products.

#### 2.1.09.07.00—Reaction kinetics in waste and engineered barrier system

This FEP was excluded by DOE. The description and screening argument given in the preliminary DOE database for this FEP are inconsistent. The FEP description reads, "Chemical reactions, such as radionuclide dissolution/precipitation reactions and reactions controlling the reduction-oxidation state, may not be [in] equilibrium in the drift and waste environment" (U.S. Department of Energy, 1999). The screening argument, however, only addresses redox reactions. Staff requires that DOE clarify the description of this FEP, provide separate screening arguments for redox kinetics, and dissolution/precipitation kinetics, and consider the effects of both types of reaction kinetics under the full range of temperatures expected to exist in the near-field environment during the performance period in their assessment of the technical basis for exclusion. According to Section 3.3.4 of the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), a major assumption of DOE's in-drift geochemical environment model is that "reactions proceed to equilibrium" because "water movement through the drift is slow compared to the reaction rates between the water and solids in the drift." Staff notes that "major assumptions" (U.S. Department of Energy, 1999) are not acceptable as screening arguments for the exclusion of any FEP. This FEP remains open until DOE provides adequate technical bases to support their "exclude" screening status. The exclusion of this FEP is unacceptable because DOE has not provided a clear technical basis and supporting calculations.

#### 2.1.09.09.00—Electrochemical effects in waste and engineered barrier system

This FEP evaluates electrochemical effects that may establish an electric potential within the drift or between materials in the drift and more distant metallic materials. Ion migration in an electric field could affect corrosion of metals in the engineered barrier system and the waste. DOE excludes this FEP based on the screening argument that "galvanic protection is considered for Yucca Mountain containers." When two dissimilar metals are in contact with each other and exposed to an electrolyte, galvanic interactions can occur. Proposed materials inside the emplacement drifts include carbon steel, Alloy 22, Type 316 NG stainless steel, and titanium grade 7. Contact between the drip shield (or waste package) and the steel sets will likely cause increased corrosion of the carbon steel. The hydrolysis of the soluble steel corrosion products may result in the formation of iron hydroxides and will also tend to make the electrolyte in the vicinity of the steel more acidic. Thus, staff is concerned that electrochemical interactions may alter the chemistry of the water in contact with the waste package and recommend the inclusion of this FEP because of the potential impact these interactions may have on the five DOE submodels related to the waste package environment. The exclusion of this FEP is unacceptable because DOE has not provided a clear technical basis and supporting calculations.

#### 2.1.10.01.00—Biological activity in waste and engineered barrier system

This FEP evaluates biological activity in the waste and engineered barrier system that may affect disposal-system performance by altering degradation processes such as waste package and waste form corrosion, affecting radionuclide transport through the formation of colloids and biofilms, and generating gases. The preliminary DOE category for this FEP was “include?” The TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) describes an in-drift microbial communities submodel that will bound the ultimate abundance of microbes and evaluate “changes in microbe abundance due to thermal loading and introduced materials.” This FEP is related to that submodel, as well as to ENFE Subissue 2. All FEPs included in DOE submodels must also be included in the DOE FEP database. Staff position on this FEP remains open until DOE commits to a screening stance and provides adequate technical bases to support that screening decision.

#### 2.1.11.03.00 Exothermic reactions in waste and engineered barrier system

While staff considers the effects of exothermic reactions in the waste and engineered barrier system on the waste package and drip shield chemical environment to be negligible compared to the effects of other coupled THC processes, DOE did not provide an adequate technical basis to reach this conclusion. Staff considers the DOE screening argument for this FEP appropriate and exclusion of this FEP from DOE analyses of the quantity and chemistry of water contacting the waste packages and waste forms is acceptable. Nevertheless, DOE must provide an adequate technical basis to support their screening decision.

#### 2.1.11.08.00—Thermal effects: chemical and microbiological changes in the waste and engineered barrier system

This DOE-excluded FEP evaluates how temperature changes in the drift may affect chemical and microbial processes in the waste and engineered barrier system. Screening arguments for the exclusion of this FEP were judged inadequate. The rates of near-field processes and microbiological activity may change in response to the thermal pulse generated by waste emplacement. The microbial communities submodel will consider “changes in microbe abundance due to thermal loading and introduced materials (TSPA-SR methods and assumptions)” for the TSPA-SR. Any FEP contained within the DOE in-drift geochemical submodels also must be listed as included within the FEP database. Staff position on this FEP remains open until DOE provides adequate technical bases to support the screening decision.

#### 2.1.12.01.00—Gas generation; 2.1.12.03.00—Gas generation (H<sub>2</sub>) from metal corrosion; 2.1.12.04.00—Gas generation (CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>S) from microbial degradation

DOE excludes these three FEPs largely because the repository will be located in the unsaturated zone and is well connected to the surface, minimizing the potential for postclosure gas buildup (U.S. Department of Energy, 1999). Gas generation could potentially affect the validity of the thermal regimes selected for the TSPA-SR analyses, which will be distinguished from one another, in part, by calculated gas compositions. In addition, exclusion of gas generation processes in the DOE analyses could result in faulty predictions about the abundance and relative proportions of various aqueous species that may be in the drift. Although mass balance calculations performed for the TSPA-VA (U.S. Department of Energy, 1998b) were used to support the assumption that “gas composition is not affected substantially

by reaction with the in-drift solids,” DOE must provide sufficient technical bases to demonstrate whether gas generation processes will be significant in the Enhanced Design Alternative-II repository design.

#### 2.1.12.05.00—Gas generation from concrete

This FEP evaluates the production of gases from the aging and degradation of concrete that may occur through radiolysis of water in the cement pore spaces and microbial growth on concrete. The screening argument given for the exclusion of this FEP is, “Ignored until importance, if any, can be quantified.” In contrast, staff believes that this FEP should be included, until its lack of importance, if any, can be quantified. The exclusion of this FEP may be acceptable, but staff position on this FEP will remain unresolved until the DOE provides adequate technical bases to support their screening decision.

#### 2.1.12.06.00—Gas transport in waste and engineered barrier system

This FEP was excluded in the preliminary FEPs database (U.S. Department of Energy, 1999) on the basis that radioactive gases are not a significant component of the future radionuclide inventory. There is no technical basis provided in the FEPs database, and there is a presupposition that the doses resulting from this transport are low. Earlier TSPA analyses by both the DOE (TRW Environmental Safety Systems, 1995) and NRC (U.S. Nuclear Regulatory Commission, 1995b), and a report prepared by the National Academy of Sciences (National Research Council, 1995) all suggest that the calculated dose from gaseous radionuclide transport is low. The most recent revision of the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) has closed out gaseous transport of  $^{14}\text{CO}_2$  as an open item.

The primary description for this FEP states, “Gas in the waste and engineered barrier system could affect the long-term performance of the disposal system. Radionuclides may be transported as gases or in gases, gas bubbles may affect flow paths, and two-phase flow conditions may be important.” It is not clear from this description whether this FEP is intended to address all gas species or just radioactive gas species. Staff were unable to identify any other FEP that explicitly addresses gas transport, but note the importance of two-phase flow in DOE TH and THC models. This FEP will remain open until DOE’s primary description is clarified. Staff recommends that DOE identify a FEP that explicitly includes transport of non-radioactive gases.

#### 2.1.13.02.00—Radiation damage in waste and engineered barrier system

This FEP evaluates strong radiation fields that “could lead to radiation damage to the waste forms and containers (commercial spent nuclear fuel, DOE spent nuclear fuel, defense high level waste), backfill, drip shield, seals, and surrounding rock.” Changes to container properties are ignored because they are expected to be small compared to corrosion. Radiation damage to backfill, seals, and rock are excluded because the effects are limited to a distance of about 30 centimeters from the source. Staff does not support the exclusion of radiation damage to container properties, backfill, and seals until adequate technical bases have been provided to support the screening arguments. Staff’s position on this FEP remains open until DOE provides adequate technical bases to support the decision to exclude portions of this FEP.

#### 2.2.08.04.00—Redissolution of precipitates directs more corrosive fluids to containers

This FEP evaluates the “redissolution of precipitates that have plugged pores as a result of evaporation of groundwater in the hot zone,” and the possibility that resulting solutions may be more corrosive than the original pore fluids. This FEP was excluded on the basis of low consequence because “plugged pores are least likely to reopen because there is no flow to dissolve the precipitates.” However, comments such as “need clarification of this FEP” appear in the text for the screening argument, suggesting that DOE has not yet committed to the scope of this FEP. The exclusion of this FEP may be acceptable, but the text should be revised to clearly indicate the scope of this FEP, DOE’s stance on the screening, and the technical bases.

Additional review comments relevant to ENFE Subissue 2, the integrated subissue on quantity and chemistry of water contacting the waste packages and waste forms, and acceptance criterion 1 are listed below.

The TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) indicates that changes in water chemistry owing to seepage/backfill interactions will be evaluated throughout the thermal evolution of the proposed repository. The evolution of water compositions due to backfill interactions is a key component of the seepage/backfill interactions submodel (CRWMS M&O, 1999a) but is only tangentially covered by secondary FEP 2.1.04.02.09 (Water Chemistry, Tunnel Backfill). If backfill is included in the Enhanced Design Alternative-II repository design, DOE should clearly define a FEP(s) that adequately captures all the FEPs contained within the seepage/backfill interactions submodel for the TSPA-SR.

The list of FEPs in Table 8 of the preliminary draft Precipitates/Salts Analysis and Model Report (E0105) (CRWMS M&O, 2000h) does not adequately reflect the relevant phenomena. Three of the four FEPs in the list deal with backfill, and it is stated in Table 22 of the preliminary draft Analysis and Model Report that two of the three are not addressed in the Analysis and Model Report. All FEPs considered in the preliminary draft Precipitates/Salts Analysis and Model Report must be represented in the FEP database.

Coupled THC processes affect the two integrated subissues relevant to ENFE Subissue 2 and represent an integral part of DOE’s conceptual model for performance assessment (CRWMS M&O, 1999a). Groundwater composition is sensitive to coupled THC conditions along the flow path and is expected to evolve as a function of time and space following waste emplacement. DOE’s spatial and temporal abstractions must appropriately address physical couplings (THMC). If any THMC couplings are excluded from DOE’s abstractions, then adequate technical bases must be provided. DOE therefore needs to provide adequate technical bases and justification for their major assumption (CRWMS M&O, 1999a) that THC processes can be decoupled, evaluated separately, and then recoupled without adversely affecting dose. Appropriate technical bases may stem from activities such as independent modeling, laboratory or field data, or sensitivity studies.

Based on the description of the seepage/cement submodel in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), calculations for the TSPA-SR should be expanded to account for variations in solution composition due to interactions with materials in the repository other than cement. When the Analysis and Model Report E0055 (Seepage/Cement Interaction) becomes available, it will be reviewed in the context of the effects of coupled THC processes on the waste package and drip shield chemical environment. DOE’s calculations on

seepage/cement interactions for the TSPA-SR should take into consideration the Enhanced Design Alternative-II repository design, including any grouted rock bolts, and the time-temperature regime being considered. The current TSPA-SR reference design does not include a cement liner for the repository tunnels, and DOE does not need to include this design feature in its TSPA abstraction. Should future design changes include a liner, however, DOE would need to provide data on the liner material and laboratory or analog data that can be used to evaluate its effect on repository performance.

The in-drift gas flux and composition submodel for the TSPA-VA (U.S. Department of Energy, 1998b) assumed that gas composition is not affected substantially by reaction with the in-drift solids. This assumption was supported by mass balance calculations (U.S. Department of Energy, 1998b). This approach will be acceptable for the TSPA-SR, but a new technical basis must be provided that is consistent with the Enhanced Design Alternative-II repository design. It is expected that new technical bases will be provided in the Analysis and Model Report E0035 (In-Drift Gas Flux & Composition) and reflected in DOE's screening of related FEPs, such as 2.1.12.01.00 (Gas generation), 2.1.12.03.00 (Gas generation from metal corrosion), and 2.1.12.04.00 (Gas generation from microbial degradation). Staff will review the Analysis and Model Report E0035 (In-Drift Gas Flux & Composition) when it becomes available.

In the TSPA-VA (U.S. Department of Energy, 1998b), several discrete time regimes were defined to have constant chemical conditions so the description of the NFGE evolution would consist of a set of step changes in system conditions. Temperature and gas compositions were used to define these regimes. If a similar approach is taken for the TSPA-SR, DOE must provide a new technical basis for distinguishing between regimes that account for new design features and are consistent with results from the in-drift gas flux and composition submodel calculations.

The preliminary draft In-Drift Precipitates/Salts Analysis and Model Report (CRWMS M&O, 2000h) results do not adequately reflect the range of environmental conditions that could lead to rapid drip shield degradation. Recent studies<sup>2</sup> indicate that localized and general corrosion rates for a Ti-0.15Pd drip shield are highly sensitive to dissolved F<sup>-</sup> concentrations, but are relatively insensitive to Cl<sup>-</sup> concentrations. Corrosion rates increase steeply above a critical value of ~50 millimolar F<sup>-</sup> ions. Assuming constant corrosion rates over time, the drip shield would be breached within tens of years if F<sup>-</sup> concentrations were to reach the values predicted for wetting/evaporation cycles at Yucca Mountain (Shoesmith, 2000). . In addition to tracking pH, chloride, and ionic strength through different repository scenarios, the extent to which precipitates and aqueous complexes sequester available fluoride should be explicitly evaluated, and the results should be presented in the Precipitate/Salts Model Lookup Tables.

#### **5.4.2.2.1.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 2**

Sufficient data on design features (including drip shield, backfill, waste packages, cladding, other engineered barrier components, and thermal loading), geology, hydrology, geochemistry, and geomechanics of the unsaturated zone and drift environment (e.g., field, laboratory, and

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<sup>2</sup>Brossia, C.S. and G.A. Cragolino, *Effects of environmental and metallurgical conditions on the passive and localized dissolution of Ti-0.15Pd*, *Corrosion Science* (submitted for publication)

natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing the abstraction of the quantity and chemistry of water contacting waste packages and waste forms in the performance assessment. The data are also sufficient to assess the degree to which FEPs related to the quantity and chemistry of water contacting waste packages and waste forms and which affect compliance with postclosure performance objectives have been characterized and to determine whether the technical bases provided for exclusion of these FEPs are adequate. Where adequate data do not exist, other information sources such as expert elicitation have been appropriately incorporated into the abstraction process. Specifically,

- DOE demonstrates that sufficient data were collected on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, to establish initial and boundary conditions, including temporal and spatial variations in conditions, for conceptual models and simulations of THMC coupled processes that affect the waste package and drip shield chemical environment
- Where sufficient data do not exist, the definition of parameter values and conceptual models is based on appropriate other sources such as expert elicitation conducted in accordance with NUREG-1563 (Kotra, et al., 1996)
- DOE provides sufficient data for sound bases for the exclusion of certain observed phenomena in its conceptual models
- DOE collects sufficient information to formulate the conceptual approach(es) for analyzing water contact with the waste packages upon drip shield failure
- DOE provides sufficient data to complete a nutrient and energy inventory calculation, if it has been used to justify the exclusion of the potential for microbial activity affecting the waste package and drip shield chemical environment, or has been used to abstract microbial effects
- If microbial activity could be sufficient to allow microbially induced corrosion of the waste package or drip shield, DOE provides sufficient data to determine the time-history of temperature, humidity, and dripping to constrain microbially induced corrosion and microbial effects such as production of organic by-products
- DOE performs sensitivity, if needed, or uncertainty analyses (including consideration of alternative conceptual models) to test for the necessity of additional data

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The TSPA-SR reference design calls for the drip shield to be installed after waste emplacement is complete. DOE has provided a limited sensitivity analyses of the effects of a drip shield in the design selection report (CRWMS M&O, 1999b). There is little, if any, laboratory or field information available on the performance of the titanium drip shield, however, and at the current time, the status of the data sufficiency for this engineered barrier component is very uncertain. DOE will need to provide additional data on design features and the technical bases for the description of these features in DOE's total system performance assessment abstraction.

In the Final Report TSPA Peer Review Panel (Whipple, et al., 1999), large uncertainties were noted in DOE-modeled results because of assumptions made by analysts using MING. Specifically, the report cites DOE efforts to bound microbial growth as a function of biomass production based on nutrient or energy limitations, which require “important, perhaps unknowable, assumptions.” The Panel concluded:

[T]his complex set of assumptions and information requirements used to analyze the contribution of microbial growth leads to a highly uncertain and speculative estimate. Taken as a whole, the Panel does not see the above approach as a clear path to answer the concerns regarding microbial activity. The answers to these questions are not likely to come from more sophisticated models and analysis, which require unavailable information, but rather from bounding analysis and the selection of materials which resist microbial activity.

Staff notes the difficulty in placing reasonable bounds on the parameters needed to resolve topics related to microbial activity. However, the path to resolving these issues at a potential repository at Yucca Mountain is through bounding analysis of microbiological effects on chemistry coupled with the selection of materials resistant to microbiological activity.

#### **5.4.2.2.1.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 2**

Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstraction of quantity and chemistry of water contacting waste packages and drip shields, such as the pH, chloride concentration, and amount of water flowing in and out of the breached waste package, are consistent with site characterization data, design data, laboratory experiments, field measurements, and natural analog data, are technically defensible and reasonably account for uncertainties and variabilities. The technical bases for the parameter values used in the abstraction are provided. Specifically,

- Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the quantity and chemistry of water contacting waste packages and drip shields calculations in the performance assessment are technically defensible and reasonable, based on data from the Yucca Mountain region (e.g., results from large-block and drift-scale heater and niche tests) and a combination of techniques that may include laboratory experiments, field measurements, natural analog research, and process-level modeling studies
- DOE demonstrates that parameters derived from process-level models used for the quantity and chemistry of water contacting waste packages and drip shields are consistent with site characterization data, laboratory experiments, field measurements, and natural analog information
- DOE provides technical bases for parameter ranges, probability distributions or bounding values. DOE demonstrates that the parameter values are derived from site-specific data or provides an analysis to demonstrate that the assumed parameter values do not under-predict repository performance

- DOE demonstrates that input values used in the quantity and chemistry of water contacting engineered barriers (e.g., drip shield, waste package, and cladding) calculations and abstractions are consistent with the initial and boundary conditions and the assumptions of the conceptual models and design concepts for the Yucca Mountain site, such as waste package and engineered barrier system design (including backfill, drip shield, ground support, and cladding), waste package degradation (corrosion and mechanical disruption), cladding degradation, deep percolation flux, important THMC coupling effects, the thermal reflux model, the thermal-loading strategy (including effects of ventilation), natural system masses and fluxes, and other design features that may affect performance
- DOE establishes that reasonable or conservative ranges of parameters or functional relations are used to determine effects of coupled THMC processes on the waste package and drip shield chemical environment
- DOE shows that the parameters used to define initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THMC effects on the waste package and drip shield chemical environment are consistent with available data
- DOE provides confirmation that the correlations between input values, if any exist, have been appropriately established in the total system performance assessment
- DOE demonstrates that parameter values, assumed ranges, probability distributions, and bounding assumptions reasonably account for uncertainties
- DOE adequately considers the uncertainties in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THMC coupled processes that affect the waste package and drip shield chemical environment
- DOE adequately represents uncertainty in parameter development for conceptual models, process-level models, and alternative conceptual models considered in developing the abstraction of quantity and chemistry of water contacting waste packages and drip shields, either through sensitivity analyses or conservative limits
- DOE demonstrates how parameters used to describe flow through the engineered barrier system bounds the effects of backfill and excavation-induced changes

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Results shown in Table 3 of preliminary draft Analysis and Model Report E0105 (Precipitates Salts Analyses) (CRWMS M&O, 2000h) require additional explanation. The concentration factor for this evaporation experiments is 956, and sodium increased by a factor of 958, which is an indication of conservative behavior of sodium and a good chemical analysis. However,  $\text{Cl}^-$  concentration increased by a factor of only 700, which seems most likely to indicate bad chemical analysis. Aqueous  $\text{SiO}_2$  increased by a factor of 1600, which is inconsistent with a

concentration factor of 956. Variations in pH were not reported. Use of these data to validate models, the primary goal of which is to predict ionic strength,  $\text{Cl}^-$  concentration, and pH, is unreasonable.

DOE must reconcile inconsistencies in the preliminary draft Analysis and Model Report on Precipitates Salts Analyses (E0105). Although it is reasonable to limit the low relative humidity model components, the rationale that “other components are almost entirely precipitated at lower ionic strength in evaporated J-13 Well water” (p. 32) is inconsistent with the data in Table 3 of the preliminary draft Analysis and Model Report showing elevated  $\text{F}^-$  and  $\text{SiO}_2$  concentrations in evaporated J-13 water (CRWMS M&O, 2000h).

Staff does not accept the claim in the preliminary draft Analysis and Model Report E0105 (Precipitates Salts Analyses) that model validation criteria are met in every case. The preliminary draft Analysis and Model Report reasonably notes major discrepancies between model results and data used to judge “validity.” The validation criterion that the low relative humidity model should provide results that agree with the high relative humidity model where the two models are joined (p. 63) is risky if the high relative humidity model is imperfect (which it is). The validation criteria that  $\text{Cl}^-$  concentration, ionic strength, and pH are conservative or within an order of magnitude (p. 63) are also risky because conservatism is difficult to determine. For example, low-ionic strength may be conservative for colloid transport but high-ionic strength may be conservative for corrosion. A risk-informed approach should be used to evaluate the conservatism of assumptions regarding  $\text{Cl}^-$  concentration, ionic strength and pH. One acceptable approach, for example, would be to perform sensitivity studies on the effects that variable  $\text{Cl}^-$  concentration, ionic strength and pH have on key FEPs, and then evaluate the impact that these effects have on repository performance.

Two major assumptions DOE plans to implement in the TSPA-SR are relevant to both the ENFE Subissue 2 and the integrated subissue on quantity and chemistry of water contacting waste packages and waste forms. These assumptions are: (i) kinetics can be ignored because all reactions proceed to equilibrium, and (ii) coupled THC processes can be reasonably approximated by decoupling and then recoupling submodels. DOE must provide technical bases demonstrating that parameter values, assumed ranges, probability distributions, and bounding assumptions reasonably account for uncertainties.

The preliminary draft Precipitates/Salts Analyses Analysis and Model Report (E0105) (CRWMS M&O, 2000h) assumes equilibrium chemical conditions (p. 22), except some minerals are not permitted to precipitate. These are reasonable assumptions for a model. However, it is incorrect to conclude that “the assumption of equilibrium conditions will not affect the uncertainty in the model” (p. 22). Indeed many of the discrepancies between the model and experimental data examined in an effort to validate the model are ascribed to kinetic effects. A major assumption of DOE’s approach for the TSPA-SR is that reactions proceed to equilibrium (CRWMS M&O, 1999a). If DOE elects to proceed under the assumption of chemical equilibrium, then uncertainties associated with this assumption must be adequately considered.

DOE has not clearly documented the basis for the parameter probability distribution functions. This may be because data are not otherwise readily available. It thus appears as if informal expert elicitation is used to define the parameter ranges. Development of parameter distributions through elicitation is acceptable, but DOE should conduct and document these

expert elicitations in accordance with the guidance in Kotra, et al. (1996) or other acceptable methods.

#### **5.4.2.2.1.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 2**

Alternative modeling approaches consistent with available data (e.g., design features, field, laboratory, and natural analog) and current scientific understanding are investigated, and results and limitations are appropriately factored into the abstraction of quantity and chemistry of water contacting waste packages and drip shields. DOE provided sufficient evidence that alternative conceptual models have been considered, that the models are consistent with available data and current scientific understanding, and that the effect of these alternative conceptual models on total system performance has been evaluated. Specifically,

- DOE investigates alternative modeling approaches consistent with available data and current scientific knowledge, and appropriately considers their results and limitations of these approaches in developing the abstraction of quantity and chemistry of water contacting waste packages and drip shields
- DOE adequately considers the effects of THMC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products in their assessment of alternative conceptual models. DOE considers: (i) thermohydrologic effects on gas and water chemistry, (ii) hydrothermally driven geochemical reactions such as zeolitization of volcanic glass, which could affect water chemistry and waste package and drip shield environmental conditions, (iii) dehydration of hydrous phases liberating moisture that may affect the waste package and drip shield chemical environment, (iv) effects of microbial processes on the waste package and drip shield chemical environment, and (v) changes in water chemistry that may result from the release of corrosion products from the waste package and drip shield and interactions between engineered materials and groundwater, which, in turn, may affect the waste package and drip shield chemical environment, in their assessment of alternative conceptual models
- If DOE uses an equivalent continuum models (ECMs), then DOE demonstrates that the models do not under-predict overall performance
- DOE provides a description which includes a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The ability of DOE to adequately track the evolution in water composition between different in-drift geochemical submodels and RIP mixing cells is of fundamental importance to the resolution of ENFE Subissue 2. DOE should provide reasonable technical bases for the infiltrating water composition(s) used in each in-drift geochemical submodel. Staff also support DOE's plans to perform sensitivity studies to learn the effects of different infiltrating water

compositions on the in-drift geochemical model output (CRWMS M&O, 1999a) and expect the results of these analyses to be placed in the context of the performance assessment.

DOE must adequately consider the effects of THMC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products in their assessment of alternative conceptual models. Hence, addressing the “major [DOE] assumption” that coupled processes can be reasonably approximated by decoupling and then recoupling the processes is a significant, but necessary, task associated with acceptance criterion 4. Technical bases should be provided demonstrating that parameter uncertainties associated with this assumption have been appropriately bounded and do not lead to an overestimation of repository performance.

Staff review of the preliminary draft Salts/Precipitates analysis Analysis and Model Report (E0105) (CRWMS M&O, 2000h) has resulted in several questions regarding model uncertainty. These are described in the following three paragraphs.

The approach used to develop the PT4 database for modeling the behavior of electrolyte solutions is questionable for a number of reasons. One reason is the manner in which Pitzer parameters from different sources were taken and used. The PT4 database is a modification of the PIT database, with added parameters from the Harvie-Moller-Weare database, from the Reardon (1990) paper, and, in the absence of parameter values, from analogy with other aqueous species [e.g.,  $\text{SiO}_2(\text{aq})$  parameters assumed equivalent to those involving  $\text{CO}_2(\text{aq})$ ]. The strength of the ion-interaction approach to calculations of mineral-solution equilibria, as used by Harvie, Moller, and Weare (1984), from whose work the Harvie-Moller-Weare database was taken, lies in large part in the internal consistency of the ion-interaction parameters and the Gibbs free energy of formation of the solid phases. Harvie, Moller, and Weare (1984) evaluated the ion-interaction parameters and compared the experimental and calculated solubilities to develop a list of recommended interaction parameters and solid phase properties. Such evaluation and comparison were not done by the authors of the PT4 database. The PIT database itself has not undergone any internal consistency check—the parameters in the database were derived from several sources.

Also, to allow calculations up to 100 °C, the PT4 database used the median value of the known values for  $d\beta^0/dT$ ,  $d\beta^1/dT$ ,  $d\beta^2/dT$ , and  $dC^{\eta}/dT$  for the unknown values of those parameters. There is no reason why this assumption would work, and it is stated in the preliminary draft Analysis and Model Report that how this assumption affects the accuracy of the model results is not directly investigated. Furthermore, although the temperature derivatives of the cation-anion parameters were given values in the PT4 database, there is no indication from the preliminary draft Analysis and Model Report that the temperature dependence of the  $\eta$  and  $\eta$  terms was considered. The PIT database does not include values for  $d\eta/dT$  and  $d\eta/dT$ , although both parameters undoubtedly vary with temperature. For calculations to 200 °C, Pabalan and Pitzer (1987) found it was adequate to set the value of  $\eta$  to its 25 °C value and assume a simple temperature dependent equation for  $\eta$ .

Finally, the idea of combining the ion pairing concept with the ion-interaction approach is misguided. The Pitzer model is valid from dilute solutions to high ionic strength. It accounts for strong ion interactions—called ion pairing in the alternative approach—through its empirical parameters, although formation of complexed species (e.g.,  $\text{HCO}_3^-$ ,  $\text{HSO}_4^-$ ) can be explicitly included in the model if the ion-pair interaction is great enough. There are no rules implicit in

the Pitzer model to decide whether an ion-association reaction should be recognized. The decision is essentially an empirical one, based on achieving the best fit to available thermodynamic data, rather than most closely reflecting the molecular reality of the solution (Clegg and Whitfield, 1991). The choice of explicit inclusion of complexed species requires parameters to be included in the database to account for interaction of that species with all other species in the database. Haphazard inclusion of complexed species in the database only results in uncertainties in the model results, the magnitude of which cannot be determined without careful verification work. Therefore, staff recommends that the DOE salts/precipitates analysis include a rigorous analysis of model uncertainties, or bound the effects of the uncertainties on repository performance.

#### **5.4.2.2.1.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 2**

Output from the abstraction of quantity and chemistry of water contacting waste packages and drip shields is justified through comparison with output from detailed process-level models and/or empirical observations (e.g., laboratory testing, field measurements, and natural analogs). Specifically,

- DOE verifies that the outputs of the quantity and chemistry of water contacting the drip shield and waste package abstractions reasonably reproduce or bound the results of corresponding process-level models or empirical observations
- DOE demonstrates that abstracted models for coupled THMC effects on the waste package and drip shield chemical environment, are based on the same assumptions and approximations demonstrated to be appropriate for closely analogous natural or experimental systems
- DOE shows that abstracted model results were verified through comparison with outputs of detailed process-level models and empirical observations
- DOE evaluates the outputs of the abstraction against field and laboratory data and natural analogs information
- DOE appropriately adopts accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THMC effects on the waste package and drip shield chemical environment
- DOE compares abstracted model results with different mathematical models to judge robustness of results

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The preliminary draft Analysis and Model Report U0135 (Natural Analogs for Unsaturated Zone) (CRWMS M&O, 2000g) was reviewed to determine whether DOE natural analog studies have been suitably designed and implemented to be useful in supporting of total system performance assessment outputs. Although natural analog studies are applicable to a range of

performance related phenomena, this preliminary draft Analysis and Model Report focuses on studies related to flow and transport in the unsaturated zone and is most relevant to the subissues addressed in the Unsaturated and Saturated Flow under Isothermal Conditions and Radionuclide Transport IRSRs. In general, the preliminary draft Analysis and Model Report recognizes the potential significance of natural analog studies and identifies several potentially important sites and studies, but presents limited direct applications to performance assessment model support for the Yucca Mountain repository. Staff supports continuation of DOE efforts to use natural analogs for model support, but note that natural analog data must be more directly related to specific performance assessment abstractions or process-level models in order to further resolution of ENFE subissues.

#### **5.4.2.2.2 Integrated Subissue on Degradation of Engineered Barriers**

There are five generic acceptance criteria. Each generic acceptance criterion is followed by specific criteria. Only those criteria related to ENFE Subissue 2 are provided here. DOE's approach to abstract coupled THC effects on waste package and drip shield chemical environment in a total system performance assessment for the proposed repository at Yucca Mountain will be acceptable from the point of view of the integrated subissue on degradation of engineered barriers provided that each acceptance criterion is met. The review in this section focuses only on topics related to both the integrated subissue on degradation of engineered barriers and DOE abstractions of coupled THC effects on the waste package and drip shield environment. A comprehensive review of DOE abstractions on degradation of the engineered barriers for the TSPA-SR will be provided in the Container Life and Source Term IRSR Revision 3. A summary and review of DOE abstractions on degradation of the engineered barriers for the TSPA-VA (U.S. Department of Energy, 1998b) is available in the Container Life and Source Term IRSR Revision 2 (U.S. Nuclear Regulatory Commission, 1999d).

##### **5.4.2.2.2.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 2**

Important design features, physical phenomena and couplings, and consistent and appropriate assumptions have been identified and described sufficiently for incorporation into the abstraction of degradation of engineered barriers and other related abstractions in the total system performance assessment, and the technical bases are provided. The abstraction identifies and describes design features of the engineered barrier system and aspects of the degradation of engineered barriers that are important to waste isolation and includes the technical bases for these descriptions. Specifically,

- DOE identifies and considers likely corrosion and degradation modes for engineered barrier materials, including uniform corrosion, pitting corrosion, crevice corrosion, stress corrosion cracking, intergranular corrosion, microbially influenced corrosion, dry-air oxidation, and hydrogen embrittlement. DOE identifies the effects of material aging and phase stability of the engineered barrier materials and initial defects on the degradation modes for the engineered barriers
- DOE evaluates the possibility of gamma-radiolysis of the groundwater in contact with the engineered barriers and evaluated the effects of gamma-radiolysis products on the corrosion of the engineered barriers

- DOE considers whether the use of a drip shield could result in the extension of the humid-air corrosion regime, condensation formation and dripping from the under side of the drip shield, failure of the drip shield, hydrogen embrittlement and the possibility of crevice corrosion between the drip shield and the waste package

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The contents of the preliminary DOE FEP database (U.S. Department of Energy, 1999) were reviewed to identify primary FEPs related to both ENFE Subissue 2 and the integrated subissue on degradation of engineered barriers (Pickett and Leslie, 1999; U.S. Nuclear Regulatory Commission, 2000a). Table 5-4 shows the 30 relevant FEPs that were identified. Further review of the preliminary draft DOE FEP database (U.S. Department of Energy, 1999) has led to the identification of two additional FEPs (2.1.09.02.00—Interaction with corrosion products, and 2.1.06.06.00—Effects and degradation of drip shield) that are relevant to the waste package chemical environment subissue. Of the 32 FEPs relevant to ENFE Subissue 2 and the integrated subissue on the degradation of engineered barriers, 12 were either excluded or not assigned an include/exclude status by DOE's preliminary screening process. In addition, portions of the following three FEPs were excluded: FEP 1.1.07.00.00—Repository design, FEP 1.1.08.00.00—Quality control, and FEP 2.1.13.02.00—Radiation damage in waste and engineered barrier system. Of the 15 FEPs that were not included in the DOE screening process, 14 overlap with the quantity and chemistry of water contacting the waste packages and waste forms integrated subissue and are discussed in Section 5.4.2.2.1.1. The remaining excluded FEP, 2.1.09.03.00—Volume increase of corrosion products, is reviewed below.

#### 2.1.09.03.00—Volume increase of corrosion products

This FEP evaluates the possibility that the volume increase of corrosion products will compress the surrounding bentonite buffer, leading to increased mechanical stresses in the canister materials and faster degradation. This FEP was excluded on the basis that credit is not taken for the inner barrier. The technical basis for exclusion of this FEP is acceptable from the standpoint of ENFE Subissue 2.

DOE has identified and considered likely corrosion and degradation modes for waste package materials, and thus additional degradation submodels are currently not required for the waste package to resolve ENFE Subissue 2. However, the possibility of localized corrosion of the drip shield due to elevated fluoride concentrations should be evaluated by DOE.

DOE has included important design features in the waste package and drip shield degradation submodels for the TSPA-SR. If additional modifications are made in repository design, DOE submodels should reflect these changes.

#### **5.4.2.2.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 2**

Sufficient data from laboratory corrosion tests and in-service experience in pertinent industrial applications, as well as sufficient site-specific data, including data from drift-scale tests, are available to adequately define relevant parameters and conceptual models necessary for

developing the abstraction of the degradation of engineered barriers in the performance assessment. The data are also sufficient to assess the degree to which FEPs related to the degradation of engineered barriers have been characterized and to determine whether the technical bases provided for exclusion of these FEPs are adequate. Specifically,

- DOE justifies data on the degradation of the engineered barriers, including general and localized corrosion, hydrogen embrittlement, and galvanic interactions are based on laboratory measurements and tests designed to replicate the range of conditions that may occur at the Yucca Mountain site
- DOE performs sensitivity and uncertainty analyses on the effects of coupled THC processes on the engineered barrier environment to determine the necessity for additional data.
- DOE considers available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on the engineered barrier degradation
- DOE provides sufficient data on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, to establish initial and boundary conditions for conceptual models and simulations of THC coupled processes that affect degradation of engineered barriers
- DOE considers a nutrient and energy inventory calculation that is sufficient to determine the potential for microbial activity that could impact degradation of engineered barriers. Should microbial activity be sufficient to allow microbially induced corrosion of the engineered barriers, DOE constrains the consequences of microbially induced corrosion

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE has not considered a nutrient and energy inventory calculation that is sufficient to determine the potential for microbial activity that could impact degradation of engineered barriers. The enhancement factor for microbially induced corrosion was calculated from the results of exposures to sterile and inoculated solutions (TRW Environmental Safety Systems, Inc., 2000a,d). However, no information is provided on the possible preferential dissolution of alloying elements as a result of microbial activity. The determination of enhancement factors for microbially induced corrosion are based on few data obtained in short-term exposures. The possibility for preferential dissolution of alloying elements needs to be evaluated. In addition, the effect of temperature on the value of enhancement factor for microbially induced corrosion was not reported and should be considered. If specific conditions predicted by the in-drift geochemical model abstraction, such as water composition or temperature, favor microbially induced corrosion, these must also be considered in evaluations of the microbially induced corrosion enhancement factor.

As noted in Section 5.4.2.2.1.1, DOE has not provided sufficient data on the environmental conditions that could lead to rapid drip shield degradation. While precipitates and aqueous complexes could sequester much of the available  $F^-$ , DOE has not yet, but should, evaluate the extent of this sequestering. DOE should conduct further studies to identify and characterize the conditions of enhanced drip shield degradation, place reasonable constraints on the aqueous

conditions at Yucca Mountain that might lead to drip shield degradation, and use this information to define appropriate conceptual models for developing the abstraction of the degradation of engineered barriers in the performance assessment.

#### **5.4.2.2.2.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 2**

Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstraction of degradation of engineered barriers are consistent with data available from laboratory corrosion tests and in-service experience in pertinent industrial applications, as well as with site-specific data, including data from drift-scale tests, are technically defensible, and reasonably account for uncertainties and variabilities. The technical bases for the parameter values used in the abstraction are provided. Specifically,

- DOE identifies those engineered barrier degradation processes that are important to repository performance, and (i) adequately determines the broad range of environmental conditions in terms of temperature, applied stresses, redox conditions, and chemical composition of the water contacting engineered barriers that promote the various degradation processes, (ii) adequately bounds the likely range of environmental conditions within the waste package emplacement drifts, (iii) identifies reasonable or conservative ranges of parameters or functional relationships to define the chemical composition and the redox conditions of the engineered barrier environment and their evolution with time, and (iv) identifies uncertainty in data, due to both temporal and spatial variations affecting the engineered barrier physical and chemical environment
- DOE justifies that input values used in the engineered barrier degradation calculations within total system performance assessment are reasonable based on data from the Yucca Mountain region, such as heater test results, and other applicable laboratory tests and natural analogs
- DOE justifies the initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on the degradation of engineered barriers and demonstrates that they are consistent with available data
- DOE properly considers, in its evaluation of coupled THC processes, the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THC coupled processes that affect degradation of the engineered barriers

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE expects infiltrating water compositions to evolve as a result of reactions with engineered barrier materials and variations in near-field temperature (see Section 5.4.2.1.2). Yet, water compositions used by DOE to evaluate general aqueous corrosion rates do not appear to be representative of either measured or modeled water compositions from Yucca Mountain. Although DOE may have used more corrosive conditions to determine the aqueous corrosion

rate for the TSPA-SR, DOE's approach must also include an evaluation of corrosion rates that considers site-specific data. Site-specific data may include water compositions that were calculated in the corrosion products or precipitates/salts submodels or that were measured in the heater tests. If DOE can show that the current [non-site-specific] approach bounds predictions of dose, no further action would be required.

#### **5.4.2.2.2.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 2**

Alternative modeling approaches consistent with available data and current scientific understanding are investigated, and results and limitations are appropriately factored into the abstraction of the degradation of engineered barriers. DOE provides sufficient evidence that alternative conceptual models have been considered, that the models are consistent with available data (from laboratory corrosion tests and field measurements) and current scientific understanding, and that the effect of these alternative conceptual models on total system performance has been evaluated. Specifically,

- DOE considers plausible alternative models and justifies approaches used in the degradation of engineered barriers abstractions
- DOE incorporates appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems as described in the following examples. The effects of THC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products include: (i) thermohydrologic effects on gas and water chemistry, (ii) hydrothermally driven geochemical reactions such as zeolitization of volcanic glass, which could affect water chemistry and engineered barrier environmental conditions, (iii) dehydration of hydrous phases liberating moisture that may affect the engineered barrier environment, (iv) effects of microbial processes on the engineered barrier environment, and (v) changes in water chemistry that may result from the release of corrosion products from the engineered barriers and interactions between cementitious materials and groundwater, which, in turn, may affect the degradation of engineered barriers

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Staff has no review comments relevant to this acceptance criterion as of May 15, 2000.

#### **5.4.2.2.2.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 2**

Output from the abstraction of the degradation of engineered barriers is justified through comparison with output from detailed process-level models and empirical observations arising from laboratory tests and field measurements. Specifically,

- DOE demonstrates that the mathematical models for degradation of engineered barriers are consistent with conceptual models based on inferences about the near-field

environment, field data and natural alteration observed at the site, and expected engineered materials

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Staff has no review comments relevant to this acceptance criterion as of May 15, 2000.

#### **5.4.3 Subissue 3: Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Chemical Environment for Radionuclide Release**

DOE's approach to assess the effects of coupled THC processes on the chemical environment for radionuclide release must meet the following generic criteria for each relevant integrated subissue: (i) integration, (ii) data and model justification, (iii) data uncertainty, (iv) model uncertainty, and (v) model support. Quality assurance is handled in a separate section of the Yucca Mountain Review Plan.

The ENFE subissue on effects of coupled THC processes on the chemical environment for radionuclide release provides input to two integrated subissues (i) Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms and (ii) Radionuclide Release Rates and Solubility Limits (U.S. Nuclear Regulatory Commission, 2000a). The abstraction of radionuclide release requires input from the two abstractions relevant to this subissue (U.S. Nuclear Regulatory Commission, 2000a). The effects of coupled THC processes need to be considered in the evaluation of each abstraction. As part of this evaluation, the acceptance criteria listed in Section 5.4.3.2 will be used to review DOE's Process Model Reports and the supporting Analysis and Model Reports as they become available. This evaluation will also include a review of the relevant FEPs, those included and those excluded from DOE's total system performance assessment.

The review will focus on DOE's Waste Form Degradation Process Model Report and the supporting Analysis and Model Reports. The information provided in those Process Model Report and Analysis and Model Reports is expected to encompass the information required to assess the effects of coupled THC processes on the chemical environment for radionuclide release. DOE's Waste Form Degradation Process Model Report describes the models, abstractions, and analyses that address the following major topics related to waste form degradation: (i) inventory, (ii) commercial spent nuclear fuel cladding degradation, (iii) commercial spent nuclear fuel waste form degradation, (iv) DOE spent nuclear fuel, Navy fuel, plutonium disposition wastes degradation, (v) high-level waste glass degradation, (vi) dissolved radionuclide concentration limits, (vii) colloid-assisted radionuclide concentration limits, (viii) in-package chemistry, and (ix) in-package source term and radionuclide transport abstraction. An evaluation of DOE's abstraction of radionuclide inventory [topic (i)] is presented in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 2000a) and Container Life and Source Term IRSR (Revision 3, in preparation). An evaluation of the DOE abstraction of in-package source term and radionuclide transport [topic (ix)] will be presented in Section 5.4.4.

