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 2

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Change History of "Issue Resolution Status Report (IRSR), Key Technical Issue: Evolution of the Near-Field Environment (ENFE)"

Revision #	Section/ Paragraph	Date	Modification
Rev0	all	September 1997	None. Initial issue.
Rev1	2.0/all	August 1998	Redefinition/clarification of subissues; addition of new subissue on near-field criticality
Rev1	3.0/1, 2 & 3	August 1998	Expanded discussion of relationship to repository performance
Rev1	3.1/1, 2	August 1998	Updated statement of DOE hypotheses based on DOE Repository Safety Strategy
Rev1	3.2/2, 3	August 1998	Expanded discussion of relationship between ENFE IRSR and other IRSR and repository performance
Rev1	3.2.1/all	August 1998	Minor modifications and clarification of subissue on seepage and flow
Rev1	3.2.2/all	August 1998	Minor modifications and clarification of subissue on waste package (WP) chemical environment
Rev1	3.2.3/all	August 1998	Minor modifications and clarification of subissue on chemical environment for radionuclide release
Rev1	3.2.4/all	August 1998	Minor modifications and clarification of subissue on effects of coupled processes on radionuclide transport (RT)
Rev1	3.2.5/all	August 1998	New section on potential nuclear criticality in the near field
Rev1	3.3/1	August 1998	Minor modification to reflect expanded discussions in Sections 3.3.3 - 3.3.5
Rev1	3.3.3/all	August 1998	New section on TSPA-VA Methods and Assumptions
Rev1	3.3.4/1	August 1998	Minor modification of Section 3.3.3 in Rev0
Rev1	3.3.5/all	August 1998	Added extensive discussion of Section 3.3.4 in Rev0

Revision #	Section/ Paragraph	Date	Modification
Rev1	3.3.6/1	August 1998	The same as Section 3.3.5 in Rev0
Rev1	3.4/1, 3	August 1998	Minor modifications to reflect results of sensitivity studies
Rev1	3.4.1/all	August 1998	Updated section reflecting sensitivity study results
Rev1	3.4.2/all	August 1998	Updated section reflecting sensitivity study results
Rev1	3.4.3/all	August 1998	Updated section reflecting sensitivity study results
Rev1	3.4.4/all	August 1998	Updated section reflecting sensitivity study results and addition of new source term model
Rev1	4.0/1, 2	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
Rev1	4.1/1	August 1998	Minor modification to reflect revised subissue definition
Rev1	4.1.1/all	August 1998	Reorganized and expanded AC to reflect TSPA IRSR AC; added Review Methods (RM); added AC to reflect performance confirmation program; revised AC to improve specificity
Rev1	4.1.2.1/3, 9	August 1998	Added reference on near-field analog and important DOE reference
Rev1	4.1.2.2/2 - 4	August 1998	Minor modifications to reflect recent references
Rev1	4.1.2.3/all	August 1998	Section added on microbial effects on seepage and flow
Rev1	4.2.1/all	August 1998	Reorganized and expanded AC to reflect TSPA IRSR AC; added RM; added AC to reflect performance confirmation program; added AC to address potential microbial effects; revised AC to improve specificity

Revision #	Section/ Paragraph	Date	Modification
Rev1	4.2/1	August 1998	Minor modification to reflect revised subissue definition
Rev1	4.2.2/1	August 1998	Minor modification to reflect revised subissue definition
Rev1	4.2.2.1/all	August 1998	Reorganized to clarify discussion; provided additional technical basis to dismiss the importance of self potentials to repository performance
Rev1	4.2.2.2/all	August 1998	Reduced discussion on WP corrosion processes found in Section 4.2.2.1 Rev0 to reflect expanded discussion in CLST IRSR Rev1; revised discussion to reflect new WP design
Rev1	4.2.2.3/2, 3	August 1998	Minor modification of Section 4.2.2.2 Rev0
Rev1	4.2.2.4/all	August 1998	Section added on microbial effects on WP chemical environment
Rev1	4.3/1	August 1998	Minor modification to reflect revised subissue definition
Rev1	4.3.1/all	August 1998	Reorganized and expanded AC to reflect TSPA IRSR AC; added RM; added AC to reflect performance confirmation program; added AC to address potential microbial effects; revised AC to improve specificity
Rev1	4.3.2/1	August 1998	Minor modification to reflect revised subissue definition
Rev1	4.3.2.1/all	August 1998	Reorganized to clarify discussion and to address cladding degradation
Rev1	4.3.2.4/all	August 1998	Section added on microbial effects on the chemical environment for radionuclide release
Rev1	4.4/1	August 1998	Minor modification to reflect revised subissue definition