For this revision of the ENFE IRSR, the review in this section will focus on DOE models, abstractions, and analyses for topics (ii) to (viii) and on the following subset of relevant Analysis and Model Reports: E0065—Analysis and Model Report on In-Drift THC Analysis,

F0055—Analysis and Model Report on Commercial Spent Nuclear Fuel Degradation Model, F0095—Analysis and Model Report on Dissolved Concentration Limits, F0105—Analysis and Model Report on Colloid Source Term, Los Alamos National Laboratory, F0110—Analysis and Model Report on Colloid Source Term, Argonne National Laboratory, F0115—Analysis and Model Report on Colloid Source Term Abstraction, F0065—Analysis and Model Report on Other Waste Form Abstraction, F0130—Analysis and Model Report on In-Waste Package Chemistry Summary, and F0170—Analysis and Model Report on In-Waste Package Chemistry Abstraction. Also included in the review are the following Analysis and Model Reports on FEPs: N0080—FEPs for the Near-Field Environment, F0050—FEP Screen Cladding, F0185—In-Waste Package FEPs Screening, E0015—Engineered Barrier System FEPs Degradation Modes Analysis, and E0110—Engineered Barrier System Degradation Modes and FEPs Abstractions.

Most of these Analysis and Model Reports and the Waste Form Degradation Process Model Report were not available for review prior to May 15, 2000. In the following sections, the resolution status based on the Viability Assessment design and TSPA-VA are considered to be current, except where the relevant Analysis and Model Report is available. As appropriate, resolution status based on information contained in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) and in Revision 3 of the Repository Safety Strategy (CRWMS M&O, 2000a) has been included where the approaches differ from that used in TSPA-VA. It is important to note that the TSPA-SR methods and assumptions report and the Repository Safety Strategy report do not contain details on how the different modeling approaches will be implemented. Our review of DOE's approach will be documented as the Analysis and Model Reports and Process Model Reports become available. Information that may become available during DOE quality assurance audits will be incorporated as appropriate.

#### **5.4.3.1 U.S. Department of Energy Approach**

##### **5.4.3.1.1 Features, Events, and Processes Screening**

The approach DOE used for the Viability Assessment did not include a formal screening process for FEPs. As a result, many important design features, physical phenomena, and couplings that cause temporal and spatial variations in conditions were not evaluated in a performance assessment framework. Thus, the TSPA-VA models neglect, without explanation, known temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release.

Recently, DOE developed a more formal documentation of the FEPs identification and screening process for the TSPA-SR (Swift, et al., 1999). The contents of the preliminary draft DOE FEP database (U.S. Department of Energy, 1999) have been reviewed, and those related to the ENFE subissue on effects of coupled THC processes on the chemical environment for radionuclide release and to the integrated subissues on (i) quantity and chemistry of water contacting waste packages and waste forms, and (ii) radionuclide release rates and solubility limits have been identified (Pickett and Leslie, 1999; U.S. Nuclear Regulatory Commission, 2000a). Those FEPs are presented in Table 5-6.

**Table 5-6. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to subissue 3 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on quantity and chemistry of water contacting waste packages and waste forms and radionuclide release rates and solubility limits**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Radionuclide Release Rates and Solubility Limits
1.1.02.03.00	Undesirable materials left	Exclude		X
1.1.07.00.00	Repository design	Include (exclude deviations from design)	X	X
1.1.08.00.00	Quality control	Include (exclude defects and deviations)	X	X
1.1.12.01.00	Accidents and unplanned events during operation	Exclude	X	X
1.1.13.00.00	Retrievability	Include		
1.2.06.00.00	Hydrothermal activity	Exclude	X	
2.1.01.02.00	Codisposal/colocation of waste	Include	X	X
2.1.01.03.00	Heterogeneity of waste forms	Include	X	X
2.1.02.01.00	DSNF degradation, alteration, and dissolution	Include	X	X
2.1.02.02.00	CSNF alteration, dissolution, and radionuclide release	Include	X	X
2.1.02.03.00	Glass degradation, alteration, and dissolution	Include	X	X
2.1.02.04.00	Alpha recoil enhances dissolution	Exclude		X
2.1.02.05.00	Glass cracking and surface area	Exclude	X	X
2.1.02.06.00	Glass recrystallization	Exclude		X
2.1.02.07.00	Gap and grain release of cesium, iodine	Include		X

**Table 5-6. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to subissue 3 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on quantity and chemistry of water contacting waste packages and waste forms and radionuclide release rates and solubility limits (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Radionuclide Release Rates and Solubility Limits
2.1.02.08.00	Pyrophoricity	Exclude	X	X
2.1.02.09.00	Void space (in glass container)	Exclude	X	X
2.1.02.13.00	General corrosion of cladding	Exclude	X	
2.1.02.14.00	MIC of cladding	Exclude	X	
2.1.02.15.00	Acid corrosion of cladding from radiolysis	Exclude (?)	X	
2.1.02.16.00	Localized corrosion (pitting) of cladding	Include	X	
2.1.02.17.00	Localized corrosion (crevice corrosion) of cladding	Include	X	
2.1.02.18.00	High dissolved silica content of waters enhances corrosion of cladding	Include	X	
2.1.02.19.00	Creep rupture of cladding	Exclude	X	
2.1.02.20.00	Pressurization from helium production causes cladding failure	Exclude	X	
2.1.02.21.00	Stress corrosion cracking of cladding	Include	X	
2.1.02.22.00	Hydride embrittlement of cladding	Include	X	
2.1.02.23.00	Cladding unzipping	Include	X	
2.1.02.24.00	Mechanical failure of cladding	Include	X	

**Table 5-6. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to subissue 3 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on quantity and chemistry of water contacting waste packages and waste forms and radionuclide release rates and solubility limits (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Radionuclide Release Rates and Solubility Limits
2.1.02.25.00	DSNF cladding degradation	Include	X	
2.1.03.06.00	Internal corrosion of waste container	Include	X	
2.1.03.11.00	Container form	Include	X	
2.1.03.12.00	Container failure (long-term)	Include	X	
2.1.04.01.00	Preferential pathways in the backfill	Include	X	
2.1.04.02.00	Physical and chemical properties of backfill	Include	X	X
2.1.04.03.00	Erosion or dissolution of backfill	Exclude	X	X
2.1.04.05.00	Backfill evolution	Include	X	X
2.1.06.01.00	Degradation of cementitious materials in drift	Include	X	
2.1.06.03.00	Degradation of the liner	Include	X	X
2.1.06.04.00	Flow through the liner	Exclude	X	X
2.1.06.06.00	Effects and degradation of drip shield		X	
2.1.06.07.00	Effects at material interfaces	Include	X	X
2.1.08.07.00	Pathways for unsaturated flow and transport in the waste and EBS	Include?	X	X
2.1.08.08.00	Induced hydrological changes in the waste and EBS	Include	X	X

**Table 5-6. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to subissue 3 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on quantity and chemistry of water contacting waste packages and waste forms and radionuclide release rates and solubility limits (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Radionuclide Release Rates and Solubility Limits
2.1.08.11.00	Resaturation of repository	Include	X	
2.1.09.01.00	Properties of the potential carrier plume in the waste and EBS	Include	X	X
2.1.09.02.00	Interaction with corrosion products	Exclude?		X
2.1.09.04.00	Radionuclide solubility, solubility limits, and speciation in the waste form and EBS	Include	X	X
2.1.09.06.00	Reduction-oxidation potential in waste and EBS	Include	X	X
2.1.09.07.00	Reaction kinetics in waste and EBS	Exclude	X	X
2.1.09.08.00	Chemical gradients/enhanced diffusion in waste and EBS	Include	X	X
2.1.09.10.00	Secondary phase effects on dissolved radionuclide concentrations at the waste form	Include	X	X
2.1.09.11.00	Waste-rock contact	Exclude	X	X
2.1.09.12.00	Rind (altered zone) formation in waste, EBS, and adjacent rock	Include	X	X
2.1.09.13.00	Complexation by organics in waste and EBS	Include		X
2.1.09.14.00	Colloid formation in waste and EBS	Include		X
2.1.09.15.00	Formation of true colloids in waste and EBS	Include		X

**Table 5-6. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to subissue 3 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on quantity and chemistry of water contacting waste packages and waste forms and radionuclide release rates and solubility limits (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Radionuclide Release Rates and Solubility Limits
2.1.09.16.00	Formation of pseudo-colloids (natural) in waste and EBS	Include		X
2.1.09.17.00	Formation of pseudo-colloids (corrosion products) in waste and EBS	Include		X
2.1.09.18.00	Microbial colloid transport in the waste and EBS	Include		X
2.1.10.01.00	Biological activity in waste and EBS	Include ?	X	X
2.1.11.01.00	Heat output/temperature in waste and EBS	Include	X	X
2.1.11.02.00	Nonuniform heat distribution/edge effects in repository	Include	X	X
2.1.11.03.00	Exothermic reactions in waste and EBS	Exclude	X	X
2.1.11.04.00	Temperature effects/coupled processes in waste and EBS	Include	X	X
2.1.11.08.00	Thermal effects: chemical and microbiological changes in the waste and EBS	Exclude	X	X
2.1.11.09.00	Thermal effects on liquid or two-phase fluid flow in the waste and EBS	Include	X	X
2.1.11.10.00	Thermal effects on diffusion (Soret effect) in waste and EBS	Exclude	X	X
2.1.12.01.00	Gas generation	Exclude	X	X
2.1.12.02.00	Gas generation (He) from fuel decay	Exclude	X	X

**Table 5-6. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to subissue 3 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on quantity and chemistry of water contacting waste packages and waste forms and radionuclide release rates and solubility limits (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms	Radionuclide Release Rates and Solubility Limits
2.1.12.03.00	Gas generation (H <sub>2</sub> ) from metal corrosion	Exclude	X	X
2.1.12.04.00	Gas generation (CO <sub>2</sub> , CH <sub>4</sub> , H <sub>2</sub> S) from microbial degradation	Exclude	X	X
2.1.12.06.00	Gas transport in waste and EBS	Exclude		X
2.1.12.07.00	Radioactive gases in waste and EBS	Exclude		X
2.1.12.08.00	Gas explosions	Exclude		X
2.1.13.01.00	Radiolysis	Include	X	X
2.1.13.02.00	Radiation damage in waste and EBS	Include, Exclude (backfill, seals, rock)		X
2.2.07.06.00	Episodic/pulse release from repository	not given		X
2.2.08.01.00	Groundwater chemistry/composition in UZ and SZ	Include	X	
3.1.01.01.00	Radioactive decay and ingrowth	Include		X
SZ = saturated zone                      UZ = unsaturated zone                      MIC = microbially induced corrosion EBS = engineered barrier system      CSNF = commercial spent nuclear fuel                      DSNF = Department of Energy spent nuclear fuel				

#### 5.4.3.1.2 Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms

For the TSPA-VA, DOE developed a set of five models to represent the near-field geochemical environment (U.S. Department of Energy, 1998b): (i) a description of the gas, water, and colloid composition coming into the drift, (ii) the composition of gas phase relative to the major gas sinks in the drift, (iii) the evolution of water composition reacting with major materials within the drift and the drift gas phase, (iv) a description of the stability and quantity of clay and iron-oxide colloids in the drift, and (v) the in-drift microbial communities. With the exception of the microbial communities and colloid model, these models were used to predict the chemistry of water contacting the waste packages and waste forms. Reaction of in-drift water and gas was calculated for different points along the flow path of water. As a result, calculations of water and gas reacting with the concrete drift liner (not present in the Enhanced Design Alternative-II design), iron corrosion products, and spent nuclear fuel were completed. Calculation of the water composition was completed as a function of time, using six discrete periods: three during the boiling regime (0 to 2,000 years) and three periods that extend beyond to 100,000 years. Based on the different degradation rates of the different engineered materials in the Viability Assessment design (concrete, waste package, and spent nuclear fuel), different sequential reactions (e.g., incoming water, concrete, waste package, and then waste form) were used in the different time periods (U.S. Department of Energy, 1998b). The two-dimensional thermal-hydrologic model at the mountain-scale provided both the air-mass fraction and gas fluxes through the drift as a function of time. These results included the effects of boiling and gas flow on the mix of air and steam in the gas phase, but none of the chemical interactions with the host rock. While 15 geochemical parameters describing the chemistry of water were calculated at various locations and at several different time periods, only the pH, ionic strength, and total carbonate concentrations were used in performance assessment calculations. The chemistry of water contacting the waste package calculated in these models was not used in the waste package degradation model (U.S. Department of Energy, 1998b). In addition, the chemistry of water expected inside the waste package was not used in the cladding degradation models.

For the TSPA-SR, DOE's methods and assumptions report (CRWMS M&O, 1999a) indicates that an in-drift geochemical environment model will be developed for the total system performance assessment that will provide a quantitative description of the changing compositional conditions under which waste forms degrade and precipitate as secondary phases and radionuclides mobilize from the waste form. The objectives of the in-drift geochemical model are to determine the changes in water chemistry resulting from the interaction of engineered barrier materials with water seeping into the drift and to provide this information to total system performance assessment analyses. The model takes into account the variation in seepage and drainage fluxes, the effects of temperature changes on chemical equilibria, and physical processes such as evaporation and condensation.

The TSPA-SR in-drift geochemical environment model separates the major potential contributors to water compositional changes into nine subsystem models. The separate models will be used to assess the relative capacities of the different processes to alter compositions of various chemical phases at various times. The nine subsystem models are: (i) seepage/backfill interactions, (ii) precipitates/salts analysis, (iii) corrosion products, (iv) seepage/invert interactions, (v) seepage/cement interactions, (vi) in-package chemistry, (vii) in-drift gas flux and composition, (viii) microbial communities, and (ix) in-drift colloids and concentrations. The

relationships between the submodels, their coupling, and spatial locations of potential importance are described in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a).

Of particular interest for this review is the submodel on in-package chemistry, which is integral to the commercial spent nuclear fuel and DOE spent nuclear fuel dissolution rate calculations. To estimate the in-package environment for the TSPA-SR, a number of assumptions will be made in the in-package chemistry model. These assumptions are that the aqueous solution fills all the voids in the waste package and that solutions that drip into the package for ~50,000 years will have the composition of J-13 Well water (CRWMS M&O, 2000i). The drip rate is assumed to range from 0.015 to 0.15 cubic meters per year and dripping water is assumed to enter and exit the waste package at the same rate and not interact to any significant degree with the waste package walls as it enters the waste package. However, the water is assumed to react with several waste package components.

Two representative waste packages were modeled, a commercial spent nuclear fuel package and a DOE spent nuclear fuel/high-level waste glass codisposal package. Commercial spent nuclear fuel waste packages are assumed to be made up of several reactive components: aluminum alloy, 304 L low-carbon stainless steel, A516 carbon steel, borated and nonborated 316 stainless steel containing  $GdPO_4$ , and zirconium-clad fuel rods. Commercial spent nuclear fuel is primarily  $UO_2$ . No interaction between the Zircaloy cladding and the internal environment is assumed, although sensitivity analyses includes the percentage of fuel area exposed by breached cladding. Codisposal wastes comprise a DOE spent nuclear fuel canister surrounded by five containers of high-level waste glass. The codisposal waste package was assumed to have the properties of a fast flux test facility waste package and have six reactive components: A516 carbon steel, 316 stainless steel (with and without  $GdPO_4$ ), 304L low-carbon steel, high-level waste glass, mixed oxide fuel (made up of Pu, U, and Np oxide), and  $UO_2$  fuel.

In the preliminary draft in-package chemistry model, water is assumed to fill the void volume, and the waste package internal components are lumped into equivalent masses per unit volume for calculating the reaction products. EQ3/6 is used to calculate the time evolution of solution composition as a result of these interactions (CRWMS M&O, 2000i). The specific partial pressures of  $CO_2$  and  $O_2$  of the repository atmosphere are set to  $10^{-3.0}$  and  $10^{-0.7}$  atmosphere, respectively. A range of degradation rates was used for each component of the waste package, and current densities were calculated from the assumed molar dissolution rates using Faraday's law.

The results of the EQ3/6 calculations indicate that reaction of waste package components with incoming fluids results in dramatic changes in solution chemistry. The pH decreases inside the waste package due to dissolution of stainless steel components, specifically due to the chromium oxidation to  $Cr^{6+}$  species. The pH increases due to the dissolution of the uranium oxide fuels, aluminum alloy, and high-level waste glass. Solution pH represents a dynamic balance between proton-producing and proton-consuming reactions. Relatively high rates of the former leads to transiently low pH, whereas relatively high rates of the latter cause solution pH to be transiently high. Solution ionic strength for codisposal waste package effluents varied between 0.003 and ~5.8 molar, but ionic strengths of commercial spent nuclear fuel waste package effluents never exceeded 1.7 molar.

Direct use of a complex code such as EQ6 within the TSPA-SR analysis calculations was not practical. Thus, DOE used abstraction of in-package processes based on a series of multiple linear regression analyses of the output from the EQ3/6 simulations. Processes at times more than 1,000 years were abstracted separately from those at greater than 10,000 years. Time-dependent parameters included in the abstraction are pH, Eh, ionic strength, total aqueous carbonate concentration, chloride concentration, and fluoride concentration. The pH constitutes the most important in-package parameter. Thus, the time discretization used by DOE for all of the abstracted parameters were based on changes in pH. The process models used constant values of O<sub>2</sub> and CO<sub>2</sub> fugacities for all times, thus the abstracted gas fugacities were also set to constant values for the modeled duration.

DOE recognizes some of the limitations of the in-package chemistry calculations in the preliminary draft Waste Form Degradation Process Model Report and the complexities in considering the geometrical effects (CRWMS M&O, 2000j). The assumption that the waste package void volume is completely filled with water and the reactions occur between an equivalent lumped mass of waste package components and this water without any feedback between corrosion and chemistry is cited as a conservative approach. The possibility of formation of NO<sub>2</sub> and nitric acid by radiolysis was discounted because of the buffering capacity of the corrosion products from the waste package components and the possible outgassing of NO<sub>2</sub> from the breached waste package. The neglect of evaporative effects is believed to be conservative because the higher amount of water will lead to greater leaching of radionuclides, but it is recognized that neglecting the formation of saline solutions by evaporation may be nonconservative in terms of dissolution rate of the waste form.

#### **5.4.3.1.3 Radionuclide Release Rates and Solubility Limits**

DOE's approach to abstraction of radionuclide release in the TSPA-VA was based on the assumption that the waste packages were breached. After the waste packages are breached, radionuclides are not available for release and transport until (i) failure of the fuel cladding or high-level waste canister, (ii) degradation of the solid waste form and (iii) mobilization of radionuclides into aqueous solution or an aqueous colloidal suspension. Mobile radionuclides are transported out of the degraded waste package and through the engineered barrier system to the geosphere via one of two mechanisms: (i) movement of dissolved or colloidal material via diffusion or (ii) movement of dissolved or colloidal material via advection (U.S. Department of Energy, 1998b). The components of the waste form degradation model used by the DOE include the initial inventory, degradation of the cladding on commercial spent nuclear fuel, dissolution rates of the waste forms, solubility constraints on radionuclide mobilization, and formation of colloids and secondary mineral phases.

The important input into the waste form degradation model in the TSPA-VA included the inventory of radionuclides. In addition, the temperature at the waste package surface, relative humidity at the waste package surface, and liquid saturation in the invert beneath the waste package, all derived from thermal-hydrologic modeling results, were inputs to the models. Results from waste package degradation, cladding degradation, water ingress into waste packages, the amount of exposed fuel surface caused by cladding degradation, and the near-field geochemical conditions were also used as input to the waste form degradation model.

Based on information provided in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), the general approach to be used by DOE for the TSPA-SR will remain the same as for the TSPA-VA approach. However, the analyses will take into account the potential effects of the new repository design, and a more elaborate evaluation of the in-drift geochemical environment will be completed. Most of the Process Model Reports and Analysis and Model Reports that provide the details on DOE models, abstractions, and analyses were not available for review prior to May 15, 2000. Therefore, the summary of DOE's approach given in the following sections is mostly based on the TSPA-VA (U.S. Department of Energy, 1998b), its supporting technical basis document (CRWMS M&O, 1998d), and on preliminary draft versions of Process Model Reports and Analysis and Model Reports that were available.

#### **5.4.3.1.3.1 Commercial Spent Nuclear Fuel Clad Degradation**

A review of DOE models and abstractions regarding the behavior of commercial spent nuclear fuel is presented in the Container Life and Source Term IRSR Revision 2 (U.S. Nuclear Regulatory Commission, 1999d). An important component of these models is the degradation of the Zircaloy cladding used in commercial water-cooled nuclear reactors. The cladding can act as a barrier to the release of radionuclides. For the TSPA-VA, DOE included the consideration of cladding as an additional metallic barrier (U.S. Department of Energy, 1998b). Cladding will also be included as a barrier in the process models of TSPA-SR (CRWMS M&O, 1999a). Several potential degradation mechanisms of Zircaloy cladding, such as uniform and localized corrosion, creep, delayed hydride cracking, hydrogen embrittlement, mechanical failure owing to rockfall, stress corrosion cracking, and fuel and cladding oxidation, can impair that beneficial action (U.S. Nuclear Regulatory Commission, 1999d; Cragolino, et al., 1999). DOE evaluated these possible failure modes, but neglected most of them as a cause of cladding failure under disposal conditions on the basis of auxiliary analyses (CRWMS M&O, 1998d). The exceptions are uniform corrosion and mechanical failure as a result of rockfall once the waste package loses structural integrity. In addition, it is assumed that a small fraction, equal to 0.1 percent of the commercial spent nuclear fuel rods, has failed during reactor operations. This type of failure is classified as "juvenile" cladding failure (U.S. Department of Energy, 1998b). Also, the small percentage (about 1.5 percent) of the commercial spent nuclear fuel fabricated with stainless steel cladding is considered to fail as soon as the waste package fails because it is assumed that the rate of corrosion of stainless steel is comparatively rapid with respect to that of Zircaloy cladding.

For the TSPA-SR, DOE developed a preliminary draft model to evaluate cladding degradation as part of the waste form degradation model (CRWMS M&O, 2000j) to determine the rate at which the commercial spent nuclear fuel is exposed to in-package environment. This model represents a significant improvement with respect to that presented in the TSPA-VA. The degradation of the commercial spent nuclear fuel cladding is assumed to occur in two stages. The first stage corresponds to rod failure as a result of perforation of the cladding and the second one due to the progressive exposure of the spent fuel matrix as a result of splitting (unzipping) of the cladding due to oxidation of the irradiated  $\text{UO}_2$  pellets either by air and moisture or by an aqueous environment.

Cladding perforation may occur prior or after waste package emplacement. The initial condition of the cladding and the percentage of rods perforated is evaluated taking into account data obtained from reactor operation, pool storage, dry storage, and transportation, including fuel

handling. A distribution of initially perforated Zircaloy fuel rods expressed as a cumulative complementary distribution function was developed from the available data. All the commercial spent nuclear fuel clad with stainless steel instead of Zircaloy (assumed to be ~1.1 percent of the total) is assumed to be initially perforated.

Cladding perforation after waste package emplacement is assumed to be caused by creep, stress corrosion cracking, mechanical failure due to seismic events, and localized corrosion. To evaluate the possibility of creep and stress corrosion cracking under disposal conditions, the temperature history of the cladding during storage and transportation and the evolution of temperature after waste package emplacement was estimated, as well as the distribution of internal pressure and the corresponding hoop stresses (CRWMS M&O, 2000j). An abstraction was developed to provide the fraction of rods that failed by creep as a function of the peak waste package surface temperature as a controlling variable. The causative species for stress corrosion cracking is considered to be iodine, a fission product found in the pellet-cladding gap. Iodine concentration was conservatively assumed to be above a value needed to cause stress corrosion cracking. Also, a critical stress level of 180 megapascal is selected as a threshold stress for stress corrosion cracking to occur, which is a relatively high value to be attained by more than a few rods.

Local corrosion of the commercial spent nuclear fuel cladding is assumed to be promoted by fluoride anion on a relatively small area of cladding (about 10 millimeters of rod length). The fraction of fuel cladding surface on different fuel rods inside the same waste package is assumed proportional to the volume of water entering the waste package in a flow-through scenario. This is considered to be a bounding analysis because it implicitly assumed a 100-percent efficiency in the chemical reaction of fluoride with Zircaloy.

Another process leading to perforation of the cladding is mechanical failure due to seismic events when their frequency is of the order of  $1 \times 10^{-6}$  per year. This type of event perforates the cladding and initiates unzipping. It is assumed rockfall will not lead to mechanical failure of the cladding based on the argument that collapse of the waste package takes hundreds of thousands of years to occur.

After cladding perforation, the inventory of radionuclides in the gap and in the grain boundaries of the irradiated fuel pellets is considered to experience fast release. The gap inventory of iodine and cesium is predicted to be released in proportion to the fission gas release fractions, while that in the grain boundaries is estimated from release experiments using intact and defective (i.e., with slits and holes) fuel rod samples.

The time to unzip a fuel rod under dry and wet conditions is estimated as a function of waste package temperature. For wet unzipping, time is also a function of the in-package chemistry, defined for this purpose in terms of pH, partial pressure of  $O_2$ , and  $CO_3^{2-}$  concentration. Although the approach is considered conservative and includes consideration of uncertainties, DOE argues it is not as conservative as in previous total system performance assessments.

#### **5.4.3.1.3.2 Commercial Spent Nuclear Fuel Waste Form Degradation**

A review of DOE models and abstractions of waste form degradation can be found in the Container Life and Source Term IRSR Revision 2 (U.S. Nuclear Regulatory Commission,

1999d). In the TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b), a key hypothesis is that the commercial spent nuclear fuel intrinsic dissolution rate will determine the aqueous release of soluble radionuclides contained within the commercial spent nuclear fuel matrix. The same hypothesis will be used for the TSPA-SR (CRWMS M&O, 1999a). The product of the intrinsic dissolution rate and the wetted surface area provides the source term for radionuclide transport through the unsaturated zone. High-solubility radionuclides, such as Tc-99 or I-129, are assumed to be released congruently with the dissolution of the matrix, whereas uranium and low-solubility radionuclides, such as Pu-239 or Am-241, are assumed to be reprecipitated on the surface of the irradiated UO<sub>2</sub> as secondary minerals. Consequently, the release of low-solubility radionuclides from the engineered barrier system is constrained by the solubility limit of the solid phase containing the radionuclide.

The commercial spent nuclear fuel dissolution rates have been measured using a wide range of techniques, including flow-through experiments using both spent fuel and UO<sub>2</sub> pellets, static tests in autoclaves, and unsaturated drip tests using spent fuel pellets contained in zirconium holders. However, only the data from the flow-through tests are used to derive the preliminary draft dissolution rate model for performance assessment (CRWMS M&O, 2000j, k). Two regression equations are used in the abstraction of commercial spent nuclear fuel dissolution rate described in the preliminary draft waste form Process Model Report (CRWMS M&O, 2000j). For pH greater than 7, the dissolution rate is given by

$$\text{Log(Rate)} = 4.69 - \frac{1085}{T} + 0.12\log_{10}[\text{CO}_3]_{\text{Total}} + 0.32\log_{10}[\text{P}_{\text{O}_2}] \quad (5-1)$$

and for pH less than or equal to 7, the equation is

$$\text{Log(Rate)} = 7.13 - \frac{1085}{T} + 0.32\log_{10}[\text{P}_{\text{O}_2}] - 0.41\text{pH} \quad (5-2)$$

The dissolution rate is expressed in milligrams per square meter per day, temperature (T) in Kelvin, carbonate concentration in moles per liter, and oxygen partial pressure in atmosphere. The abstracted equations are derived such that the rates from the two equations are equal at pH 7. Equation (5-1) is an empirical regression model that is only loosely based on irreversible thermodynamic reasoning (Stout and Leider, 1998). The regression coefficient, adjusted R<sup>2</sup>, for Equation (5-1) is 0.5014 (CRWMS M&O, 2000k), indicating the model does not represent a significant portion of the experimental data. A more elaborate model, with cross terms and a term involving burn-up, was proposed by Stout and Leider (1998) and exhibited a much better statistical fit to the data (adjusted R<sup>2</sup> = 0.8174).

Equation (5-2) is derived by assuming that the dependence of dissolution rate on oxygen partial pressure and temperature is the same at pH values below 7 as it is above 7. The term involving carbonate is neglected based on the reasoning that surface adsorption of carbonate ions is negligible below this pH. Additionally, one experimental data point at pH 3 and the calculated rate at pH 7 from Equation (5-1) are used to derive the slope of the pH dependence

for pH values between 3 and 7. The model is then compared to other rate measurements and found to predict higher rates than the experiments, thus, justifying its use as a “bounding” model.

Unsaturated drip tests have been performed during the past eight years (Wronkiewicz, et al., 1992). The tests involved spent fuel contained in Zircaloy holders exposed to dripping water or moist environment. The drip rates used, 0.0078 liters per year to 0.078 liters per year, are much lower than those assumed in the in-package calculations (1.5 to 150 liters per year). The drip rates used should be scaled to surface area of reacting media exposed. Based on 1 square centimeter of fuel surface, the low end of the drip rate would correspond to about 8 centimeters per year of dripping. However, this scaling relationship is at present poorly understood. It may depend on the manner in which dripping water contacts the fuel (Wronkiewicz, et al., 1992). The release rates of various radionuclides are monitored. The release rate of Tc-99 is used to derive the intrinsic dissolution rate of the spent nuclear fuel. The dissolution rates measured in the high drip rate tests are close to that predicted by Equation (5-1), if a surface roughness factor of three is assumed in the drip tests (CRWMS M&O, 2000k). This is surprising because, unlike the flow-through tests used to derive Equation (5-1), the drip tests encourage corrosion product formation and would be expected to yield lower dissolution rates. The low drip rate tests exhibited lower dissolution rates. The drip tests show that Np-237 and Pu-239 are retained in the corrosion product after an initial period of high release (CRWMS M&O, 2000j).

#### **5.4.3.1.3.3 U.S. Department of Energy-Owned Spent Nuclear Fuel, Navy Fuel, and Plutonium Disposition Wastes Degradation**

DOE spent nuclear fuel has more than 250 potential spent nuclear fuel form types for disposal. These were grouped into 16 categories based on similar performance characteristics (CRWMS M&O, 1998d). Performance analyses of the first 13 categories conducted in 1997 indicated only Categories 1, 4, 5, 6, 8, and 11 contributed significantly to dose, and the dose from a composite of 13 categories was about two orders of magnitude below that of the commercial spent nuclear fuel in the repository (CRWMS M&O, 1998d). Based on the 1997 analyses, the TSPA-VA analyses used a surrogate DOE spent nuclear fuel with a radionuclide inventory based on a weighted average of the inventories of Categories 1, 4, 5, 6, 8, and 11. A conservative assumption was built into the surrogate model by using a dissolution rate based on the uranium metal fuel dissolution model. The metallic spent fuel dissolution rate is faster than the rate for the nonmetallic fuels.

For the TSPA-VA, the model for the degradation of metallic fuel was an Arrhenius kinetic equation using parameters derived from assessments of spent nuclear fuel and high-level waste (Rechard, 1995). When the temperature in the repository is below 100 °C, water was assumed to be present on the waste form (wet oxidizing conditions), and humid air conditions are assumed at all other times.

DOE notes in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) that there were little applicable experimental data to substantiate the parameter values selected for the TSPA-VA, and that the TSPA-SR will use a degradation rate and a corresponding effective surface area that bounds the experimental data on N-reactor fuel collected by DOE over a range of conditions in the last few years. In the preliminary draft waste form Process Model

Report, three types of dissolution models are proposed for each fuel type: (i) upper-limit models that assume that complete dissolution of the fuel occurs at any time step after breach of the container, and all radionuclides are available for instantaneous release; (ii) conservative models that use the higher end of the dissolution rate data or, where no data are available, considers a multiplier of data for a similar fuel; and (iii) best estimate models constructed from a reasonable database of the specific fuel type.

For the TSPA-SR, the upper limit model for each fuel type is used, except for the Navy fuel (CRWMS M&O, 2000j). Alternatively, the N-reactor fuel is used as the surrogate for all DOE spent nuclear fuel, and the conservative or best basis models for this fuel is used to represent all DOE spent nuclear fuel, including the Navy fuel. The best estimate dissolution rate for the N-reactor fuel is given by

$$\text{Rate} \left( \frac{\text{mg}}{\text{m}^2 \text{day}} \right) = 1.75 \times 10^5 \left( \frac{D_{\text{matrix}}}{D_{\text{U-metal}}} \right) \quad (5-3)$$

where  $D_{\text{matrix}}$  is the density of the spent nuclear fuel matrix, and  $D_{\text{U-metal}}$  is the density of uranium metal. The rate for the conservative model is a factor of ten higher than calculated from Equation (5-3).

The Analysis and Model Report on DOE spent nuclear fuel degradation was not available for review at the time this revision of the ENFE IRSR was prepared. A review of this Analysis and Model Report is necessary to fully understand the technical bases for the dissolution model used. While the Navy fuel is considered to be bound by either the commercial spent nuclear fuel or the N-reactor fuel, the dissolution rate data on this fuel is not publicly available. Therefore, the inclusion of this fuel in the DOE spent nuclear fuel bounding case using N-reactor fuel is tentative, pending further information. However, use of the upper limit model does not require further experimental data.

#### 5.4.3.1.3.4 High-Level Waste Glass Degradation

DOE recognizes that glass waste forms are thermodynamically unstable during long time periods and will alter on contact with water. Radionuclides can be mobilized from the glass waste by a variety of processes, including degradation and alteration of the glass, phase separation, congruent dissolution, precipitation of silicates, coprecipitation of other minerals including iron corrosion products, and selective leaching. Not all these processes are incorporated into DOE's abstraction of glass degradation. DOE states in the TSPA-VA technical basis document (CRWMS M&O, 1998d) that the entire phenomenology of the glass alteration process cannot be incorporated directly into a performance assessment model because it would necessitate tracking too many system variables and because several of the relationships between glass reaction rate and solution chemistry cannot be quantified with available experimental data.

Instead, DOE generated a simplified glass model amenable for incorporation into performance assessment calculations. This model incorporates the parameters DOE believed most

important for controlling the rate of glass reaction: (i) temperature, (ii) fluid pH, (iii) fluid silica concentration, and (iv) reactive glass surface. In the TSPA-VA, DOE used a simplified rate equation for waste glass dissolution, expressed (in mass units of grams) as

$$\text{Rate} = S \left[ k \left( 1 - \frac{Q}{K} \right) + k_{\text{long}} \right] \quad (5-4)$$

where

Rate =	alteration rate of glass (g/yr)
S	= surface area (m <sup>2</sup> )
k	= glass surface alteration rate constant (g/m <sup>2</sup> /y), a function of temperature and pH of the solution
Q	= concentration of dissolved silica (g/m <sup>3</sup> water)
K	= a quasi-thermodynamic parameter for glass equal to the silica saturation value for the glass (g/m <sup>3</sup> water)
k <sub>long</sub>	= long-term dissolution rate determined from test data

The values of k and k<sub>long</sub> are determined from experimental data. For the TSPA-VA, an expression for k as a function of pH and temperature was derived by fitting experimental data for a simplified glass composition (SRL-165), which had the most complete set of data at the time of the TSPA-VA. The value of k<sub>long</sub> is based on glass corrosion tests at 90 °C and is given the same temperature dependency as k (CRWMS M&O, 1998d). The value of K, the silica saturation for waste glass, is assumed equal to the equilibrium constant for amorphous silica. This assumption was made by DOE in the absence of a mechanistic quantitative understanding of how K varies with glass composition and experimental test conditions.

For the TSPA-SR, the basic form of rate expression adopted by DOE (CRWMS, 2000j) to describe the dissolution of waste glass immersed in water is

$$\text{Rate} = S \left\{ k_0 \cdot 10^{\eta \cdot \text{pH}} \cdot \exp\left(\frac{-E_a}{RT}\right) \cdot \left[ 1 - \frac{Q}{K} \right] \right\} \quad (5-5)$$

where

k <sub>0</sub>	=	intrinsic dissolution rate, which depends only on glass composition, (g/m <sup>2</sup> /yr)
η	=	pH-dependence coefficient
E <sub>a</sub>	=	effective activation energy (kJ/mol)
R	=	gas constant [8.314 J/(mol•K)]
T	=	temperature (Kelvin)

Equation (5-5) contains two main parts: the forward rate, k<sub>0</sub>•10<sup>η•pH</sup>•exp(-E<sub>a</sub>/RT), which represents the dissolution rate in the absence of feedback effects of dissolved silica (and other aqueous species), and the reaction affinity term, 1-(Q/K), which quantifies the feedback effects. Because of the complexity in defining parameters and associated uncertainties, a simpler

bounding approach was adopted which combined  $1 - (Q/K)$  with  $k_0$ , and the following abstraction was developed for aqueous degradation of high-level waste:

$$\text{Rate} = S \left\{ k_{\text{eff}} \cdot 10^{\eta} \cdot \text{pH} \cdot \exp\left(\frac{-E_a}{RT}\right) \right\} \quad (5-6)$$

where

$$k_{\text{eff}} = k_0 \cdot (1 - Q/K)$$

Equation (5-6) reduces the abstracted high-level waste glass degradation model to an equation involving four parameters ( $\eta$ ,  $E_a$ ,  $S$ , and  $k_{\text{eff}}$ ) and two variables ( $\text{pH}$  and  $T$ ). The forward rate was measured under flow-through conditions in which the affinity term was maintained near one. The test results indicated that the  $\text{pH}$  and temperature effects were independent of glass composition within the range of the glass compositions tested and, therefore, the same parameter values were used for all waste glasses. The  $\text{pH}$  dependence indicated a “V”-shaped curve. The  $k_{\text{eff}}$  was determined based on experimental observations of the behavior of the affinity term. Several options were evaluated to conservatively bound three stages of glass corrosion. Based on this evaluation, data from the PCT-A test were used to obtain bounding values for  $k_{\text{eff}}$ . The exposed surface area was calculated by assuming that all surfaces corrode at the same rate when exposed to water, and that surface area remains constant during the corrosion process.

Because of the discontinuity in the  $\text{pH}$  behavior, separate rate expressions were obtained for the acid range and the alkaline range, as shown by Equations (5-7) and (5-8).

For the low  $\text{pH}$  range ( $\text{pH} < \text{pH}_m$ ),

$$\frac{\text{Rate}}{S} = (\text{gm} \cdot \text{m}^2 / \text{day}) = 10^{(9 \pm 1)} \cdot 10^{(-0.6 \pm 0.1)\text{pH}} \cdot \exp\left(\frac{-58 \pm 10}{RT}\right) \quad (5-7)$$

For the high  $\text{pH}$  range ( $\text{pH} \geq \text{pH}_m$ ),

$$\frac{\text{Rate}}{S} = (\text{gm} \cdot \text{m}^2 / \text{day}) = 10^{(6.9 \pm 0.5)} \cdot 10^{(0.4 \pm 0.1)\text{pH}} \cdot \exp\left(\frac{-80 \pm 10}{RT}\right) \quad (5-8)$$

where

$$\begin{aligned} \text{pH}_m &= \text{pH at which the minimum dissolution rate occurs} \\ &= 2.1 + 1,149/T \end{aligned}$$

#### 5.4.3.1.3.5 Dissolved Radionuclide Concentration Limits

For the TSPA-VA, and also for the TSPA-SR, DOE's approach to calculate bounds on the aqueous concentration of radionuclides in groundwater that reacted with the waste form is to initially derive the concentrations from the waste form dissolution model. Subsequently, a comparison is made between the waste form dissolution-based aqueous radionuclide concentration and a value determined by the solubility limit for each radionuclide considered. If the solubility-limited value is lower for a given radionuclide than its concentration derived from the waste form dissolution, the aqueous concentration is set to the solubility-limited value and the difference in mass is assumed to precipitate out of solution. The solubility-limited values place constraints on the aqueous concentration of the particular radionuclide element considered, with each isotope of that element present in proportion to its isotopic abundance (CRWMS M&O, 1998d).

The concentration limit usually is the solubility limit of the solid phases that contain the radioisotopes (either solid phases with the radioisotope as the dominant element or solid phases with trace amounts of the radionuclide, as in coprecipitated species). For one radioelement (neptunium), DOE claims that experiments have shown much lower concentrations in solution compared to the calculated value. In this case, DOE may use a lower solubility limit to bound the aqueous concentration of neptunium. The solid phases that form depend on the temperature, redox conditions, and chemical composition of the groundwater. Because of uncertainty in the values for these variables in the waste package and near-field environment, a wide range of possible radionuclide concentration limits exists.

For TSPA-VA, the distribution of each radioisotope transported was primarily based on an elicitation of experts both inside and outside the Yucca Mountain Project conducted in 1993 (Wilson, et al., 1994; CRWMS M&O, 1998d). The assumptions behind the expert panel's development of the distributions are (i) the unsaturated zone water composition is bounded by the composition of J-13 Well water and that of Well UE-25p#1, (ii) the solubility limits will be determined by the far-field groundwater environment, (iii) the environment is oxidizing, and (iv) future climate changes will cause groundwater compositional changes. Several additional sources were used to further constrain distributions of solubility limits for the radionuclides used in the TSPA-VA analysis. Also, the solubility-limited concentration distribution for neptunium used in previous performance assessment calculations was revised based on an analysis by Sassani and Siegmann (1998).

For the TSPA-SR, the calculation of dissolved concentration limits builds upon three primary feeds: (i) estimates of in-package fluid chemistry (pH, Eh, ionic strength, carbonate concentration), (ii) measured (and estimated) thermodynamic parameters describing the stabilities of aqueous species and solid radioisotope phases, and (iii) a determination of the likely solubility controlling phases for the radionuclides of concern (CRWMS M&O, 2000j). DOE's approach to estimate in-package fluid chemistry is discussed in Section 5.3.1.2. For the TSPA-SR analysis, the pure phases were chosen because, in general, they yield higher dissolved concentrations compared to coprecipitated phases. The specific phase selected for a particular radionuclide is based on information from geologic and/or experimental observations or crystallochemical arguments. Where no information can be gleaned from field or experimental observations, the most amorphous and hydrated form of the radionuclide, believed to be the most soluble, was selected. For uranium, schoepite was assumed to be the

solubility controlling phase. For neptunium, plutonium, americium, and nickel, the solubility controlling solids chosen were  $\text{Np}_2\text{O}_5$  [or  $\text{Np}(\text{OH})_4(\text{am})$  under reducing conditions],  $\text{Pu}(\text{OH})_4(\text{am})$ ,  $\text{AmOHCO}_3$ , and  $\text{NiO}$ , respectively.

The amount of thermodynamic data available for the different radionuclides, the sensitivity of the solubilities to fluid chemistry, and the importance of the different radionuclides to total system performance are variable. Thus, DOE used three approaches to implement solubility limits within the TSPA-SR analysis: (i) multi-termed functions of chemistry for uranium, neptunium, and americium; (ii) distributions for plutonium, palladium, lead, and nickel; and (iii) constant bounding values for technetium, iodine, thorium, cesium, strontium, chloride, carbon, niobium, zirconium, radium, and tin. The concentration of uranium for TSPA-SR analysis is calculated using an equation fit to EQ3NR-derived schoepite solubility as a function of pH,  $\text{CO}_2$  fugacity, and temperature. The solubility of neptunium for TSPA-SR analysis is calculated from a pH-dependent equation. A log-uniform distribution is assigned for plutonium solubility, with a minimum of  $1.0 \times 10^{-9}$  and a maximum of  $1.9 \times 10^{-4}$  molar, based on EQ3NR calculations of  $\text{Pu}(\text{OH})_4$  solubility at pH 8 and a range of Eh and  $\text{CO}_2$  fugacity. An equation with pH and  $\text{CO}_2$  fugacity terms is used to calculate americium concentrations for TSPA-SR analysis, and the solubilities of actinium, curium, and samarium are assumed identical to that of americium. The solubilities of technetium, carbon, chloride, iodine, and cesium are set to 1.0 molar, which lets the waste inventory control their release, because no solubility limiting solids are predicted to form for these radioelements. The solubility of strontium is also set to 1.0 molar to simplify the analysis. A log-uniform distribution is proposed for nickel solubility, with a minimum of  $1.4 \times 10^{-6}$  and a maximum of 3.1 molar. For lead solubility, a log-uniform distribution is recommended for TSPA-SR, with a minimum of  $1.0 \times 10^{-10}$  and a maximum of  $1.0 \times 10^{-5}$  molar. In the case of palladium solubility, a log-uniform distribution is recommended, with a minimum of  $1.0 \times 10^{-10}$ , a maximum of  $1.0 \times 10^{-5}$ , and a mean of  $3.2 \times 10^{-8}$  molar. Constant values of  $1.0 \times 10^{-7}$ ,  $2.3 \times 10^{-6}$ ,  $5.0 \times 10^{-8}$ ,  $1.0 \times 10^{-5}$ , and  $6.8 \times 10^{-10}$  molar, respectively, for the solubilities of nickel, radium, tin, thorium, and zirconium are recommended in the preliminary draft Waste Form Process Model Report (CRWMS M&O, 2000j).