Revision #	Section/ Paragraph	Date	Modification
Rev1	4.4.1/all	August 1998	Reorganized and expanded AC to reflect TSPA IRSR AC; added RM; added AC to reflect performance confirmation program; added AC to address potential microbial effects; revised AC to improve specificity
Rev1	4.4.2/1	August 1998	Minor modification to reflect revised subissue definition
Rev1	4.4.2.1/all	August 1998	Reorganized to clarify discussion; reduced discussion of RT processes to reflect expanded discussion in RT IRSR Rev0
Rev1	4.4.2.4/all	August 1998	Section added on microbial effects on RT
Rev1	4.5/all	August 1998	New subissue on potential near-field criticality with RM, AC, and technical bases identified
Rev1	5.0/all	August 1998	Reorganized and clarified to reflect expanded discussions in Section 5.1 - 5.5
Rev1	5.1/all	August 1998	Revised site characterization analysis ope items' status and their description to refle performance assessment approach; removed Comments 29 and 92 as previously resolved; closed Comment 90; determined that Comments 81 and 84 are no longer pertinent to ENFE resolution
Rev1	5.2/all	August 1998	Revised Section 5.3 Rev0 to reflect staff review of DOE's thermal testing program; closed Comment 3 of NRC letter on DOE thermohydrology program
Rev1	5.3/all	August 1998	Major revision of Section 5.2 Rev0 to reflect: (1) DOE's comments on Rev0 and their proposed TSPA-VA approach; and (2) staff's concerns on DOE's TSPA
Rev1	5.4/all	August 1998	New sections summarizing subissue resolution achieved in the revision, and those coupled thermal-hydrologic-chemical processes potentially affecting repository performance
Rev1	5.5/all	August 1998	Revised Section 5.4 Rev0 to reflect progress in issue resolution

Revision #	Section/ Paragraph	Date	Modification
Rev1	6.0/all	August 1998	Provide References for Rev1
Rev2	1.0/all	May 1999	Revised to reflect changes in NRC and DOE programs
Rev2	2.0/1	May 1999	Revised to reflect the proposed 10 CFR Part 63
Rev2	3.4.1.2/all	May 1999	Updated discussion to reflect publication of sensitivity studies
Rev2	3.4.2.2/all	May 1999	Updated discussion to reflect results publication of sensitivity studies
Rev2	3.4.3.2/2	May 1999	Updated discussion to reflect results publication of sensitivity studies
Rev2	3.4.4.2/all	May 1999	Updated discussion to reflect results using TPA 3.2 and publication of sensitivity studies
Rev2	4.1.2.1/1	May 1999	Revised to reflect DOE comment on Rev1
Rev2	4.3.2.2/2	May 1999	Revised to reflect DOE comment on Rev1
Rev2	4.3.2.4/3	May 1999	Revised to reflect DOE comment on Rev1
Rev2	4.5.1/1	May 1999	Revised to reflect acceptance criteria in TSPA IRSR Rev1
Rev2	4.5.2.1/6,7	May 1999	Revised to reflect DOE comments on Rev1
Rev2	4.5.2.2/4	May 1999	Revised to reflect DOE comment on Rev1
Rev2	5.0/all	May 1999	Updated discussion of status of resolution
Rev2	5.1/all	May 1999	Updated discussion of site characterization and study plan comments to reflect review of TSPA-VA
Rev2	5.2/2	May 1999	Revised to reflect resolution of all comments
Rev2	5.3/1	May 1999	Revised to reflect DOE comment on Rev1
Rev2	5.4/all	May 1999	Revised Rev1 to reflect application of acceptance criteria to review of TSPA-VA