DOE recognizes that inherent limitations in the database lead to uncertainties in calculated solubilities. However, these limitations are claimed to lead to only a factor of two uncertainty when the ionic strength exceeds about 0.7 molar. DOE claims this uncertainty is small relative to other uncertainties within the system. Also, DOE made improvements in its calculation of dissolved concentration limits compared to the approach taken for the TSPA-VA in response to concerns expressed by the TSPA-VA peer review panel (Whipple, et al., 1999). For example, the thermodynamic data for neptunium was thoroughly reviewed and a regression model was developed such that the solubility limit could be coupled to the estimated in-package chemical environment. Also, formation of uranium secondary phases that could potentially incorporate neptunium and reduce neptunium solubility was not incorporated into DOE's abstraction of dissolved concentration limits because experiments studying this mechanism were not conclusive. The preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j) states that distributions of plutonium, thorium, and nickel were carefully reevaluated taking into account expected in-package chemical conditions, but the preliminary draft Analysis and Model Report on Summary of Dissolved Concentration Limits (F0095; CRWMS M&O, 2000l) indicates DOE's in-package chemistry analyses were not completed in time to be fully utilized in the evaluation of solubility limits. Also, solubility limits for other key radionuclides (Tc, I, C, Nb, Zr, Pa, Pb, Ra, Sn, Cs, and Cl) were reevaluated and fixed at bounding values. According to DOE, the solubility values determined by informal expert

elicitation conducted in 1993 (Wilson, et al., 1994) were used only to corroborate the newly evaluated distributions and fixed values (CRWMS M&O, 2000j).

#### 5.4.3.1.3.6 Colloid-Assisted Radionuclide Concentration Limits

DOE's abstraction of colloidal radionuclide release is reported in the preliminary draft version of Analysis and Model Report F0115 (CRWMS M&O, 2000m). The abstraction uses empirical data on release and colloid stability to formulate a dependence of colloidal radionuclide release on in-package ionic strength and pH. The preliminary draft report uses literature and Yucca Mountain Project data to support its construction of an algorithm for calculating colloid-associated radionuclide concentrations in solutions leaving the waste package. No credit is taken for colloid retardation within the waste package. Direct inputs for conceptual models and parameters are supplied by two supporting preliminary draft Analysis and Model Reports (F0105 and F0110; discussed below), as well as from a small number of literature sources. The abstraction takes output from in-package geochemical models and uses pH, ionic strength, and dissolved radionuclide concentration to calculate colloid concentrations, irreversibly colloid-bound radionuclide concentrations, and reversible colloid binding of radionuclides. The results are combined to provide a total colloid-associated source term for a given radionuclide; presently, the abstraction is applied only to plutonium and americium, though the americium abstraction is incomplete. [The preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j) provides more information on americium modeling in the form of  $K_d$  values for reversible colloid attachment, but does not provide parameters for irreversible attachment.] The abstraction classifies colloids as waste form, groundwater (preexisting), or iron oxyhydroxide (from corrosion). Waste-form colloids and their associated irreversibly bound radionuclides are assumed to form only in the presence of high-level waste glass. In contrast, the abstraction for spent fuel waste forms includes only reversible attachment onto iron oxyhydroxide and groundwater colloids, due to limited laboratory evidence for generation of waste-form colloids during spent fuel corrosion (CRWMS M&O, 2000j). Note that the following discussion was based on a preliminary draft version of the abstraction Analysis and Model Report; details may change before final release of Revision 00, but these changes are expected to have been incorporated into the final Waste Form Degradation Process Model Report.

The following key inputs are used in the preliminary draft colloid release abstraction (CRWMS M&O, 2000m):

- solution ionic strength, pH, and radionuclide concentration, from separate total system performance assessment in-package geochemical calculations
- effect of ionic strength on water concentration of waste-form colloidal plutonium, including a maximum colloidal plutonium concentration of  $6 \times 10^{-8}$  molar at ionic strength  $< 0.01$  molar and a minimum of  $1 \times 10^{-11}$  molar at ionic strength  $> 0.05$  molar; from data in preliminary draft Analysis and Model Report F0110 (CRWMS M&O, 2000n)
- maximum ionic strength stability limits for waste-form colloids as a function of pH, ranging from 0.01 molar at pH 2, to 0.05 molar at pH  $\geq 6$ ; based on montmorillonite data from Tombacz, et al. (1990) and on preliminary draft Analysis and Model Report F0110 (CRWMS M&O, 2000n)

- maximum ionic strength stability limits for iron oxyhydroxide colloids as a function of pH, ranging from 0.05 molar at pH < 6 and > 11 to a minimum of 0.01 molar at pH 8 to 9; from Liang and Morgan (1990)
- relationship between ionic strength and mass of groundwater colloids, ranging between a minimum of  $3 \times 10^{-6}$  and a maximum of  $3 \times 10^{-2}$  milligrams per liter; from the TSPA-VA technical basis document (CRWMS M&O, 1998d; figure 4-30)
- range of distribution coefficients for reversible sorption onto colloids, with a geometric mean of  $1 \times 10^4$  milliliters per gram for plutonium and  $1 \times 10^5$  milliliters per gram for americium; based on data in preliminary draft Analysis and Model Report F0105 (CRWMS M&O, 2000o).

The order of calculation is as follows:

- (1) water concentration of radionuclide irreversibly sorbed to waste-form colloids, using ionic strength and pH (applied only to plutonium and only in the presence of high-level waste glass)
- (2) mass concentration of waste-form colloids, using experimental relationship between concentrations of colloids and radionuclide irreversibly sorbed to them (applied only to high-level waste glass)
- (3) radionuclide reversibly sorbed to waste-form colloids, using distribution coefficient (applied only to high-level waste glass)
- (4) mass concentration of iron oxyhydroxide colloids, using ionic strength and pH
- (5) radionuclide reversibly sorbed to iron oxyhydroxide colloids, using distribution coefficient
- (6) mass concentration of groundwater colloids, using ionic strength
- (7) radionuclide reversibly sorbed to groundwater colloids, using distribution coefficient
- (8) summed colloidal radionuclide concentration and summed colloid mass concentration output to exterior of waste package

With respect to the approach to reversible attachment of radionuclides to colloids, the modeling approach presented in this preliminary draft Analysis and Model Report (CRWMS M&O, 2000m) does not represent a significant departure from the approach adopted in the TSPA-VA (CRWMS M&O, 1998d). The preliminary draft Analysis and Model Report abstraction is, however, an improvement. The TSPA-VA abstraction—applied only to plutonium—used modeled in-package ionic strength to constrain colloid concentration, then applied a  $K_d$  to calculate dissolved concentrations. The relationship between colloid concentration and ionic strength was based on groundwater data. Different classes of colloids were assigned different  $K_d$ s, but it is not clear from the TSPA-VA technical basis document (CRWMS M&O, 1998d) how their individual colloid concentrations were calculated. The preliminary draft abstraction Analysis and Model Report (CRWMS M&O, 2000m) uses the groundwater ionic strength relationship only for groundwater colloids and uses laboratory and literature data for the ionic

strength effect on concentrations of colloids derived from engineered materials and waste. In all cases, pH dependence was incorporated into the ionic strength relationship. In addition, more recent  $K_d$  data from Los Alamos National Laboratory are applied. The new abstraction, therefore, represents a marked refinement in that model parameters are more specific to colloid type and repository conditions.

The new approach to modeling irreversible colloid attachment, in contrast, does differ fundamentally from the Viability Assessment approach. In the TSPA-VA (CRWMS M&O, 1998d), the flux of irreversibly-attached plutonium was calculated as simply a fraction of the reversible flux; the fraction was based on Nevada Test Site observation of plutonium transport (Kersting, et al., 1999) and was given a log-uniform distribution covering six orders of magnitude. The preliminary draft Analysis and Model Report (CRWMS M&O, 2000m), in contrast, uses direct measurements of colloid-associated plutonium from laboratory experiments on waste forms to derive model expressions for concentrations of both colloids and irreversibly-bound colloidal radionuclides.

Clearly, the preliminary draft Analysis and Model Report abstraction of colloidal release modeling represents an appropriate enhancement over the TSPA-VA approach.

Preliminary draft Analysis and Model Report F0105 (CRWMS M&O, 2000o) describes literature data and Yucca Mountain project laboratory studies on colloid stability and colloid sorption of radionuclides. The preliminary document provides direct input to the preliminary draft abstraction Analysis and Model Report (CRWMS M&O, 2000m) in the form of a range of sorption coefficients, or  $K_d$ s, to be used in modeling reversible attachment of plutonium and americium to colloids. The  $K_d$ s were based on batch colloid sorption experiments conducted at Los Alamos National Laboratory. Preliminary draft Analysis and Model Report F0105 has not yet been received in final form, and is expected to be substantially revised.

This preliminary draft Analysis and Model Report includes an extensive discussion on literature relevant to reversible binding of radionuclides to colloids, including discussion of kinetics, followed by a report on previously unavailable laboratory data on plutonium and americium attachment to mineral colloids. The experiments were designed to observe adsorption and desorption rates, as well as the effect of colloid concentration. Dependence on pH and ionic strength of sorption onto colloids is not addressed. The key outputs to the preliminary draft abstraction Analysis and Model Report are distributions of  $K_d$  values for plutonium and americium to be used in modeling reversible attachment. This approach appears valid, as long as sufficient uncertainty is applied to the  $K_d$  range to account for deviations of repository conditions from those in the laboratory. As discussed above, discussion of uncertainty applied to parameter ranges needs better attention in the preliminary draft abstraction Analysis and Model Report.

In the preliminary draft Analysis and Model Report F0105, no technical basis is provided for the use of the mass-based coefficient  $K_d$  rather than the surface-area based coefficient  $K_A$ . In addition, the report omits important experimental details—such as pH variation during the experiments—needed to support the applicability of the results. Many of the preliminary draft Analysis and Model Report deficiencies are expected to be addressed in the final Revision 00 version.

The preliminary draft Argonne National Laboratory Analysis and Model Report (CRWMS M&O, 2000n) contains literature and previous Argonne National Laboratory data from static and drip corrosion tests on high-level waste glass and spent nuclear fuel. These data support a model of irreversible plutonium colloid attachment that is used in the colloid source term preliminary draft abstraction Analysis and Model Report (CRWMS M&O, 2000m). The direct inputs to the adopted abstraction—all based on Argonne National Laboratory work—are (i) a relationship between colloidal plutonium concentration and ionic strength based on static high-level waste glass corrosion tests, (ii) effect of ionic strength on colloid stability, and (iii) a direct relationship between colloidal plutonium concentration and colloid concentration. The adopted abstraction uses data only from the high-level waste glass tests, but spent nuclear fuel results are included in the development of a model in the Argonne National Laboratory preliminary draft Analysis and Model Report (CRWMS M&O, 2000n) that will be used in the preliminary draft abstraction Analysis and Model Report as an alternative model.

#### **5.4.3.2 U.S. Nuclear Regulatory Commission Staff Evaluation**

Staff analysis of DOE's approach to abstraction of the effects of coupled THC processes on the chemical environment for radionuclide release was conducted based on the acceptance criteria presented in Sections 5.4.3.2.1 and 5.4.3.2.2. In Section 5.4.3.2.1, staff analysis based on the acceptance criteria for the integrated subissue on quantity and chemistry of water contacting waste packages and waste forms is presented. In Section 5.4.3.2.2, staff analysis based on the acceptance criteria for the integrated subissue on radionuclide release rates and solubility limits is presented.

To facilitate discussion of resolution status, the acceptance criteria used in staff analysis are listed in Sections 5.4.3.2.1 and 5.4.3.2.2. There are five generic acceptance criteria. Each generic acceptance criteria is followed by specific criteria. Only those specific criteria related to ENFE Subissue 3 are included here. DOE's approach to abstract the effects of coupled THC processes on the chemical environment for radionuclide release will be acceptable provided that each acceptance criterion is met.

##### **5.4.3.2.1 Integrated Subissue on Quantity and Chemistry of Water Contacting Waste Packages and Waste Forms**

###### **5.4.3.2.1.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 3**

Important design features, physical phenomena and couplings, and consistent and appropriate assumptions have been identified and described sufficiently for incorporation into the abstraction of the quantity and chemistry of water contacting waste packages and waste forms in the performance assessment and other related abstractions in the total system performance assessment, and the technical bases are provided. The features, phenomena and couplings, and assumptions used to abstract the quantity and chemistry of water contacting waste packages and waste forms have been provided. The abstraction is consistent with the identification and description of those aspects of the quantity and chemistry of water contacting waste packages and waste forms that are important to waste isolation. The abstraction is also consistent with the technical bases for these descriptions of barriers important to waste isolation. Specifically,

- DOE spatial and temporal abstractions appropriately address the physical couplings (THMC)
- DOE provides adequate technical bases, including activities such as independent modeling, laboratory or field data, or sensitivity studies, for exclusion of any THMC couplings and FEPs

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Using the screening of the preliminary draft DOE FEP database (U.S. Department of Energy, 1999), the database provided in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) and the audit review of Pickett and Leslie (1999), 62 primary FEPs are identified as relevant to both the integrated subissue on quantity and chemistry of water contacting waste packages and ENFE Subissue 3. Of these 62, 21 are excluded by the preliminary DOE screening process. For two design related FEPs (1.1.07.00.00—Repository design; 1.1.08.00.00—Quality control), deviations from design are excluded. One additional FEP (2.1.06.06.00—Effects of degradation of drip shield) has no screening status given in the preliminary draft DOE database. A review of these 24 FEPs indicated that fifteen of those should not have been excluded or that the technical basis for exclusion is inadequate.

Staff either does not support DOE's preliminary screening or concluded there is an inadequate technical basis for exclusion of the following fifteen FEPs. The bases for the determination that these FEPs should not be excluded are discussed in the following paragraphs.

1.1.07.00.00—Repository design, 1.1.08.00.00—Quality control (deviations from design), and 1.1.12.01.00—Accidents and unplanned events during operation

These FEPs involve operational and preclosure aspects of the proposed repository and were excluded in DOE's preliminary screening on the basis of low consequence. As explained in the screening argument for FEP 1.1.03.01.00,

The TSPA is based on an assumption that the repository will be constructed, operated, and closed according to design. Deviations from design during the operational period are the subject of an extensive quality control program, and are outside the scope of the long-term performance assessment. Significant deviations that are detected during the operational period will be corrected and, therefore, are excluded from the TSPA on the basis of low probability.

Operational errors cannot be excluded without quality control procedures. Quality control procedures and operational procedures under normal and accident conditions are reviewed in specific sections of the Yucca Mountain Review Plan. Also, DOE references these FEPs with regard to the operational period; preclosure aspects of the proposed repository are reviewed under the preclosure section of the Yucca Mountain Review Plan and not in the postclosure model abstraction section that includes this integrated subissue. Nevertheless, these preclosure activities may impact post-closure repository performance. An inadequate technical basis for screening these FEPs has been provided. In addition, the screening of some of the FEPs is inconsistent with post-closure performance assessment calculations. For instance, the effects of the FEP on undesirable materials is required for the analysis of potential microbial

effects on performance (see section 4.1; CRWMS M&O, 1998a). DOE needs to provide a more robust technical basis for exclusion of these FEPs, perhaps by citing project documents where these assumptions and postulated actions (i.e., actions to correct mistakes) are codified. Also DOE needs to screen these FEPs so that the screening is consistent with actual post-closure performance assessment calculations.

#### 1.2.06.00.00—Hydrothermal activity

While this FEP refers to natural hydrothermal activity and is excluded on the basis of low consequence, the preliminary draft Unsaturated Zone Flow Process Model Report (CRWMS M&O, 2000e) states that, “mineralogical data suggest that no hydrothermal alteration has occurred since the waning of Timber Mountain volcanism about 10 Ma.” This statement refers to probability and not consequence. An argument to exclude this FEP based on consequences would demonstrate that if hydrothermal activity occurred, then repository performance would not be adversely affected. In addition, the technical basis does not even address the alternate hypothesis that hydrothermal activity has occurred. This alternative hypothesis is the focus of the ongoing University of Nevada Las Vegas fluid inclusion study. At least some of the fluid inclusion evidence suggest thermal fluids (45 - 80 °C) have passed through Yucca Mountain. Although earlier work has dated the age of one elevated fluid inclusion samples as 160 ka (Dublyansky, 1998), the current fluid inclusion study has found that the fluid inclusions with measured elevated temperatures are not in the very youngest minerals that precipitated (Cline, 2000). Thus DOE should either screen this FEP based on probability, taking into account the observations from the fluid inclusion study, or screen the FEP based on consequences and demonstrate that if hydrothermal fluids did occur then the magnitude and time of the resulting expected annual dose would not be significantly changed by its omission.

Because data and analyses do not currently support the likelihood of hydrothermal activity, the exclusion of this FEP from DOE evaluation of coupled THC effects on the waste package chemical environment is acceptable.

#### 2.1.02.05.00—Glass cracking and surface area

This FEP concerns possible effects on high-level waste glass alteration and radionuclide dissolution as a result of surface area increase due to glass cooling and handling. The screening argument states that “The robust container used at Yucca Mountain means that the concerns about fragmentation (and increased leach rate) of vitrified wastes are secondary” (U.S. Department of Energy, 1999). However, as in the TSPA-VA (U.S. Department of Energy, 1998b), planned DOE modeling of high-level waste glass degradation for the TSPA-SR includes a surface area term (CRWMS M&O, 1999a). In addition, greater glass surface area may also enhance the chemical effect on water contacting the glass. Development of the surface area term in the high-level waste glass degradation abstraction should include allowance for cooling and handling effects, unless DOE provides a technical basis for neglecting them. Staff expects this FEP and the technical basis for its exclusion to be more fully addressed in the FEPs screening Analysis and Model Report F0185.

#### 2.1.02.08.00—Pyrophoricity

DOE excluded this FEP on the basis of low consequence. However, adequate technical bases are not provided to support the exclusion of this FEP. The screening argument for this FEP

relies on calculations that have not yet been performed. Results from the proposed calculations would provide an adequate basis for the screening of this FEP. The exclusion of this FEP is unacceptable because DOE has not yet provided a clear technical basis and supporting calculations.

#### 2.1.02.09.00—Void space (in glass container)

DOE has not provided a technical basis to support the exclusion of this FEP. Exclusion of this FEP is unacceptable until a satisfactory technical basis is provided.

#### 2.1.02.13.00—General corrosion of cladding

As stated in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), "...because Zircaloy provides such excellent protective properties on average, characterization of the uncertainty in its performance is important." Zirconium oxidation is presented in this database entry as a mechanism for cladding degradation. The screening argument in the preliminary database (U.S. Department of Energy, 1999) provides a description of the process rather than a rationale for its exclusion. This FEP is unresolved until DOE provides a technical basis for exclusion. Staff expects the technical basis for exclusion of this FEP to be addressed in the FEPs screening Analysis and Model Report F0050.

#### 2.1.02.14.00—Microbially induced corrosion of cladding

This FEP was excluded by DOE on the basis of low consequence and because there is no experimental evidence of enhanced microbial corrosion of zirconium metal or alloys (CRWMS M&O, 2000j). The basis for screening is probability (no evidence for the process to occur) and not consequence. If DOE wants to exclude this FEP based on consequences then DOE will need to provide calculations demonstrating that if microbial corrosion of cladding occurred then the repository performance would not be adversely impacted. This FEP is unresolved until DOE provides a sufficient technical basis for exclusion of this FEP.

#### 2.1.02.15.00—Acid corrosion of cladding from radiolysis

The database screening of this FEP is tentative; the TSPA disposition field states, "Not a major issue, could use analysis at later time" (U.S. Department of Energy, 1999). Otherwise, there is no screening argument for a process that is potentially significant (Cragnolino, et al., 1999) and this specific mechanism is not addressed in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a). This FEP is unresolved until DOE provides a technical basis for exclusion. Staff expects the technical basis for exclusion of this FEP to be addressed in the FEPs screening Analysis and Model Report F0050.

#### 2.1.02.19.00—Creep rupture of cladding

Although the preliminary draft DOE database (U.S. Department of Energy, 1999) gives the status of this FEP as excluded, the preliminary draft Process Model Report on Waste Form Degradation (CRWMS M&O, 2000j) indicates creep rupture is considered in DOE's analysis of cladding degradation. Staff will evaluate the DOE abstraction of this FEP described in the relevant Analysis and Model Reports. DOE should make the FEP database consistent with the actual screening DOE chooses for the FEP.

#### 2.1.06.06.00—Effects and degradation of drip shield

No screening status was given for this FEP in the preliminary draft DOE database (U.S. Department of Energy, 1999). However, staff believes the drip shield and its degradation could potentially affect the flow and chemistry of water contacting the waste forms. Staff expects this FEP to be addressed in the FEPs screening Analysis and Model Report F0185.

#### 2.1.09.07.00—Reaction kinetics in waste and engineered barrier system

The FEP description reads, “Chemical reactions, such as radionuclide dissolution/precipitation reactions and reactions controlling the reduction-oxidation state, may not be in equilibrium in the drift and waste environment” (U.S. Department of Energy, 1999). DOE’s preliminary screening argument states, “Subtleties of the reduction-oxidation reactions are only of interest if they actually significantly affect transport. Use of experimentally derived  $K_d$ s already averages over interactions (and material heterogeneity). Specific effects of redox kinetics are therefore excluded from the TSPA on the basis of low consequence.” The screening argument appears to address only the effect of kinetics on redox reactions affecting radionuclide transport. The kinetics of dissolution/precipitation appear not to be considered. Thus, DOE’s screening argument constitutes an oversimplification of chemical processes in the waste and engineered barrier system. In addition, the sole secondary entry—2.1.09.07.01—is “included”, and the argument says that kinetics “... are to be included in detailed modeling of chemical interactions using EQ3/6.” In discussing the implementation in the TSPA-SR of the in-drift geochemical environment model, the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a, Sections 3.3.5 and 3.3.7) refers to the use of kinetic rate constants. Therefore, it appears that this FEP will not be excluded, and staff expects this to be reported in the FEPs screening Analysis and Model Report F0185. DOE should ensure that the database is consistent with the screening decision of the FEP in the performance assessment.

#### 2.1.11.08.00—Thermal effects: chemical and microbiological changes in the waste and engineered barrier system

This FEP is broadly defined as follows: “Temperature changes may affect chemical and microbial processes in the waste and engineered barrier system” (U.S. Department of Energy, 1999). The screening argument refers to other entries—2.1.10.01.00 and subentries in 2.1.09.00.00—but does not provide a rationale for exclusion. None of the primary entries cited in the screening argument deal specifically with thermal effects. The discussions of in-drift geochemical environment models in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a, Section 3.3) make it clear that temperature effects on chemical processes affecting radionuclide release will be implemented in the TSPA-SR. It, therefore, appears that this FEP will not be excluded, and staff expects this FEP to be considered in DOE’s screening of FEPs with respect to radionuclide release in Analysis and Model Report F0185. DOE should ensure that the database is consistent with the screening decision of the FEP in the performance assessment.

#### 2.1.12.01.00—Gas generation

The FEP description states, in part, “Gas generation might lead to pressurization of the repository, produce multiphase flow, and affect radionuclide transport. This FEP aggregates all types of gas generation into a single category” (U.S. Department of Energy, 1999). The

preliminary screening argument states that “Since the repository would be in the unsaturated zone, which is well-connected to the surface, gas produced by whatever reaction is expected to escape or at least be only temporarily confined beneath the condensate zone above the drifts.” Exclusion of this FEP implies that gases generated in the near field will have no impact on the chemical environment for radionuclide release. Clearly, gases can affect water chemistry, in turn affecting release processes. The conclusion that gas will escape with no effect on the near-field chemical environment needs a technical basis, particularly because the possibility of temporary confinement is acknowledged. With respect to FEP screening related to radionuclide release, staff expects this FEP and the technical basis for its exclusion to be addressed in Analysis and Model Report F0185.

The status of the remaining nine FEPs excluded by the DOE screening process is considered resolved and an adequate technical basis for their exclusion has been provided by DOE. The bases for this determination are discussed in the following paragraphs.

#### 2.1.02.20.00—Pressurization from He production causes cladding failure

The staff considers this FEP will not significantly influence the effects of coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release. This FEP was excluded by the DOE based on its analysis that accumulation of He sufficient to potentially cause pressurization will not occur until after the repository has cooled down. Staff considers DOE’s screening argument for this FEP is appropriate and the technical basis for exclusion of this FEP from DOE’s evaluation of coupled THC effects on the chemical environment for radionuclide release is acceptable.

#### 2.1.04.03.00—Erosion or dissolution of backfill

The FEP description in the preliminary draft DOE database (U.S. Department of Energy, 1999) states, “Solid material in buffer or backfill is carried away by flowing groundwater, either by erosion of particulate matter or by dissolution.” The FEP pertains to a physical process, which will not significantly impact the chemical environment for radionuclide release. Staff considers the technical basis for exclusion of this FEP from DOE’s evaluation of coupled THC effects on the chemical environment for radionuclide release acceptable, especially since backfill has been removed from the Enhanced Design Alternative II design.

#### 2.1.06.04.00—Flow through the liner

DOE excludes this FEP because the Enhanced Design Alternative-II design eliminates the concrete liner that was included in the TSPA-VA design. At this time, staff considers the technical basis for excluding this FEP in DOE’s evaluation of coupled THC effects on the chemical environment for radionuclide release as acceptable.

#### 2.1.09.11.00—Waste-rock contact

This FEP refers to contact between the waste form and rock due to mechanical failure of the drip shields and waste packages, for example from rockfall and drift collapse. This FEP was excluded by the DOE on the basis that interactions of the water with rocks and with Fe of the container are expected to be more important in controlling waste form degradation compared to contact between waste form and rock. In addition, DOE’s preliminary draft analyses of in-

package chemistry (CRWMS M&O, 2000i) indicates the changes in chemical environment for radionuclide release are dominated by interaction of water, not with the rocks, but with the waste package components. Staff considers DOE's screening argument for this FEP is appropriate, and technical basis for exclusion of this FEP from the DOE evaluation of coupled THC effects on the chemical environment for radionuclide release is acceptable.

#### 2.1.11.03.00—Exothermic reactions in waste and engineered barrier system

DOE (U.S. Department of Energy, 1999) states, "A repository at Yucca Mountain is planned to be "hot". Maximum rock temperature in the drift walls are expected to reach 165–185 °C. The temperature changes suggested in this FEP are inconsequential by comparison." Staff also considers the effects of exothermic reactions in the waste and engineered barrier system on the chemical environment for radionuclide release negligible compared to the effects of other coupled THC processes. Staff considers the DOE screening argument for this FEP appropriate, and exclusion of this FEP from the DOE analyses of radionuclide release is acceptable.

#### 2.1.11.10.00—Thermal effects on diffusion (Soret effect) in waste and engineered barrier system

The FEP description in the preliminary draft DOE database (U.S. Department of Energy, 1999) states, "The Soret effect is a diffusion process caused by a thermal gradient. In liquids having both light and heavy molecules (or ions), the heavier molecules tend to concentrate in the cold region. Temperature differences in the waste and engineered barrier system may result in a component of diffusive solute flux that is proportional to the temperature gradient." This FEP has been excluded by the DOE because the contribution of the Soret effect to radionuclide transport through the engineered and natural barriers is likely to be small in comparison to other chemical processes. This is an appropriate screening argument, particularly given the relatively heavy radioelements and the lower thermal gradient attributed to the Enhanced Design Alternative-II design, and the staff has no further questions on this FEP or the technical basis for its exclusion.

#### 2.1.12.02.00—Gas generation (He) from fuel decay

The screening argument in the preliminary draft DOE database (U.S. Department of Energy, 1999) states, "He production is a result of alpha-decay of actinides, so it is a consequence of the decay process. From estimates of alpha decay of commercial spent nuclear fuel, He could add 10 MegaPascal over 10<sup>6</sup> years. Current pressure is about 32 MegaPascal within the cladding. It is not expected that this increased pressure will affect the cladding failure rate." Staff considers the effects of this FEP on the chemical environment for radionuclide release negligible compared to the effects of other coupled THC processes. The technical basis for exclusion of this FEP from the DOE analyses of radionuclide release is acceptable.

#### 2.1.12.03.00—Gas generation (H<sub>2</sub>) from metal corrosion

The screening argument in the preliminary draft DOE database (U.S. Department of Energy, 1999) states, "A repository in the unsaturated zone in Yucca Mountain is expected to be connected to the atmosphere and to be operating under oxic conditions. Gas generated by metal corrosion will interact with the containers or escape from the drifts." Staff considers H<sub>2</sub>

gas generation from corrosion of stainless steel and Alloy 22 negligible compared to other processes such as radiolysis. Exclusion of the effects of this FEP on the chemical environment for radionuclide release from the DOE analyses is acceptable. The technical basis for exclusion of the effects of this FEP on the chemical environment for radionuclide release from the DOE analyses is acceptable.

#### 2.1.12.04.00—Gas generation (CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>S) from microbial degradation

DOE's preliminary description of this FEP (U.S. Department of Energy, 1999) states, "Microbial breakdown of cellulosic material, and possibly plastics and other synthetic materials, will produce mainly CO<sub>2</sub>, but also other gases. The rate of microbial gas production will depend upon the nature of the microbial populations established, the prevailing conditions (temperature, pressure, geochemical conditions), and the substrates present." DOE excludes this FEP on the basis that there is no plan to dispose of organic materials at Yucca Mountain. Staff considers the effects of this FEP on the chemical environment for radionuclide release negligible compared to the effects of other coupled THC processes. The technical basis for exclusion of this FEP from the DOE analyses of radionuclide release is acceptable.

The preliminary draft FEP database developed by the DOE (U.S. Department of Energy, 1999) provides a more formalized documentation of the FEPs screening and identification process that will be used for the DOE site recommendation. Pickett and Leslie (1999) provide an early audit review of the DOE treatment of FEPs. Future DOE analysis of FEPs should meet the acceptance criteria for scenario analysis in the TSPAI IRSR and in the Yucca Mountain Review Plan. DOE must provide an adequate technical basis for omitting all FEPs that may significantly affect the chemical environment for radionuclide release. This FEPs analysis will be an important action for DOE to take to facilitate resolution of this subissue. Staff will review the FEPs Analysis and Model Reports and the other Analysis and Model Reports to determine if the relevant FEPs have been adequately incorporated into the DOE analyses and whether an adequate basis has been provided for all FEPs that have been excluded from the performance assessment calculations.

- The DOE abstraction is consistent with the detailed information on waste package design and other engineered features

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE described the general design features associated with the Enhanced Design Alternative-II design of the waste package in the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j). The quantities of the various waste package components assumed in their calculations are identified in the associated preliminary draft Analysis and Model Report (CRWMS M&O, 2000i). DOE abstraction of in-package chemistry appears consistent with the detailed information on waste package design, but resolution of the subissue with respect to this acceptance criterion requires further review of DOE's Analysis and Model Reports and Process Model Reports.

- DOE identifies and adequately considers the effects of the drip shield and backfill on the quantity and chemistry of water contacting waste packages and waste forms, including the potential for condensate formation and dripping from the underside of the shield

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE has not considered the effects of the drip shield and backfill on the quantity and chemistry of water contacting the waste form. Since the License Application Design Selection process, DOE has removed backfill from the reference design (CRWMS M&O, 2000b), thus, the potential effect of backfill on the quantity and chemistry water is no longer an issue. However, the potential effect of the drip shield has not been considered, and DOE will need to provide justification for neglecting the effect of condensate formation and dripping from the underside of the drip shield on the quantity and chemistry of water contacting the waste form.

- DOE identifies the range of environmental conditions to be expected inside of breached waste packages, on the cladding, and contacting the waste forms
- DOE uses important design features, including waste package design and material selection, backfill, drip shield, ground support, cladding, thermal-loading strategy, and degradation processes, to determine the initial and boundary conditions for calculations of the quantity and chemistry of water contacting waste forms
- DOE identifies and considers likely modes of corrosion for container materials, including dry-air oxidation, humid-air corrosion, and aqueous corrosion processes, such as general corrosion, localized corrosion, microbial induced corrosion, stress corrosion cracking, and hydrogen embrittlement, as well as the effect of galvanic coupling, in determining the quantity and chemistry of water entering the waste packages
- DOE identifies and considers likely modes of failure for cladding, including aqueous corrosion processes, such as localized corrosion, stress corrosion cracking, and hydrogen embrittlement, creep, delayed hydride cracking, unzipping from oxidation of fuel, and mechanical failure from rockfall, in determining the quantity and chemistry of water contacting waste forms
- DOE consistently addresses the effect of corrosion products on the chemistry of water contacting the waste forms, and the effect on waste package corrosion of the quantity and chemistry of water contacting waste packages in all relevant abstractions
- DOE consistently addresses the role of parameters such as the pH and carbonate concentration of water and the effect of released radionuclides on the chemistry of water contacting the waste packages and waste forms in all relevant abstractions

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The range in composition of fluids inside breached waste packages was predicted by DOE based on reaction-path modeling, taking into account the interaction of the incoming fluid with waste package components and waste forms. The calculations considered important design features of the waste package and material selection, considered the formation of alteration minerals, and addressed the roles of pH, carbonate concentration, and release radionuclides (mainly uranium) on the chemistry of water contacting the waste forms. However, the calculations ignored the potential effect of the drip shield, ground support, and thermal-loading

strategy on the quantity and chemistry of water contacting waste forms. The J-13 Well water composition was used for the water entering the waste package, and there was no consideration of early-stage, high-temperature phenomena such as evaporation and condensation because the waste package is not expected to breach during the thermal period. The feedback between pH and cladding corrosion was also not included based on the assumption that pH will not become low enough to significantly increase clad failure rates.

In general, the staff finds the approach taken by DOE in determining the range of in-package chemistry acceptable. Staff agrees that, if corrosion is the only mechanism for degradation of waste packages, then breach of waste packages during the thermal period will not be significant and high-temperature phenomena need not be considered in determining the initial conditions for the in-package chemistry model. However, the potential for juvenile failure and for mechanical disruption of waste packages exists, and DOE will need to demonstrate the probability of these other mechanisms is not high enough to warrant evaluating the consequences of these other processes.

Furthermore, the in-package chemistry model is based on assumed corrosion rates for waste package component materials. No technical justification was provided for the assumed rates, and the likely modes of corrosion that account for the rates were not identified. DOE needs to provide sufficient information on the likely modes of corrosion that could affect the quantity and chemistry of water entering waste packages and contacting waste forms.

DOE's abstraction of in-package chemistry assumed general corrosion of cladding is insignificant under the chemical conditions expected inside reacting waste packages. Staff believes cladding degradation will not significantly influence the in-package fluid chemistry, but could significantly affect the amount of fuel exposed and the quantity of water contacting the waste form. Cladding degradation mechanisms, including initial failures, localized corrosion, and mechanical damage, are addressed in the preliminary draft cladding degradation Analysis and Model Report but it was not available to the authors of the preliminary draft Process Model Report on Waste Form Degradation (CRWMS M&O, 2000j). Thus, for the abstraction of in-package chemistry, DOE investigated a range of clad damage and commercial spent nuclear fuel exposure (100, 20, and 1 percent). The staff considers this approach of using a range of fuel exposure acceptable. However, DOE will need to evaluate whether the results presented in the preliminary draft cladding degradation Analysis and Model Report are significantly different from the assumed clad damage and commercial spent nuclear fuel exposure to affect the results of the in-package chemistry abstraction.

- DOE demonstrates that the conditions and assumptions used to generate look-up tables or regression equations are consistent with all other conditions and assumptions in the total system performance assessment for abstracting the quantity and chemistry of water contacting waste packages and waste forms

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The assumptions and conditions used to generate regression equations for in-package chemistry presented in the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j) and in the supporting preliminary draft Analysis and Model Reports

(CRWMS M&O, 2000i) appears to be consistent. Consistency with other Analysis and Model Reports and Process Model Reports will be checked.

- DOE consistently addresses the effect of distribution of flow on the amount of water contacting the waste forms in all relevant abstractions
- DOE consistently addresses the size and distribution of penetrations of waste containers that affect the quantity and chemistry of water that contacts the waste form in all relevant abstractions

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE's in-package chemistry calculations used input fluid fluxes of 1.5 to 150 liters per year, which are supposed to be in general agreement with bounding calculations for flow through a drip shield discussed in the preliminary draft Engineered Barrier System Process Model Report (CRWMS M&O, 2000b). Staff will review the relevant Process Model Reports and Analysis and Model Reports to determine if the quantity of water contacting waste packages and waste forms has been addressed consistently in all relevant abstractions.

#### **5.4.3.2.1.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 3**

Sufficient data on design features (including drip shield, backfill, waste packages, cladding, other engineered barrier components, and thermal loading), geology, hydrology, geochemistry, and geomechanics of the unsaturated zone and drift environment (e.g., field, laboratory, and natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing the abstraction of the quantity and chemistry of water contacting waste packages and waste forms in the performance assessment. The data are also sufficient to assess the degree to which FEPs related to the quantity and chemistry of water contacting waste packages and waste forms and which affect compliance with postclosure performance objectives have been characterized and to determine whether the technical bases provided for exclusion of these FEPs are adequate. Where adequate data do not exist, other information sources such as expert elicitation have been appropriately incorporated into the abstraction process. Specifically,

- DOE demonstrates that sufficient data were collected on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, to establish initial and boundary conditions, including temporal and spatial variations in conditions, for conceptual models and simulations of THMC coupled processes that affect the chemical environment for radionuclide release
- Where sufficient data do not exist, the definition of parameter values and conceptual models is based on appropriate other sources such as expert elicitation conducted in accordance with NUREG-1563 (Kotra, et al., 1996)
- DOE collects sufficient information to formulate the conceptual approach(es) for analyzing water contact with the cladding and waste forms upon waste package failure

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

A preliminary review of the preliminary draft DOE Analysis and Model Reports and Process Model Reports indicates there are sufficient data available to adequately define relevant parameters and conceptual models necessary to develop the abstraction of the quantity and chemistry of water contacting waste forms in the performance assessment. Sufficient data to establish initial and boundary conditions for conceptual models and simulations of coupled THC processes that affect the chemical environment for radionuclide release also appears to be available. However, resolution of the subissue with respect to this acceptance criterion requires further review of the pertinent DOE Analysis and Model Reports and Process Model Reports.

- DOE provides sufficient data or sound bases for the inclusion or exclusion of certain observed phenomena in its conceptual models

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The evaluation of phenomena excluded by the DOE in its conceptual models was discussed previously in acceptance criterion 1.

- DOE provides sufficient data to complete a nutrient and energy inventory calculation, if it has been used to justify the exclusion of the potential for microbial activity affecting the chemical environment for radionuclide release
- If microbial activity could be sufficient to affect repository performance, DOE provides sufficient data to assess microbial effects such as production of organic by-products and microbially enhanced dissolution of the high-level waste glass form

STATUS OF RESOLUTION: CLOSED

DOE did not attempt to evaluate the potential effect of microbial activity on the chemical environment for radionuclide release. The process was screened out in the DOE FEPs analysis presented in the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j) based on limited nutrient availability limiting biological activity, the absence of experimental evidence of enhanced microbial corrosion of zirconium metal or alloys, and the offsetting beneficial effect of microbial activity increasing colloid size and increasing filtration. As indicated in acceptance criterion 1, NRC staff considers the technical basis for exclusion of microbial activity from the DOE evaluation of coupled THC effects on the chemical environment for radionuclide release as acceptable.

- DOE performs sensitivity, if needed, or uncertainty analyses (including consideration of alternative conceptual models) to test for the necessity of additional data

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Large uncertainties are recognized in the thermodynamic and kinetic data and in the simplified approach used for the abstraction of in-package chemistry. Some attempts have been made to

account for uncertainties in the model parameters. For example, uncertainty in the dissolution rates of waste package materials is dealt with using high and low values. Drip rates onto the waste package ranged from 1.5 to 150 liters per year. On the other hand, the composition of water entering the waste package was assumed to be that of J-13 Well water and was not varied. In addition, the simulation temperature was set at 25 °C, neglecting the potential effect of elevated temperatures on the basis that many of the thermodynamic parameters are not strongly sensitive to temperature over the range 25 to 100 °C. Thus, the effect of using other initial water compositions or higher temperatures on the calculated in-package chemistry is unknown. DOE should conduct additional process-level modeling to ascertain the chemistry of in-package water has been bounded by the approach presented in the preliminary draft Waste Form Degradation Process Model Report and supporting Analysis and Model Reports.

#### **5.4.3.2.1.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 3**

Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstraction of quantity and chemistry of water contacting waste packages and waste forms, such as the pH, chloride concentration, and amount of water flowing in and out of the breached waste package, are consistent with site characterization data, design data, laboratory experiments, field measurements, and natural analog data, are technically defensible, and reasonably account for uncertainties and variabilities. The technical bases for the parameter values used in the total system performance assessment abstraction are provided. Specifically,

- Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the quantity and chemistry of water contacting waste forms calculations in the performance assessment are technically defensible and reasonable, based on data from the Yucca Mountain region (e.g., results from large-block and drift-scale heater and niche tests) and a combination of techniques that may include laboratory experiments, field measurements, natural analog research, and process-level modeling studies
- DOE demonstrates that parameters derived from process-level models used for the quantity and chemistry of water contacting waste forms are consistent with site characterization data, laboratory experiments, field measurements, and natural analog information
- DOE provides technical bases for parameter ranges, probability distributions or bounding values. DOE demonstrates that the parameter values are derived from site-specific data or provides an analysis to demonstrate that the assumed parameter values do not under-predict repository performance
- DOE demonstrates that input values used in the quantity and chemistry of water contacting engineered barriers (e.g., drip shield, waste package, and cladding) calculations in the total system performance assessment are consistent with the initial and boundary conditions and the assumptions of the conceptual models and design concepts for the Yucca Mountain site, such as waste package and engineered barrier system design (including backfill, drip shield, ground support, and cladding), waste package degradation (corrosion and mechanical disruption), cladding degradation, deep percolation flux, important THMC coupling effects, the thermal reflux model, the thermal-

loading strategy (including effects of ventilation), natural system masses and fluxes, and other design features that may affect performance

- DOE establishes that reasonable or conservative ranges of parameters or functional relations are used to determine effects of coupled THMC processes on the chemical environment for radionuclide release
- DOE shows that the parameters used to define initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THMC effects on the chemical environment for radionuclide release, are consistent with available data
- DOE provides confirmation that the correlations between input values, if any exist, have been appropriately established in the DOE total system performance assessment
- DOE demonstrates that parameter values, assumed ranges, probability distributions and bounding assumptions reflect the range of environmental conditions to be expected inside breached waste packages
- DOE demonstrates that parameter values, assumed ranges, probability distributions, and bounding assumptions reasonably account for uncertainties
- DOE adequately considers the uncertainties in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THMC coupled processes that affect the chemical environment for radionuclide release
- DOE adequately represents uncertainty in parameter development for conceptual models, process-level models, and alternative conceptual models considered in developing the abstraction of quantity and chemistry of water contacting waste forms, either through sensitivity analyses or conservative limits

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The dissolution rate assumed for Type 316 stainless steel and the borated stainless steel is one order of magnitude lower than measured experimentally (Kirchheim, et al., 1989). Additionally, the lower dissolution rate assumed for the borated stainless steel compared to Type 316 stainless steel is counterintuitive. The presence of boron, in the form of second phase particles of borides, would be expected to result in higher corrosion rate, especially in local zones around the boride particles. Better technical bases for these assumptions need to be provided.

The in-package chemistry and the dissolution rate of spent fuel are influenced by the flux of water entering the waste package. However, the quantity of water assumed to drip into the waste package for calculating the in-package chemistry is two to three orders of magnitude greater than the drip rate of water used in experimental studies conducted at Argonne National Laboratory. While the Argonne National Laboratory test results are not used for model abstraction, and the rate of fuel dissolution is not used in calculating the in-package chemistry, the Argonne National Laboratory test results are being used to confirm model abstractions and increase confidence in the assumptions used for waste form degradation models.

The rate of dissolution of commercial spent nuclear fuel used to calculate the in-package environment is significantly different from the rate of dissolution used for calculating radionuclide release. However, the commercial spent nuclear fuel dissolution rate as proposed is a conservative upper bound for acidic dissolution because its predicted dissolution rate was higher than that reported by Grambow and others (CRWMS M&O, 2000k).

#### **5.4.3.2.1.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 3**

Alternative modeling approaches consistent with available data (e.g., design features, field, laboratory, and natural analog) and current scientific understanding are investigated and results and limitations are appropriately factored into the abstraction of quantity and chemistry of water contacting waste packages and waste forms. DOE provided sufficient evidence that alternative conceptual models have been considered, that the models are consistent with available data and current scientific understanding, and that the effect of these alternative conceptual models on total system performance has been evaluated. Specifically,

- DOE investigates alternative modeling approaches consistent with available data and current scientific knowledge, and appropriately considers their results and limitations of these approaches in developing the abstraction of quantity and chemistry of water contacting waste forms
- DOE adequately considers the effects of THMC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products in their assessment of alternative conceptual models. DOE considers: (i) thermohydrologic effects on gas and water chemistry, (ii) hydrothermally driven geochemical reactions such as zeolitization of volcanic glass, which could affect flow pathways, water chemistry and waste package environmental conditions, (iii) dehydration of hydrous phases liberating moisture that may affect the chemical environment for radionuclide release, (iv) effects of microbial processes on the chemical environment for radionuclide release, (v) changes in water chemistry that may result from the release of corrosion products from the waste package and interactions between engineered materials and groundwater, which, in turn, may affect the chemical environment for radionuclide release, and (vi) changes in boundary conditions (e.g., drift shape and size) and hydrologic properties relating to the response of the geomechanical system to thermal loading, in their assessment of alternative conceptual models
- DOE provides a description which includes a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Radiolysis could have complex effects on the release of radionuclides from spent nuclear fuel (Shoesmith and Sunder, 1992; Christensen, and Bjergrakke, 1987; Christensen, Sunder, and Shoesmith, 1994). In aqueous solution, radiolysis produces oxidizing species such as hydrogen peroxide ( $H_2O_2$ ), hydroxyl radical (OH), and perhydroxyl radical ( $HO_2$ ), and reducing

species such as hydrated electron ( $e^-$ ), hydrogen atom (H), and hydrogen molecule ( $H_2$ ) (Spinks and Woods, 1990). In an air-water-vapor system, nitrogen fixation products—nitrogen acids, nitrogen oxides, and ammonia—would also be formed in addition to the radiolytic products formed in the aqueous system (Burns, et al., 1982; Reed and Van Konynenburg, 1987; Willis and Boyd, 1976). In the preliminary draft DOE analyses (CRWMS M&O, 2000j), gamma radiolysis effects are neglected on the basis that the radiation level in the waste package would not be significant after about 1000 years, and the probability of breach is quite low before 1,000 years. For the dissolution of spent nuclear fuel ( $UO_2$ ), the effects of gamma and alpha radiation have been extensively studied in aqueous solutions (Shoesmith, et al., 1985; Shoesmith and Sunder, 1992; Christensen, and Bjergrakke, 1987; Christensen, Sunder, and Shoesmith, 1994; Christensen, 1991). Shoesmith, Sunder and Tait (1997) conducted experiments in aerated non-complexing solutions and in aerated carbonate solutions under gamma irradiation conditions. They concluded that the effect of gamma irradiation becomes insignificant below ~1 to ~10 gray per hour. In a more recent review, Shoesmith (1999) predicted that the gamma radiolysis effect on fuel dissolution rate will be negligible after a few hundred years on the basis that the gamma would decay to an insignificant level (below ~ 1 to 10 gray per hour).

Sunder, Shoesmith, and Miller (1997) conducted experiments on the oxidation of  $UO_2$  nuclear fuel in aqueous solution by the products of alpha radiolysis of water. They predicted that the corrosion of nuclear fuel, supported solely by the alpha radiolysis of water, could be significant for time periods of ~2000 years for the enriched pressurized water reactor spent fuel. Corbel, et al. (2000) also found that the release of uranium in aerated water is strongly enhanced by alpha irradiation. Shoesmith (1999), however, recently predicted that the alpha radiolysis will have no significant effect on fuel corrosion and can be ignored. This is because the conditions at Yucca Mountain will be perpetually aerated and the oxidizing products produced by the alpha radiolysis may not have significant impact on the redox conditions. On the other hand, Shoesmith did caution that the influence of alpha radiolysis in tight wet cracks in the fuel remains to be determined.

The unsaturated drip tests with spent nuclear fuel (CRWMS M&O, 2000j; Stout and Leider, 1998; Finch, et al., 1999) may have given an indication on the effects of radiation on the dissolution rate of uranium fuel in the moist-air system. These investigators observed that the overall leaching rate of radionuclides is higher with the higher burn-up fuel than with the lower burn-up fuel. However, the conditions for the unsaturated tests were not designed to study the effects of irradiation; the parameters related to radiation effects, such as dose rate, air to water volume ratio, and air partial pressure were not controlled. After the failure of the waste package, the effect of alpha, gamma, and beta irradiation on the dissolution of uranium fuel in humid air may still be significant. DOE needs to assess the effect of alpha, gamma and beta radiation on the nuclear fuel under the humid air environment after the failure of the waste package.

DOE assumed that the outer overpack, Alloy 22, does not react with the dripping water and, therefore, does not affect the in-package chemistry. The other waste package components are assumed to dissolve uniformly such that they can be lumped into a single mass for purposes of chemical calculations. The chromium in the stainless steel is assumed to dissolve in its hexavalent form. While this assumption is conservative, lumping all the components into a single mass for estimating the chemistry may lead to highly nonconservative estimates of pH values. For example, the lowest pH in the EQ6 calculations is approximately 3.6 (CRWMS

M&O, 2000i). This assumption, while it simplifies calculations of in-package chemistry, needs further justification.

At issue is the spatial variation in chemistry that is likely to occur in the waste package and that is likely to result in local pH values considerably more acidic than calculated, based on a volume averaged mass. The pH in crevices and other tight spaces differs from bulk pH values because the dissolution reactions become spatially separated from the reduction reactions. Alternative models that consider the electrochemical reactions coupled to transport processes should be considered. For example, Cavanaugh, et al. (1983) reported that the pH values in corroding cavities of stainless steels range from 0 to 2, with the pH increasing with increasing molybdenum and decreasing chromium concentration. The pH in crevices of aluminum alloys can be either acidic (pH 4) or alkaline (pH 9) depending on the initial pH and surface condition. Therefore, the pH generated by localized dissolution of aluminum would be most likely influenced by the pH resulting from the corrosion of other components. Because the internal geometry of the waste package will have many tightly packed regions, local pH may affect the dissolution rate of spent fuel locally and, hence, the local release rate of highly soluble radionuclides such as Tc-99.

#### **5.4.3.2.1.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 3**

Output from the abstraction of quantity and chemistry of water contacting waste forms is justified through comparison with output from detailed process-level models and/or empirical observations (e.g., laboratory testing, field measurements, natural analogs).

- DOE verifies that the outputs of the quantity and chemistry of water contacting waste forms abstraction reasonably reproduce or bound the results of corresponding process-level models or empirical observations
- DOE demonstrates that abstracted models for coupled THMC effects on the chemical environment for radionuclide release, are based on the same assumptions and approximations demonstrated to be appropriate for closely analogous natural or experimental systems
- DOE shows that abstracted model results were verified through comparison with outputs of detailed process-level models and empirical observations
- DOE evaluates the outputs of the abstraction against field and laboratory data and natural analogs information
- DOE appropriately adopts accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THMC effects on the chemical environment for radionuclide release
- DOE compares abstracted model results with different mathematical models to judge robustness of results

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The abstractions for the in-package environment are derived from a detailed process-level model that used the EQ3/6 code. The predictions of this code have not been verified by empirical observations. DOE recognized the difficulties in modeling the detailed effects of geometry and corrosion reactions on the in-package chemistry. Because the corrosion reactions of metallic materials and spent fuel are electrochemical in nature, a lumped mass chemical reaction approach, such as that embodied in the EQ/6 code, may not be adequate. Detailed reactive transport models that incorporate electrochemical reactions, while more appropriate for this case than the EQ3/6 code, can be difficult to implement in a system that consists of multiple materials and geometries. Experiments to simulate certain aspects of waste package geometry and materials may aid in gaining confidence in the model abstractions.

#### **5.4.3.2.2 Integrated Subissue on Radionuclide Release Rates and Solubility Limits**

Detailed analyses of DOE abstractions, models, and analyses of radionuclide release rates and solubility limits are provided in the Container Life and Source Term IRSR Revision 2 (U.S. Nuclear Regulatory Commission, 1999d). Of relevance to the ENFE IRSR is the effect of coupled THC processes on the chemical environment affecting radionuclide release rates and solubility limits. Staff analysis of DOE's approach to abstraction of coupled THC effects on the chemical environment affecting radionuclide release and solubility limits is presented in the following sections.

##### **5.4.3.2.2.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 3**

Important design features, physical phenomena and couplings, and consistent and appropriate assumptions have been identified and described sufficiently for incorporation into the abstraction of radionuclide release rates and solubility limits in the total system performance assessment, and the technical bases are provided. The features, phenomena and couplings, and assumptions used to abstract release of radionuclides from waste forms inside the waste package and the transport and release of radionuclides from the engineered barrier system have been provided. The abstraction is consistent with the identification and description of those aspects of radionuclide release rates and solubility limits that are important to waste isolation. The abstraction is also consistent with the technical bases for these descriptions of barriers important to waste isolation. Specifically,

- As part of its FEPs analysis in developing the abstraction, DOE identifies and considers processes for cladding performance, spent nuclear fuel degradation, high-level waste glass dissolution, degradation of other radioactive wastes, the compatibility of spent nuclear fuel and high-level waste glass and other radioactive waste forms with internal components of the waste package, and the release of radionuclides from the engineered barrier system and provides adequate technical bases for the exclusion of any of these processes

- DOE provides adequate technical bases for exclusion of any potentially important couplings and FEP

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Using the screening decisions documented in the U.S. Department of Energy (1999) preliminary draft database provided in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) and the audit review of Pickett and Leslie (1999), 57 primary FEPs are identified as relevant to both the integrated subissue on radionuclide release rates and solubility limits and to this ENFE subissue. Of these 57, 22 are excluded by the DOE screening process. Of the 22 excluded FEPs, 15 overlap with the excluded FEPs identified as relevant to the integrated subissue on quantity and chemistry of water contacting waste packages and waste forms and are discussed in Section 5.4.3.2.1.1. For two design-related FEPs (1.1.07.00.00—Repository design and 1.1.08.00.00—Quality control), deviations from design are excluded; these FEPs are discussed in Section 5.4.3.2.1.1. For one additional FEP (2.1.13.02.00—Radiation damage in waste and engineered barrier system), the FEP component related to backfills, seals, and rock is excluded. In addition, a FEP (2.2.07.06.00—Episodic/pulse release from repository) has no screening status given in the preliminary draft DOE database. A review of these nine FEPs excluded by the DOE screening process (not including the 15 discussed previously in Section 5.4.3.2.1.1) is presented below.

Staff either does not support DOE's preliminary screening or concluded there is an inadequate technical basis for exclusion of the following five FEPs. The bases for the determination that these FEPs should not be excluded are discussed in the following paragraphs.

#### 1.1.02.03.00—Undesirable materials left

This FEPs involves operational and preclosure aspects of the proposed repository and was excluded in DOE's preliminary screening on the basis of low consequence. As explained in the screening argument for FEP 1.1.03.01.00,

The TSPA is based on an assumption that the repository will be constructed, operated, and closed according to design. Deviations from design during the operational period are the subject of an extensive quality control program, and are outside the scope of the long-term performance assessment. Significant deviations that are detected during the operational period will be corrected and, therefore, are excluded from the TSPA on the basis of low probability.

Also, DOE references this FEP with regard to the operational period; preclosure aspects of the proposed repository are reviewed under the preclosure section of the Yucca Mountain Review Plan and not in the postclosure model abstraction section that includes this integrated subissue. Nevertheless, pre-closure activities may impact post-closure repository performance. An inadequate technical basis for screening this FEPs has been provided. In addition, the screening of this FEPs is inconsistent with post-closure performance assessment calculations. For instance, the effects of this FEP is required for the analysis of potential microbial effects on performance (see section 4.1; CRWMS M&O, 1998a). DOE needs to provide a more robust technical basis for exclusion of this FEP, perhaps by citing project documents where these assumptions and postulated actions (i.e., actions to correct mistakes) are codified. Also DOE

needs to screen this FEP so that the screening is consistent with actual post-closure performance assessment calculations.

#### 2.1.02.04.00—Alpha recoil enhances dissolution

The preliminary screening argument provided by DOE (U.S. Department of Energy, 1999) to exclude this FEP states, “Preferential dissolution because of alpha recoil is a problem for dating waters and may influence the apparent dissolution rates and solubility of certain daughters. However, the alpha-produced daughters still have the same chemical solubility limits as those produced elsewhere. Changes in isotopic abundance for a solute do not appear to be significant enough to matter.” Staff considers insufficient technical basis was provided by DOE to screen out this FEP, particularly with respect to the potential effect of alpha recoil on the structural integrity of the waste form that could enhance the degradation rate. Exclusion of this FEP is unacceptable until a satisfactory technical basis is provided.

#### 2.1.09.02.00—Interaction with corrosion products

This FEP is concerned with effects on radionuclide mobility of corrosion products of metallic components. The preliminary draft database specifically refers to sorption, desorption, coprecipitation, and dissolution processes. The database preliminary screening of this FEP is tentative; the TSPA disposition field states that this effect will be “included only in the integrated source term and associated sorption parameters” (U.S. Department of Energy, 1999). In addition, the screening argument states that “Interaction of contaminants with corrosion products is expected to control mobilization and speciation of the contaminants.” Finally, the TSPA-SR methods and assumptions report states that “changes to water chemistry due to corrosion products will be provided as input to the TSPA drip shield, waste package, and waste form degradation models” (CRWMS M&O, 1999a, Section 3.3.2). It is apparent that this FEP will not be excluded, and staff expects this to be reported in the FEPs screening Analysis and Model Report F0185. In addition, staff considers this FEP relevant to all seven DOE abstractions covered in this review. DOE should ensure that the database is consistent with the disposition of the FEP in the total system performance assessment.

#### 2.1.12.08.00—Gas explosions

DOE excludes this FEP largely because the repository will be located in the unsaturated zone and be well connected to the surface, thus minimizing the potential for postclosure gas buildup, and because there is a lack of an ignition source. Based on the information available at this time, this exclusion may be acceptable if the DOE provides a technical basis for the screening argument, particularly because the possibility exists for temporary confinement of gas. Technical support for the lack of an ignition source will be a key part of the basis for exclusion.

#### 2.2.07.06.00—Episodic/pulse release from repository

No screening argument was provided by DOE to support exclusion of this FEP. Staff considers exclusion of this FEP unacceptable until an adequate technical basis is provided.

The status of the remaining four FEPs excluded by DOE’s preliminary screening process is considered resolved. The bases for this determination are discussed in the following paragraphs.

#### 2.1.02.06.00—Glass recrystallization

DOE excluded this FEP on the basis that (i) glass recrystallization is a slow process, (ii) glass recrystallization is only possible if high glass temperature is maintained during a prolonged period, (iii) glass recrystallization is unlikely to occur below 400 °C, and (iv) the waste glass at a Yucca Mountain repository will not be exposed to temperatures of 400 °C. The staff considers DOE's rationale appropriate and the technical basis for exclusion of this FEP from DOE's evaluation of coupled THC effects on the chemical environment for radionuclide release as acceptable.

#### 2.1.12.06.00—Gas transport in waste and engineered barrier system and

#### 2.1.12.07.00—Radioactive gases in waste and engineered barrier system

The screening argument provided by DOE (U.S. Department of Energy, 1999) states that radioactive gases of concern at Yucca Mountain are  $^{14}\text{CO}_2$ ,  $^{14}\text{CH}_4$ , radioactive fission gases (Ar, Xe, Kr), and radon. For a repository in the unsaturated zone, DOE states that  $\text{CO}_2$ ,  $\text{CH}_4$ , and fission gases will in part escape to the atmosphere, whereas the residence time for radon is long enough for decay so only further daughters might matter. DOE states that FEPs related exclusively to the behavior of radioactive gases have been excluded from the performance assessment on the basis of low consequence because radioactive gases are not a significant component of the future radionuclide inventory of the proposed Yucca Mountain repository. Staff finds the technical basis acceptable and considers exclusion of this FEP from the DOE abstractions of the effects of coupled THC processes on the chemical environment for radionuclide release acceptable.

#### 2.1.13.02.00—Radiation damage in waste and engineered barrier system

DOE's preliminary draft database (U.S. Department of Energy, 1999) indicates this FEP is included, except for radiation damage on backfill, seals, and rock. Staff considers the potential effects of radiation damage of backfill, seals, and rock on the chemical environment for radionuclide release will be negligible compared to the effects of other THC processes. Staff considers acceptable excluding radiation damage on backfill, seals, and rock from DOE's analyses of radionuclide release. In addition, the technical basis for exclusion of this FEPs is sufficient.

The preliminary draft FEP database developed by DOE (U.S. Department of Energy, 1999) provides a more formalized documentation of the FEPs screening and identification process that will be used for DOE's site recommendation. Pickett and Leslie (1999) provide an early audit review of DOE's treatment of FEPs. Future DOE analysis of FEPs should meet the acceptance criteria for scenario analysis in the TSPA IRSR and in the Yucca Mountain Review Plan. DOE must provide an adequate technical basis for all FEPs. Because the chemical environment for radionuclide release may affect repository performance, this FEPs analysis will be an important action for DOE to take to facilitate resolution of this subissue. Staff will review the FEPs Analysis and Model Reports and the other Analysis and Model Reports to determine if the relevant FEPs have been adequately incorporated into the DOE analyses and whether an adequate technical basis for exclusion has been provided for each ENFE-relevant excluded FEP.

- DOE's abstraction is consistent with the detailed information on waste package design and other engineered features
- DOE reasonably accounts for the range of environmental conditions, including water composition and water and vapor movement, expected inside breached waste packages and in the engineered barrier environment surrounding the waste package (e.g., temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release rates and solubility limits)

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The model DOE developed to evaluate commercial spent nuclear fuel cladding degradation represents a significant improvement with respect to that presented in the TSPA-VA. DOE considered most of the degradation mechanisms that may affect the integrity of the cladding under disposal conditions. However, the effect of chloride anions is not considered in DOE's analysis of localized corrosion and stress corrosion cracking of cladding. Instead, the role of fluoride as a species promoting accelerated corrosion in local areas is emphasized (CRWMS M&O, 2000j). Chloride is known to induce pitting corrosion (Brossia and Cragnolino, 2000) if a potential above the repassivation potential—easily attained in the presence of radiolysis products, such as  $H_2O_2$  or other oxidizing species—is reached. The same electrochemical and environmental conditions promote stress corrosion cracking in the presence of sufficiently high hoop stresses.

In the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j), the analysis of the flow and volume of water contacting the fuel rods to evaluate local attack of the cladding by fluoride is limited and requires additional justification. Inconsistencies occur regarding the evaluation of the in-package pH because a low pH is assumed for the attack by fluoride, whereas it is not taken into account in the concentration in solution of  $Fe^{3+}$  ions that may promote oxidizing conditions conducive for pitting in chloride solutions.

The abstraction used for commercial spent nuclear fuel dissolution rate considers the pH, total carbonate, oxygen partial pressure, and temperature. A significant improvement in the TSPA-SR modeling of waste form dissolution compared to the TSPA-VA approach is the consideration of in-package chemistry alteration by reaction with waste package components. However, as mentioned in Section 5.4.3.2.1, the in-package chemistry calculations should be supported by empirical observations and more detailed process-level models. For example, the abstractions do not explicitly consider some interactions between various waste package internal components and spent nuclear fuel. Spent fuel, because of its hyperstoichiometry, is a p-type semiconductor and, hence, capable of sustaining electrochemical reactions. Thus, galvanic interactions between cladding and fuel, basket materials and fuel, and other components need to be evaluated.

The pH coefficients for the high-level waste glass dissolution rate were determined using buffered solutions prepared with deionized water. The analysis ignores the presence of corrosion products, such as  $FeOOH$ ,  $FeCl_2$ , and  $FeCl_3$ , resulting from dissolution of waste package internal components that could influence glass corrosion processes and the pH dependence of the dissolution rate. Also,  $\beta$ - $FeOOH$  (akaganeite) can occlude significant concentrations of chloride within its lattice structure.

The staff agrees with the general approach to modeling colloid release as described in preliminary draft Analysis and Model Report F0115 (CRWMS M&O, 2000m) and the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j). The abstraction of colloid release as described in preliminary draft Analysis and Model Report F0115 (Colloid Source Term Abstraction) (CRWMS M&O, 2000m) attempts to quantify the effects of variable in-package chemical conditions on colloidal radionuclide release using limited, site-specific data supplemented by information from the literature. However, the preliminary draft colloid abstraction Analysis and Model Report does not provide sufficient justification that the abstraction is unlikely to neglect effects that could increase the colloidal radionuclide concentration. For example, the corrosion test results discussed in preliminary draft Analysis and Model Report F0110 (CRWMS M&O, 2000i) were conducted at 90 °C and the colloid sorption tests in preliminary draft Analysis and Model Report F0105 (CRWMS M&O, 2000o) at room temperature. However, the preliminary draft colloid abstraction Analysis and Model Report did not discuss possible temperature effects, and did not critically assess the potential effects on colloid behavior of chemical parameters other than ionic strength and pH (e.g., oxidation potential and alkalinity; U.S. Nuclear Regulatory Commission, 1999f). The colloid abstraction does not explicitly address a sufficient range of possible coupled and uncoupled effects on colloid stability and radionuclide attachment to ensure that the abstraction is adequate for the range of possible repository conditions.

DOE recognizes that the solubility of an element varies as the environmental conditions within a repository changes, and evaluation of solubility limits requires knowledge of changes in this environment and the dependence of radionuclide solubility on the environment. In the preliminary draft Analysis and Model Report F0095 (Summary of Dissolved Concentration Limits) (CRWMS M&O, 2000l), DOE states that analysis of the in-package chemistry and the analysis of solubility limits were conducted in parallel. Thus, the results of the in-package chemistry calculations were not fully utilized in evaluating the solubility limits. DOE identified several areas of improvement being considered for future revision of the dissolved concentration limit Analysis and Model Report: (i) a uniform, accepted thermodynamic database for EQ3/6, (ii) laboratory experiments to reduce conceptual uncertainties, especially the controlling solids for plutonium, (iii) better defined in-package chemistry, (iv) detailed analyses for most elements, as those done for neptunium, plutonium, and uranium, (v) more EQ3NR calculations to give better abstractions of radionuclide solubility limits, and (vi) evaluation of solubility limits for high-level waste glass waste package. Staff will review this additional information when it become available.

- The abstraction is consistent with technical bases, data and models in the spatial and temporal distribution of flow abstraction, the quantity and chemistry of water contacting waste package and waste forms abstraction, the degradation of engineered barriers abstraction, and the mechanical disruption of engineered barriers abstraction

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The colloid release model is well integrated into the total system performance assessment by drawing input from in-package chemical models and providing output to models of in-drift radionuclide concentrations. The results of the in-package chemistry calculations were not fully utilized in evaluating the solubility limits.

Staff review of DOE's approach with respect to this acceptance criterion is not complete. Staff will review the relevant Process Model Reports and Analysis and Model Reports to determine if DOE's abstraction of radionuclide release rates and solubility limits are consistent with the technical bases, data, and models relevant to the other integrated subissues.

#### **5.4.3.2.2.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 3**

Sufficient data on design features (including drip shield, backfill, waste packages, waste forms, other engineered barrier components, and thermal loading), geology, hydrology, and geochemistry of the unsaturated zone and drift environment (e.g., field, laboratory, and natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing the abstraction of radionuclide release rates and solubility limits used in the performance assessment. The data are also sufficient to assess the degree to which FEPs related to radionuclide release rates and solubility limits and which affect compliance with the postclosure performance objectives have been characterized and to determine whether the technical bases provided for exclusion of these FEPs are adequate. Where adequate data do not exist, other information sources such as expert elicitation have been appropriately incorporated. Specifically,

- DOE demonstrates that sufficient data have been collected on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, to establish initial and boundary conditions, including temporal and spatial variation, for conceptual models and simulations of THC coupled processes, including the potential for microbial processes that may affect radionuclide release for this abstraction

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Insufficient data are available on the reactivity of commercial spent nuclear fuel cladding to chloride solutions. Chloride is known to induce pitting corrosion (Brossia and Cragolino, 2000) if a potential above the repassivation potential is reached. Additional data are needed for DOE to demonstrate that, with the chloride concentrations expected inside waste packages, cladding degradation and consequent radionuclide release will not be enhanced.

Insufficient information has been provided on the potential effect of corrosion products on high-level waste glass degradation. The initial and boundary conditions used in calculating high-level waste glass degradation should account for the potential effect of corrosion products resulting from dissolution of waste package components.

- Where DOE uses data supplemented by models to support abstraction of solubility limits, the anticipated range of proportions and compositions of phases under the various physicochemical conditions expected are supported by experimental data (U.S. Nuclear Regulatory Commission, 1984). DOE adequately evaluates run product compositions for liquids and solids and determines radionuclide solubilities from under-saturation and over-saturation. If DOE does not use experimental reversal to determine the solubility limit, an approach from over-saturation is used

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Staff will review the Analysis and Model Report on Pure Phase Solubility Limits—LANL (CRWMS M&O, 2000p) when it becomes available in final form.

- DOE justifies the use of test results not specifically collected from the Yucca Mountain site for cladding performance, spent nuclear fuel, high-level waste glass, other radioactive waste forms intended for disposal, and other engineered barrier system components such as drip shield and backfill, for the environmental conditions expected to prevail
- Where sufficient data do not exist, the definition of parameters values and conceptual models is based on appropriate other sources such as expert elicitation conducted in accordance with NUREG-1563 (Kotra, et al., 1996)
- Sensitivity or uncertainty analyses are adequate to determine the possible need for additional data

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Staff review with respect to these acceptance criteria is not complete.

#### **5.4.3.2.2.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 3**

Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstraction of radionuclide release rates and solubility limits are consistent with site characterization, design data, laboratory experiments, field measurements, and natural analog data. Specifically,

- Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstractions of radionuclide release rates and solubility limits in the TSPA are technically defensible and reasonable based on data from the Yucca Mountain region, laboratory tests, and natural analogs
- DOE demonstrates the use of reasonable or conservative ranges of parameters or functional relations to determine effects of coupled THC processes such as secondary mineral formation and localized corrosion products on radionuclide release. These values are consistent with the initial and boundary conditions and the assumptions for the conceptual models and design concepts at the Yucca Mountain site. For example, estimations used in each abstraction are based on the thermal-loading strategy, including effects of ventilation; engineered barrier system design; and natural system masses and fluxes
- DOE includes experimental and conceptual uncertainty in determining the range in solubility limits

- Uncertainty is adequately represented in parameter development for conceptual models, process models, and alternative conceptual models considered in developing the abstraction of radionuclide release rates and solubility limits, either through sensitivity analyses or use of conservative limits
- DOE adequately considers the uncertainties in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THC coupled processes that affect radionuclide release

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

As stated in acceptance criterion 1, the results of the in-package chemistry calculations were not fully utilized in evaluating the solubility limits and the evaluation of dissolved concentration limits did not fully account for the variation in-package chemistry. The effect of chloride anions was not considered in the DOE analysis of localized corrosion and stress corrosion cracking of the commercial spent nuclear fuel cladding. The effect of corrosion products on high-level waste glass dissolution was not considered.

- DOE addresses and provides adequate technical bases for the exclusion of colloids in the abstraction

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

No technical basis was provided for the inclusion of colloidal effects only on plutonium and americium release and for the exclusion of americium from irreversible attachment modeling. The exclusion of a colloidal release component for other radionuclides needs to be supported.

- DOE uses an appropriate range of time-history of temperature, humidity, and dripping to constrain the probability for microbial effects, such as production of organic by-products that act as complexing ligands for actinides and microbial-enhanced dissolution of the high-level waste glass form

STATUS OF RESOLUTION: CLOSED

As noted in Section 5.4.3.1.2.2, DOE did not attempt to evaluate the potential effect of microbial activity on the chemical environment for radionuclide release. The process was screened out in DOE's FEPs analysis presented in the preliminary draft Waste Form Degradation Process Model Report (CRWMS M&O, 2000j) based on limited nutrient availability limiting biological activity, the absence of experimental evidence of enhanced microbial corrosion of zirconium metal or alloys, and the offsetting beneficial effect of microbial activity increasing colloid size and increasing filtration. The staff considers the technical basis for exclusion of microbial activity from DOE's evaluation of coupled THC effects on the chemical environment for radionuclide release sufficient.

#### **5.4.3.2.2.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 3**

Alternative modeling approaches consistent with available data and current scientific understanding are investigated and results and limitations are appropriately factored into the abstraction of radionuclide release rates and solubility limits. In its technical basis, the DOE has provided sufficient evidence that alternative conceptual models have been considered, that the models are consistent with available data (e.g., design features, field, laboratory, and natural analog) and current scientific understanding, and that the effect of these alternative conceptual models on total system performance has been evaluated. Specifically,

- Conceptual model uncertainties are adequately defined and documented and effects on conclusions regarding performance are properly assessed
- DOE investigates alternative modeling approaches consistent with available data and current scientific knowledge, and appropriately considers their results and limitations in developing the total system performance assessment abstraction
- In considering alternative conceptual models for radionuclide release rates and solubility limits, DOE uses appropriate models, tests, and analyses that are sensitive to the processes modeled for both natural and engineering systems
- DOE appropriately considers the effects of THC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products on radionuclide release

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

As pointed out earlier, the evaluation of dissolved concentration limits did not fully account for the variation of in-package chemistry. The effect of chloride anions was not considered in DOE's analysis of localized corrosion and stress corrosion cracking of the commercial spent nuclear fuel cladding. The effect of corrosion products on high-level waste glass dissolution was not considered.

#### **5.4.3.2.2.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 3**

Output from the abstraction of radionuclide release rates and solubility limits is justified through comparison with output from detailed process-level models and empirical observations (e.g., laboratory testing, field measurements, natural analogs).

- DOE verifies that the outputs of radionuclide release rates and solubility limits abstractions reasonably reproduce or bound the results of corresponding process-level models, empirical observations, or both

- DOE bases abstracted models for coupled THC effects on radionuclide release on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems
- DOE adopts well-documented procedures that have been accepted by the scientific community to construct and test the numerical models used to simulate coupled THC effects on radionuclide release

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE compared its abstracted commercial spent nuclear fuel dissolution model with unsaturated drip tests, batch tests, and a range of literature results and claim that the model and uncertainty range adequately accounted for, or overestimated, all dissolution rate data. DOE also states that a comparison of the phases produced in the unsaturated drip tests compare well with that of natural analogs.

Staff analyses of DOE's abstractions of coupled THC effects on radionuclide release rates and solubility limits based on these acceptance criteria will be updated based on a more thorough review of the relevant Analysis and Model Reports and Process Model Reports, once they are available.

#### **5.4.4 Subissue 4: Effects of Coupled Thermal-Hydrologic-Chemical Processes on Radionuclide Transport**

DOE's approach to assess the effects of coupled THC processes on the chemical environment for radionuclide release must meet the following generic criteria for each relevant integrated subissue: (i) integration, (ii) data and model justification, (iii) data uncertainty, (iv) model uncertainty, and (v) model support. Quality assurance is handled in a separate section of the Yucca Mountain Review Plan.

Two integrated subissues require input from the ENFE KTI within the scope of the ENFE subissue on radionuclide transport through engineered and natural barriers. The abstractions are (i) Radionuclide Release Rates and Solubility Limits and (ii) Radionuclide Transport in the Unsaturated Zone.

Radionuclide transport in the unsaturated zone is addressed in more detail in the Radionuclide Transport IRSR (U.S. Nuclear Regulatory Commission, 1999g). The description of radionuclide transport within a performance assessment framework requires input from the two integrated subissues relevant to this subissue (U.S. Nuclear Regulatory Commission, 2000a). The processes listed previously in Section 4.4 need to be considered in the evaluation of each abstraction. As part of this evaluation, we will use the acceptance criteria in Section 5.4.4.2 to review DOE's Process Model Reports and the supporting Analysis and Model Reports as they become available. This evaluation will also include a review of the relevant FEPs excluded from DOE's total system performance assessment and their technical basis for their exclusion.

The following Analysis and Model Reports have been identified in DOE's Analysis and Model Report/Process Model Report schedule as relevant to the ENFE subissue on radionuclide

transport through engineered and natural barriers: E0050—Analysis and Model Report on the Engineered Barrier System Radionuclide Transport Model, E0095—Analysis and Model Report on the Engineered Barrier System Radionuclide Transport Abstraction, E0120—Analysis and Model Report on the Description of the Near-Field Environment Basecase, and U0060—Analysis and Model Report on the Radionuclide Transport Model. The relevant Analysis and Model Reports on FEPs include U0170—FEPs for Unsaturated Zone Flow and Transport, N0080—FEPs for the Near-Field Environment, E0015—Engineered Barriers FEPs Degradation Modes Analysis (CRWMS M&O, 2000q), and E0110—Engineered Barrier System Degradation Modes and FEPs Abstraction. With the exception of preliminary draft E0015 and the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b), most of these Analysis and Model Reports and the relevant Process Model Reports were not available for review. Preliminary draft Analysis and Model Reports that are available and may be relevant to this subissue include: E0020—In-Drift Corrosion Products (CRWMS M&O, 2000r), E0000—Invert Diffusion Properties Model (CRWMS M&O, 2000s), and E0065—In-Drift Thermal-Hydrological-Chemical Model (CRWMS M&O, 2000f). Of these, preliminary draft E0065 (CRWMS M&O, 2000f) does not include chemical processes in Revision 00 and will not be discussed further.

As appropriate, status based on information contained in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) has been included where the approaches differ from that used in the TSPA-VA. It is important to note that the TSPA-SR methods and assumptions report does not contain details on how the different modeling approaches will be implemented. We will document our review using the acceptance criteria listed in Sections 5.4.4.2.1 and 5.4.4.2.2 as the Analysis and Model Reports and Process Model Reports become available for review.

#### **5.4.4.1 U.S. Department of Energy Approach**

##### **5.4.4.1.1 Features, Events, and Processes Screening**

The approach that DOE used for the Viability Assessment did not include a formal screening process for FEPs. As a result, many important design features, physical phenomena, and couplings were not evaluated in a performance assessment framework. Recently, DOE developed a more formal documentation of the FEPs identification and screening process for the TSPA-SR. The contents of the preliminary draft DOE FEP database (U.S. Department of Energy, 1999) have been reviewed and those related to the radionuclide release rates and solubility limits, and the radionuclide transport in the unsaturated zone, integrated subissues and the ENFE subissue on radionuclide transport in the near field have been identified (Pickett and Leslie, 1999; U.S. Nuclear Regulatory Commission, 2000a). Available updated screening analyses are provided in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) and the supporting preliminary draft Engineered Barrier System FEPs and Degradation Modes Analysis (CRWMS M&O, 2000q). The results of DOE's preliminary screening are presented in Table 5-7.

**Table 5-7. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 4 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on radionuclide release rates and solubility limits and radionuclide transport through the unsaturated zone**

<b>Features, Events, and Processes Number</b>	<b>Features, Events, and Processes Name</b>	<b>Preliminary DOE Screening</b>	<b>Radionuclide Release Rates and Solubility Limits</b>	<b>Radionuclide Transport in the Unsaturated Zone</b>
1.1.02.03.00	Undesirable materials left	Exclude	X	
1.1.07.00.00	Repository design	Include <sup>1</sup>	X	
1.1.08.00.00	Quality control	Include <sup>1</sup>	X	
1.1.12.01.00	Accidents and unplanned events during operation	Exclude	X	
1.1.13.00.00	Retrievability	Include <sup>2</sup>	X	
1.2.04.02.00	Igneous activity causes changes to rock properties	Include		X
1.2.08.00.00	Diagenesis	Exclude		X
2.1.04.02.00	Physical and chemical properties of backfill	Include	X	
2.1.04.03.00	Erosion or dissolution of backfill	Exclude	X	
2.1.04.05.00	Backfill evolution	Include	X	
2.1.04.08.00	Diffusion in backfill	Exclude	X	
2.1.04.09.00	Radionuclide transport through backfill	Exclude	X	
2.1.06.01.00	Degradation of cementitious materials in drift	Include <sup>2</sup>	X	X
2.1.06.02.00	Effects of rock reinforcement materials	Include <sup>2</sup>	X	
2.1.06.03.00	Degradation of the liner	Exclude	X	
2.1.06.04.00	Flow through the liner	Exclude	X	
2.1.06.05.00	Degradation of invert and pedestal	Include	X	

**Table 5-7. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 4 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on radionuclide release rates and solubility limits and radionuclide transport in the unsaturated zone (continued)**

<b>Features, Events, and Processes Number</b>	<b>Features, Events, and Processes Name</b>	<b>Preliminary DOE Screening</b>	<b>Radionuclide Release Rates and Solubility Limits</b>	<b>Radionuclide Transport in the Unsaturated Zone</b>
2.1.06.06.00	Effects and degradation of drip shield	Include <sup>2</sup>	X	
2.1.06.07.00	Effects at material interfaces	Exclude	X	
2.1.08.04.00	Condensation forms on backs of drifts	Include <sup>2</sup>	X	
2.1.08.05.00	Flow through invert	Include	X	
2.1.08.06.00	Wicking in waste and engineered barriers	Include <sup>2</sup>	X	
2.1.08.07.00	Pathways for unsaturated flow and transport in waste and engineered barriers	Include	X	
2.1.08.08.00	Induced hydrological changes in waste and engineered barriers	Include	X	
2.1.08.11.00	Resaturation of repository	Include <sup>2</sup>	X	
2.1.09.01.00	Properties of the potential carrier plume in waste and engineered barriers	Include	X	X
2.1.09.02.00	Interaction with corrosion products	Include	X	
2.1.09.04.00	Radionuclide solubility, solubility limits, and speciation in waste form and engineered barriers	Include <sup>3</sup>	X	
2.1.09.05.00	In-drift sorption	Exclude	X	
2.1.09.06.00	Reduction-oxidation potential in waste and engineered barriers	Include	X	
2.1.09.07.00	Reaction kinetics in waste and engineered barriers	Exclude	X	
2.1.09.08.00	Chemical gradients/enhanced diffusion in waste and engineered barriers	Exclude	X	
2.1.09.12.00	Rind (altered zone) formation in waste, engineered barriers, and adjacent rock	Include	X	X

**Table 5-7. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 4 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on radionuclide release rates and solubility limits and radionuclide transport in the unsaturated zone (continued)**

<b>Features, Events, and Processes Number</b>	<b>Features, Events, and Processes Name</b>	<b>Preliminary DOE Screening</b>	<b>Radionuclide Release Rates and Solubility Limits</b>	<b>Radionuclide Transport in the Unsaturated Zone</b>
2.1.09.13.00	Complexation by organics in waste and engineered barriers	Exclude	X	
2.1.09.14.00	Colloid formation in waste and engineered barriers	Include <sup>2</sup>	X	
2.1.09.16.00	Formation of pseudo-colloids (natural) in waste and engineered barriers	Include <sup>2</sup>	X	
2.1.09.17.00	Formation of pseudo-colloids (corrosion products) in waste and engineered barriers	Include <sup>2</sup>	X	
2.1.09.18.00	Microbial colloid transport in waste and engineered barriers	Exclude	X	
2.1.09.19.00	Colloid transport and sorption in waste and engineered barriers	Exclude	X	
2.1.09.20.00	Colloid filtration in waste and engineered barriers	Exclude	X	
2.1.09.21.00	Suspensions of particles larger than colloids	Exclude	X	X
2.1.10.01.00	Biological activity in waste and engineered barriers	Include	X	
2.1.11.01.00	Heat output/temperature in waste and engineered barriers	Include	X	
2.1.11.02.00	Nonuniform heat distribution/edge effects in repository	Include <sup>3</sup>	X	
2.1.11.04.00	Temperature effects/coupled processes in waste and engineered barriers	Include	X	
2.1.11.08.00	Thermal effects: chemical and microbiological changes in waste and engineered barriers	Include	X	
2.1.11.09.00	Thermal effects on liquid or two-phase fluid flow in waste and engineered barriers	Include	X	
2.1.11.10.00	Thermal effects on diffusion (Soret effect) in waste and engineered barriers	Exclude	X	
2.1.12.01.00	Gas generation	Exclude	X	

**Table 5-7. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 4 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on radionuclide release rates and solubility limits and radionuclide transport in the unsaturated zone (continued)**

<b>Features, Events, and Processes Number</b>	<b>Features, Events, and Processes Name</b>	<b>Preliminary DOE Screening</b>	<b>Radionuclide Release Rates and Solubility Limits</b>	<b>Radionuclide Transport in the Unsaturated Zone</b>
2.1.12.03.00	Gas generation (H <sub>2</sub> ) from metal corrosion	Exclude	X	
2.1.12.04.00	Gas generation (CO <sub>2</sub> , CH <sub>4</sub> , H <sub>2</sub> S) from microbial degradation	Exclude	X	
2.1.12.06.00	Gas transport in waste and engineered barriers	Exclude	X	
2.1.12.07.00	Radioactive gases in waste and engineered barriers	Include	X	
2.1.12.08.00	Gas explosions	Exclude	X	
2.1.13.01.00	Radiolysis	Include	X	
2.2.01.02.00	Thermal and other waste and engineered barriers-related changes in the adjacent host rock	Include		X
2.2.01.04.00	Elemental solubility in excavation disturbed zone	Include <sup>3</sup>	X	
2.2.01.05.00	Radionuclide transport in excavation disturbed zone	Include <sup>3</sup>	X	X
2.2.07.06.00	Episodic/pulse release from repository	Include	X	
2.2.08.01.00	Groundwater chemistry/composition in UZ and SZ	Include		X
2.2.08.02.00	Radionuclide transport occurs in a carrier plume in geosphere	Include		X
2.2.08.03.00	Geochemical interactions in geosphere (dissolution, precipitation, and weathering) and effects on radionuclide transport	Include		X
2.2.08.04.00	Redissolution of precipitates directs more corrosive fluids to containers	Include <sup>2</sup>	X	
2.2.08.05.00	Osmotic processes	Exclude		X
2.2.08.06.00	Complexation in geosphere	Include		X

**Table 5-7. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 4 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on radionuclide release rates and solubility limits and radionuclide transport in the unsaturated zone (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Radionuclide Release Rates and Solubility Limits	Radionuclide Transport in the Unsaturated Zone
2.2.08.07.00	Radionuclide solubility limits in the geosphere	Exclude <sup>3</sup>	X	X
2.2.08.08.00	Matrix diffusion in geosphere	Include		X
2.2.08.09.00	Sorption in UZ and SZ	Include		X
2.2.08.10.00	Colloidal transport in geosphere	Include		X
2.2.09.01.00	Microbial activity in geosphere	Include		X
2.2.10.01.00	Repository-induced thermal effects in geosphere	Include		X
2.2.10.06.00	Thermo-chemical alteration (solubility, speciation, phase changes, and precipitation/dissolution)	Include		X
2.2.10.07.00	Thermo-chemical alteration of the Calico Hills unit	Include		X
2.2.10.09.00	Thermo-chemical alteration of the Topopah Spring basal vitrophyre	Include		X
2.2.11.01.00	Naturally occurring gases in geosphere	Exclude		X
2.2.11.03.00	Gas transport in geosphere	Exclude		X
3.1.01.01.00	Radioactive decay and ingrowth	Include <sup>3</sup>	X	
See Footnotes and abbreviations on next page				

**Table 5-7. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 4 of the Evolution of the Near-Field Environment Key Technical Issue and to the integrated subissues on radionuclide release rates and solubility limits and radionuclide transport in the unsaturated zone (continued)**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening	Radionuclide Release Rates and Solubility Limits	Radionuclide Transport in the Unsaturated Zone
SZ = saturated zone FEP = feature, event, and process	UZ = unsaturated zone Process Model Report = process and model report	EBS = engineered barrier system		
<p><sup>1</sup>Deviations from design and operational error are excluded.</p> <p><sup>2</sup>Included FEPs identified in Table 1-3 of preliminary draft EBS Degradation, Flow, and Transport Process Model Report (Civilian Radioactive Waste Management System, Management and Operating Contractor, 2000b), but not in audit review of Pickett and Leslie (1999).</p> <p><sup>3</sup>Identified in audit review of Pickett and Leslie (1999) but not in Table 1-3 of the preliminary draft EBS Degradation, Flow, and Transport Process Model Report (Civilian Radioactive Waste Management System, Management and Operating Contractor, 2000b). Screening of Swift, et al. (1999) used.</p>				

#### 5.4.4.1.2 Radionuclide Release and Solubility Limits

The engineered barrier system transport conceptual model outlined in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) is similar to that used for radionuclide transport through the concrete invert in TSPA-VA, with 1-D diffusion and/or advective transport. The approach is one of bounding analyses rather than detailed modeling. The preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) provides additional details on the modeling approach and the results of sensitivity analyses, but the technical bases for much of the TSPA-SR is included in the supporting Analysis and Model Reports that are not yet available. In addition, much of the modeling analysis is based on the Enhanced Design Alternative-II design that included backfill (CRWMS M&O, 1999b) rather than the recently adopted TSPA-SR reference design that removed backfill (CRWMS M&O, 2000b).