Revision #	Section/ Paragraph	Date	Modification
Rev2	5.4.1/all	May 1999	Revised Rev1 to reflect application of acceptance criteria to review of TSPA-VA
Rev2	5.4.2/all	May 1999	Revised Rev1 to reflect application of acceptance criteria to review of TSPA-VA
Rev2	5.4.3/all	May 1999	Revised Rev1 to reflect application of acceptance criteria to review of TSPA-VA
Rev2	5.4.4/all	May 1999	Revised to Rev1 reflect application of acceptance criteria to review of TSPA-VA
Rev2	5.4.5/all	May 1999	Revised to Rev1 reflect application of acceptance criteria to review of TSPA-VA
Rev2	5.5/all	May 1999	Revised Rev1 to reflect progress in issue resolution based on application of acceptance criteria to review of TSPA-VA
Rev2	6.0/all	May 1999	Updated References for Rev2

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: Center for Nuclear Waste Regulatory Analyses-generated original data contained in this report meets quality assurance requirements described in the Center 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 (QA) guidance contained in the Center QA manual.

1.0 INTRODUCTION

Site characterization activities are specified in the U.S. Nuclear Regulatory Commission's (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 times to completion, allows for the early identification and resolution of potential licensing issues. Moreover, NRC's 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 informal, pre-licensing 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 the NRC licensing review.

Thus, consistent with NRC's regulations and a 1992 agreement with DOE, staff-level issue resolution can be achieved during the prelicensing consultation period. However, resolution at the staff level would not preclude the issue being raised and considered during licensing proceedings. Issue resolution at the staff level during prelicensing is achieved when the staff has no further questions or comments (i.e., open items), at a point in time, regarding how the DOE program is addressing an issue. There may be some cases where resolution at the staff level may be limited to documenting a common understanding regarding differences in the NRC and the DOE technical positions. Pertinent, additional information could raise new questions or comments regarding a previously-resolved issue.

NRC's high-level radioactive waste (HLW) program was realigned during fiscal year (FY) 1996-1997. The realignment was in response to: (i) a reduction in Congressional budget appropriations for NRC in FY 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 HLW program was realigned to focus pre-licensing work on those topics most critical to the post-closure 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 FY 1996 Annual Progress Report (see Sagar, 1997).]

The current Division of Waste Management (DWM) approach is to focus most activities on issue resolution of the respective KTIs, at the staff level. DWM activities have been re-prioritized to improve the integration of, and streamline, 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. This is accomplished by identifying KTIs, integrating their activities into a risk-informed approach, and evaluating their significance for post-closure 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 will 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 *Annual Progress Report* (e.g., Sagar, 1997), which summarized the significant technical work toward resolution of all KTIs during the preceding 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 (LA).

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 U.S. Department of Energy, 1998a), that is, it's approach to the Viability Assessment. Section 4.0 provides our review methods and acceptance criteria, which indicate the technical basis for resolution of the subissue and that will be used by us in subsequent reviews of DOE submittals. These acceptance criteria are guidance for the staff and, indirectly, for DOE as well. Our technical basis for the acceptance criteria is also explained in detail to further document the rationale for our decisions. Section 5.0 concludes the IRSR with the status of resolution, indicating those items resolved at the staff level or those items remaining open. These open items will be tracked by us, and resolution will be documented in future IRSRs.

The IRSRs are the basis for our review of information in DOE's Viability Assessment (VA; U.S. Department of Energy, 1998b). Our comments on the VA are intended to facilitate DOE's efforts to focus its program and develop a high-quality LA. We reviewed the preliminary design concept, the total system performance assessment (TSPA), the LA 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 TSPA (U.S. Nuclear Regulatory Commission, 1999b). Detailed comments on the VA are provided in this revision of the IRSR. Section 5 of the IRSR documents the application of review methods and acceptance criteria to the VA and updates the status of resolution.

2.0 KEY TECHNICAL ISSUE AND SUBISSUES

Both NRC and DOE are evaluating the potential Yucca Mountain (YM) repository using a performance assessment (PA) approach. The NRC site-specific regulations for the proposed YM repository have been issued as the proposed 10 CFR Part 63 (U.S. Nuclear Regulatory Commission, 1999a). The proposed standards are performance-based. The near-field environment is defined from the perspective of NRC 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 YM. Thermal-hydrologic (TH) couplings and thermal-mechanical couplings are addressed primarily in the Thermal Effects on Flow (U.S. Nuclear Regulatory Commission, 1998a) and Repository Design and Thermal Mechanical Effects (U.S. Nuclear Regulatory Commission, 1998b) IRSRs, 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 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 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 evaluations of the effects of the ENFE on repository performance. The scope of ENFE KTI work includes review of 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 (QA) 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 DOE in 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 (WPs); long WP lifetime; slow rate of radionuclide release; and concentration reduction of radionuclides 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 WPs. In addition, nuclear criticality has been identified as a potential disruptive process to the repository system (U.S. Department of Energy, 1998a).

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

- Coupled THC effects on seepage and flow
- WP chemical environment
- Chemical environment for radionuclide release
- Effects of THC processes on radionuclide transport through engineered and natural barriers
- Coupled THC processes affecting potential nuclear criticality in the near field

The scope of Revision 1 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 total system performance assessment (TSPA). 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 (ESF) (Bish, et al., 1996). In addition, an increasing amount of data is becoming available from the ESF (e.g., Paces, et al., 1996), from thermal testing, and additional data are anticipated from exploration of the east-west drift at YM [Civilian Radioactive Waste Management System Management and Operating Contractor (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., 1996a). 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 performance assessment should not provide over-optimistic estimates of performance. The adequacy of DOE's treatment of coupled processes in the performance assessment will be evaluated, guided by this constraint. 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.

In summary, Revision 1 of the IRSR addressed all the subissues and provided acceptance criteria. These criteria will be used to judge DOE's evaluations presented in the VA and LA regarding the effects of the evolution of the near-field environment on repository performance. We will evaluate in this and the subsequent versions of the IRSR whether DOE's assessment of the effects of ENFE on repository performance includes important physical phenomena and processes, consistent assumptions and definitions, consideration of alternative models, adequate abstraction of process models, appropriate expert judgements, and quality assurance concerns.

3.0 IMPORTANCE TO REPOSITORY PERFORMANCE

The consequences of coupled geochemical processes can affect several aspects of the proposed YM HLW 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 WP 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 PA modules would change as a result of the coupled processes. Likewise, the effects of coupled processes will modify values of parameters used in WP, waste form (radionuclide release), and flow and transport modules of PA 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 PA 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, since the effects of coupled process have not yet been sufficiently evaluated in DOE or NRC 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 WP 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 of the DOE Repository Safety Strategy (U.S. Department of Energy, 1998a). 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 PA 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 the VA 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 YM site was presented in its 1988 Site Characterization Plan (U.S. Department of Energy, 1988). DOE updated that 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). The updated safety strategy reflects recent site characterization information, new WP 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 WPs 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). The updated strategy continues to rely on engineered and natural barriers to contain and isolate the waste from the public. Four system attributes are the most important for predicting the performance of engineered and natural barriers. These system attributes are: (i) limited water contacting WPs; (ii) long WP 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).

A number of working hypotheses have been developed by DOE to guide testing of the most important post-closure 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 WP can be limited. For the second system attribute, long WP 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 (RH), and that corrosion of the inner barrier is slow. The safety strategy (U.S. Department of Energy, 1998a) notes that environmental characteristics of the WP 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 effected by the evolution of the near-field environment. Although unspecified among DOE hypothese criticality is noted in the safety strategy as a potential disruptive scenario.