Depending on the scenario used for wetting the waste form, it is likely that the TSPA-SR will use a bounding analysis for transport out of the waste package, assuming no sorption on waste package corrosion products (CRWMS M&O, 1999a). Additional bounding assumptions are (i) after waste package failure, the waste form is covered by a thin liquid film (CRWMS M&O, 2000b), regardless of the in-package conditions; (ii) advective release from the waste package is proportional to the total axial length of open patches relative to the total axial length of the waste package; (iii) for diffusive transport through the invert, the diffusivity coefficient is based on the self-diffusivity of water and corrected for porosity and saturation using Archie's Law; and (iv) no sorption onto the invert materials (i.e.,  $K_d = 0$ ). In the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), it is mentioned that if in-package transport has a large effect on performance, then a simple model will be developed, but it is not clear in the methods and assumptions report how the impact on performance will be evaluated or how in-package transport will be modeled.

For the TSPA-SR, DOE proposes to move the zero concentration boundary into the host rock where the engineered barrier system transport model is coupled to the unsaturated zone flow and transport model (CRWMS M&O, 1999a). Low solubility radionuclides are presumed to precipitate at the engineered barrier system boundary and are removed slowly as a function of seepage flux. The reference design proposed for the TSPA-SR (CRWMS M&O, 1999b) calls for a titanium drip shield that will have significant effects on the flow of water through the engineered barriers.

In Revision 3 of DOE's Repository Safety Strategy (CRWMS M&O, 2000a), DOE identifies 27 key postclosure performance attributes of the repository system to be described in the TSPA-SR. This list includes seven principal factors DOE believes are most important to postclosure performance. Of DOE's principal factors, retardation of radionuclide migration in the unsaturated zone is relevant to ENFE Subissue 4. Of the other remaining 20 factors, four are relevant to ENFE Subissue 4: (i) in-package radionuclide transport, (ii) transport through the drift invert, (iii) colloid-facilitated transport in the unsaturated zone, and (iv) coupled processes-effects on unsaturated zone transport.

In the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), DOE describes the approach that will be used to develop the model abstractions for radionuclide transport through the engineered and natural barrier systems. For the principal factor and other factors related to

ENFE Subissue 4, DOE's engineered barrier system radionuclide transport model (E0050) will address the effects on performance of radionuclide transport through the engineered barriers (CRWMS M&O, 1999a).

In the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a), input to the engineered barrier system radionuclide transport model is provided by four different abstraction models: (i) Water Distribution and Removal Abstraction, (ii) In-Drift Geochemical Environment Model, (iii) Waste Package Degradation Abstraction Model, and (iv) Waste Form Degradation.

In the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b), only the water distribution and removal model (E0090) is identified as providing input to the engineered barrier system radionuclide transport model (E0050).

The preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report includes sensitivity analyses for a number of parameters affecting ENFE Subissue 4, including sorption properties of engineered barrier system materials (CRWMS M&O, 2000b). In addition to sorption properties for the invert, these parameters include a sensitivity analysis of the influence of colloid properties on radionuclide transport. Additional analyses are provided to evaluate the potential effects of a drainage system on transport through the engineered barriers.

Output from the engineered barrier system radionuclide transport model will be used as input to the unsaturated zone transport model. Coupling between the engineered barrier system and unsaturated zone transport models is to be more explicit in the TSPA-SR analysis than was the case in TSPA-VA (CRWMS M&O, 1999a, Section 3.1.2.5).

#### **5.4.4.1.3 Radionuclide Transport Through the Unsaturated Zone**

The following Analysis and Model Reports identified in DOE's Analysis and Model Report/Process Model Report schedule as relevant to radionuclide transport through the unsaturated zone were not available for review prior to May 15, 2000: E0120—Analysis and Model Report on the Description of the Near-Field Environment Basecase, U0060—Analysis and Model Report on the Radionuclide Transport Model, U0170—FEPs for Unsaturated Zone Flow and Transport, N0080—FEPs for the Near-Field Environment.

The Integrated Site Model Process Model Report (CRWMS M&O, 1999c) provides input to the TSPA-SR and contains the TSPA representations of geology, rock properties, and mineralogy distributions. For the rock properties model (RPM3.1), borehole petrophysical logs were converted to porosity estimates and volumetric water content. Geostatistical estimation (kriging) and simulation (Monte Carlo) techniques were used to evaluate uncertainty in property values. Porosity data were used as a surrogate to derive other values such as bulk density and saturated hydraulic conductivity using a regression ( $R^2$ ) correlation coefficient. Kriging is used to produce a model of hydrous phase mineral alterations that constrain the distribution of saturated hydraulic conductivity. The mineralogy model (MM 3.0) provides distributions of 10 mineral groups including the zeolitic minerals contained in the Calico Hills unit, which may be an important natural barrier. The integrated site model provides input into the preliminary draft unsaturated zone flow and transport model (CRWMS M&O, 2000e).

The preliminary draft integrated unsaturated zone flow and transport model for TSPA-SR comprises four major components: (i) unsaturated zone flow, (ii) drift seepage, (iii) drift-scale coupled THC processes, and (iv) unsaturated zone transport (CRWMS M&O, 2000e). The latter two components are relevant to radionuclide transport in the unsaturated zone.

Results of the drift-scale THC process model suggest that impacts of THC processes on fracture and matrix properties (such as those affecting radionuclide transport) would be negligible (see Section 5.2.3 of CRWMS M&O, 2000e).

Results of the unsaturated zone transport process model suggest that matrix diffusion and sorption can be effective retardation mechanisms; vitric Calico Hills layers provide retardation via flow through the matrix; Pu-239 daughter products are important to transport; and colloidal transport is sensitive to filtration (Section 5.2.4 in CRWMS M&O, 2000e). Results from particle tracking simulations indicate that the dual-porosity method is more conservative than the dual-permeability method where longer radionuclide transport times are predicted due to matrix diffusion processes.

#### **5.4.4.2 U.S. Nuclear Regulatory Commission Staff Evaluation**

##### **5.4.4.2.1 Integrated Subissue on Radionuclide Release Rates and Solubility Limits**

There are five generic acceptance criteria. Each generic acceptance criterion is followed by specific criteria. Only those specific criteria related to ENFE Subissue 4 are provided here. DOE's approach to abstract coupled THC effects on radionuclide transport through engineered and natural barriers in a total system performance assessment for the proposed repository at Yucca Mountain will be acceptable provided that each acceptance criterion is met.

##### **5.4.4.2.1.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 4**

Important design features, physical phenomena and couplings, and consistent and appropriate assumptions have been identified and described sufficiently for incorporation into the abstraction of radionuclide release rates and solubility limits in the total system performance assessment, and the technical bases are provided. The features, phenomena and couplings, and assumptions used to abstract release of radionuclides from waste forms inside the waste package and the transport and release of radionuclides from the engineered barrier system have been provided. The abstraction is consistent with the identification and description of those aspects of radionuclide release rates and solubility limits that are important to waste isolation. The abstraction is also consistent with the technical bases for these descriptions of barriers important to waste isolation. Specifically,

- DOE reasonably accounts for the range of environmental conditions, including water composition and water and vapor movement, expected inside breached waste packages and in the engineered barrier environment surrounding the waste package (e.g., temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release rates and solubility limits)

- DOE provides adequate technical bases for exclusion of any potentially important couplings and FEP
- The abstraction is consistent with technical bases, data and models in the spatial and temporal distribution of flow abstraction, the quantity and chemistry of water contacting waste package and waste forms abstraction, the degradation of engineered barriers abstraction, and the mechanical disruption of engineered barriers abstraction

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Using the distribution of entries from the preliminary draft U.S. Department of Energy (1999) database provided in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) and the audit review of Pickett and Leslie (1999), 48 primary FEPs are identified as being related to both the radionuclide release rates and solubility limits integrated subissue and this ENFE subissue. The screening status of these FEPs is updated in Table 5-6 to include the results of DOE's screening analysis summarized in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b). Of these 48, 23 are excluded by DOE's preliminary screening process, mostly on the basis of low consequence. For two design-related FEPs deviations from design are excluded. In addition, the FEP on retrievability was excluded in the preliminary draft database, but is included in the draft process model report. Review and evaluation of the technical bases for excluding these 26 FEPs, provided below, will focus on the relevant Analysis and Model Reports, including U0170—FEPs for Unsaturated Zone Flow and Transport, N0080—FEPs for the Near-Field Environment, E0015—Engineered Barriers FEPs Degradation Modes Analysis (CRWMS M&O, 2000q), and, E0110—Engineered Barriers Degradation Modes and FEPs Abstraction. As of May 15, 2000, only E0015 was available for review. A review of preliminary draft E0015 (CRWMS M&O, 2000q) indicates that beyond the preliminary screening criteria of Swift, et al. (1999), the basis for FEPs inclusion is incorporated by reference to process model Analysis and Model Reports that are not yet available. The screening status has been updated (CRWMS M&O, 2000b,q) and will be further revised as appropriate when Analysis and Model Reports become available.

We determined that the technical basis provided was sufficient to support the screening argument, and the staff agrees with, DOE's preliminary exclusion of five FEPs. The remaining 21 FEPs either should not be excluded based on the current preliminary technical basis, or an insufficient technical basis has been provided to evaluate the screening, chosen by DOE. The bases for these determinations are discussed in the following paragraphs.

Staff judges that DOE exclusion of the following five FEPs is currently acceptable.

2.1.04.03.00—Erosion or dissolution of backfill; 2.1.04.08.00—Diffusion in backfill;  
2.1.04.09.00—Radionuclide transport through backfill

Because of recent changes in the reference design (CRWMS M&O, 2000b), the TSPA-SR reference design no longer includes backfill. It should be noted that 2.1.04.09.00 was included in CRWMS M&O (2000q), suggesting inconsistency between the Process Model Report and the supporting documents. Nevertheless, based on the information available at this time, the

exclusion of these FEPs is acceptable. Should future design changes call for backfill, the screening of these FEPs will need to be reexamined.

2.1.06.03.00—Degradation of the liner; 2.1.06.04.00—Flow through the liner

DOE excludes these FEPs because the TSPA-SR reference design eliminates the concrete liner that was included in the TSPA-VA design (CRWMS M&O, 2000b). For this reason, DOE does not need to include this design feature in its TSPA abstraction, and the exclusion of these FEPs is acceptable. Should future design changes include a liner, however, DOE would need to provide data on the liner material and laboratory or analog data that can be used to evaluate its effect on repository performance.

Staff either does not support DOE's preliminary screening, concluded that there is an inadequate technical basis to support exclusion, or determined there are inconsistencies between various documents concerning the exclusion of the following 21 FEPs.

1.1.02.03.00 Undesirable materials left; 1.1.12.01.00—Accidents and unplanned events during operation

These FEPS involve operational and preclosure aspects of the proposed repository and were excluded in DOE's preliminary screening on the basis of low consequence. As explained in the screening argument for FEP 1.1.03.01.00,

The TSPA is based on an assumption that the repository will be constructed, operated, and closed according to design. Deviations from design during the operational period are the subject of an extensive quality control program, and are outside the scope of the long-term performance assessment. Significant deviations that are detected during the operational period will be corrected and, therefore, are excluded from the TSPA on the basis of low probability.

Operational errors cannot be excluded without quality control procedures. Quality control procedures and operational procedures under normal and accident conditions are reviewed in specific sections of the Yucca Mountain Review Plan. Also, DOE references these FEPs with regard to the operational period; preclosure aspects of the proposed repository are reviewed under the preclosure section of the Yucca Mountain Review Plan and not in the postclosure model abstraction section that includes this integrated subissue. Nevertheless, these preclosure activities may impact post-closure repository performance. An inadequate technical basis for screening these FEPs has been provided. In addition, the screening of some of the FEPs is inconsistent with post-closure performance assessment calculations. For instance, the effects of the FEP on undesirable materials is required for the analysis of potential microbial effects on performance (see section 4.1; CRWMS M&O, 1998a). DOE needs to provide a more robust technical basis for exclusion of these FEPs, perhaps by citing project documents where these assumptions and postulated actions (i.e., actions to correct mistakes) are codified. Also DOE needs to screen these FEPs so that the screening is consistent with actual post-closure performance assessment calculations.

1.1.07.00.00—Repository design; 1.1.08.00.00—Quality Control; 1.1.13.00.00—Retrievability

These three FEPs are included in the DOE screening, but deviations from design and operational error are excluded on the basis of low consequence. As explained in the screening argument for FEP 1.1.03.01.00,

The TSPA is based on an assumption that the repository will be constructed, operated, and closed according to design. Deviations from design during the operational period are the subject of an extensive quality control program. Significant deviations that are detected during the operational period will be corrected and, therefore, are excluded from the TSPA on the basis of low probability.

The excluded portions of these FEPs involve operational and preclosure aspects of the proposed repository. Also, as design features, DOE indicates that these FEPs are included, but are not a part of the TSPA-SR (CRWMS M&O, 2000b). Nevertheless, these pre-closure activities may impact post-closure repository performance. An inadequate technical basis for screening these FEPs has been provided. DOE needs to provide a more robust technical basis for exclusion of these FEPs, perhaps by citing project documents where these assumptions and postulated actions (i.e., actions to correct mistakes) are codified.

2.1.06.07.00—Effects at material interfaces

DOE excludes this FEP in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) on the basis of low consequence. This FEP was included in preliminary draft CRWMS M&O (2000q), however, suggesting inconsistency between the Process Model Report and the supporting documents. No technical basis is provided in the preliminary draft Process Model Report. The exclusion of this FEP will remain open until DOE provides a clear technical basis and the supporting calculations.

2.1.09.05.00—In-drift sorption

This FEP was included in preliminary draft CRWMS M&O (2000q), suggesting inconsistency between the preliminary draft Process Model Report and the supporting documents. DOE provides sensitivity analyses using uniformly distributed  $K_d$  values of 0 to 1, 1 to 5, 5 to 10, 10 to 50, and 50 to 100 milliliters per gram, indicating the strong dependence of calculated transport through the invert on sorption coefficients (CRWMS M&O, 2000b). In the summary provided in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report, however, this FEP is excluded on the basis of low consequence. Because of the uncertainty in radionuclide sorption on the invert material (crushed tuff), DOE assumes no retardation in the invert beneath the waste package. Given the short travel path through the invert (0.5 to 0.7 meter; CRWMS M&O, 2000b) relative to the entire flow path to the critical group (~20 kilometers), this assumption is not likely to underestimate dose to the critical group. The technical basis for exclusion of this FEP is acceptable. Should the DOE change the assumption of no retardation in the invert material, a technical basis will be necessary for the sorption parameters used in the TSPA abstraction. DOE should resolve the inconsistency in the Process Model Report and supporting document concerning the screening of this FEP.

#### 2.1.09.07.00—Reaction kinetics in waste and engineered barriers

DOE's screening focuses on the effect of redox kinetics on  $K_o$ s, and excludes them on the basis of low consequence. Exclusion of this FEP is not acceptable and remains open. The effect of kinetics should be more generally applied to chemical processes affecting transport through the engineered and natural barriers, and should be included. DOE should provide a technical basis for exclusion of this FEP or include its effects on performance in the performance assessment.

#### 2.1.09.08.00—Chemical gradients/enhanced diffusion in waste and engineered barriers

DOE's summary in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) excludes this FEP on the basis of low consequence. This FEP was included in preliminary draft CRWMS M&O (2000q), however, suggesting inconsistency between the Process Model Report and the supporting documents. Because no technical basis is provided in the preliminary draft Process Model Report and because the analysis in Revision 3 of the DOE Repository Safety Strategy (CRWMS M&O, 2000a) suggests that dose is sensitive to diffusion through the invert, the exclusion of this FEP remains open. DOE should provide a technical basis in the supporting Analysis and Model Report E0110—Engineered Barriers Degradation Modes and FEPs Abstraction. The technical basis for exclusion of this FEP will be reevaluated when the E0110 and the supporting Analysis and Model Reports are available. DOE should resolve the inconsistency in the Process Model Report and supporting document concerning the screening of this FEP.

#### 2.1.09.13.00—Complexation by organics in waste and engineered barriers

This FEP is excluded by DOE on the basis of low consequence (CRWMS M&O, 2000b). Review of the supporting preliminary draft Analysis and Model Report (CRWMS M&O, 2000q) indicates that an important reason for exclusion was low organic concentration at Yucca Mountain. While this may be true under ambient conditions, no technical basis is provided. In addition, the availability of nutrients and the effect of coupled THC processes in the engineered barrier system may affect the microbial population. DOE may be using a conservative assumption of instantaneous transport from the waste package, but because no technical basis is provided on the effect of organics on transport through the engineered barriers, the exclusion of this FEP remains open. DOE needs to provide a technical basis and the calculations necessary to support a screening on low consequence.

#### 2.1.09.18.00—Microbial colloid transport in the waste and engineered barriers and

#### 2.1.09.19.00—Colloid transport and sorption in the waste and engineered barriers

DOE excluded these FEPs on the basis of low consequence. These FEPs were included in preliminary draft CRWMS M&O (2000q), however, suggesting inconsistency between the preliminary draft Process Model Report and the supporting documents. Because DOE includes colloid formation FEPs in its screening analysis, particle transport through the engineered barrier system into the unsaturated zone is plausible. Exclusion of these FEPs may be acceptable, but will remain open until DOE provides a more complete technical basis and calculations to support an assumption of low consequence. DOE should resolve the inconsistency in the Process Model Report and supporting document concerning the screening of this FEP.

#### 2.1.09.20.00—Colloid filtration in the waste and engineered barriers

Colloid filtration is a process that would limit colloid transport through the engineered barriers. Exclusion of this FEP is, therefore, a bounding case and would not tend to underestimate dose to the critical group. The technical basis for exclusion is appropriate, although it is perhaps at odds with the exclusion of the other FEPs related to colloid transport. DOE should be consistent in screening related FEPs or demonstrate that the inconsistency does not lead to calculations of repository performance that are not bounded.

#### 2.1.09.21.00—Suspensions of particles larger than colloids

In the preliminary draft FEP database (U.S. Department of Energy, 1999), the exclusion of particles larger than colloids was based on the assumption that although particles may be transported through fractures in the unsaturated zone, low groundwater velocities through the saturated zone would lead to particle settling. Qualitative comparison to colloid size distributions from wells in the Yucca Mountain region is also used as part of the exclusion rationale. The screening basis in the preliminary draft Process Model Report is given as low consequence (CRWMS M&O, 2000b, table 1-3), while the screening basis in the supporting FEPs Analysis and Model Report is low probability (CRWMS M&O, 2000q, Attachment III), suggesting inconsistency in the screening analysis. Because DOE includes colloid formation FEPs in its screening analysis, and because of the large amounts of iron particles that may be introduced in the engineered barriers, particle transport through the engineered barrier system into the unsaturated zone is plausible. Exclusion of this FEP may be acceptable, but will remain open until DOE provides a more complete technical basis and the calculations to support an assumption of low consequence.

#### 2.1.11.10.00—Thermal effects on diffusion (Soret effect) in waste and engineered barriers

The contribution of the Soret effect to radionuclide transport through the engineered and natural barriers is likely to be small in comparison to other chemical processes such as sorption/desorption and has been excluded by DOE. This is an appropriate screening argument, particularly given the relatively heavy radioelements and the lower thermal gradient attributed to the TSPA-SR reference design. However, DOE gives a low consequence screening basis in the preliminary draft Process Model Report (CRWMS M&O, 2000b) while listing a low probability basis in the supporting preliminary draft FEPs Analysis and Model Report (CRWMS M&O, 2000q). The exclusion of this FEP is likely to be acceptable, but DOE needs to reconcile the apparent discrepancy between the Process Model Report and the supporting Analysis and Model Report.

#### 2.1.12.01.00—Gas generation, 2.1.12.03.00—Gas generation (H<sub>2</sub>) from metal corrosion, and 2.1.12.04.00—Gas generation (CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>S) from microbial degradation

DOE excludes these three FEPs largely because the repository will be located in the unsaturated zone and is well connected to the surface, minimizing the potential for postclosure gas buildup (U.S. Department of Energy, 1999). In addition, FEP 2.1.12.04.00 is included in the preliminary draft FEPs Analysis and Model Report (CRWMS M&O, 2000q), indicating inconsistency between the preliminary draft Process Model Report (CRWMS M&O, 2000b) and the supporting document. The exclusion of these FEPs may be acceptable if DOE provides a technical basis for this screening argument, particularly because DOE acknowledges that the

possibility exists for temporary confinement of generated gases, and these might still have a chemical impact on radionuclide transport through the engineered and natural barrier systems. DOE will also need to reconcile the apparent discrepancy between the preliminary draft Process Model Report and the supporting preliminary draft Analysis and Model Report. The exclusion of these FEPs remains open until DOE provides a clear technical basis.

#### 2.1.12.06.00—Gas transport in waste and engineered barriers

This FEP was excluded in the preliminary draft FEP database (U.S. Department of Energy, 1999) on the basis that radioactive gases are not a significant component of the future radionuclide inventory. There is no technical basis provided in the preliminary draft FEP database, and there is a presupposition that the doses resulting from this transport are low. Earlier TSPA analyses by both the DOE (TRW Environmental Safety Systems, Inc., 1995) and NRC (Wescott, et al., 1995) and a report prepared by the National Academy of Sciences (National Research Council, 1995) all suggest that the calculated dose from gaseous radionuclide transport is low. The most recent revision of the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) closed out gaseous transport of  $^{14}\text{CO}_2$  as an open item.

The primary description for this FEP states, “Gas in the waste and engineered barrier system could affect the long-term performance of the disposal system. Radionuclides may be transported as gases or in gases, gas bubbles may affect flow paths, and two-phase flow conditions may be important.” It is not clear from this description whether this FEP is intended to address all gas species or just radioactive gas species. Staff were unable to identify any other FEP that explicitly addresses gas transport, but note the importance of two-phase flow in DOE TH and THC models. This FEP will remain open until DOE’s primary description is clarified. Staff recommends that DOE identify a FEP that explicitly includes transport of non-radioactive gases.

#### 2.1.12.08.00—Gas explosions

DOE excludes this FEP largely because the repository will be located in the unsaturated zone and is well connected to the surface, minimizing the potential for postclosure gas buildup, and due to the lack of an ignition source. Based on the information available at this time, this exclusion may be acceptable if DOE provides a technical basis for the screening argument, particularly because the possibility exists for temporary confinement of gas. The exclusion of this FEP remains open until DOE provides a clear technical basis.

#### 2.2.08.07.00—Radionuclide solubility limits in the geosphere

DOE’s screening argument assumes that radionuclide solubility limits in the geosphere may be different from those in the engineered barriers. Solubility limits in the geosphere will be determined by interaction between the contaminant plume and the host rock. This argument makes valid points, but does not provide an explicit case for exclusion. At this time, the exclusion of this FEP is uncertain and remains open until DOE provides a clear technical basis.

The preliminary draft FEP database, developed by the DOE (U.S. Department of Energy, 1999; Swift, et al., 1999) and updated in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b), provide a more formalized documentation of the FEPs screening and identification process that will be used for the DOE

Site Recommendation. Pickett and Leslie (1999) provides an early audit review of DOE's treatment of FEPs. Many of the excluded FEPs were screened by DOE on the basis of low consequence, but the technical basis is not available for review at this time. These excluded FEPs will remain open until DOE provides a clear technical basis and the supporting calculations. Future DOE analysis of FEPs should meet the acceptance criteria for scenario analysis in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) and in the Yucca Mountain Review Plan. This FEPs analysis will be an important action for DOE to take to facilitate resolution on this subissue.

In DOE performance assessment abstractions, the FEPs are not dealt with on a one-by-one basis. For simplification, the included FEPs for this integrated subissue have been grouped into a number of categories based on the relationship among the FEPs descriptions provided by Swift, et al. (1999): (i) invert, (ii) hydrologic processes, (iii) chemical processes, (iv) colloid formation, (v) biological activity, (vi) thermal processes, (vii) radiolysis, and (viii) radioactive decay. An additional 11 included FEPs were identified as important to transport through the engineered barrier system (CRWMS M&O, 2000b). For completeness, these are also provided in Table 5-7. The evaluation of the acceptability of the DOE abstractions is presented using these categorizations.

Due to late changes (mid-January 2000) to the TSPA-SR reference design, most of the analyses presented in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) include backfill and do not conform to the current no backfill design. In future revisions of the Analysis and Model Reports and Process Model Reports, DOE will either need to provide additional analyses to evaluate the effects of no backfill on the engineered barrier system or provide a technical basis for the applicability of the earlier calculations. Possible effects include temperature, flow rates, and water chemistry.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available prior to May 15, 2000, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.1.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 4**

Sufficient data on design features (including drip shield, backfill, waste packages, waste forms, other engineered barrier components, and thermal loading), geology, hydrology, and geochemistry of the unsaturated zone and drift environment (e.g., field, laboratory, and natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing the abstraction of radionuclide release rates and solubility limits used in the performance assessment. The data are also sufficient to assess the degree to which FEPs related to radionuclide release rates and solubility limits and which affect compliance with the postclosure performance objectives have been characterized and to determine whether the technical bases provided for exclusion of these FEPs are adequate. Where adequate data do

not exist, other information sources such as expert elicitation have been appropriately incorporated. Specifically,

- DOE demonstrates that sufficient data have been collected on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, to establish initial and boundary conditions, including temporal and spatial variation, for conceptual models and simulations of THC coupled processes, including the potential for microbial processes that may affect radionuclide release for this abstraction
- Sensitivity or uncertainty analyses are adequate to determine the possible need for additional data

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The TSPA-SR reference design calls for the drip shield to be installed after waste emplacement is complete. DOE provided a limited sensitivity analyses of the effects of a drip shield in the design selection report (CRWMS M&O, 1999b) and in the Repository Safety Strategy, Revision 3 (CRWMS M&O, 2000a). There is little, if any, laboratory or field information available on the performance of the titanium drip shield. Also at the current time the status of the data sufficiency for this engineered barrier component is uncertain. DOE will need to provide additional data on design features and the technical bases for the description and implementation of these features in the total system performance assessment abstraction.

The current TSPA-SR reference design does not include a cement liner for the repository tunnels. For this reason, DOE does not need to include this design feature in its abstraction, and no additional information is necessary. Should future design changes include a liner, however, DOE would need to provide data on the liner material and laboratory or analog data that can be used to evaluate its effect on repository performance.

Although transport through the invert is most sensitive to sorption, DOE will assume no retardation (i.e.,  $K_d = 0$  for all radionuclides) in the crushed tuff invert (CRWMS M&O, 2000b). The transport path through the invert is short (0.5 to 0.7 meters) relative to the entire path length to the critical group (~20 kilometers), and it is unlikely that this assumption will underestimate dose. This approach is acceptable.

There is general agreement that data to support assumptions in the models of the in-drift geochemistry used in the TSPA-VA are lacking (U.S. Nuclear Regulatory Commission, 1998g, 1999f; Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999). Concrete has been largely eliminated from the DOE design. Although this limitation will reduce some of the uncertainty in the effects of engineered materials on radionuclide transport through the engineered barriers, several FEPs related to interaction with corrosion products, the effects of reinforcement materials, and degradation of cementitious materials are to be included in the TSPA-SR model abstraction (CRWMS M&O, 2000b). For example, interaction of water with the titanium drip shield called for in the TSPA-SR reference will affect in-drift geochemistry and, therefore, radionuclide transport through the near-field environment. The modified water case should be part of the basecase in DOE's performance assessment analyses, and, regardless of

the design, sufficient data are necessary to establish initial and boundary conditions for conceptual models.

DOE included several FEPs related to colloid formation. Limited experimental or site characterization information exists on colloid concentrations in the near field, and only the empirical relationship to ionic strength is used for particle concentration. The particle concentration data used in developing the colloid distributions are based on data from saturated zone wells in tuffs and alluvium, and the relationship to in-drift colloids where introduced materials will have a strong effect is uncertain. Some inconsistency occurs in the DOE model for the formation of iron pseudocolloids. DOE notes data that indicate the irreversibility of plutonium sorption on iron colloids (CRWMS M&O, 2000b, Sections 3.1.2.6.1 and 3.1.2.6.6) and corrosion products (CRWMS M&O, 2000r, Section 6.5.2.4). The treatment of colloids in the abstraction, however, calls for only reversible sorption onto corrosion products and natural groundwater colloids (CRWMS M&O, 2000b, Section 3.2.1.1.5). Because there is a likely to be a significant amount of corrosion products (CRWMS M&O, 2000r), and because irreversible sorption on colloids potentially will have a stronger impact on performance, DOE will need to provide additional support on colloid formation.

As discussed previously, the preliminary draft FEPs screening analysis in CRWMS M&O (2000b) indicates that although colloid formation is included, FEPs related to colloid transport will be screened from the TSPA-SR on the basis of low consequence. This is not a bounding assumption as indicated by DOE (CRWMS M&O, 2000b, Section 3.1.2.6), and data supporting the exclusion of colloid transport are not yet available. For example, in contrast to the TSPA-VA where only colloid transport of plutonium was considered, the colloid model will be applied to both plutonium and americium (CRWMS M&O, 2000b) in the TSPA-SR. Data are not yet available, however, to evaluate the effect of colloids on transport through the near field. Diffusion of colloids through the invert is neglected in the TSPA-SR conceptual model (CRWMS M&O, 2000b, Section 3.1.2.6.5), but the possibility of advective transport is not described in detail. Given the short flow path through the invert and the low diffusion coefficients for particles, this omission is unlikely to underestimate the effect on dose. However, given the potential for large enhancements of radionuclide solubility due to colloid formation (CRWMS M&O, 2000b, Section 3.1.2.6.4) and the possibility of enhanced transport velocity (CRWMS M&O, 2000b, Section 3.2.1.2), a significant amount of uncertainty remains in DOE's treatment of colloid transport.

The in-drift microbial communities model has been judged by others to be at a preliminary stage (Whipple, et al., 1999). The model was only used to provide first-order limits on potential microbial effects (CRWMS M&O, 1998d). The model approach required by the acceptance criterion from previous ENFE IRSR (U.S. Nuclear Regulatory Commission, 1999f) was used by DOE. The staff has no further concerns regarding the nutrient and energy inventory calculations approach used by DOE to determine the potential for microbial activity that could impact the transport of radionuclides.

DOE elected to exclude microbial colloid transport on the basis of a relatively large size and filtration. This exclusion is inconsistent with the exclusion of colloid filtration in the engineered barrier system (CRWMS M&O, 2000b) discussed in the previous acceptance criterion. Filtration in the unsaturated zone may be possible, but no data are provided on either the size of the microbes likely in the near-field environment or on the pore apertures in the engineered barriers. Neglecting the potential impact of microbial processes on radionuclide transport will

require justification. If DOE plans to screen on the basis of a small microbial population (low consequence), DOE should use the time-history of temperature, humidity, and dripping (CRWMS M&O, 1997b), or other appropriate approach, to constrain the microbial effects, such as production of organic by-products that act as complexing ligands for actinides (McKinley, West, and Grogan, 1985). If colloid filtration is to be invoked, DOE should also provide information on pore apertures and likely microbial colloid sizes to support size exclusion and reconcile the FEPs screening analysis.

Radiolysis to lower solution pH is likely to be most effective in a thin water film adjacent to the waste package. Further from the waste package, the effects on radionuclide transport will be reduced by dilution of the low pH solution. Because of the short transport paths involved, DOE may be able to use sensitivity analyses and bounding calculations to support omitting radiolysis from its conceptual models of radionuclide transport through the engineered barriers.

DOE has not clearly documented the basis for the parameter probability distribution functions. This may be because data are not otherwise readily available. It thus appears as if informal expert elicitation is used to define the parameter ranges. Development of parameter distributions through elicitation is acceptable, but DOE should conduct and document these expert elicitations in accordance with the guidance in Kotra, et al. (1996) or other acceptable methods.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available prior to May 15, 2000, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.1.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 4**

Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstraction of radionuclide release rates and solubility limits are consistent with site characterization, design data, laboratory experiments, field measurements, and natural analog data. Specifically,

- Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstractions of radionuclide release rates and solubility limits in the total system performance assessment are technically defensible and reasonable based on data from the Yucca Mountain region, laboratory tests, and natural analogs
- DOE demonstrates the use of reasonable or conservative ranges of parameters or functional relations to determine effects of coupled THC processes such as secondary mineral formation and localized corrosion products on radionuclide release. These values are consistent with the initial and boundary conditions and the assumptions for the conceptual models and design concepts at the Yucca Mountain site. For example, estimations used in each abstraction are based on the thermal-loading strategy, including effects of ventilation; engineered barriers design, including drift liner, backfill, and drip-shield; and natural system masses and fluxes

- Uncertainty is adequately represented in parameter development for conceptual models, process models, and alternative conceptual models considered in developing the abstraction of radionuclide release rates and solubility limits, either through sensitivity analyses or use of conservative limits
- The parameters used to describe flow through and out of the engineered barrier system sufficiently bound the effects of backfill, if used, excavation-induced changes, and thermally induced mechanical changes that affect flow

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The data used to support models and parameter values for the abstraction of the effects of the TSPA-SR reference design features such as the titanium drip shield are only summarized in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b). Detailed analyses are not available, and the resolution status cannot be determined with any certainty. Review of the relevant Analysis and Model Reports will be necessary to evaluate the acceptability of approaches used by DOE.

Although DOE's approach used in the TSPA-VA to model transport through the engineered barrier system is appropriate, the parameter values used in the TSPA-VA are not acceptable as presented for the TSPA-SR reference design. The TSPA-VA design was based on a concrete invert; this design has been superseded by a crushed tuff invert with steel ground supports in the TSPA-SR (CRWMS M&O, 2000b). DOE will need to continue to evaluate these parameters, especially in consideration of the new conditions related to the selection of the TSPA-SR reference design. Because of the short transport path lengths through the invert, DOE may be able to use a combination of sensitivity analyses and bounding calculations to demonstrate the effect of transport through the invert on repository performance.

In the TSPA-VA, DOE completed sensitivity analyses for transport through a concrete invert using the 5th and 95th percentile  $K_d$  values with all other parameters set at basecase expected values. Their results suggest no influence on dose out to 10,000 years, and little influence on total dose at longer times. Even the extreme case of zero retardation ( $K_d = 0$ ) resulted only in a factor of two increase in dose starting at about 40,000 years, with the effect decreasing at times greater than about 250,000 years. One-dimensional transport analyses at the fifth percentile summarized in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) indicate that  $50 < K_d < 100$  milliliters per gram can delay breakthrough by about 500 years relative to the nonretarded case, but does not carry the calculations out to dose. Due to uncertainty in defining  $K_d$  values for the invert materials and describing flow through the invert for thousands of years, DOE has chosen for the TSPA-Site Recommendation to neglect sorption on invert materials ( $K_d = 0$ ) for all radionuclides. This is a bounding assumption, and is unlikely to underestimate dose to the critical group. DOE's sensitivity analyses indicate that 1-D transport through the invert is least sensitive to molecular diffusion. Supporting data are limited, DOE indicates a temperature effect, and there is a strong dependence of effective diffusivity on saturation in the invert (CRWMS M&O, 2000s).

In the TSPA-VA analyses, some unrealistic model results from the thermal-chemical evaluations indicated that a review of the parameters used in the process-level models is

warranted. Andradite (a garnet mineral), diopside, and wollastonite are typically elevated temperature skarn minerals. With reference to the appearance of garnet and tremolite in simulations, it is noted that “their presence in the simulations is likely due to limitations of the current thermodynamic data or to incompletely adequate solid-solution models, especially for clay minerals and zeolites” (Hardin, 1998, p. 5-49). In addition, only local equilibrium models were developed for in-drift water chemistry evaluations. DOE continues to identify these data as a source of uncertainty in the in-drift geochemical environment model for the TSPA-SR (CRWMS M&O, 1999a). The challenges associated with compiling an internally consistent and validated thermodynamic and kinetic database for relevant mineral phases are recognized, however, this leads to uncertainty with the acceptability of DOE’s model results.

Experimental and field evidence suggest that plutonium attachment to colloids can vary from relatively fast and reversible to effectively irreversible (CRWMS M&O, 1998d, Section 6.4.2.3). The DOE notes, however, that the parameters necessary to describe the processes controlling plutonium attachment are not all available, and uncertainty exists in the parameters used in the TSPA-VA abstracted models. For example, the plutonium  $K_d$ s used in modeling colloid transport are based on experimental data reported by Lu, et al. (1998) and ranges approximately seven orders of magnitude (100 to  $10^9$  milliliters per gram). By including the higher sorption coefficients in its colloid transport model, DOE has taken a more conservative approach. However, DOE did not demonstrate in TSPA-VA that the conditions of the experiments of Lu, et al. (1998) are relevant to expected conditions at Yucca Mountain. By including lower values in their range, DOE potentially 'dilutes' the conservative nature of the colloid transport calculation. DOE should either continue to conduct sensitivity analyses to demonstrate that higher values don't have a worse effect on overall performance, or provide a stronger technical basis that they have indeed captured the upper likely range of  $K_d$ s for colloids. Also, DOE will need to provide similar technical basis or sensitivity calculations for other radionuclides considered in the colloid transport of other radionuclides (Am, Th) that may be considered in TSPA-SR.

In the TSPA-VA, DOE did not use reasonable or conservative range of parameters to determine the effects of coupled THC processes on the transport of radionuclides. Model parameters, particularly  $K_C$  and the irreversibly sorbed fraction in the colloid transport abstraction, were not well constrained and will require additional experiments or other analyses to establish reasonable bounding limits. In addition, the modeling approach was limited to plutonium in the TSPA-VA. DOE indicated this approach will be applied to other strongly sorbing radionuclides that may be subject to colloid transport, and information is presented for americium colloid transport (CRWMS M&O, 2000b). This approach is appropriate, but given that current analysis indicates that colloid transport FEPs will be screened out, DOE should clearly demonstrate that colloidal transport of these and other radionuclides is not important to repository performance.

In the TSPA-VA, DOE did not consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on radionuclide transport. This is not acceptable for this acceptance criterion. DOE should consider contributions to data uncertainty from both temporal and spatial variability, especially given the recent changes to exclude backfill from the TSPA-SR reference design. For example, most of the analyses presented in preliminary draft CRWMS M&O (2000b) seem to be based on the presence of backfill. These new design features and their contributions to uncertainty should be implemented in DOE’s abstractions of the effects of coupled THC processes on radionuclide transport. The TSPA-SR

methods and assumptions report (CRWMS M&O, 1999a) indicates that spatial and temporal variations in results from the drift-scale heater test are now considered in the process-level modeling studies, which may move towards meeting this acceptance criterion.

DOE should demonstrate that reasonable or conservative ranges of parameters were used to determine effects of coupled THC processes on radionuclide transport in the abstracted models. This demonstration may be achieved using experimental, site, and natural analog data to aid verification of abstracted models and data and parameters used in the process-level models. DOE should provide rationales for parameter values, assumed ranges, and probability distributions. If DOE uses bounding assumptions, it should be technically defensible and reasonably account for uncertainties. DOE should collect and use data on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, to establish initial and boundary conditions for conceptual models and simulations of THC coupled processes that are consistent with the Enhanced Design Alternative-II design features.

We concur with DOE's use of sensitivity analyses to evaluate the relative importance of diffusion in the invert, sorption in the unsaturated zone to repository performance, and bounding the potentially nonconservative interactions. This determination is especially true given the differences in the engineered barrier system contained in the TSPA-SR reference design and the recent decision to exclude backfill. DOE should use sensitivity and uncertainty analyses of their abstracted model to determine if additional new data are needed to support the abstracted model.