3.2 IMPORTANCE OF SUBISSUES TO TOTAL REPOSITORY SYSTEM PERFORMANCE

The ENFE KTI is currently considered to be an important factor in repository performance. The consequences of coupled THC processes may affect many aspects of repository performance. In addition, the same coupled process could affect different 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, 1996), 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 analysis 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.

We have developed a strategy for assessing the performance of the potential HLW repository at YM (U.S. Nuclear Regulatory Commission, 1998c). The TSPA Methodology 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 IRSR supports the TSPA Methodology IRSR and the overall compliance determination by describing the information needed in key performance areas and by pursuing issue resolution in those areas. Those elements that are important to TSPA of a facility at the YM site are defined as Key Elements of System Abstraction (KESA)s. Therefore, the approach that we will use to independently evaluate the DOE TSPA will focus the KESA. The KESAs 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 seven key elements of the engineered and natural subsystems. The seven KESA that the ENFE influences are: (i) WP corrosion (temperature, humidity, and chemistry); (ii) quantity and chemistry of water contacting waste forms; (iii) radionuclide release rates and solubility limits; (iv) fracture versus matrix flow; (v) spatial distribution of flow; (vi) retardation in fractures; and (vii) retardation in the saturated zone. Just as the effects of a single coupled process may affect more than one aspect of repository performance, both coupled processes and KESA may be incorporated in several of the ENFE subissues or in other KTIs. The acceptance criteria in this IRSR are designed to ensure that information necessary to describe the effects of coupled near-field THC processes on key elements of subsystem abstractions is acceptable. It should be noted that the acceptance criteria in this IRSR are subsidiary to and designed to complement the broader-level acceptance criteria for the abstraction of the KESA found in the TSPA Methodology IRSR (U.S. Nuclear Regulatory Commission, 1998c). The consideration of near-field coupled processes may also influence whether DOE has met acceptance criteria located in IRSRs other than TSPA methodology [e.g., Container Life and Source Term (CLST; U.S. Nuclear Regulatory Commission, 1998d), Radionuclide Transport (RT; U.S. Nuclear Regulatory Commission, 1998e), Thermal Effects on Flow (TEF; U.S. Nuclear Regulatory Commission, 1998a)]. The importance of coupled THC effects on performance for each subissue and the relationship between the KESA and each subissue are described more fully in the following sections.

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 (UZ) is the first subissue of the ENFE KTI. The three KESA (see Appendix A) that are influenced by the ENFE within the scope of the seepage subissue are: (i) fracture versus matrix flow; (ii) spatial and temporal distribution of flow; and (iii) quantity and chemistry of water that affects the engineered barrier system. There are three main coupled processes that will occur in the near-field environment that have the potential to affect performance. Each of these processes needs to be considered in the evaluation of each of the KESA. 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 coupled process 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 YM contain abundant water, commonly 10 percent of the rock volume (e.g., data from Flint, 1996). 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 this process to performance has been recognized by DOE (Bish, et al., 1996). The spatially varying distribution of the zeolitic horizons in YM, 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 TEF IRSR. Flow through these thermally-affected zeolitic horizons will also be influenced y 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 process that may affect performance of the repository is coupled THC processes that affect 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 TEF IRSR, 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 coupled process within the subissue of seepage and flow that may affect performanc of the repository is likely to be spatially limited to near the drifts of the repository. The referen design description of the ground control system indicates 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 YM 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_2. 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 will likely depend on the spatially variable mechanical stability of the geologic medium [this topic is addressed in the Repository Design and Thermal Mechanical Effects (RDTME) 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.