DOE has not clearly documented the basis for the parameter probability distribution functions. This may be because data are not otherwise readily available. It thus appears as if informal expert elicitation is used to define the parameter ranges. Development of parameter distributions through elicitation is acceptable, but DOE should conduct and document these expert elicitations in accordance with the guidance in Kotra, et al. (1996) or other acceptable methods.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available for review prior to May 15, 2000, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.1.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 4**

Alternative modeling approaches consistent with available data and current scientific understanding are investigated and results and limitations are appropriately factored into the abstraction of radionuclide release rates and solubility limits. In its technical basis, DOE has provided sufficient evidence that alternative conceptual models have been considered, that the models are consistent with available data (e.g., design features, field, laboratory, and natural analog) and current scientific understanding, and that the effect of these alternative conceptual models on the total system performance has been evaluated. Specifically,

- In considering alternative conceptual models for radionuclide release rates and solubility limits, DOE uses appropriate models, tests, and analyses that are sensitive to the processes modeled for both natural and engineering systems
- DOE appropriately considers the effects of THC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products on radionuclide release

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The proposed approach to modeling radionuclide transport through the engineered barrier system is consistent with models used to simulate transport through the natural barrier system. DOE recognized in the TSPA-VA, however, that some modeling assumptions may not be conservative. A similar modeling approach is proposed for the TSPA-SR (CRWMS M&O, 1999a, 2000b), with a zero-concentration boundary condition imposed in the host rock rather than at the edge of the engineered barriers. The conceptual model will need to be modified further to reflect the TSPA-SR reference design and refined to demonstrate the effects of changing chemical conditions on transport. As necessary, updated calculations will need to be provided to reflect the removal of backfill from the reference design.

The staff agrees that the short transport length through the invert relative to the total transport length suggests that retardation in the invert is likely to have little effect on overall system performance in terms of the magnitude of the dose. However, if diffusion is the dominant mechanism, the delay caused by transport through the invert is potentially a substantial portion of a 10,000-year regulatory period. Thus, the model will need to provide technical support for omitting transport through the invert in the TSPA-SR and be consistent with expected engineered materials, particularly given the recent change in invert materials for the reference design.

The approach developed by DOE is appropriate for abstracting the effects of colloids on radionuclide transport through the engineered barriers. DOE recommended a more detailed approach (CRWMS M&O, 1998d, Section 8.5.2.5) be used for the in-drift colloid model, but this approach requires additional laboratory, field, and site-scale observations to develop the necessary model parameters, and it does not appear that it will be included in the TSPA-SR Revision 00 (CRWMS M&O, 1999a). Additional potential refinements noted by DOE for the TSPA-SR include extending the colloid transport abstraction to other strongly sorbing radionuclides (e.g., americium) and adding filtration capability. These enhancements of the colloid transport model are appropriate, but DOE acknowledges that these improvements are not likely to be completed in time for the TSPA-SR (CRWMS M&O, 1999a). DOE will need to implement these enhancements before the staff can evaluate the acceptability of these conceptual models. This is particularly important if the staff is to evaluate the DOE's exclusion of these FEPs based on low consequence.

In the Near-Field/Altered-Zone Models Report (Hardin, 1998), numerous explicit statements of THC model inadequacy are noted. For example, in the description of the limitations of J-13 Well water as a starting composition, and in noting effects of condensation, water-rock interactions, nonisothermal chemistry, and engineered materials, Hardin states, "the models described here represent only a part of the whole-system processes, but they are

representative of the kind of thermodynamic modeling that can be applied to additional parts of the system" (Hardin, 1998, p. 5-48). We concur with this observation. Present models are recognized to be inadequate, particularly because that the most recent changes to the reference design (e.g., no backfill) are apparently not reflected in the supporting technical basis.

The major limitation recognized by both NRC and DOE staffs is that although conceptually the complexity of coupled THC processes is recognized, many aspects of these processes are greatly simplified or omitted in the TSPA-VA analyses. Effects of the coupled THC processes on radionuclide transport were omitted. Neglect of these processes contributes to model uncertainty without proper support and justification is not acceptable. DOE should use appropriate models, tests, and analyses sensitive to the THC couplings being considered for both natural and engineering systems. Information provided in the TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) and in the preliminary draft Engineered Barrier System Degradation, Flow, and Transport Process Model Report (CRWMS M&O, 2000b) indicate that DOE is attempting to address this acceptance criterion for Site Recommendation, but that many of the THC processes will continue to be uncoupled.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available at the time of writing this revision of the ENFE IRSR, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.1.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 4**

Output from the abstraction of radionuclide release rates and solubility limits is justified through comparison with output from detailed process-level models and empirical observations (e.g., laboratory testing, field measurements, natural analogs). Specifically,

- DOE verifies that the outputs of radionuclide release rates and solubility limits abstractions reasonably reproduce or bound the results of corresponding process-level models, empirical observations, or both
- DOE bases abstracted models for coupled THC effects on radionuclide release on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems
- DOE adopts well-documented procedures that have been accepted by the scientific community to construct and test the numerical models used to simulate coupled THC effects on radionuclide release

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

The general lack of data to support and test critical assumptions in mathematical models used in the TSPA-VA has been noted by others (U.S. Nuclear Regulatory Commission, 1998g, 1999f; Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999). The TSPA-SR methods and assumptions report (CRWMS M&O, 1999a) indicates that natural analog

information will be used to support the performance assessment analyses. The staff will need to review how DOE uses analog data to verify the TSPA-SR conceptual models and model results. DOE should conduct additional exercises using experimental, site, and natural analog data to support their models. Additional discussion of the model validation process is provided in Eisenberg, et al. (1999). DOE should use the results from both their laboratory and field heater test program to test their abstracted models for consistency with observations. Models used should produce results reasonably representative of the systems modeled.

DOE provided a preliminary inventory of introduced materials and discussion of conceptual models of the effects of corrosion products on engineered barrier system transport (CRWMS M&O, 2000r). This preliminary draft Analysis and Model Report (E0020—In-Drift Corrosion Products) is limited to general discussion and does not provide either a technical basis, bounding analysis, or sensitivity analyses of the effects on repository performance. Detailed models and experimental study of uranium sorption on waste package corrosion products (iron oxides) and cementitious materials have been described in the near-field models report of Hardin (1998), but the results were not included in the TSPA-VA analysis. It is anticipated that these results, and additional experimental and modeling data, will be developed as part of the preparation for the TSPA-SR, particularly with the new materials introduced in the TSPA-SR reference design, should DOE take any credit for retardation within the engineered barrier system. The proposed work described in Hardin (1998) includes site-specific studies and laboratory and modeling analyses for other waste package materials, concretes, and cementitious materials. Given the TSPA-SR reference design changes, the tests on concrete and cementitious materials should be de-emphasized.

Data from DOE field heater tests and natural analog studies were not available for the TSPA-VA, and information is not yet available on the specific details of how the information will be incorporated in the TSPA-SR. DOE made a qualitative comparison to experimental and field observations in developing the TSPA-VA models and providing parameter estimates for near-field transport (e.g., CRWMS M&O, 1998d, Section 6.4.2.3). Limited site-specific colloid data has been used to constrain particle size distribution functions (CRWMS M&O, 2000b, Section 3.1.2.6.2), but the applicability of these saturated zone data to in-drift conditions is still uncertain. The resulting abstracted models were preliminary in nature, however, and, in general, have not been verified quantitatively against natural analog or experimental data. Parameter values have been determined for a limited range in chemical conditions. Thus, model results have not been tested against laboratory experiments or field data, and model support for the effects of coupled THC processes on the transport of radionuclides remains uncertain. As part of DOE's effort to verify their abstracted model of the effects of coupled THC effects on radionuclide transport, DOE should use well-documented procedures to construct and test the numerical models. DOE should also use the results from their laboratory experiments, field heater test program, and natural analog program to test the abstracted models for consistency with observations. DOE should conduct exercises using experimental, site, and natural analog data to aid verification of their models. Models used should produce results reasonably representative of the systems modeled.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available at the time of writing this revision of the ENFE IRSR, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.2 Integrated Subissue on Radionuclide Transport Through the Unsaturated Zone**

This integrated subissue will be considered in more detail as part of the Radionuclide Transport IRSR. DOE's approach to abstract coupled THC effects on radionuclide transport through the unsaturated zone in a total system performance assessment for the proposed repository at Yucca Mountain will be acceptable provided that each acceptance criterion is met.

##### **5.4.4.2.2.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 4**

Important design features, physical phenomena and couplings, and consistent and appropriate assumptions have been identified and described sufficiently for incorporation into the abstraction of radionuclide transport in the unsaturated zone and other related abstractions in the total system performance assessment, and the technical bases are provided. The abstraction identifies and describes aspects of radionuclide transport in the unsaturated zone that are important to waste isolation and includes the technical bases for these descriptions. Specifically,

- The DOE description is adequate and the conditions and assumptions in the abstraction of radionuclide transport in the unsaturated zone are readily identified and consistent with the body of data presented in the description
- DOE provides adequate technical bases for exclusion of any potentially important couplings and FEPs

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Using the distribution of entries from the preliminary draft DOE FEP database (U.S. Department of Energy, 1999) provided in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 2000a) and the audit review of Pickett and Leslie (1999), 24 primary FEPs are identified as being related to both the radionuclide transport through the unsaturated zone integrated subissue and this ENFE subissue. Of these 24, six are excluded by the preliminary DOE screening process. Of the six excluded FEPs, two overlap with the radionuclide release rates and solubility limits integrated subissue and were discussed previously in Section 5.4.4.1.1. Review and evaluation of the technical bases for excluding the remaining four FEPs will focus on the relevant Analysis and Model Reports, including U0170—FEPs for Unsaturated Zone Flow and Transport and N0080—FEPs for the Near-Field Environment. As of May 15, 2000, these two Analysis and Model Reports are not available for review.

A preliminary evaluation (Pickett and Leslie, 1999) of the resolution status of the six excluded FEPs (not including the two discussed previously in Section 5.4.4.1.1), based on the preliminary DOE screening provided in the DOE FEP database (U.S. Department of Energy, 1999), is provided here. It is important to note that the resolution status will be updated when the Analysis and Model Reports become available.

#### 1.2.08.00.00—Diagenesis

The screening argument used by DOE to exclude this FEP is based on the definition of diagenesis as natural diagenesis and does not include those processes related to the emplacement of waste in the proposed repository. As long as this distinction is maintained, the technical basis for exclusion is appropriate, and the staff has no additional questions at this time.

#### 2.1.09.21.00—Suspensions of particles larger than colloids (see Section 5.4.4.2.1.1)

#### 2.2.08.05.00—Osmotic processes

Like the Soret effect, the contributions of osmotic processes to radionuclide transport through the engineered and natural barriers are likely to be small in comparison to other chemical processes such as sorption/desorption and have been excluded by DOE. This is an appropriate screening argument, particularly in the absence of a credible semipermeable membrane, and the staff has no further questions on the technical basis for exclusion of this FEP.

#### 2.2.08.07.00—Radionuclide solubility limits in the geosphere (see Section 5.4.4.2.1.1)

#### 2.2.11.01.00—Naturally-occurring gases in the geosphere and 2.2.11.03.00—Gas transport in the geosphere

Similar to other FEPs related to gas generation (see Section 5.4.4.1.1), DOE's screening arguments for these two FEPs expect naturally-occurring gases to escape to the atmosphere through a well connected unsaturated zone, preventing buildup in the repository. Chemical changes due to gas generation are expected to be small relative to the controls exerted by other components of the repository system. Exclusion of these FEPs may be acceptable if DOE provides additional technical bases to demonstrate that potential chemical effects are limited and will not adversely affect performance. The second FEP (Gas transport in the geosphere) also includes potential radionuclide transport. As discussed in Section 5.4.4.1.1, preliminary TSPA analyses (TRW Environmental Safety Systems, Inc., 1995; Wescott, et al., 1995) and other analyses (National Research Council, 1995; U.S. Nuclear Regulatory Commission, 2000a) has indicated that gaseous radionuclide transport is not a significant contributor to dose. Exclusion of this aspect of the FEP on gas transport in the geosphere is appropriate, and the staff has no further questions. DOE should provide a technical basis for omitting these processes or include these effects in the performance analyses via abstracted models or bounding assumptions.

An important step in resolving items associated with FEPs is the FEP database with its more formal documentation of the technical basis for exclusion FEPs from the DOE total system performance assessment analyses (U.S. Department of Energy, 1999; Swift, et al., 1999). DOE should continue to develop its FEPs analysis methodology to meet the acceptance criteria for scenario analysis in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 2000a) and the Yucca Mountain Review Plan. This FEPs analysis will be an important action for DOE to take to facilitate resolution on this subissue.

As noted previously in Section 5.4.4.1.1, DOE's performance assessment abstractions do not deal with FEPs on a one-by-one basis. Similar to the approach used in Section 5.4.4.1.1, the included FEPs for this integrated subissue have been grouped into a number of categories based on the relationship among the FEPs descriptions provided by Swift, et al. (1999): (i) igneous effects on rock properties, (ii) degradation of engineered barrier system components, (iii) properties/geochemistry of groundwater, (iv) geochemical transport processes, (v) colloid transport, (vi) biological activity, and (vii) thermal and thermo-chemical effects on engineered barrier system and geosphere. The evaluation of the acceptability of the DOE abstractions is presented using this categorization.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available at the time of writing this revision of the ENFE IRSR, these acceptance criteria remain open. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.2.2 Acceptance Criterion 2—Data and Model Justification for Evolution of the Near-Field Environment Subissue 4**

Sufficient data on the geology, hydrology, and geochemistry of the unsaturated zone (e.g., field, laboratory, and natural analog data) are available to adequately define relevant parameters and conceptual models necessary for developing the abstraction of radionuclide transport in the unsaturated zone in the total system performance assessment. The data are also sufficient to assess the degree to which FEPs related to radionuclide transport in the unsaturated zone have been characterized and to determine whether the technical bases provided for exclusion of these FEPs are adequate. Specifically,

- DOE demonstrates that sufficient data have been collected on the characteristics of the natural system and engineered system, as it affects the natural system, to establish initial and boundary conditions, and temporal and spatial variations, for the abstraction of radionuclide transport, including the potential effects from coupled THC processes in the unsaturated zone
- The data on the geology, hydrology, microbial ecology, and geochemistry of the unsaturated zone, including the influence of structural features and stratigraphy, used in the abstraction are based on techniques that may include laboratory experiments, site-specific field measurements, natural analog research, and process-level modeling studies
- Sensitivity or uncertainty analyses used to support DOE's abstraction are adequate to determine the possible need for additional data. If additional data are needed, DOE has identified specific plans to acquire necessary information

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Two of the FEPs on degradation of engineered barrier system materials are focused on cementitious materials in drift and the drift liner. These features of the TSPA-VA design are either greatly reduced (cementitious materials) or not present in the TSPA-SR reference design

(liner). Some cementitious materials such as grouted rock bolts and concrete-lined ventilation shafts and tunnels are likely to be used in constructing the repository. The quantity of materials and effects of these materials on radionuclide transport in the TSPA-SR reference design will need to be addressed by DOE.

Radionuclide transport through the unsaturated zone may be affected by interaction of water with other features of the engineered barrier system such as the titanium drip shield, ground support structures, and steel inverts. Regardless of the design, the DOE will need to provide sufficient data to establish initial and boundary conditions for conceptual models.

In the TSPA-VA, DOE's near-field geochemical environment model focused on the central part of the repository. This focus was rationalized on the basis that the magnitude of coupled processes effects is likely to be largest there (CRWMS M&O, 1998d; Section 4.4.2.1). Neglecting processes at the edges of the emplacement zone may not be conservative. DOE should justify neglecting processes at the margin of the emplacement area.

No experimental or site characterization information exists on colloid concentrations in the near field, however, and only the empirical relationship to ionic strength is used for particle concentration. In addition, the colloid model is only applied to plutonium transport. For these reasons, a significant amount of uncertainty remains in DOE's treatment of colloid transport. The TSPA-SR methods and assumptions report indicates enhancements such as filtration and colloid transport of americium are being considered for the colloid transport model, but DOE will need to provide additional data to provide the technical support for these data.

Neglecting the potential impact of microbial processes on radionuclide transport will require justification. DOE should use the time-history of temperature, humidity, and dripping (CRWMS M&O, 1997b), or other appropriate approach, to constrain the probability for microbial effects such as production of organic by-products that act as complexing ligands for actinides (McKinley, West, and Grogan, 1985). While DOE (U.S. Department of Energy, 2000; Comment 20) indicated that microbial colloids could be included in transport models, the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e, Section 1.2.3, Assumption 3) assumes the quantities of organic matter and microbes are sufficiently small that they are unlikely to be of importance. DOE needs to provide a consistent explanation for the importance of microbes and colloids relative to effects on radionuclide transport.

If DOE chooses to rely on the Calico Hills or Prow Pass vitric units containing zeolite minerals beneath the repository to retard radionuclide transport, then DOE must demonstrate that the effects of THC processes will not adversely affect the properties of the natural barrier. For example, potentially adverse effects from temperature (above 70 °C) or changes in solution chemistry (e.g., increasing sodium) that can alter highly sorptive zeolites to nonsorptive analcime, as described by Los Alamos National Laboratory (Chipera and Bish, 1997; Bish, et al., 1996), must be shown by the DOE to be negligible, or the degraded capacity of the barrier should be included into the total system performance assessment.

Bish, et al. (1996) write, "the breakdown of clinoptilolite at Yucca Mountain due to destabilization at higher temperature and/or different fluid compositions may have a significant impact on the performance of a high-level radioactive waste repository at Yucca Mountain. The potential effects...include increased porosity and permeability, significant release of water

contained in the clinoptilolite structure, and loss of an important sorption barrier to radionuclide migration, especially neptunium.”

The above statement of concern is inconsistent with the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e, Section 3.12.5). The draft report states that, “at the top of the Calico Hills stratigraphic unit, the predicted maximum temperature rises to 70 to 75 °C after 2,000 to 7,000 years. This suggests that the temperature-induced property changes will be insignificant in the Calico Hills because such changes are expected to occur if Calico Hills zeolite temperatures remain above 90 °C for prolonged periods of time.” Furthermore, the presentation at the joint NRC and DOE Key Technical Issue meeting (Barr, 2000) presented the temperature profiles at 1,000 years (before the maximum temperatures reach the top of the Calico Hills unit). However, even at 1,000 years it appears that 70 °C is reached at this unit, particularly in the northern part of the repository block.

If DOE plans to consider natural sorptive barriers such as zeolites, further explanation and data are needed to resolve conflicting information. In responding to Revision 2 of the ENFE IRSR DOE states, “DOE believes that THC-induced changes in the repository flow system are minor (U.S. Department of Energy, 2000, p. 5).” However, this statement does not address many factors such as mineral alteration due to temperature effects or changes in the hydrologic and geochemical system affecting radionuclide transport. Staff agrees with the statement in the preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e, Section 1.2.3) that qualitative arguments require quantitative verification to assume that thermo-hydrologic flow processes would have a minimal effect on radionuclide transport.

Furthermore, it is not clear how the modeling using the code TOUGHREACT has been applied to the Calico Hills unit. Further justification and data are needed to resolve the issue regarding THC effects on sorptive zeolites.

The characteristics of the sorptive units determined outside of the repository footprint, such as at Busted Butte, must be demonstrated to be analogous to those beneath the planned repository horizon. The Integrated Site Model Process Model Report (CRWMS M&O, 1999c) mineralogy model indicates sorptive zeolite abundance is more variable to the north and south of the planned repository in the Calico Hills non welded unit than the variability existing at Busted Butte. While extensive testing has been performed at Busted Butte, no information is available or currently planned to determine characteristics of the units beneath the repository. Therefore, should DOE take credit for processes at Busted Butte as an analog to processes affecting radionuclide retardation in the Calico Hills unit beneath the repository horizon, then further justification is needed. The degree of characterization of sorptive units beneath the repository, or the amount of analogous information used to support the characteristics assumed for units beneath the repository, should be commensurate with the degree of credit taken for the barrier in DOE’s total system performance assessment.

DOE has not clearly documented the basis for the parameter probability distribution functions. This may be because data are not otherwise readily available. It thus appears as if informal expert elicitation is used to define the parameter ranges. Development of parameter distributions through elicitation is acceptable, but DOE should conduct and document these expert elicitations in accordance with the guidance in Kotra, et al. (1996) or other acceptable methods.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available prior to May 15, 2000, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.2.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 4**

Parameter values, assumed ranges, probability distributions, and bounding assumptions used in the abstraction of radionuclide transport in the unsaturated zone are consistent with site characterization data, are technically defensible, and reasonably account for uncertainties and variabilities. The technical bases for the parameter values used in the abstraction are provided. Specifically,

- For those radionuclides whose transport in the unsaturated zone is important to performance, DOE has adequately assessed the effects of coupled THC processes on the fraction of flow traveling through both fractures and matrix in the unsaturated zone, and the flow velocities through both fractures and matrix in the unsaturated zone
- If the DOE safety case relies on matrix diffusion in the unsaturated zone, rock fracture-matrix and solute diffusion parameters include both temporal and spatial variations associated with coupled THC processes that may affect performance
- Uncertainty is adequately represented, including both temporal and spatial variations in parameter development for conceptual models, process-level models, and ACMs considered in developing the abstraction of radionuclide transport including the potential effects from coupled THC processes in the unsaturated zone, either through sensitivity analyses or conservative limits

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

DOE should use reasonable or conservative ranges of parameters to determine effects of coupled THC processes on radionuclide transport in the unsaturated zone. DOE should provide information from exercises using experimental, site, and natural analog data to support abstracted models and data and parameters used in the process-level models. Parameters used in process-level models should produce results reasonably representative of these systems. DOE should use technically defensible parameter values, assumed ranges, and probability distributions. If DOE uses bounding assumptions, the bounding assumptions should be technically defensible and reasonably account for uncertainties.

DOE should consider contributions to data uncertainty from both temporal and spatial variability. One approach adopted by DOE to reduce this uncertainty is the lower temperatures proposed for the TSPA-SR reference design. The contributions of these design changes to uncertainty should be implemented in their abstracted models of the effects of coupled THC processes on the transport of radionuclides. Presentations at the In-drift Geochemical Environment Workshop and information provided in the TSPA-SR methods and assumptions

report (CRWMS M&O, 1999a) indicate that spatial and temporal variations in results from the drift-scale heater test are now being considered in the process-level modeling studies.

The preliminary draft Unsaturated Zone Flow and Transport Process Model Report (CRWMS M&O, 2000e, Section 3.11) suggests matrix diffusion and sorption in the rock matrix may be important retardation processes. While sorption in fractures is not considered viable in the unsaturated zone due to lack of characterization of fracture properties such as geometry, matrix diffusion at the fracture-matrix interface either may be considered (Section 3.11.2.5) or may not be considered (Section 3.11.9.1) viable by DOE to be effective in retarding radionuclides.

If DOE chooses to take credit for matrix diffusion in the unsaturated zone, then THC coupled processes affecting matrix diffusion processes must be evaluated (such as reduced fracture surface area by mineral coatings or nonstagnant flow and transport at the fracture-matrix boundary) and shown not to affect matrix diffusion and sorption processes.

Experimental and field evidence suggest that plutonium attachment to colloids can vary from relatively fast and reversible to effectively irreversible (CRWMS M&O, 1998d, Section 6.4.2.3; Lu, et al., 1998). DOE notes, however, that the parameters necessary to describe the processes controlling plutonium attachment are not all available. Uncertainty is also in the parameters used in the TSPA-VA abstracted models. For example, the plutonium  $K_d$ s used in modeling colloid transport are based on experimental data reported by Lu, et al. (1998) and approximately range seven orders of magnitude (100 to  $10^9$  milliliters per gram). By including the higher sorption coefficients in its colloid transport model, DOE has taken a more conservative approach. However, DOE did not demonstrate in TSPA-VA that the conditions of the experiments of Lu, et al. (1998) are relevant to expected to conditions at Yucca Mountain. By including lower values in their range, DOE potentially 'dilutes' the conservative nature of the colloid transport calculation. DOE should either continue to conduct sensitivity analyses to demonstrate that higher values don't have a worse effect on overall performance, or provide a stronger technical basis that they have indeed captured the upper likely range of  $K_d$ s for colloids. Also, DOE will need to provide similar technical basis or sensitivity calculations for other radionuclides considered in the colloid transport of other radionuclides (Am, Th) that may be considered in TSPA-SR. Due to lack of data, DOE's TSPA-VA abstraction neglects colloid filtration. The connection among the different parts of DOE's colloid transport abstraction (engineered barrier system, unsaturated zone, saturated zone) is not clear. For these reasons, the acceptability of DOE's approach to colloid transport in the TSPA-SR is uncertain. DOE's Analysis and Model Reports and Process Model Reports will need to provide a clear link between the different DOE transport models and establish the relationship between parameter value ranges and conditions at Yucca Mountain.

Some mineral phases, anticipated on the basis of alteration in the vicinity of Yucca Mountain (e.g., smectite, illite, and zeolites), are absent from the models of coupled thermal-chemical effects. These limitations are generally recognized by DOE. The lower temperatures of the TSPA-SR reference design may contribute to reduced uncertainty for mineral precipitation and dissolution. Regardless of the repository design, however, DOE should consider uncertainties in the characteristics of the natural and engineered materials in their abstraction of the effects of coupled THC processes on the transport of radionuclides. Both NRC and DOE continue to recognize the challenges associated with compilation of an internally consistent and validated thermodynamic and kinetic database for relevant mineral phases (CRWMS M&O, 1999a). DOE attempted to reduce some of this uncertainty by reducing the maximum repository temperature

in the TSPA-SR (CRWMS M&O, 1999b, 2000b). This approach is appropriate, but temperatures are still above ambient, and the temperature effects on geochemical processes affecting radionuclide transport are still uncertain. DOE will need to provide technical bases for the consistency of the thermodynamic and kinetic data used in its TSPA abstraction, or demonstrate through sensitivity analyses that the uncertainty and its impact on repository performance can be bounded.

DOE has not clearly documented the basis for the parameter probability distribution functions. This may be because data are not otherwise readily available. It thus appears as if informal expert elicitation is used to define the parameter ranges. Development of parameter distributions through elicitation is acceptable, but DOE should conduct and document these expert elicitations in accordance with the guidance in Kotra, et al. (1996) or other acceptable methods.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available at the time of writing this revision of the ENFE IRSR, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.2.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 4**

Alternative modeling approaches consistent with available data and current scientific understanding are investigated and results and limitations are appropriately factored into the abstraction of radionuclide transport in the unsaturated zone. DOE has provided sufficient evidence that alternative conceptual models have been considered, that the models are consistent with available data (e.g., field, laboratory, and natural analog) and current scientific understanding, and that the effect of these alternative conceptual models on the total system performance has been evaluated. Specifically,

- DOE investigates alternative modeling approaches that are consistent with available data and current scientific knowledge, and appropriately considers their results and limitations in developing the abstraction of radionuclide transport, including the potential effects from coupled THC processes in the unsaturated zone
- In considering alternative conceptual models for radionuclide transport in the unsaturated zone, DOE uses appropriate models, tests, and analyses that are sensitive to the processes, including coupled THC processes, modeled. For example, for radionuclide transport through fractures, DOE adequately considers alternative modeling approaches to develop its understanding of fracture distributions and ranges of fracture properties in the unsaturated zone so that the limitations of understanding of fracture distributions and properties are appropriately abstracted and propagated through the process-level and performance assessment models

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

Significant uncertainty remains with regard to site specific conditions and the potential for colloidal transport of strongly sorbing radionuclides. DOE identified colloid transport of other radionuclides (e.g., americium) and colloid filtration as potential enhancements to the TSPA-SR colloid transport abstraction, but indicates these enhancements may not be ready in time for the TSPA-SR Revision 00. Whichever type of approach is used for the colloid transport model will require additional laboratory, field, and site-scale observations to define the uncertainty in the necessary model parameters or additional sensitivity analyses to demonstrate the bounding nature of the current model approach. DOE will also need to demonstrate that the model is applicable to colloid transport through the unsaturated zone.

In the Near-Field/Altered-Zone Models Report (Hardin, 1998), numerous explicit statements of model inadequacy are noted. For example, in the description of the limitations of J-13 Well water as a starting composition, and in noting effects of condensation, water-rock interactions, nonisothermal chemistry, and engineered materials, Hardin states, "the models described here represent only a part of the whole-system processes, but they are representative of the kind of thermodynamic modeling that can be applied to additional parts of the system" (Hardin, 1998, p. 5-48). We concur with this observation. Present models are recognized to be inadequate. Nevertheless, DOE will need to demonstrate that thermodynamic models have been integrated into the TSPA-SR to the extent they impact performance or are required to demonstrate certain FEPs can be excluded. The thermodynamic modeling will need to be consistent with available site characterization data and design data. DOE may also use sensitivity analyses and bounding calculations to demonstrate that the effect of the remaining uncertainty on repository performance can be constrained.

In the TSPA-VA, DOE did not assess the effects of coupled THC processes on radionuclide transport in the unsaturated zone. Thus, DOE did not provide a reasonable description of the mathematical models included in its analyses of coupled THC effects on the transport of radionuclides. This does not meet this acceptance criterion. To a large extent, the TSPA-SR (CRWMS M&O, 1999a) will continue to decouple THC processes. Additional data and sensitivity analyses will be necessary to demonstrate that the decoupled approach used by DOE is appropriate and bounds radionuclide transport through the unsaturated zone.

The Integrated Site Model Process Model Report (CRWMS M&O, 1999c) indicates that the rock properties model provides a statistical basis for property surfaces (such as porosity) and bounds a much smaller area (to reflect the limited availability of data) than the geology framework model or the mineralogy model. DOE asserts that the smaller area reduces model uncertainty for the integrated site model. In general, the treatment of model uncertainty and validation is extremely terse in the mineralogy model compared to the geology framework and rock properties models. If DOE chooses to rely on the Integrated Site Model to provide input to compare sorptive zeolites and vitric minerals at Busted Butte to minerals beneath the repository horizon and to provide input to process models and the total system performance assessment, a better statistical basis is needed for the comparison.

One of the most important caveats for the rock properties model is that these data represent matrix properties and do not account for small-scale fractures or lithophysal cavities that may affect flow and transport. Therefore, matrix hydraulic conductivity does not include conductivity attributable to flow through lithophysal cavities because the porosity derived values do not measure these cavities. DOE's treatment of model uncertainty should address this limitation.

Sorptive zeolites occur in variable amounts below the repository horizon in four distinct stratigraphic groups separated by nonzeolitic intervals. Zeolitization increases from northeast to southwest in the Calico Hills Formation with a striking decrease from east to west. The unit exposed at Busted Butte and beneath the Experimental Studies Facility may be more zeolitized than to the west (where the repository will be located) as depicted in the integrated site model. For instance, the upper layer of the Calico Hills at Busted Butte contains approximately twenty percent zeolite, while the southern part of the Experimental Studies Facility contains approximately three percent zeolite. Model uncertainty in the mineralogy model needs a statistical basis for providing more reliable input to performance assessment models including unsaturated zone flow and transport.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available prior to May 15, 2000, these acceptance criteria remain open at this time. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.4.2.2.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 4**

Output from the abstraction of radionuclide transport in the unsaturated zone is justified through comparison with output from detailed process-level models and/or empirical observations (e.g., laboratory testing, field measurements, natural analogs). Specifically,

- DOE verifies that the outputs of radionuclide transport, including the potential effects from coupled THC processes, in the unsaturated zone abstractions reasonably produce or bound the results of corresponding process-level models, empirical observations, or both
- DOE bases abstracted models for radionuclide transport in the unsaturated zone on the same assumptions and approximations shown to be appropriate for closely analogous natural, including natural alteration observed at the site, or experimental systems
- DOE adopts well-documented procedures that have been accepted by the scientific community to construct and test the mathematical and numerical models used to simulate radionuclide transport through the unsaturated zone
- DOE provides sensitivity analyses or bounding analyses to support the abstraction of radionuclide transport in the unsaturated zone that cover ranges consistent with site data, field or laboratory experiments and tests, and natural analog research

STATUS OF RESOLUTION: OPEN. Further review of DOE's Analysis and Model Reports and Process Model Reports is needed.

As noted previously in Section 5.4.4.2.1.5, there was a general lack of data to support and test critical assumptions in mathematical models used by DOE in the TSPA-VA. Additional exercises using experimental, site, and natural analog data are necessary to support DOE models and abstractions. DOE should use the results from both laboratory and field heater test

program to test their abstracted models for consistency with observations. Models used should produce results reasonably representative of the systems modeled.

To the extent that models are generally regarded as preliminary, model verification for the effects of coupled THC processes on the transport of radionuclides is premature. As part of the continuing DOE effort to support their abstracted model of the effects of coupled THC effects on radionuclide transport in the unsaturated zone, DOE should use well-documented procedures to construct and test the numerical models.

DOE made a qualitative comparison to experimental and field observations in developing the TSPA-VA models and providing parameter estimates for near-field transport (e.g., CRWMS M&O, 1998d, Section 6.4.2.3). The resulting abstracted models are preliminary in nature, however, and, in general, have not been evaluated quantitatively against natural analog or experimental data. Parameter values have been determined for a limited range in chemical conditions. Thus, model results have not been tested against laboratory experiments or field data, and this acceptance criterion is not met. It is anticipated that these results, and additional experimental and modeling data that reflect the TSPA-SR reference design, will be developed as part of the preparation for the TSPA-SR, to the extent necessary. These tests, if conducted and properly documented, are likely to meet this acceptance criterion.

The TSPA-VA sensitivity analyses indicated that retardation in the unsaturated zone only provided a small contribution to repository performance. Nevertheless, the Repository Safety Strategy (CRWMS M&O, 2000a) considers radionuclide migration in the unsaturated zone as one of the principal factors of the post-closure safety case. DOE should continue its work investigating more detailed process models of colloid transport and radionuclide sorption in the unsaturated zone. These efforts should be commensurate with the degree of credit DOE asserts for radionuclide retardation reducing potential doses. These models should consider not only J-13 water, but also evolved water and gas phases that are the output of coupled THC calculations and consistent with the TSPA-SR reference design.

Because a number of final, DOE-approved Process Model Reports and Analysis and Model Reports were not available for review prior to May 15, 2000, these acceptance criteria remain open. Reviews of the Process Model Reports and Analysis and Model Reports, when they become available, are necessary to close the subissue.

#### **5.4.5 Subissue 5: Effects of Coupled Thermal-Hydrologic-Chemical Processes on Potential Nuclear Criticality in the Near-Field**

Nuclear criticality is not considered as an integrated subissue by NRC (U.S. Nuclear Regulatory Commission, 1998c, 2000a). The subject of nuclear criticality was not addressed in our comments on the DOE TSPA-VA (U.S. Nuclear Regulatory Commission, 1999b). Criticality, however, is considered in the scenario analysis subissue of the TSPA IRSR Revision 2 (U.S. Nuclear Regulatory Commission, 2000a). The approach taken in Revision 2 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1999f) was to focus on the use of sensitivity and uncertainty analyses to determine whether criticality could affect repository performance.

The scenario analysis acceptance criteria in the TSPA IRSR Revision 2 allow scenarios to be screened out of a performance assessment based on probability, consequence, or regulatory

arguments (U.S. Nuclear Regulatory Commission, 2000a). DOE will complete a scenario analysis and will determine whether criticality needs to be abstracted into a performance assessment. If criticality is abstracted by DOE, their abstraction of criticality within the near field would need to meet the acceptance criteria for abstraction listed below. DOE's efforts to assess the effects of criticality on repository performance are addressed in the Container Life and Source Term, ENFE, and Radionuclide Transport IRSRs. The Container Life and Source Term IRSR (U.S. Nuclear Regulatory Commission, 1999d) will evaluate criticality within the waste package, whereas the ENFE IRSR addresses criticality within the near-field environment. Consistent with the rest of the ENFE IRSR, the near field is defined as that area within and surrounding the emplacement drifts affected by coupled THC processes. The Radionuclide Transport IRSR (U.S. Nuclear Regulatory Commission, 1999g) only addresses criticality that could occur under isothermal conditions or at greater distances from the repository.

The Disposal Criticality Analysis Methodology Topical Report (U.S. Department of Energy, 1998e) describes the approach DOE will use to evaluate the potential for and consequences of repository criticality as an input to both the repository design process and the total system performance assessment conducted for a potential license application. DOE's approach consists of four steps. First, DOE will identify physical and chemical configurations that have potential for leading to criticality. Second, DOE will perform criticality analyses on each configuration to determine its potential for criticality according to a well-defined criterion. Third, DOE will calculate criticality probabilities for those configurations exceeding the criterion. Fourth, DOE will estimate consequences for all configurations. Probability and consequence screening will be conducted as part of the repository design process using specific criteria. In addition, the probability and consequence modeling results will be passed onto the performance assessment program for DOE for exclusion of the FEP or abstraction into the performance assessment. For this reason, data and models to be employed in criticality analyses for the design process are relevant also for the total system performance assessment in the event that criticality is not screened out. The topical report (U.S. Department of Energy, 1998e) lists nine specific "configuration classes" for the near field. Details of the scenario by which a particular configuration could lead to criticality are provided in the report. The methodology report defines the "near field" as external to the waste package but inside the drift wall (U.S. Department of Energy, 1998e).

Our review of the overall approach presented in the methodology topical report (U.S. Department of Energy, 1998e) is documented in a safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b). DOE's topical report provides a more detailed description of DOE's approach for analyzing near-field criticality (U.S. Department of Energy, 1998e). DOE is proposing to estimate the probability of a particular criticality scenario and then to calculate its consequences. The consequence analyses are limited to considering effects on radionuclide inventory and repository thermal profile (U.S. Department of Energy, 1998e).

We concluded in Revision 2 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1999f) that DOE's approach to dealing with near-field criticality satisfies NRC's scenario analysis approach and is acceptable. Staff review of DOE's treatment of near-field criticality in the TSPA-VA is discussed below. Based on the discussion in the ENFE IRSR Revision 2 and the safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) on DOE's topical report, this subissue of effects of coupled THC processes on potential nuclear criticality in the near field is considered closed, pending confirmation. Confirmation is dependent on

- Closure of open items identified in our safety evaluation report on DOE's topical report
- DOE documentation of the technical basis for exclusion of criticality FEPs from the total system performance assessment, if they are so excluded

The safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) lists 28 open items on which NRC acceptance of the methodology relies. Open items with particular relevance to the ENFE KTI are

- The consequence criteria for transient and external criticalities are not addressed in the topical report
- DOE has not presented a model validation methodology or work scope for external criticality models
- In developing the methodology for steady-state criticality consequences, DOE must consider other types of moderators, especially with respect to external criticality
- DOE must also include other types of steady-state criticality consequences, especially with respect to internal criticality, in its consequence analysis approach
- DOE needs to develop and present for NRC acceptance the modeling approach for external steady-state criticality consequence

In general, DOE's approach to abstract the effects of coupled THC processes on potential criticality in the near field must meet the following generic acceptance criteria: (i) integration, (ii) data and model justification, (iii) data uncertainty, (iv) model uncertainty, and (v) model support. Staff reiterates that, although the analysis described in the topical report (U.S. Department of Energy, 1998e) is intended to support design, the data and models employed may be relevant to the total system performance assessment.

#### **5.4.5.1 U.S. Nuclear Regulatory Commission Staff Evaluation**

##### **5.4.5.1.1 Acceptance Criterion 1—Integration for Evolution of the Near-Field Environment Subissue 5**

DOE evaluates in-package criticality or external-to-package criticality within the emplacement drift and provides an adequate technical basis for screening these events. If either event is included in the performance assessment, then DOE uses acceptable technical bases for selecting the design criteria that mitigate any potential impact of in-package criticality on the repository performance, identifies the FEPs that may increase the reactivity of the system inside the waste package, identifies the configuration classes and configurations that have potential for nuclear criticality, and includes changes in (i) thermal conditions and degradation of engineered barriers in the abstraction of the quantity and chemistry of water contacting the waste packages and waste forms and (ii) changes in radionuclide inventory and thermal conditions in the abstraction of radionuclide release.

STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

The TSPA-VA presented a simplified analysis of nuclear criticality (U.S. Department of Energy, 1998b, Section 4.4.4; CRWMS M&O, 1998d, Section 10.6). Consistent with the methodology proposed in the topical report on disposal criticality (U.S. Department of Energy, 1998e), DOE first evaluated the likelihood of criticality, and then assessed its effect on repository performance. Two external to the waste package criticality mechanisms—one near field and one far-field—are addressed in the TSPA-VA (U.S. Department of Energy, 1998b). DOE concluded, based on a more detailed discussion, that the external criticality mechanisms are exceedingly unlikely. The near-field scenario involved transport of fissile material (uranium and plutonium) out the bottom of a breached waste package. The scenario then envisioned accumulation of fissile material in the invert and rock material in the bottom of the drift via sorption or precipitation (CRWMS M&O, 1998d, Section 10.6.2.2.1). DOE used high solubilities for uranium (6,000 parts per million) and plutonium (78 parts per million), estimated for high-pH conditions, in its evaluation of the scenario. This assumption is conservative. Calculations by DOE suggest that this mechanism is incapable of resulting in mass concentrations of uranium and plutonium sufficient for criticality for high-level waste glass logs.

The consequences of an external criticality for performance from commercial spent nuclear fuel are discussed briefly (U.S. Department of Energy, 1998b; CRWMS M&O, 1998d). The only consequence considered by DOE in the TSPA-VA resulted from an increase in radionuclide inventory. An example analysis from an earlier report of a single critical event in fractured tuff showed an increase in radioactivity in the reactor of only 14 percent (CRWMS M&O, 1998e). The excess radioactivity decays to three percent after 20,000 years. (CRWMS M&O, 1998e). Due to the remote probability of the event and the negligible effect on total radionuclide inventory, DOE concluded that external criticality has insignificant effect on repository performance (U.S. Department of Energy, 1998d, p. O-3).

DOE did not consider the effects on the near field from heat output from criticality. DOE's analyses of criticality internal to the waste package at 15,000 years after emplacement shows that local heat output may be raised by a factor of 20. This heat output is similar to that when the waste had aged only 100 years (CRWMS M&O, 1998d, Fig. 10-73). The resulting temperature elevation may contribute to coupled THC processes that affect radionuclide release and transport. An external criticality, particularly in the near field, may also result in such a thermal pulse. DOE will consider effects on repository thermal profile (U.S. Department of Energy, 1998e). In the safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) on the DOE Disposal Criticality Analysis Methodology Topical Report (U.S. Department of Energy, 1998e), NRC approved of the DOE approach to scenario analysis (e.g, identification of configurations and configuration classes) for the criticality analysis supporting design. FEPs related to ENFE and criticality are listed in Table 5-8. Final screening of criticality for the total system performance assessment will be conducted separately. During the presentation on the ENFE (Barr, 2000) at the joint NRC and DOE technical exchange on Key Technical Issues DOE indicated that the FEPs associated with criticality in the near field environment will be excluded. Pending DOE's documentation of the technical bases for screening of criticality FEPs, staff has no questions concerning scenario screening for potential nuclear criticality in the near field at this time.

Coupled processes associated with the criticality event could provide feedback to the transport process responsible for the critical configuration. This interaction between criticality and coupled processes should be considered in future scenario analyses. Should DOE abstract near-field criticality in a future performance assessment, then it should consider all the relevant

**Table 5-8. U.S. Department of Energy features, events, and processes preliminary draft database (Revision 00b) (U.S. Department of Energy, 1999) entries relevant to Subissue 5 of the Evolution of the Near-Field Environment Key Technical Issue**

Features, Events, and Processes Number	Features, Events, and Processes Name	Preliminary DOE Screening
2.1.14.01.00	Criticality in waste and engineered barrier system	Include
2.1.14.09.00	Near-field criticality, fissile material deposited in near-field pond	Include
2.1.14.10.00	Near-field criticality, fissile solution flows into drift low point	Include
2.1.14.11.00	Near-field criticality, fissile solution is adsorbed or reduced in invert	Exclude
2.1.14.12.00	Near-field criticality, filtered slurry or colloidal stream collects on invert surface	Include
2.1.14.13.00	Near-field criticality associated with colloidal deposits	Exclude
2.1.14.14.00	Out-of-package criticality, fuel/magma mixture	Exclude
2.2.14.03.00	Far-field criticality, sorption on clay/zeolite in Topopah Spring basal vitrophyre	Exclude
2.2.14.07.00	Far-field criticality, dryout produces fissile salt in a perched water basin	Include
2.2.14.08.00	Far-field criticality associated with colloidal deposits	?

FEPs. For instance, DOE should evaluate the impact of heat generated from a criticality on repository performance. DOE indicated it will do this as part of their disposal criticality analysis (U.S. Department of Energy, 1998e).

Should DOE abstract near-field criticality in a future performance assessment, then important mass transfer and mass transport processes and mechanisms for formation of both a critical mass and configuration, which are plausible for the Yucca Mountain near-field environment, should be considered. Also, DOE models should reasonably account for known temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality. If potentially important couplings in the near field are neglected, DOE should provide a technical basis for doing so. Finally, the bases used for simplifying modeling assumptions and approximations should be documented and justified. Pending closure of open items identified in the safety evaluation report (Nuclear Regulatory Commission, 2000b) and DOE documentation of screening of criticality FEPs, we have no further questions regarding integration for near-field criticality.

#### **5.4.5.1.2 Acceptance Criterion 2—Data and Model Justification Evolution of the Near-Field Environment Subissue 5**

If external-to-package criticality is included in the total system performance assessment, then available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on the potential for nuclear criticality were considered.

STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

DOE's treatment of external criticality in the TSPA-VA was preliminary and incomplete, but as discussed in Section 5.4.5, the methodology topical report (U.S. Department of Energy, 1998e) suggests that future criticality analyses will be systematic and comprehensive. In addition to the proposed criticality analysis methodology (U.S. Department of Energy, 1998e), DOE listed future and ongoing investigations that will improve the ability to evaluate the external criticality issue (U.S. Department of Energy, 1998d). These technical investigations are focused on colloid formation, stability, and transport because colloids are viewed as a means for transport of plutonium out of the waste package.

Should DOE abstract near-field criticality in a future performance assessment, it should use data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on the potential for nuclear criticality in the near-field environment in their abstraction. DOE should also consider site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect nuclear criticality in the near-field environment. Finally, DOE should collect sufficient data on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material to establish initial and boundary conditions for conceptual models and simulations of THC coupled processes that may affect nuclear criticality in the near-field environment. Pending closure of open items identified in the safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) and DOE documentation of screening of criticality FEPs, we have no further questions regarding data and model justification for near-field criticality.

#### **5.4.5.1.3 Acceptance Criterion 3—Data Uncertainty for Evolution of the Near-Field Environment Subissue 5**

If external-to-package criticality is included in the total system performance assessment, then reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on potential nuclear criticality. Parameter values, assumed ranges, probability distributions, and bounding assumptions were technically defensible and reasonably accounted for uncertainties.

STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

All DOE criticality analyses to date have been preliminary with respect to planned analyses for any potential license application. Thus, a systematic data evaluation has not been performed. Sensitivity studies in an analysis of external plutonium criticality (CRWMS M&O, 1998e) identified two uncertain parameters that had the greatest effect on criticality potential: fracture aperture and waste form dissolution rate.

Should DOE abstract near-field criticality in a future performance assessment, it should adopt reasonable or conservative ranges of parameters or functional relations to determine effects of coupled THC processes on potential nuclear criticality in the near-field environment. DOE should also use a conservative range for fracture aperture and waste form dissolution rate and an appropriate range of input parameters for calculating the effective neutron multiplication factor ( $k_{\text{eff}}$ ). Also, DOE should consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality. DOE should consider the uncertainties in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THC coupled processes that affect potential nuclear criticality. Finally, DOE should use initial conditions, boundary conditions, and computational domain in sensitivity analyses involving coupled THC effects on potential nuclear criticality in the near-field environment that are consistent with available data. Pending closure of open items identified in the safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) and DOE documentation of screening of criticality FEPs, we have no further questions regarding data uncertainty for near-field criticality.

#### **5.4.5.1.4 Acceptance Criterion 4—Model Uncertainty for Evolution of the Near-Field Environment Subissue 5**

If external-to-package criticality is included in the total system performance assessment, then DOE provides a reasonable description of the mathematical models included in its analyses of coupled THC effects on potential nuclear criticality, evaluates their model results and limitations, and considers alternative modeling approaches consistent with available data and current scientific understanding.

STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

Should DOE abstract near-field criticality in a future performance assessment, DOE should investigate alternative modeling approaches that are consistent with available data and current scientific understanding. Also, DOE should provide a reasonable description of the mathematical models included in its analyses of coupled THC effects on potential nuclear criticality. Neutronics models for spent fuel inventory and criticality will be used by DOE for quantitative modeling in the criticality analysis. DOE will use degradation models that include corrosion and geochemistry (e.g., EQ3/6). In addition, DOE will use Monte Carlo-based codes for estimating criticality probabilities and criticality consequence. Finally, DOE will use their performance assessment code to model performance impact (U.S. Department of Energy, 1998e). These models should be adequately described. Pending closure of open items identified in the safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) and DOE documentation of screening of criticality FEPs, we have no further questions regarding model uncertainty for near-field criticality.

#### **5.4.5.1.5 Acceptance Criterion 5—Model Support for Evolution of the Near-Field Environment Subissue 5**

If external-to-package criticality is included in the total system performance assessment, then the mathematical models for coupled THC effects on potential nuclear criticality were consistent

with conceptual models based on inferences about the near-field environment, field data, and natural alteration observed at the site, and expected engineered materials.

#### STATUS OF RESOLUTION: CLOSED, PENDING CONFIRMATION

Should DOE abstract near-field criticality in a future performance assessment, the mathematical models it uses should be consistent with conceptual models based on inferences about the near-field environment, field data and natural alteration observed at the site, and expected engineered materials. Also, DOE should adopt accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on potential nuclear criticality. The methodology topical report (U.S. Department of Energy, 1998e) outlines validation methodologies for all codes and discusses how uncertainties will be compensated with conservatism. For example, a critical limit—the value of  $k_{\text{eff}}$  at which a system is considered potentially critical—will be established that accounts for model and data uncertainties (U.S. Department of Energy, 1998d, Section 3.4), and conservative assumptions will be made in constructing external criticality configurations (U.S. Department of Energy, 1998e, Section 3.3). Finally, DOE should verify abstracted model results through comparison to outputs of detailed process models and empirical observations. Pending closure of open items identified in the safety evaluation report (U.S. Nuclear Regulatory Commission, 2000b) and DOE documentation of screening of criticality FEPs, we have no further questions regarding model support for near-field criticality.

## 6.0 REFERENCES

- Altman, W.D., J.P. Donnelly, and J.E. Kennedy, *Peer Review for High-level Nuclear Waste Repositories*, NUREG-1297, U.S. Nuclear Regulatory Commission, Washington, D.C., 1988a.
- Altman, W.D., J.P. Donnelly, and J.E. Kennedy, *Qualification of Existing Data for High-Level Nuclear Waste Repositories*, NUREG-1298, U.S. Nuclear Regulatory Commission, Washington, D.C., 1988b.
- Ames, L.L., Jr., *Some zeolite equilibria with alkali metal cations*, American Mineralogist, Vol. 49, pp. 127–145, 1964.
- Amy, P.S., and D.L. Haldeman (eds.), *The Microbiology of the Terrestrial Deep Subsurface*, CRC Press, Lewis Publishers, Boca Raton, Florida, 1997.
- Angell, P., et al., *Evolution of the Near-Field Environment in the Proposed High-Level Waste Repository at Yucca Mountain—A Review of Hypotheses*, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1996.
- Apted, M.J., *Natural analogs for the predictive reliability of the engineered barrier system for high-level waste*, B.I. Come and N.A. Chapman (eds.), Fourth Natural Analogue Working Group Meeting and Poços de Caldas Project, EUR 13014 EN, Commission of the European Communities, Luxembourg, 1990.
- Arthur, R.C., and W.M. Murphy, *An analysis of gas-water-rock interactions during boiling in partially saturated tuff*, Sciences Géologiques Bulletin, Vol. 42, pp. 313–327, 1989.
- Atkins, M., A.N. Beckley, and F.P. Glasser, *Influence of cement on the near-field environment and its specific interactions with U and iodine*, Radiochimica Acta, Vol. 44/45, pp. 255–261, 1988.
- Atkins, M., et al., *Assessment of the performance of cement-based composite material for radioactive waste immobilization*, V.M. Oversby and P.W. Brown (eds.), Scientific Basis for Nuclear Waste Management XIII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 176, pp. 117–127, 1990.
- Atkins, M., F.P. Glasser, and A. Kindness, *Phase relations and solubility modeling in the CaO-SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-MgO-SO<sub>3</sub>-H<sub>2</sub>O system: For application to blended cements*, T. Abrajano, Jr., and L.H. Johnson (eds.), Scientific Basis for Nuclear Waste Management XIV, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 212, pp. 387–394, 1991.
- Atkins, M., D. Damidot, and F.P. Glasser, *Performance of cementitious systems in the repository*, A. Barkatt and R.A. Van Konynenburg (eds.), Scientific Basis for Nuclear Waste Management XVII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 333, pp. 315–326, 1994.

Atkinson, A., N.M. Everitt, and R.M. Guppy, *Time dependence of pH in a cementitious repository*, W. Lutze and R.C. Ewing (eds.), Scientific Basis for Nuclear Waste Management XII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 127, pp. 439–446, 1989.

Baca, R.G., and M.S. Jarzempa, *Detailed Review of Selected Aspects of Total System Performance Assessment—1995*, AI 5708-761-711, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Bales, R.C., et al., *Bacteriophage transport in sandy soil and fractured tuff*, Applied and Environmental Microbiology, Vol. 55, pp. 2,061–2,067, 1989.

Bard, A.J., and L.R. Faulkner, *Electrochemical Methods: Fundamentals and Applications*, John Wiley & Sons, New York, New York, 1980.

Barkatt, A., E.E. Saad, R. Adiga, W. Sousanpour, A. Barkatt, M.A. Adel-Hadadi, J.A. O’Keefe, and S. Alterescu, *Leaching of Natural and Nuclear Waste Glasses in Sea Water*, Applied Geochemistry, Vol. 4, pp. 593-603, 1989.

Barr, D., *Evolution of the Near-Field Environment: NRC/DOE Technical Exchange on Yucca Mountain Pre-Licensing Issues*, Yucca Mountain Site Characterization Office, U.S. Department of Energy Office of Civilian Radioactive Waste Management, April 25, 2000.

Bates, J.K., et al., *Colloid formation during waste form reaction: Implications for nuclear waste disposal*, Science, Vol. 256, pp. 649–651, 1992.

Bates, J.K., et al., *Yucca Mountain Project—Argonne National Laboratory, Annual Progress Report, FY 1994*, ANL-94/42, Argonne National Laboratory, Argonne, Illinois, 1995.

Berner, U., *A Thermodynamic Description of the Evolution of Pore Water Chemistry and Uranium Speciation During the Degradation of Cement*, PSI-Bericht NR. 62, Paul Scherrer Institute, Wurenlingen und Villigen, Switzerland, 1990.

Bertetti, F.P., R.T. Pabalan, and M.G. Almendarez, *Studies of neptunium<sup>V</sup> sorption on quartz, clinoptilolite, montmorillonite, and  $\alpha$ -alumina*, E.A. Jenne (ed.), Adsorption of Metals by Geomedia, Academic Press, Inc., New York, New York, pp. 131-148, 1998.

Bish, D.L., *Smectite Dehydration and Stability: Applications to Radioactive Waste Isolation at Yucca Mountain, Nevada*, LA-11023-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1988.

Bish, D.L., *Evaluation of Past and Future Alterations in Tuff at Yucca Mountain, Nevada, Based on the Clay Mineralogy of Drill Cores USW G-1, G-2, and G-3*, LA-10667-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1989.

Bish, D.L., *Study Plan for Kinetics and Thermodynamics of Mineral Evolution at Yucca Mountain, Nevada*, YMP-LANL-SP 8.3.1.3.3.2, Los Alamos National Laboratory, Los Alamos, New Mexico, 1993.

Bish, D.L., and S.J. Chipera, *Revised Mineralogic Summary of Yucca Mountain, Nevada*, LA-11497-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1989.

Bish, D.L., and J.L. Aronson, *Paleogeothermal and paleohydrologic conditions in silicic tuff from Yucca Mountain, Nevada*, Clays and Clay Minerals, Vol. 41(2), pp. 148–161, 1993.

Bish, D.L., et al., *Summary and Synthesis Report on Mineralogy and Petrology Studies for the Yucca Mountain Site Characterization Project*, WBS Element 1.2.3.2.1.1 and 1.2.3.2.1.2, Studies 8.3.1.3.2.1, 8.3.1.3.2.2, and 8.3.1.3.3.2, Los Alamos National Laboratory, Los Alamos New Mexico, 1996.

Blesa, M.A., P.J. Morando, and A.E. Regazzoni, *Chemical Dissolution of Metal Oxides*, CRC Press, Boca Raton, Florida, 1994.

Boden, P.J., *Effect of Concentration, Velocity and Temperature, Corrosion: Metal/Environment*, L.L. Shreir, R.A. Jarman, and G.T. Burstein (eds.), Reactions, Third Edition, Vol. 1, pp. 2:3–2:30, Butterworth-Heinemann, Boston, Massachusetts, 1994.

Bourcier, W.L., et al., *Solution Compositional Effects on Dissolution Kinetics of Borosilicate Glass*, Proceedings of the Seventh International Symposium on Water-Rock Interaction, A.A. Balkema, Rotterdam, Netherlands, pp. 81-84, 1992.

Bowers, T.S., and R.G. Burns, *Activity diagrams for clinoptilolite: Susceptibility of this zeolite to further diagenetic reactions*, American Mineralogist, Vol. 75, pp. 601–619, 1990.

Bowman, C.D., and F. Venneri, *Underground Supercriticality from Plutonium and Other Fissile Material*, LA-UR 94-4022A, Los Alamos National Laboratory, Los Alamos, New Mexico, 1995.

Bradbury, M.H., and F.-A. Sarott, *Sorption Databases for the Cementitious Near-Field of a L/ILW Repository for Performance Assessment*, PSI Bericht Nr. 95-06, Paul Scherrer Institute, Wurenlingen, Switzerland, 1995.

Brossia, C.S. and G.A. Cragolino, *Effects of environmental, electrochemical, and metallurgical variables on the passive and localized dissolution of Ti Grade 7*, CORROSION/2000, Paper #211, NACE International, Houston, Texas, 2000.

Brossia, S., D. Dunn, and N. Sridhar, *Effects of Environmental Factors on Container Life*, CNWRA 98-008, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1998.

Brown, D.A., and B.L. Sherrif, *The Effects of Microbial Activity on the Natural and Engineered Barriers of a High-Level Nuclear Waste Repository*, SKI Report 98:24, Swedish Nuclear Power Inspectorate, Stockholm, Sweden, 1998.

Brown, R.L., R.S. Bowman, and T.L. Kieft, *Microbial effects on nickel and cadmium sorption and transport in volcanic tuff*, Journal of Environmental Quality, Vol. 23, No. 4, pp. 723–729, 1994.

Broxton, D.E., D.L. Bish, and R.G. Warren, *Distribution and chemistry of diagenetic minerals at Yucca Mountain, Nye County, Nevada*, Clays and Clay Minerals, Vol. 35(2), pp. 89-110, 1987.

Buddemeier, R.W., and J.R. Hunt, *Transport of colloidal contaminants in groundwater: Radionuclide migration at the Nevada Test Site*, Applied Geochemistry, Vol. 3, pp. 535–548, 1988.

Burns, W.G., et al., *Effects of radiation on the leach rates of vitrified radioactive waste*, Journal of Nuclear Materials, Vol. 107, pp. 245–270, 1982.

Byers, F.M., Jr., *Petrochemical Variation of Topopah Spring Tuff Matrix with Depth (Stratigraphic Level), Drill Hole USW G-4, Yucca Mountain, Nevada*, LA-10561-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1985.

Campbell, A.C., and K.M. Krupka, *Application of geochemical data and modeling in performance assessments of low-level radioactive waste disposal facilities*, Symposium on Hydrogeologic and Geochemical Aspects of Waste Disposal, Waste Management '97, WM Symposia, Inc., Tucson, Arizona, 1997.

Canavan, G.H., et al., *Comments on "Nuclear Excursions" and "Criticality Issues"*, LA-UR-95-0851, Los Alamos National Laboratory, Los Alamos, New Mexico, 1995.

Carlos, B., *Fracture-Coating Minerals in the Topopah Spring Member and Upper Tuff of Calico Hills from Drill Hole J-13*, LA-11504-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1989.

Carlos, B.A., D.L. Bish, and S.J. Chipera, *Fracture-lining minerals in the lower Topopah Spring Tuff at Yucca Mountain*, Proceedings of the Second International Conference on High Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, pp. 486–493, 1991.

Carlos, B.A., et al., *Fracture-lining manganese oxide minerals in silicic tuff, Yucca Mountain, Nevada*, Chemical Geology, Vol. 107, pp. 47–69, 1993.

Cavanaugh, M.A., et al., *The anodic dissolution of a Ni-base superalloy*, Corrosion, Vol. 39, pp. 144-150, 1983.

Chan, K.S., *A micromechanical model for predicting hydride embrittlement in nuclear fuel cladding material*, Journal of Nuclear Materials, Vol. 227, pp. 220–236, 1996.

Chigira, M., and M. Watanabe, *Silica precipitation behavior in a flow field with negative temperature gradients*, Journal of Geophysical Research, Vol. B8(99), pp. 15,539–15,548, 1994.

Chipera, S.J., and D.L. Bish, *Equilibrium modeling of clinoptilolite-analcime equilibria at Yucca Mountain, Nevada*, Clays and Clay Minerals, Vol. 45(2), pp. 226-239, 1997.

Choi, J.S., and T.H. Pigford, *Underground criticality in geologic disposal*, W.J. Gray and I.R. Triay (eds.), Scientific Basis for Nuclear Waste Management XX, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 465, 1997.

Choppin, G.R., *Chemistry of actinides in the environment*, Radiochimica Acta, Vol. 43, pp. 82–83, 1988.

Choppin, G.R., and J.N. Mathur, *Hydrolysis of actinyl(VI) cations*, Radiochimica Acta, Vol. 52/53, pp. 25–28, 1991.

Christensen, H, *Radiation Induced Dissolution of UO<sub>2</sub>*, T.A. Abrajano and L.H Johnson (eds.), Scientific Basis for Nuclear Waste Management XIV, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 212, pp. 213-220, 1991.

Christensen, H. and E. Bjergrakke, *Radiation Induced Dissolution of UO<sub>2</sub>*, J.K. Bates and W.B. Seefeldt (eds.), Scientific Basis for Nuclear Waste Management X, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 84, pp. 115-122, 1987.

Christensen, H., S. Sunder and D.W. Shoosmith, *Oxidation of nuclear fuel (UO<sub>2</sub>) by the products of water radiolysis: development of kinetic model*, Journal of Alloys and Compounds, Vol. 213/214, pp. 93–99, 1994.

Christofi, N., and J.C. Philp, *European microbiology related to the subsurface disposal of nuclear waste*, Amy, P.S., and D.L. Haldeman (eds.), The Microbiology of the Terrestrial Deep Subsurface, CRC Press, Lewis Publishers, Boca Raton, Florida, 1997.

Civilian Radioactive Waste Management System, Management and Operating Contractor (CRWMS M&O), *Final Report on the Enhanced Characterization of the Repository Block (ECRB) Planning Effort, Revision 0, Volume 1 of 3*, B00000000-01717-5700-00008, Las Vegas, Nevada, 1997a.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Final Report Waste Package Degradation Expert Elicitation, Revision 0, August 15, 1997*, prepared by Geomatrix Consultants, Inc., for TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1997b.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Tracers, Fluids, and Material Data Reporting and Management*, Yucca Mountain Site Characterization Project Procedure YAP-2.8Q, Revision 2, January 9, 1998, MOL.19980219.0941, Las Vegas, Nevada, 1998a.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Determination of Importance Evaluation for the ESF Enhanced Characterization of the Repository Block Cross Drift, Revision 2*, BABEAF000-01717-2200-00011, Las Vegas, Nevada, 1998b.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Near-Field/Altered Zone Coupled Effects Expert Elicitation, May 29, 1998*, prepared by Geomatrix Consultants, Inc., for TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1998c.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Total System Performance Assessment—Viability Assessment (TSPA-VA) Analyses Technical Basis Document, Revision 01*, B00000000–01717–4301, North Las Vegas, Nevada, 1998d.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Report on External Criticality of Plutonium Waste Forms in a Geologic Repository, Revision 01*, BBA000000–01717–5705–00018, North Las Vegas, Nevada, 1998e.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Total System Performance Assessment—Site Recommendation Methods and Assumptions, Revision 00, ICN 01*, TDR–MGR–MD–000001, North Las Vegas, Nevada, 1999a.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *License Application Design Selection Report, Revision 01, ICN 01*, B00000000–01717–4600–00123, North Las Vegas, NV, 1999b.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Integrated Site Model (ISM) Process Model Report, Revision 0*, TDR-NBS-GS-000002, North Las Vegas, Nevada, 1999c.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Repository Safety Strategy: Plan to Prepare the Postclosure Safety Case to Support Yucca Mountain Site Recommendation and Licensing Considerations, Revision 3*, TDR–WIS–RL–000001, North Las Vegas, Nevada, 2000a.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Engineered Barrier System Degradation, Flow, and Transport Process Model Report, Revision 00, Preliminary Draft*, TDR–EBS–MD–000006, North Las Vegas, Nevada, 2000b.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Seepage Model for Performance Assessment Including Drift Collapse, Revision 00, Preliminary Draft*, MDL–NBS–HS–000002, North Las Vegas, Nevada, 2000c.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Abstraction of Drift Seepage, Revision 00, Preliminary Draft*, ANL-NBS-MD-000005, North Las Vegas, Nevada, 2000d.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Unsaturated Zone Flow and Transport Process Model Report, Revision 00, Preliminary Draft*, TDR-NBS-HS-000002, North Las Vegas, Nevada, 2000e.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *In-Drift Thermal-Hydrological-Chemical Model, Revision 00, Preliminary Draft*, ANL-EBS-MD-000026, North Las Vegas, Nevada, 2000f.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Natural Analogs for the Unsaturated Zone, Revision 00, Preliminary Draft*, ANL-NBS-HS-000007, North Las Vegas, Nevada, 2000g.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *In-Drift Precipitates/Salts Analysis, Revision 00, Preliminary Draft*, ANL-EBS-MD-000045, North Las Vegas, Nevada, 2000h.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *In-Package Chemistry Abstraction for TSPA-LA, Revision 00, Preliminary Draft*, ANL-EBS-MD-000037, North Las Vegas, Nevada, 2000i.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Waste Form Degradation Process Model Report, Revision 00, Preliminary Draft*, TDR-WIS-MD-000001, North Las Vegas, Nevada, 2000j.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *CSNF Waste Form Degradation Summary Abstraction, Revision 00, Preliminary Draft*, ANL-EBS-MD-000015, North Las Vegas, Nevada, 2000k.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Summary of Dissolved Concentration Limits, Revision 00, Preliminary Draft*, ANL-WIS-MD-000010, North Las Vegas, Nevada, 2000l.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Waste-Form Colloid-Associated Concentration Limits: Abstraction and Summary, Revision 00A, Preliminary Draft*, ANL-WIS-MD-000012, North Las Vegas, Nevada, 2000m.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Colloid-Associated Radionuclide Concentration Limits: ANL, Revision 00, Preliminary Draft*, ANL-EBS-MD-000020, North Las Vegas, Nevada, 2000n.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Colloid Stability and Attachment/Detachment Properties, Revision 00A, Preliminary Draft*, ANL-EBS-MD-000021, North Las Vegas, Nevada, 2000o.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Pure Phase Solubility Limits--LANL, Revision 00B, Preliminary Draft*, ANL-EBS-MD-000017, North Las Vegas, Nevada, 2000p.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Engineered Barrier System Features, Events, and Processes and Degradation Modes Analysis, Revision 00, Preliminary Draft*, ANL-EB-MD-000035, North Las Vegas, Nevada, 2000q.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *In-Drift Corrosion Products, Revision 00, Preliminary Draft*, ANL-EB-MD-000041, North Las Vegas, Nevada, 2000r.

Civilian Radioactive Waste Management System, Management and Operating Contractor, *Invert Diffusion Properties Model, Revision 00, Preliminary Draft*, ANL-EB-MD-000031, North Las Vegas, Nevada, 2000s.

Clegg, S.L., and M. Whitfield, *Activity coefficients in natural waters*, in K.S. Pitzer (ed.), Activity Coefficients in Electrolyte Solutions, CRC Press, Boca Raton, Florida, pp. 279-434, 1991.

Cline, J., *Yucca Mountain Fluid Inclusion Thermochronology Project Press Release*, June 21, 2000, University of Nevada, Las Vegas, 2000.

Coburn, S.K., *Corrosion in Fresh Water*, in Metals Handbook, Ninth Edition, Volume 1: Properties and Selection, Irons and Steels, American Society for Metals, Metals Park, Ohio, pp. 733-738, 1978.

Codell, R.B., and W.M. Murphy, *Geochemical model for C-14 transport in unsaturated rock*, Proceedings of the Third International Conference on High-Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, pp. 1,959-1,965, 1992.

Corbell, C., et al., *Uranium oxide mass loss rate in water for an interface under alpha irradiation*, R.W. Smith and D.W. Shoesmith (eds.), Scientific Basis for Nuclear Waste Management XXIII, Materials Research Society, Warrendale, Pennsylvania, 2000 (in press).

Corwin, R.F., and D.B. Hoover, *The self potential method in geothermal exploration*, Geophysics, Vol. 44(2), pp. 226-245, 1979.

Couture, R.A., *Steam rapidly reduces the swelling capacity of bentonite*, Nature, Vol. 318, pp. 50-52, 1985.

Cowan, G.A., *A natural fission reactor*, Scientific American, Vol. 235, pp. 36-47, 1975.

Cox, B., *Environmentally-induced cracking of zirconium alloys—A review*, Journal of Nuclear Materials, Vol. 170, pp. 1–23, 1990.

Cragolino, G.A., and J.R. Galvele, *Anodic behavior and pitting of zirconium and Zircaloy-4 in aqueous solutions of sodium chloride*, R.P. Frankenthal and J. Kruger (eds.), Passivity of Metals, The Electrochemical Society, Princeton, New Jersey, pp. 1,053–1,057, 1978.

Cragolino, G.A., et al., *Hanford Tank Waste Remediation System Familiarization Report*, CNWRA 97-001, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Cragolino, G.A., et al., *Assessment of Performance Issues Related to Alternate Engineered Barrier System Materials and Design Options*, CNWRA 99-003, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1999.

Davis, J.A., and D.B. Kent, *Surface complexation modeling in aqueous geochemistry*, in M.F. Hochella, Jr., and A.F. White (eds.), Reviews in Mineralogy, Volume 23, Mineral-Water Interface Geochemistry, Mineralogical Society of America, Washington, D.C., pp. 177–260, 1990.

Delany, J.M., *Reaction of Topopah Spring Tuff with J-13 Water: A Geochemical Modeling Approach Using EQ3/6 Reaction Path Code*, UCRL-53631, Lawrence Livermore National Laboratory, Livermore, California, 1985.

de Marsily, G., *Quantitative Hydrogeology: Groundwater Hydrology for Engineers*, Academic Press, Inc., New York, New York, 1986.

Dubessy, J., et al., *Radiolysis evidenced by H<sub>2</sub>-O<sub>2</sub>-bearing fluid inclusions in three U deposits*, Geochimica et Cosmochimica Acta, Vol. 52, pp. 1,155–1,167, 1988.

Dublyansky, Y.V., *Fluid inclusion studies of samples from the Exploratory Study Facility, Yucca Mountain, Nevada, Draft Report*, October 1998, Blacksburg, Virginia, 1998.

Dunn D.S., Y.-M. Pan, and G.A. Cragolino, *Effects of Environmental Factors on the Aqueous Corrosion of High-Level Radioactive Waste Containers—Experimental Results and Models*, CNWRA 99-004, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1999.

Dunn, D.S., Y.-M. Pan, and G.A. Cragolino, *Stress corrosion cracking, passive and localized corrosion of Alloy 22 High-Level Radioactive Waste Containers*, CORROSION/2000, Paper #206, NACE International, Houston, Texas, 2000.

Dunn, D.S., et al., *Corrosion of iron under alternating wet and dry conditions*, The Journal of Science and Engineering Corrosion, Vol. 56, pp. 470-481, 2000.

Ebert, W.L., and J.K. Bates, *A comparison of glass reaction at high and low SA/V:PCT versus MCC-1*, Proceedings of Third International Conference on High-Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, Vol. 1, pp. 934–942, 1992.

Ehrlich, H.L., *Geomicrobiology*, third edition, Marcel Dekker, Inc., New York, New York, 1996.

Eisenberg, N.A., et al., *Regulatory Perspectives on Model Validation in High-Level Radioactive Waste Management Programs: A Joint NRC/SKI White Paper*, NUREG-1636, U.S. Nuclear Regulatory Commission, Washington, D.C., 1999.

Farmer, J.C., and R.D. McCright, *Localized Corrosion and Stress Corrosion Cracking of Candidate Materials for High-Level Radioactive Waste Disposal Containers in U.S.: A Critical Literature Review*, W. Lutze and R.C. Ewing (eds.), Scientific Basis for Nuclear Waste Management XII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 127, pp. 359–371, 1989.

Farmer, J.C., et al., *Survey of Degradation Modes of Candidate Materials for High-Level Radioactive-Waste Disposal Containers, Vol. 3, Localized Corrosion and Stress Corrosion Cracking of Austenitic Alloys*, UCID–21362, Lawrence Livermore National Laboratory, Livermore, California, 1988.

Finch, R.J., et al., *Oxidative corrosion of spent fuel in vapour and dripping groundwater at 90 °C*, D.J. Wronkiewicz and J.H. Lee, (eds.), Scientific Basis for Nuclear Waste Management XXII, Materials Research Society, Warrendale, Pennsylvania, Symposium Proceedings 556, pp. 431–438, 1999.

Finn, P.A., et al., *Colloidal products and actinide species in leachate from spent nuclear fuel*, Radiochimica Acta, Vol. 66/67, pp. 189–195, 1994a.

Finn, P.A., et al., *The effect of fuel type in saturated spent fuel tests*, Proceedings of the Fifth International Conference on High Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, Vol. 2, pp. 1,080–1,087, 1994b.

Flint, L.E., *Characterization of Hydrogeologic Units Using Matrix Properties at Yucca Mountain, Nevada*, Water-Resources Investigation Report 97-4243, U.S. Geological Survey, Denver, Colorado, 1997.

Geesey, G., *Review of the Potential for Microbially Influenced Corrosion of High-Level Nuclear Waste Containers*, CNWRA 93-014, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1993.

Glasser, F.P., et al., *The chemical environment in cements*, C.M. Jantzen, J.A. Stone, and R.C. Ewing (eds.), Scientific Basis for Nuclear Waste Management VIII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 44, pp. 849–858, 1985.

Glasser, F.P., D.E. Macphee, and E.E. Lachowski, *Solubility modeling of cements: Implications for radioactive waste immobilization*, J.K. Bates and W.B. Seefeldt (eds.), Scientific Basis for Nuclear Waste Management X, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 84, pp. 331–341, 1987.

Goldberg, S., H.S. Forster, and E.L. Heick, *Flocculation of illite/kaolinite and illite/montmorillonite mixtures as affected by sodium adsorption ratio and pH*, Clays and Clay Minerals, Vol. 39, pp. 375–380, 1991.

Grambow, B., *Spent Fuel Dissolution and Oxidation: An Evaluation of Literature Data*, SKB Technical Report 89-13, Swedish Nuclear Fuel and Waste Management Co., Stockholm, Sweden, 1989.

Gray, W.J., H.R. Leider, and S.A. Steward, *Parametric study of LWR spent fuel dissolution kinetics*, Journal of Nuclear Materials, Vol. 190, pp. 46–52, 1992.

Grogan, H.A., and I.G. McKinley, *An approach to microbiological modeling: application to the near field of a Swiss low/intermediate level waste repository*, Technical Report 89-06, Nagra, Baden, Switzerland, 1990.

Haldeman, D.L., et al., *The Effects of Diesel Exhaust on the Microbiota within a Tuffaceous Tunnel System*, UCRL-ID-125176, Lawrence Livermore National Laboratory, Livermore, California, 1996.

Hardin, E.L., *Near-Field/Altered-Zone Models Report*, UCRL-ID-129179, Lawrence Livermore National Laboratory, Livermore, California, 1998.

Hardin, E.L., and D.A. Chestnut, *Synthesis Report on Thermally Driven Coupled Processes*, UCRL-ID-128495, Lawrence Livermore National Laboratory, Livermore, California, 1997.

Harvie, C.E., N. Moller, and J.H. Weare. *The prediction of mineral solubilities in natural waters: The Na-K-Mg-Ca-Cl-SO<sub>4</sub>-OH-HCO<sub>3</sub>-CO<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O system to high ionic strengths at 25 °C*. Geochimica et Cosmochimica Acta, Vol. 48, pp. 723-751, 1984.

Heimann, R.B., *Interaction of cement and radioactive waste forms in multicomponent systems tests at 200 °C Part 2: Mineralogical changes of cement*, Cement Concrete Research, Vol. 18, pp. 554–560, 1988.

Hersman, L.E., *Summary and Synthesis of Biological Sorption and Transport*, Milestone 3663, MOL.19970103.0052, Los Alamos National Laboratory, Los Alamos, New Mexico, 1996.

Hoare, J.P., *The oxygen electrode on noble metals*, P. Delahay (ed.), Advances in Electrochemistry and Electrochemical Engineering, Volume 6, Interscience Publishers, New York, New York, 1967.

Holland, H.D., and L.H. Brush, *U oxides in ores and spent fuels*, Proceedings of the Conference on High-Level Radioactive Solid Waste Forms, NUREG/CP-005, U.S. Nuclear Regulatory Commission, Washington, D.C., pp. 597–615, 1980.

Horn, J.M., and A. Meike, *Microbial Activity at Yucca Mountain*, UCRL-ID-122256, Lawrence Livermore National Laboratory, Livermore, California, 1995.

Hunter, R.J., *Foundations of Colloid Science Volume 1*, Oxford University Press, New York, New York, 1987.

Jantzen, C.M., *Nuclear waste glass durability: I. Predicting environmental response from thermodynamic (Pourbaix) diagrams*, Journal of American Ceramic Society, Vol. 75, pp. 2,433–2,448, 1992.

Jarzemba, M.S., et al., *NRC Sensitivity and Uncertainty Analyses for a Proposed HLW Repository at Yucca Mountain, Nevada, Using TPA 3.1, Results and Conclusions*, NUREG-1668, Volume 2, U.S. Nuclear Regulatory Commission, Washington, D.C., 1999.

Johnson, G.K., et al., *Thermodynamic studies of zeolites: Clinoptilolite*, Journal of Chemical Thermodynamics, Vol. 23, pp. 475–484, 1991.

Kallay, N., E. Barouch, and E. Matijevic, *Diffusional detachment of colloidal particles from solid/solution interfaces*, Advances in Colloids and Interface Science, Vol. 27, pp. 1–42, 1987.

Kehler, B.A., G.O. Ilevbare, and J.R. Scully, *Comparison of the crevice corrosion resistance of Alloys 625 and 22 in concentrated chloride solution from 60 to 95 °C*, CORROSION/2000, Paper 182, NACE International, Houston, Texas, 2000.

Kerrisk, J.F., *Reaction-Path Calculations of Groundwater Chemistry and Mineral Formation at Rainier Mesa, Nevada*, LA-10560-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1983.

Kerrisk, J.F., *Groundwater Chemistry at Yucca Mountain, Nevada, and Vicinity*, LA-10-29-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1987.

Kersting, A.B., et al., *Migration of plutonium in groundwater at the Nevada Test Site*, Nature, Vol. 397, pp. 56–59, 1999.

Kessler, J., and R. McGuire, *Yucca Mountain Total System Performance Assessment, Phase 3*, EPRI TR-107191, Electric Power Research Institute, Palo Alto, California, 1996.

Kieft, T.L., et al., *Microbial abundance and activities in relation to water potential in the vadose zones of arid and semiarid sites*, Microbial Ecology, Vol. 26, pp. 59–78, 1993.

Kieft, T.L., et al., *Factors limiting microbial growth and activity at a proposed high-level nuclear repository, Yucca Mountain, Nevada*, Applied and Environmental Microbiology, Vol. 63, No. 8, pp. 3,128–3,133, 1997.

Kirchheim, R., et al., *The passivity of iron-chromium alloys*, Corrosion Science, Vol. 29, pp. 899–917, 1989

Knauss, K., *Zeolitization of glassy Topopah Spring Tuff under hydrothermal conditions*, in J.K. Bates and W.B. Seefeldt (eds.), Scientific Basis for Nuclear Waste Management X, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 84, pp. 737–745, 1987.

Knauss, K., et al., *Hydrothermal Interaction of Topopah Spring Tuff With J-13 Water as a Function of Temperature*, UCRL-90853, Lawrence Livermore National Laboratory, Livermore, California, 1984.

Knauss, K.G., W.J. Beiriger, and D.W. Peifer, *Hydrothermal Interaction of Solid Wafers of Topopah Spring Tuff with J-13 Water at 90 ° and 150 °C Using Dickson-Type, Gold-Bag Rocking Autoclaves: Long-Term Experiments*, UCRL-53722, Lawrence Livermore National Laboratory, Livermore, California, 1987.

Knief, R.A., *Nuclear Engineering Theory and Technology of Commercial Nuclear Power*, Hemisphere Publishing Corporation, Washington, D.C., 1992.

Kohler, M., E. Wieland, and J.O. Leckie, 1992, *Metal-ligand-surface interactions during sorption of uranyl and neptunyl on oxides and silicates*, Y.K. Kharaka and A.S. Maest (eds.), Proceedings of the 7th International Symposium on Water-Rock Interaction–WRI-7, A.A. Balkema, Rotterdam, Netherlands, Vol. 1, pp. 51–54, 1992.

Kotra, J., et al., *Branch Technical Position on the Use of Expert Elicitation in the High-Level Radioactive Waste Program*, NUREG-1563, U.S. Nuclear Regulatory Commission, Washington, D.C., 1996.

Langmuir, D., *Overview of coupled processes with emphasis in geochemistry*, C.-F. Tsang (ed.), Coupled Processes Associated With Nuclear Waste Repository, Academic Press Inc., New York, New York, pp. 67–101, 1987.

Lee, J.H., J.E. Atkins, and B. Dunlap, *Incorporation of Corrosion-Time and Effects of Corrosion-Product Spalling in Waste Package Degradation Simulation in the Potential Repository at Yucca Mountain*, W.J. Gray, and I.R. Triay (eds.), Scientific Basis for Nuclear Waste Management XX, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 465, pp. 1,075–1,082, 1997.

Lessart, P., et al., *Effects of Microbial Activity on the Near and Far Fields of a Deep Repository*, EUR 17110 EN, European Communities, Luxembourg, 1997.

Liang, L., and J.J. Morgan, *Chemical aspects of iron oxide coagulation in water: laboratory studies and implications for natural systems*, Aquatic Sciences, Vol. 52, pp. 32–55, 1990.

Lichtner, P.C., *Estimate of Near-Field Chemistry Based on MULTIFLO Calculations*, AI 20-5708-561-793, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Lichtner, P.C., and J.C. Walton, *Near-Field Liquid-Vapor Transport in a Partially Saturated High-Level Nuclear Waste Repository*, CNWRA 94-022, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1994.

Lichtner, P.C., and J. Eikenberg, *Propagation of a Hyperalkaline Plume Into the Geological Barrier Surrounding a Radioactive Waste Repository*, PSI Report No. 95-01, Paul Scherrer Institute, Wurenlingen, Switzerland, 1995.

Lichtner, P.C., and M.S. Seth, *Multiphase-multicomponent nonisothermal reactive transport in partially saturated porous media*, Proceedings of the International Conference on Deep Geological Disposal of Radioactive Waste, Canadian Nuclear Society, Winnipeg, Canada, pp. 3–133 to 3–142, 1996a.

Lichtner, P.C., and M.S. Seth, *User's Manual for MULTIFLO: Part II MULTIFLO 1.0 and GEM 1.0, Multicomponent-multiphase reactive transport model*, CNWRA 96-010, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1996b.

Lichtner, P.C., and D. R. Turner, *Critique of DOE's Near-Field Environment Modeling for the Proposed high-level waste Repository at Yucca Mountain, Nevada*, AI 20-5708-561-791, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Lichtner, P.C., R.T. Pabalan, and C.I. Steefel, *Preliminary Evaluation of the Effects of Alkaline Plume Migration on the Near-Field Environment of a High-Level Waste Geologic Repository*, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Lichtner, P.C., R.T. Pabalan, and C.I. Steefel, *Model calculations of porosity reduction resulting from cement-tuff diffusive interaction*, I.G. McKinley, and C. McCombie (eds.), Scientific Basis for Nuclear Waste Management XXI, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 506, pp. 709–718, 1998.

Lichtner, P.C., G. Keating, and B. Carey, *A Natural Analogue for Thermal-Hydrologic-Chemical Coupled Processes at the Proposed Nuclear Waste Repository at Yucca Mountain, Nevada*, LA-13610-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1999.

Lieser, K.H., and C. Bauscher, *Technetium in the hydrosphere and in the geosphere. II, Influence of pH, of complexing agents and of some minerals on the sorption of technetium*, Radiochimica Acta, Vol. 44/45, pp. 125–128, 1988.

Light, W.B., et al., *Analytical models for C-14 transport in a partially saturated, fractured, porous media*, Proceedings of Nuclear Waste Isolation in the Unsaturated Zone: Focus '89, American Nuclear Society, La Grange Park, Illinois, pp. 271–277, 1989.

Lin, W., and W.D. Daily, *Hydrological properties of Topopah Spring Tuff under a thermal gradient: Laboratory results*, International Journal of Rock Mechanics and Mineral Science and Geomechanical Abstracts, Vol. 27, pp. 373–385, 1990.

Lu, N., et al., *Reversibility of Sorption of Plutonium-239 onto Colloids of Hematite, Goethite, Smectite, and Silica*, LA–UR–98–3057, Los Alamos National Laboratory, Los Alamos, New Mexico, 1998.

Machesky, M.L., D.A. Palmer, and D.J. Wesolowski, *Hydrogen ion adsorption at the rutile-water interface to 250 °C*, Geochimica et Cosmochimica Acta, Vol. 58, pp. 5,627–5,632, 1994.

Maguire, M., *The pitting susceptibility of zirconium in aqueous Cl<sup>-</sup>, Br<sup>-</sup>, and I<sup>-</sup> solutions*, R.T. Webster, and C.S. Young (eds.), Industrial Applications of Titanium and Zirconium: Third Conference, ASM STP 830, American Society for Testing and Materials, Philadelphia, Pennsylvania, pp. 175–189, 1984.