Thus, the spatial and temporal distribution of flow, the distribution of flow in fractures and matrix, and the quantity and chemistry of fluids intersecting the repository horizon will be affected as a result of the evolution of the near-field geochemical environment. To adequately describe these KESA in a TSPA (see TSPA Methodology IRSR; U.S. Nuclear Regulatory Commission, 1998c), the effects of coupled THC processes on seepage and flow will needed 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 WP chemical environment is the second subissue of the ENFE KTI. The two KESA (see Appendix A) that are influenced by the ENFE within the scope of the WP chemical environment subissue are: (i) quantity and chemistry of water that affects the engineered barrier system; and (ii) WP corrosion (temperature, humidity, and chemistry). The three coupled processes 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 WP chemical environment subissue and WP KESA, 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 WP chemical environment KESA will be presented here. One additional coupled process, the generation of natural and spontaneous self potentials, is no longer thought to affect the WP chemical environment significantly (see Section 4.2.2).

Successfully abstracting the WP corrosion process within a performance assessment framework, which is primarily addressed in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d), 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, Cl^-, oxygen content, and total carbonate in the near field of the repository and possibly other components. As a result, coupled THC process will assert a strong influence on WP 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 dry oxidation of the WP.

The number, location, and time when WPs are contacted 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 WP will be the result of coupled THC processes with natural and engineered materials, including both the WP and the concrete liner. Depending on the temperature of the WP and the drifts, and the flux of water intercepting the WP, further concentration of the fluid on the surface of the WP, due to evaporative effects, is possible. Thus, to adequately describe the KESA in a TSPA, the effects of coupled THC processes on WP 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 WP corrosion (temperature, humidity, and chemistry), 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 three KESA (see Appendix A) are addressed within the chemical environment for radionuclide release subissue and are addressed in more detail in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d). The effects of coupled processes on the KESA 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 (Cragnolino and Galvele, 1978; Maguire, 1984). Under the environmental and potential conditions leading to pitting, stress corrosion cracking (SCC) 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 extreme' 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 Regazzoni, 1994; Grambow, 1989). A major near-field environment 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 WPs, 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 WP, air mass fraction in the WP, 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 C, 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 WP degradation, can react with U(VI) 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 PA codes (Mohanty, et al., 1997; TRW Environmental Safety Systems, Inc., 1997b)

Secondary U 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 because of the volume expansion. Third, Np, Pu, and other radioactive waste species may be incorporated into secondary U phases by coprecipitation. Fourth, the secondary U phases may limit ingress of water and oxidants to unaltered wastes. Finally, the secondary U phases may control the solubility or dissolution rate of spent fuel. Thus, secondary U phases may control the source term for radionuclide (not just U) releases from the breached WPs. 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 WPs.

The second main waste form planned for the proposed repository at YM is HLW 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 YM (Murphy and Pabalan, 1994). However, they are likely to incorporate augmented quantities of components of the engineered barrier system, such as Fe and Ca. Clay minerals generally have low solubilities. Some quantity of radioactive waste pecies are likely to be incorporated through solid solution in mineral alteration products of glass waste forms. Contribution of the HLW 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 proposed YM 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 three KESA (see Appendix A) that are influenced by the ENFE within the scope of the radionuclide transport subissue are: (i) fracture versus matrix flow; (ii) retardation in fractures; and (iii) retardation in the saturated 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 KESA.