Maiti, T.C., M.R. Smith, and J.C. Laul, *Colloid formation study of U, Th, Ra, Pb, Po, Sr, Rb, and Cs in briny (high ionic strength) groundwaters: Analog study for waste disposal*, Nuclear Technology, Vol. 84, pp. 82–87, 1989.

Manaktala, H., et al., *Potential Impact of Colloids on the Long-Term Performance of a High-level Radioactive Waste Repository*, CNWRA 95-015, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1995.

Manteufel, R.D., et al., *A Literature Review of Coupled Thermal-Hydrologic-Mechanical-Chemical Processes Pertinent to the Proposed Repository at Yucca Mountain*, NUREG/CR-6021, U.S. Nuclear Regulatory Commission, Washington, D.C., 1993.

Manteufel, R.D., et al., *Total-System Performance Assessment (TPA) Version 3.0 Code: Module Description and User's Guide*, IM 20-5708-762-730, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Massari, J.W., *Degradation Sequence of Internal Basket Structure and Commercial SNF After Waste Package Breach*, Presentation at the Performance Assessment Operations In-Drift Geochemical Environment and Engineered Barrier System Transport Workshop, April 13–15, 1999, Las Vegas, Nevada, TRW Environmental Safety Systems, Inc., North Las Vegas, Nevada, 1999.

Matyskiela, W., *Silica redistribution and hydrologic changes in heated fractured tuff*, Geology, Vol. 25 (12), pp. 1,115-1,118, 1997.

McCarthy, J.F., and J.M. Zachara, *Subsurface transport of contaminants*, Environmental Sciences Technology, Vol. 23, pp. 496–502, 1989.

McCauley, R.A., *Corrosion of Ceramics*, Marcel Dekker, New York, New York, 1995.

McCright, R.D., *Engineered Materials Characteristics Report for the Yucca Mountain Site Characterization Project, Volume 3, Revision 1.1: Corrosion Data and Modeling*, UCRL-ID-119564, Lawrence Livermore National Laboratory, Livermore, California, 1998.

McDowell-Boyer, L.M., J.R. Hunt, and N. Sitar, *Particle transport through porous media*, Water Resources Research, Vol. 22, pp. 1,901–1,921, 1986.

McKinley, I.G., and I Hagenlocher, *Quantification of microbial activity in a nuclear waste repository*, Abstract for International Symposium on Subsurface Microbiology, Bath, United Kingdom, 1993.

McKinley, I.G., J.M. West, and H.A. Grogan, An analytical overview of consequences of microbial activity in a Swiss HLW repository, Technical Report 85-43, Nagra, Baden, Switzerland, 1985.

McKinley, I.G. et al., *Microbiology in nuclear waste disposal: interfaces and reaction fronts*, Fems Microbiology Reviews, Vol. 20, No. 3–4, pp. 545–556, 1997.

McVay, G.L. and C.Q. Buckwalter, *Effect of Iron on Waste-Glass Leaching*. Journal of the American Ceramic Society, Vol. 66, pp. 170-173, 1983.

Means, J.L., A.S. Maest, and D.A. Crerar, *The Organic Geochemistry of Deep Ground Waters and Radionuclide Partitioning Experiments Under Hydrothermal Conditions*, Technical Report ONWI-448, Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, Ohio, 1983.

Meijer, A., *Yucca Mountain Project Far-Field Sorption Studies and Data Needs*, LA-11671-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1990.

Mohanty, S., et al., *Engineered Barrier System Performance Assessment code: EBSPAC Version 1.1. Technical Description and User's Manual*, CNWRA 97-006, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

Murphy, W.M., *Geochemical models for gas-water-rock interactions in a proposed nuclear waste repository at Yucca Mountain, Nevada*, Proceedings of Site Characterization and Model Validation: Focus '93, American Nuclear Society, La Grange Park, Illinois, pp. 115–121, 1993.

Murphy, W.M., *Retrograde solubilities of source term phases*, W.J. Gray and I.R. Triay (eds.), Scientific Basis for Nuclear Waste Management XX, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 465, pp. 713–720, 1997.

Murphy, W.M., and R.B. Codell, *Alternate source term models for Yucca Mountain performance assessment based on natural analog data and secondary mineral solubility*, D.J. Wronkiewicz and J. Lee (eds.), Scientific Basis for Nuclear Waste Management XXII, Materials Research Society, Warrendale, Pennsylvania, Symposium Proceedings 556, pp. 551–558, 1999.

Murphy, W.M., and R.T. Pabalan, *Geochemical Investigations Related to the Yucca Mountain Environment and Potential Nuclear Waste Repository*, NUREG/CR-6288, U.S. Nuclear Regulatory Commission, Washington, D.C., 1994.

Murphy, W.M., and E.C. Percy, *Source-term constraints for the proposed repository at Yucca Mountain, Nevada, derived from the natural analog at Peña Blanca, Mexico*, C.G. Sombret (ed.), Scientific Basis for Nuclear Waste Management XV, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 257, pp. 521–527, 1992.

Murphy, W.M., and J.D. Prikryl, *High-level waste near-field processes and variations*, in B. Sagar (ed.), Report on Research Activities July-December 1995, CNWRA 95-02S, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, pp. 10–1 to 10–14, 1996.

Murphy, W.M., M.S. Jarzemba, and P.C. Lichtner, *Comments on Bowman-Venneri*, AI 20-5702-541-525, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1995.

Murphy, W.M., E.C. Percy, and D.A. Pickett, *Natural analog studies at Peña Blanca and Santorini*, Seventh CEC Natural Analogue Working Group Meeting Proceedings, Commission of the European Communities, Luxembourg, pp. 105–112, 1997.

Murphy, W.M., et al., *Reaction kinetics and thermodynamics of aqueous dissolution and growth of analcime and Na-clinoptilolite at 25 °C*, American Journal of Science, Vol. 296, pp. 128–186, 1996.

Nash, J.T., H.C. Granger, and S.S. Adams, *Geology and concepts of genesis of important types of U deposits*, B.J. Skinner (ed.), Economic Geology 75th Anniversary Volume 1905–1980, pp. 63–116, 1981.

National Research Council, *Technical Bases for Yucca Mountain Standards*, National Academy Press, Washington, D.C., 1995.

Neall, F.B., *Modeling the long-term chemical evolution of cement-groundwater systems*, in W.M. Murphy and D.A. Knecht (eds.), Scientific Basis for Nuclear Waste Management XIX, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 412, pp. 483–490, 1996.

Needes, C.R.S., M.J. Nicol, and N.P. Finkelstein, *Electrochemical model for the leaching of U dioxide: 2—alkaline carbonate media*, A.R. Burkin (ed.), Leaching and Reduction in Hydrometallurgy, The Institution of Mining and Metallurgy, London, United Kingdom, pp. 12–19, 1975.

- Newman, J., *Electrochemical Systems*, Prentice Hall, Englewood Cliffs, New Jersey, 1991.
- Nishikata, A., et al., *Monitoring of corrosion rate of carbon steel under wet/dry cycle conditions and its corrosion mechanism*, Corrosion Engineering, Vol. 43, pp. 225–234, 1994.
- Nitao, J.J., *Numerical Modeling of the Thermal and Hydrological Environment Around a Nuclear Waste Package Using the Equivalent Continuum Approximation: Horizontal Emplacement*, UCID-21444, Lawrence Livermore National Laboratory, Livermore, California, 1988.
- Nitsche, H. et al., *Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region*. Milestone Report 3010, LA-12562-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1993.
- Nitsche, H. et al., *Measured Solubilities and Speciations from Oversaturation Experiments of Neptunium, Plutonium, and Americium in UE-25 p#1 Well Water from the Yucca Mountain Region*, LA-12563-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1994.
- Ogard, A.E., and J.F. Kerrisk, *Groundwater Chemistry Along Flow Paths Between A Proposed Repository Site and the Accessible Environment*, LA-10188-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1984.
- Olofsson, U., et al., *Formation and properties of americium colloids in aqueous systems*, in S.V. Topp (ed.), Scientific Basis for Nuclear Waste Management IV, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 6, pp. 191–198, 1982a.
- Olofsson, U., et al., *Properties and mobilities of actinide colloids in geologic systems*, in W. Lutze (ed.), Scientific Basis for Radioactive Waste Management V, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 11, pp. 755–764, 1982b.
- Pabalan, R.T., and K.S. Pitzer, *Thermodynamics of concentrated electrolyte mixtures and the prediction of mineral solubilities to high temperatures for mixtures in the system Na-K-Mg-Cl-SO<sub>4</sub>-OH-H<sub>2</sub>O*, Geochimica et Cosmochimica Acta, Vol. 51, pp. 2,429–2,443, 1987.
- Pabalan, R.T., and D.R. Turner, *Sorption modeling for high-level waste performance assessment*, in B. Sagar (ed.), NRC High-Level Radioactive Waste Research at CNWRA January Through June 1993, CNWRA 92-02S, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, pp. 8–1 to 8–18, 1993.
- Pabalan, R.T., and D.R. Turner, *U(6+) sorption on montmorillonite: Experimental and surface complexation modeling study*, Aqueous Geochemistry, Vol. 2, pp. 203–226, 1997.
- Pabalan, R.T., et al., *Experimental study of U(6+) sorption on the zeolite mineral clinoptilolite*, C. Interrante and R. Pabalan (eds.), Scientific Basis for Nuclear Waste Management XVI, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 294, pp. 777–782, 1993.

Pabalan, R.T., et al., *U<sup>VI</sup> sorption onto selected mineral surfaces*, E.A. Jenne (ed.), Adsorption of Metals by Geomedia, Academic Press, Inc., New York, New York, pp. 99–130, 1998.

Paces J.D., et al., *Ages and Origins of Subsurface Secondary Minerals In the Exploratory Studies Facility*, Milestone Report 3GQH450M, U.S. Geological Survey, Denver, Colorado, 1996.

Paquette, J., and R.J. Lemire, *A description of the chemistry of aqueous solutions of U and plutonium to 200 °C using potential-pH diagrams*, Nuclear Science and Technology, Vol. 79, pp. 26–48, 1981.

Parks, P.B., T.G. Williamson, and M.L. Hyder, *Consequences of the Bowman-Venneri Nuclear Excursion Thesis on the Prospects for Placing Vitrified Canisters in Geologic Repositories*, WSRC-TR-95-0036, Westinghouse Savannah River Company, Aiken, South Carolina, 1995.

Parrott, P.J., *Design for avoiding damage due to carbonation-induced corrosion*, V.M. Malhotra (ed.), Durability of Concrete, Third International Conference, SP-145, American Concrete Institute, Detroit, Michigan, pp. 283–298, 1994.

Pearcy, E. C., et al., *Alteration of uraninite from the Nopal I deposit, Peña Blanca District, Chihuahua, Mexico, compared to degradation of spent nuclear fuel in the proposed US high-level nuclear waste repository at Yucca Mountain, Nevada*, Applied Geochemistry, Vol. 9, pp. 713–732, 1994.

Pearcy, E.C., J.D. Prikryl, and B.W. Leslie, *U transport through fractured silicic tuff and relative retention in areas with distinct fracture characteristics*, Applied Geochemistry, Vol. 10, pp. 685–704, 1995.

Pedersen, K., *Investigations of subterranean bacteria in deep crystalline bedrock and their importance for the disposal of nuclear waste*, Canadian Journal of Microbiology, Vol. 42, No. 4, pp. 382–391, 1996.

Pedersen, K., and F. Karlsson, *Investigation of Subterranean Microorganisms—Their Importance for Performance Assessment of Radioactive Waste Disposal*, SKB TR 95-10, Swedish Nuclear Fuel and Waste Management Co., Stockholm, Sweden, 1995.

Pedersen, K., M. Motamedi, and O. Karnland, *Survival of Bacteria in Nuclear Waste Buffer Materials—The Influence of Nutrients, Temperature and Water Activity*, SKB TR 95-27, Swedish Nuclear Fuel and Waste Management Co., Stockholm, Sweden, 1995.

Peters, R.R., et al., *Fracture and Matrix Hydrologic Characteristics of Tuffaceous Materials from Yucca Mountain, Nye County, Nevada*, SAND84-1471, Sandia National Laboratories, Albuquerque, New Mexico, 1984.

Pickett, D.A., and W.M. Murphy, *Isotopic constraints on radionuclide transport at Peña Blanca, Seventh CEC Natural Analogue Working Group Meeting Proceedings*, Commission of the European Communities, Luxembourg, pp. 113–122, 1997.

Pickett, D.A., and B.W. Leslie, *An Audit of the U.S. Department of Energy Treatment of Features, Events, and Processes at Yucca Mountain, Nevada, with Emphasis on the Near-Field Environment*, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1999.

Pierce, A.P., J.W. Mytton, and G.B. Gott, *Radioactive elements and their daughter products in the Texas Panhandle and other oil and gas fields in the United States*, Geology of U and Thorium, International Conference, 1955, USGS Professional Paper 300, U.S. Geological Survey, Washington, D.C., pp. 527–532, 1955.

Pierce, A.P., G.B. Gott, and J.W. Mytton, *U and helium in the Panhandle gas field, Texas, and adjacent areas*, USGS Professional Paper 454-G, U.S. Geological Survey, Washington, D.C., pp. G1–57, 1964.

Potter, E.C., and G.M.W. Mann, *Oxidation of Mild Steel in High-Temperature Aqueous Systems*, Proceedings of First International Congress on Metallic Corrosion, Butterworths, London, England, pp. 417–423, 1962.

Pruess, K., J.S Y. Wang, and Y.W. Tsang, *On thermohydrologic conditions near high-level nuclear wastes emplaced in partially saturated fractured tuff. 1. Simulation studies with explicit consideration of fracture effects*, Water Resources Research, Vol. 26, pp. 1,235–1,248, 1990.

Ransom, B., and H.C. Helgeson, *Estimation of the standard molal heat capacities, entropies, and volumes of 2:1 clay minerals*, Geochimica et Cosmochimica Acta, Vol. 58, pp. 4,537–4,547, 1994.

Reardon, E.J., *An ion interaction model for determining ion equilibria in cement/water systems*, Cement Concrete Research, Vol. 20, pp. 175–192, 1990.

Reardon, E.J., *Problems and approaches to the prediction of the chemical composition in cement/water systems*, Waste Management, Vol. 12, pp. 221–239, 1992.

Rechard, R.P., ed., *Performance Assessment of the Direct Disposal in Unsaturated Tuff of Spent Nuclear Fuel and High-Level Waste Owned by U.S. Department of Energy*, SAND94-2563, Volumes. 1–3, Sandia National Laboratories, Albuquerque, New Mexico, 1995.

Reed, D.T., and R. A. Van Konynenburg, *Effect of Ionizing Radiation on Moist Air System*, in M.J. Apter and R.E. Westerman (eds.), Scientific Basis for Nuclear Waste Management XI, Material Research Society, Pittsburgh, Pennsylvania, Symposium Proceeding 112, pp. 393–404, 1987.

Rimstidt, J.D., W.D. Newcomb, and D.L. Shettel, Jr., *A vertical thermal gradient experiment to simulate conditions in vapor dominated geothermal systems, epithermal gold deposits, and high-level radioactive repositories in unsaturated media*, Proceedings of the 4th International Symposium on Water-Rock Interaction, A.A. Balkema, Rotterdam, Netherlands, pp. 585–588, 1989.

Robert, M., and M. Terce, *Effect of gels and coatings on clay mineral chemical properties*, B. Bar-Yosef, N.J. Barrow, and J. Goldshmid (eds.), Inorganic Contaminants in the Vadose Zone: Ecological Studies Volume 74, Springer-Verlag, New York, New York, pp. 57–71, 1989.

Ryan, J.N., and P.M. Gschwend, *Colloid mobilization in two Atlantic coastal plain aquifers: Field studies*, Water Resources Research, Vol. 26, pp. 307–322, 1990.

Sagar, B., ed., *NRC High-Level Radioactive Waste Program Annual Progress Report: Fiscal Year 1996*, NUREG/CR-6513, No.1, Nuclear Regulatory Commission, Washington, D.C., 1997.

Sanchez, R., et al., *Criticality Characteristics of Mixtures of Plutonium, Silicon Dioxide, Nevada Tuff, and Water*, LA-UR-95-21130, Los Alamos, National Laboratory, Los Alamos, New Mexico, 1995.

Santanán, L., S. Raghavan, and B.A. Chin, *Zircaloy cladding rupture during repository storage*, Nuclear Technology, Vol. 97, pp. 316–322, 1992.

Sassani, D.C., and E.R. Siegmann, *Constraints on Solubility-Limited Neptunium Concentrations for Use in Performance Assessment Analyses*, Report No. 00000000-01717-2200-00191, Revision 00, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1998.

Savage, D., *Review of Potential Effects of Alkaline Plume Migration from a Cementitious Repository for Radioactive Waste*, R&D Technical Report P60, Environmental Agency, Bristol, United Kingdom, 1997.

Shoesmith, D.W., *Fuel Corrosion Processes Under Waste Disposal Conditions*, AECL-12034, AECL Technologies, Inc., Pinawa Manitoba, Canada, 1999.

Shoesmith, D.W., and S. Sunder, *The prediction of nuclear fuel (UO<sub>2</sub>) dissolution rates under waste disposal conditions*, Journal of Nuclear Materials, Vol. 190, pp. 20–35, 1992.

Shoesmith, D.W., et al., *Oxidation of CANDU UO<sub>2</sub> fuel by the alpha-radiolysis products of water*, L.O. Werme (ed.), Scientific Basis for Nuclear Waste Management IX, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 50, pp. 309–316, 1985.

Shoesmith, D.W., S. Sunder, and J.C. Tait, *Validation of Oxidative Dissolution Model for Used CANDU Fuel*, AECL-11798, COG-97-015-1, Whiteshell Laboratories, Pinawa Manitoba, Canada, 1997.

Shoesmith, D.W., et al., *The corrosion of nuclear fuel (UO<sub>2</sub>) in oxygenated solutions*, Corrosion Science, Vol. 29, pp. 1,115–1,128, 1989.

Shoesmith D. W., *Review of the Expected Behaviour of Alpha Titanium Alloys Under Yucca Mountain Conditions, Revision 00*, AECL-12089, AECL Technologies, Inc., Pinawa Manitoba, Canada, 2000.

Southwell, C.R., and A.L. Alexander, *Corrosion of Metals in Tropical Waters—Structural Ferrous Metals*, Materials Protection, Vol. 9, No. 1, pp. 14–23, 1970.

Spinks, J.W.T., and R.J. Woods, *Introduction to Radiation Chemistry*, Wiley-Interscience, New York, New York, 1976.

Spinks, J.W.T., and R.J. Woods, *Introduction to Radiation Chemistry, 3<sup>rd</sup> Edition*, John Wiley & Sons, Inc., New York, New York, 1990.

Sridhar, N., and D.S. Dunn, *Effect of applied potential on changes in solution chemistry inside crevices on type 304L stainless steel and alloy 825*, The Journal of Science and Engineering Corrosion, Vol. 50, No. 11, pp. 857–872, 1994.

Sridhar, N., P.C. Lichtner, and D. Dunn, *The Evolution of Chemistry Inside Disbonded Coatings of Cathodically Protected Steel Pipeline—Modeling and Experimental Studies*, Final Report of Internal Research Project 20-9932, Southwest Research Institute, San Antonio, Texas, 1996.

Staudigel, H., et al., *Biologically mediated dissolution of glass*, Chemical Geology, Vol. 126, pp. 147–154, 1995.

Steeffel, C., and P.C. Lichtner, *Diffusion and reaction in rock matrix bordering a hyperalkaline fluid-filled fracture*, Geochimica et Cosmochimica Acta, Vol. 58, pp. 3,595–3,612, 1994.

Stout, R.B., and H.R. Leider, *Waste Form Characteristics Report, Version 1.3*, UCRL-ID-108314, Lawrence Livermore National Laboratory, Livermore, California, 1998.

Stroes-Gascoyne, S., *Microbial studies in the Canadian nuclear fuel waste management program*, Proceedings of the International Conference on Deep Geological Disposal of Radioactive Waste, Canadian Nuclear Society, Winnipeg, Canada, pp. 3–191 to 3–202, 1996.

Stroes-Gascoyne, S., and M. Gascoyne, *The introduction of microbial nutrients into a nuclear waste disposal vault during excavation and operation*, Environmental Science & Technology, Vol. 32, No. 3, pp. 317–326, 1998.

Stroes-Gascoyne, S., et al., *Microbial Analysis of the Buffer/Container Experiment at AECL's Underground Research Laboratory*, SKB TB 96-02, Swedish Nuclear Fuel and Waste Management Co., Stockholm, Sweden, 1996.

Suarez, D.L., et al., *Effect of pH on saturated hydraulic conductivity and soil dispersion*, Soil Science Society of America Journal, Vol. 48, pp. 50–55, 1984.

Sunder, S., and H. Christensen, *Gamma radiolysis of water solutions relevant to the nuclear fuel waste management program*, Nuclear Technology, Vol. 104, pp. 403–417, 1993.

Sunder, S., D. Shoesmith, and N.H. Miller, *Oxidation and dissolution of nuclear fuel (UO<sub>2</sub>) by the products of alpha radiolysis of water*, Journal of Nuclear Materials, Vol. 244, pp. 66–74, 1997.

Sunder, S., et al., *Oxidation of UO<sub>2</sub> fuel by the products of gamma radiolysis of water*, Journal of Nuclear Materials, Vol. 190, pp. 78–86, 1992.

Swift, P., et al., *Features, Events, and Processes Screening and Scenario Development for the Yucca Mountain Total Systems Performance Assessment*, SAND98–2831C, Sandia National Laboratories, Albuquerque, New Mexico, 1999.

Thomas, D.M., and I. Gudmundsson, *Advances in the study of solids deposition in geothermal systems*, Geothermics, Vol. 18, pp. 5–15, 1989.

Thorseth, I.H., H. Furnes, and O. Tumyr, *Textural and chemical effects of bacterial activity on basaltic glass: An experimental approach*, Chemical Geology, Vol. 119, pp. 139–160, 1995.

Thorstenson, D.C., et al., *Physical and chemical characteristics of topographically affected airflow in an open borehole at Yucca Mountain, Nevada*, Proceedings of Nuclear Waste Isolation in the Unsaturated Zone: Focus '89, American Nuclear Society, La Grange Park, Illinois, pp. 256–270, 1990.

Thorstenson, D.C., et al., *Chemistry of unsaturated zone gases sampled in open boreholes at the crest of Yucca Mountain, Nevada: Data and basic concepts of chemical and physical processes in the mountain*, Water Resources Research, Vol. 34, No. 6, pp. 1,507–1,529, 1998.

Tombacz, E., et al., *The pH-dependent colloidal stability of aqueous montmorillonite suspensions*, Colloids and Surfaces, Vol. 49, pp. 71–80, 1990.

Travis, B.J., and H.E. Nuttall, *Analysis of colloid transport*, L.O. Werme (ed.), Scientific Basis for Nuclear Waste Management IX, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 50, pp. 737–745, 1985.

Travis, B.J., and H.E. Nuttall, *Two-Dimensional Numerical Simulation of Geochemical Transport in Yucca Mountain*, LA-10532-MS, Los Alamos National Laboratory, Los Alamos, New Mexico, 1987.

TRW Environmental Safety Systems, Inc., *Total System Performance Assessment—1995: An Evaluation of the Potential Yucca Mountain Repository*, B00000000-01717-2200-00136, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1995.

TRW Environmental Safety Systems, Inc., *Status/Summary Report for Fiscal Year 1996 Activities Within the Performance Assessment Overview Study on the Consequences of Cementitious Materials*, LV.PA.DCS.09/96-038, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1996.

TRW Environmental Safety Systems, Inc., *Reference Design Description for a Geologic Repository*, B00000000-01717-5707-00002, Revision 01, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1997a.

TRW Environmental Safety Systems, Inc., *Total System Performance Assessment—Viability Assessment (TSPA-VA) Methods and Assumptions, Revision 01*, B00000000-01717-2200-00193, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1997b.

TRW Environmental Safety Systems Inc., *Environment on the Surfaces of the Drip Shield and Waste Package Outer Barrier, Revision 00B, Preliminary Draft*, ANL-EB-MD-000001, TRW Environmental Safety Systems Inc., Las Vegas Nevada, 1999.

TRW Environmental Safety Systems Inc., *General Corrosion and Localized Corrosion of Waste Package Outer Barrier, Revision 00, Preliminary Draft*, ANL-EB-MD-000003, TRW Environmental Safety Systems Inc., Las Vegas, Nevada, 2000a.

TRW Environmental Safety Systems Inc., *General Corrosion and Localized Corrosion of Drip Shield, Revision 00, Preliminary Draft*, ANL-EB-MD-000004, TRW Environmental Safety Systems Inc., Las Vegas, Nevada, 2000b.

TRW Environmental Safety Systems Inc., *Hydrogen Induced Cracking of Drip Shield, Revision 00, Preliminary Draft*, ANL-EB-MD-000006, TRW Environmental Safety Systems Inc., Las Vegas, Nevada, 2000c.

TRW Environmental Safety Systems Inc., *Waste Package Degradation Process Model Report, Revision 00, Preliminary Draft*, TDR-WIS-MD-000002, TRW Environmental Safety Systems Inc., Las Vegas, Nevada, 2000d.

Tsang, Y.W., and K. Pruess, *A study of thermally induced convection near a high-level nuclear waste repository in partially saturated fractured tuff*, Water Resources Research, Vol. 23, pp. 1,958–1,966, 1987.

Turner, D.R., R.T. Pabalan, and F.P. Bertetti, *Neptunium(V) sorption on montmorillonite: An experimental and surface complexation modeling study*, Clays and Clay Minerals, Vol. 46, pp. 256–269, 1998.

U.S. Department of Energy, *Site Characterization Plan, Yucca Mountain Site, Nevada Research and Development Area, NV*, DOE/RW-0199, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, Washington, D.C., 1988.

U.S. Department of Energy, *Civilian Radioactive Waste Management Program Plan, Revision 1*, DOE/RW-0458, Revision 1, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, Washington, D.C., 1996a.

U.S. Department of Energy, *[Draft] Highlights of the U.S. Department of Energy's Updated Waste Containment and Isolation Strategy for the Yucca Mountain Site*, enclosure to a letter dated July 19, 1996, from S.J. Brocoum, U.S. Department of Energy, to M.V. Federline, U.S. Nuclear Regulatory Commission, 1996b.

U.S. Department of Energy, *Site Characterization Progress Report: Yucca Mountain, Nevada, October 1, 1996–March 31, 1997, Number 16*, DOE/RW-0501, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, Washington, D.C., 1997a.

U.S. Department of Energy, *Responses to Nuclear Regulatory Commission (NRC) Comments on DOE's Thermohydrology Testing and Modeling Program*, letter dated July 14, 1997, from S.J. Brocoum, U.S. Department of Energy, to M.J. Bell, U.S. Nuclear Regulatory Commission, Washington, D.C., 1997b.

U.S. Department of Energy, *Repository Safety Strategy: U.S. Department of Energy's Strategy to Protect Public Health and Safety After Closure of a Yucca Mountain Repository*, YMP/96-01 Revision 2, U.S. Department of Energy, Washington, D.C., 1998a.

U.S. Department of Energy, *Viability Assessment of a Repository at Yucca Mountain, Volume 3: Total System Performance Assessment*, DOE/RW-0508/V3, U.S. Department of Energy, Office of Civilian Radioactive Waste Management, North Las Vegas, Nevada, 1998b.

U.S. Department of Energy, *Viability Assessment of a Repository at Yucca Mountain, Volume 2: Preliminary Design Concept for the Repository and Waste Package*, DOE/RW-0508/V2, U.S. Department of Energy, Office of Civilian Radioactive Waste Management, North Las Vegas, Nevada, 1998c.

U.S. Department of Energy, *Viability Assessment of a Repository at Yucca Mountain, Volume 4: License Application Plan and Costs*, DOE/RW-0508/V4, U.S. Department of Energy, Office of Civilian Radioactive Waste Management, North Las Vegas, Nevada, 1998d.

U.S. Department of Energy, *Disposal Criticality Analysis Methodology Topical Report*, YMP/TR-004Q, Revision 0, North Las Vegas, Nevada, 1998e.

U.S. Department of Energy, *Yucca Mountain Features, Events, and Processes Database, Revision 00b Preliminary Version*, U.S. Department of Energy, Washington, D.C., 1999.

U.S. Department of Energy, *U.S. Department of Energy Review of U.S. Nuclear Regulatory Commission's Evolution of the Near-Field Environment Issue Resolution Status Report, Revision 2*, letter dated March 22, 2000 from S. Brocoum, U.S. Department of Energy, to C.W. Reamer, U.S. Nuclear Regulatory Commission, 2000.

U.S. Nuclear Regulatory Commission, *Determination of Radionuclide Solubility in Groundwater for Assessment of High-Level Waste Isolation*, Technical Position, November 1984, U.S. Nuclear Regulatory Commission, Washington, D.C., 1984.

U.S. Nuclear Regulatory Commission, *NRC Staff Site Characterization Analysis of the Department of Energy's Site Characterization Plan, Yucca Mountain Site, Nevada*, NUREG-1347, U.S. Nuclear Regulatory Commission, Washington, D.C., 1989.

U.S. Nuclear Regulatory Commission, *NRC Staff Evaluation of DOE Response to NRC Site Characterization Analysis of the DOE Site Characterization Plan, Yucca Mountain, Nevada*, letter dated July 31, 1991, from R. Bernero, U.S. Nuclear Regulatory Commission, to R. J. Bartlett, U.S. Department of Energy, 1991.

U.S. Nuclear Regulatory Commission, *Review of DOE Response to NRC Comments on Study Plan 8.3.1.3.2.1 Mineralogy, Petrology, and Chemistry of Transport Pathways*, memorandum dated July 2, 1992, from M.V. Federline, U.S. Nuclear Regulatory Commission, to J. Holonich, U.S. Nuclear Regulatory Commission, 1992.

U.S. Nuclear Regulatory Commission, *Review of Potential for Underground Autocatalytic Criticality*, letter dated August 7, 1995, from C.J. Paperiello, U.S. Nuclear Regulatory Commission, to L. Barrett, U.S. Department of Energy, 1995.

U.S. Nuclear Regulatory Commission, *Transmittal of the Center for Nuclear Waste Regulatory Analyses Detailed Report Related to the Audit Review of the U.S. Department of Energy's 1995 Total System Performance Assessment*, letter and enclosure dated November 5, 1996, from J.H. Austin, U.S. Nuclear Regulatory Commission, to R.A. Milner, U.S. Department of Energy, 1996.

U.S. Nuclear Regulatory Commission, *Comments on the Department of Energy Thermohydrology Testing and Modeling Program*, letter dated January 23, 1997, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1997a.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Evolution of the Near-Field Environment, Revision 0)*, enclosure to letter dated November 7, 1997, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1997b.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Thermal Effects of Flow, Revision 1)*, enclosure to letter dated October 1, 1998, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998a.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Repository Design and Thermal Mechanical Effects, Revision 1)*, enclosure to letter dated October 1, 1998, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998b.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Total System Performance Assessment and Integration, Revision 1)*, enclosure to letter dated December 8, 1998, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998c.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Container Life and Source Term, Revision 1)*, enclosure to letter dated December 1, 1998, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998d.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Evolution of the Near-Field Environment, Revision 1)*, enclosure to letter dated August 28, 1998, from N.K. Stablein, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998e.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Radionuclide Transport, Revision 0)*, enclosure to letter dated December 23, 1998, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998f.

U.S. Nuclear Regulatory Commission, *U.S. Nuclear Regulatory Commission Comments on the U.S. Department of Energy Total System Performance Assessment*, letter dated July 6, 1998, from M.J. Bell, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1998g.

U.S. Nuclear Regulatory Commission, *10 CFR Part 19 et al. : Disposal of High-Level Radioactive Wastes in a Proposed Repository at Yucca Mountain, Nevada; Proposed Rule, Federal Register*, Vol. 64, no. 34, February 22, 1999, pp. 8640 - 8676, 1999a.

U.S. Nuclear Regulatory Commission, *U.S. Nuclear Regulatory Commission Staff Review of the U.S. Department of Energy Viability Assessment for a High-Level Radioactive Waste Repository at Yucca Mountain, Nevada*, letter dated June 2, 1999, from C.J. Paperiello, U.S. Nuclear Regulatory Commission, to L.H. Barrett, U.S. Department of Energy, 1999b.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Thermal Effects on Flow, Revision 2)*, letter dated September 15, 1999, from C.W. Reamer, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1999c.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Container Life and Source Term, Revision 2)*, U.S. Nuclear Regulatory Commission, Washington, DC, 1999d.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Unsaturated and Saturated Flow Under Isothermal Conditions, Revision 2)*, letter dated July 12, 1999, from C.W. Reamer, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1999e.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Evolution of the Near-Field Environment, Revision 2)*, letter dated July 14, 1999, from C.W. Reamer, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1999f.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Radionuclide Transport, Revision 1)*, U.S. Nuclear Regulatory Commission, Washington, DC, 1999g.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Total System Performance Assessment and Integration, Revision 2)*, U.S. Nuclear Regulatory Commission, Washington, DC, 2000a.

U.S. Nuclear Regulatory Commission, *Safety Evaluation Report for Disposal Criticality Analysis Methodology Topical Report, Revision 0, June 2000*, U.S. Nuclear Regulatory Commission, Washington, DC, 2000b.

U.S. Nuclear Waste Technical Review Board, *Moving Beyond the Yucca Mountain Viability Assessment: A Report to the U.S. Congress and the Secretary of Energy*, April, 1999, Arlington, Virginia, 1999.

van Cappellen, P., et al., *A surface complexation model of the carbonate mineral-aqueous solution interface*, *Geochimica et Cosmochimica Acta*, Vol. 57, pp. 3,505–3,518, 1993.

Van Konynenburg, R.A., *Radiation Chemical Effects in Experiments to Study the Reaction of Glass in an Environment of Gamma-Irradiated Air, Groundwater, and Tuff*, UCRL-53719, Lawrence Livermore National Laboratory, Livermore, California, 1986.

Van Konynenburg, R.A., *Comments on the "Draft Paper Underground Supercriticality from Plutonium and Other Fissile Material" written by C.D. Bowman and F. Venneri (LANL)*, UCRL-ID-120990, Lawrence Livermore National Laboratory, Livermore, California, 1995.

Vaughan, P.J., *Analysis of permeability reduction during flow of heated, aqueous fluid through Westerly Granite*, C.-F. Tsang (ed.), *Coupled Processes Associated with Nuclear Waste Repositories*, Academic Press Inc., New York, New York, pp. 529–539, 1987.

Verma, K., and K. Pruess, *Thermohydrological conditions and silica redistribution near high-level nuclear waste emplaced in saturated geological formations*, Journal of Geophysical Research, Vol. 93(B2), pp. 1,159–1,173, 1988.

Walton, J.C., *Effects of evaporation and solute concentration on presence and composition of water in and around the waste package at Yucca Mountain*, Waste Management, Vol. 13, pp. 293–301, 1993.

Walton, F.B., J.P.M. Ross, and D.G. Juhnke, *The effects of simultaneous heat and mass transport on radionuclide migration*, C.M. Jantzen, J.A. Stone, and R.C. Ewing (eds.), Scientific Basis for Nuclear Waste Management VIII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 44, pp. 663–672, 1985.

Wan, J., and J.L. Wilson, *Visualization of the role of the gas-water interface on the fate and transport of colloids in porous media*, Water Resources Research, Vol. 30, pp. 11–23, 1994.

Wan, J., J.L. Wilson, and T.L. Kieft, *Influence of the gas-water interface on transport of microorganisms through unsaturated porous media*, Applied and Environmental Microbiology, Vol. 60, No. 2, pp. 509–516, 1994.

Wang, F., et al., *Long term corrosion study of waste package candidate material for the YMP; initial results*, CORROSION98, Paper 162, NACE International, Houston, Texas, 1998.

Wescott, R.G., et al., *NRC Iterative Performance Assessment Phase 2*, NUREG-1464, U.S. Nuclear Regulatory Commission, Washington, D.C., 1995.

West, J.M., *A review of progress on the geomicrobiology of radioactive waste disposal*, Radioactive Waste Management and Environmental Restoration, Vol. 19, pp. 263–283, 1995.

West, J.M., et al., *The microbiology of the Maqarin site Jordan—A natural analogue for cementitious radioactive waste repositories*, T. Murakami and R.C. Ewing (eds.), Scientific Basis for Nuclear Waste Management XVIII, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 353, pp. 181–188, 1995.

West, J.M., et al., *Alteration of repository structural materials within the first few years*, I.G. McKinley, and C. McCombie (eds.), Scientific Basis for Nuclear Waste Management XXI, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 506, pp. 503–510, 1998.

Whipple, C., et al., *Final Report: Total System Performance Assessment Peer Review Panel, February 11, 1999*, Las Vegas, Nevada, 1999.

White, A.F., H.C. Claassen, and L.V. Benson, *The effect of dissolution of volcanic glass on the water chemistry in a tuffaceous aquifer, Rainier Mesa, Nevada*, Water-Supply Paper 1535-Q, U.S. Geological Survey, Washington, D.C., 1980.

Wilder, D.G., *Near-Field Altered-Zone Environment Report Volume II*, UCRL-LR-124998, Lawrence Livermore National Laboratory, Livermore, California, 1996.

Willis, C. and A.W. Boyd, *Excitation in the Radiation Chemistry of Inorganic Gases*, International Journal Radiation Physics and Chemistry, Vol. 8, pp. 71–111, 1976.

Wilson, C.N., *Results from NNWSI Series 3 Spent Fuel Dissolution Tests*, PNL-7170, Pacific Northwest Laboratory, Richland, Washington, 1990.

Wilson, M.L., et al., *Total System Performance Assessment for Yucca Mountain—SNL Second Iteration (TSPA-93)*, SAND93-2675, Prepared for U.S. Department of Energy by Sandia National Laboratory, Albuquerque, New Mexico, 1994.

Wolery, T.J., *EQ3/6, A Software Package for Geochemical Modeling of Aqueous Systems: Package Overview and Installation Guide (Version 7.0)*, UCRL-MA-110662, Lawrence Livermore National Laboratory, Livermore, California, 1992.

Wronkiewicz, D.J., J.E. Young, and J.K. Bates, *Effects of alpha and gamma radiation on glass reaction in an unsaturated environment*, T. Abrajano, Jr., and L.H. Johnson (eds.), Scientific Basis for Nuclear Waste Management XIV, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 212, pp. 99–106, 1991.

Wronkiewicz, D.J., et al., *U release and secondary phase formation during unsaturated testing of  $UO_2$  at 90 °C*, Journal of Nuclear Materials, Vol. 190, pp. 107–126, 1992.

Wronkiewicz, D.J., et al., *Effects of radiation exposure on glass alteration in a steam environment*, C.G. Interrante, and R.T. Pabalan (eds.), Scientific Basis for Nuclear Waste Management XVI, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 294, pp. 183–190, 1993.

Yang, I.C., *Flow and transport through unsaturated rock—data from two test holes, Yucca Mountain, Nevada*, Proceedings of the Third International Conference on High Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, pp. 732–737, 1992.

Yang, I.C., C.A. Peters, and D.C. Thorstenson, *Carbon isotopic data from test hole USW UZ-1, Yucca Mountain, Nevada*, Proceedings of the Fourth International Conference on High Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, pp. 401–406, 1993.

Yang, I.C., G.W. Rattray, and P. Yu, *Interpretation of Chemical and Isotopic Data from Boreholes in the Unsaturated Zone at Yucca Mountain, Nevada*, Water Resources Investigations Report 96-4058, U.S. Geological Survey, Denver, Colorado, 1996.

Yang, I.C., et al., *Hydrochemical Investigations and Geochemical Modeling in Characterizing the Unsaturated Zone at Yucca Mountain, Nevada [Draft]*, U.S. Geological Survey, Denver, Colorado, 1996.

Zachara J.M., and J.P. McKinley, *Influence of hydrolysis on the sorption of metal cations by smectites: Importance of edge coordination reactions*, *Aquatic Sciences*, Vol. 55, pp. 250–261, 1993.

## **APPENDIX A**

### **FIGURE ILLUSTRATING ELEMENTS OF THE NUCLEAR REGULATORY COMMISSION TOTAL SYSTEM PERFORMANCE ASSESSMENT**

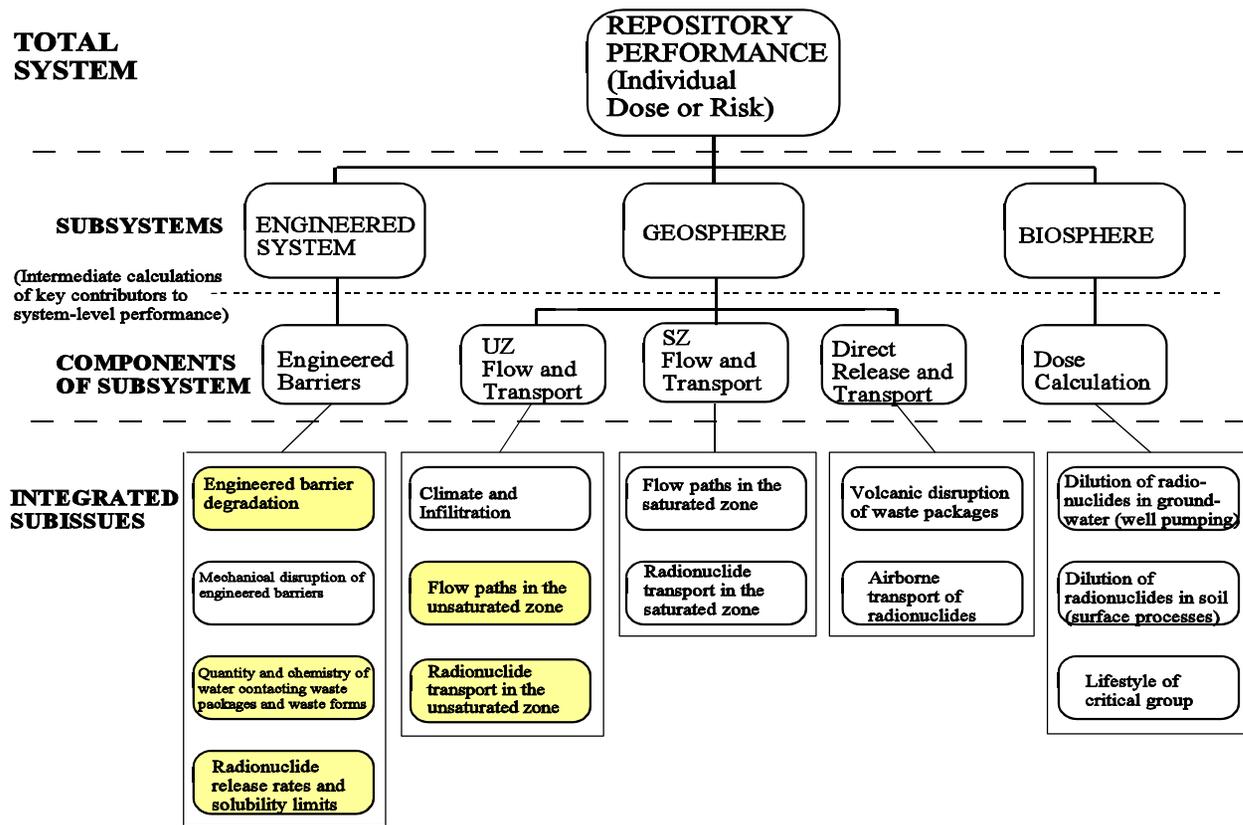


Figure 2. Flow diagram for total system performance assessment. This key technical issue, Evolution of the Near-Field Environment, provides input to the highlighted key elements.