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 UO_2^{2+}, NpO_2^{+}, and 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, 1996). 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 Cs^+, Sr^{2+}, and 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, 1996). The stability of the sorbant minerals, the amount of sorbant 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 (TcO_4^-) and does not sorb strongly, whereas, under reducing conditions, 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 $^{14}CO_2$ 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 proposed YM 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 HLW creates a potential for a sustained neutron chain reaction (criticality event). Such an event could arise if there is failure of the HLW 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 proposed YM 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 $Pu\text{-}SiO_2\text{-}H_2O$ and $Pu\text{-tuff-}H_2O$, 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 or 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 YM repository have been included in performance assessment studies conducted prior to the Viability Assessment. Those recent PA studies that have addressed the effects of coupled geochemical processes include: (i) TSPAs 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 TSPA 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., 1996a); (iv) one TSPA prepared by NRC—Iterative Performance Assessment, Phase 2 (IPA Phase 2) (Wescott, et al., 1995); and (v) sensitivity studies conducted by the NRC and Center staffs using the TPA code (Manteufel, et al., 1997). The manner in which these studies incorporate effects of coupled processes is summarized in the following sections. DOE has 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^-, F^-, and total carbonate concentrations. However, in TSPA-93, Eh and Cl^- concentration were not used. The temperature, fractional time the WPs are wet, the pH, F^-, 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 reports 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 kW/acre) may cause near-field conditions to extend throughout the UZ" (Wilson, et al., 1994; page 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; page 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; page 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 (Ra) and tin (Sn). 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 Ra and 10 times the upper limit of the TSPA-95 range for Sn. 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 PA (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 YM 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 Jarzemba, 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 DOE 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 WP, 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, pCO_2, CO_3^{2-}, Cl^-, F^-, Si, PO_4^{3-}, and SO_4^{2-}. Section 6.3 of TRW Environmental Safety Systems, Inc., (1997b) provides a fairly comprehensive description of coupled thermal-hydrologic-chemical 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). In its review, the NRC/Center will judge the TSPA-VA, in part, by comparison to the description provided in Section 6.3 of TRW Environmental Safety Systems, Inc., (1997b).

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 an UZ-TH 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 EBS materials (e.g., WP, 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 (EPRI) has evaluated some of the processes that the ENFE KTI has responsibility to address. EPRI used a code called Integrated Multiple Assumptions and Release Calculations (IMARC) 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 WP 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., 1996a). Preliminary consequence sensitivity analyses were conducted using a TSPA model (the RIP code) by modifying the TSPA-95 base case (parameter set as 83 MTU/acre, backfill, high infiltration rate, climatic variation, drips on the WP 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_ds, for all radionuclides to zero for distances of 10 and 100 m into the unsaturated zone (UZ), and for the entire UZ. 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 Np, Pu, 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 base case results. A single realization, expected-value calculated dose at the accessible environment, at 10^4, 10^5, and 10^6 yr was compared to the base case. In addition, the complementary cumulative distribution function (CCDF) of 100 realizations of calculated dose at the accessible environment at times of 10^4 and 10^5 yr were compared to the base case.

For the 10^4 yr time frame, the results indicate essentially no change from the base case for both scenarios where there is no retardation up to 100 m into the UZ. For unretarded transport through the entire UZ, 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 yr for the second scenario.

For the 10^5 yr time frame and 10 m of unretarded UZ transport, there is essentially no difference for the first scenario compared to the base case. 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 yr. For unretarded transport through 100 m and the entire UZ, 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 UZ pathway, Np-237 contributes the highest doses after ~20,000 yr and ~10,000 yr in the first and second scenarios, respectively. In the latter case, the peak dose is about 300 times the base case value, and Pu-239 is the second highest dose contributor at ~60,000 to 100,000 yr, comparable to the dose contribution from Th-229.

For the 10^6 yr time frame, the results for the first scenario indicate that the long-term Np-237 peak dose is: (i) relatively unchanged for 10 m of unretarded transport; (ii) about twice the base case for 100 m unretarded transport; and (iii) about 4 times the base case for unretarded transport throughout the UZ. 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 m of unretarded UZ pathway. For unretarded transport throughout the entire UZ, the major dose contributors are Pu-239 and Pu-242 after about 150,000 yr.

In all the cases simulated, except for the second scenario in which transport is unretarded for the entire UZ, there is little change to peak doses at 10,000 yr 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 yr time frames, respectively. For unretarded transport through 100 m and the entire UZ, Np-237 peak doses are about 0.5 and 1.5 orders of magnitude, respectively, higher than the base case for the first scenario, and about 1.0 and 2.5 orders-of-magnitude, respectively, higher for the second scenario.

For the 10^6 yr 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 UZ compared to the base case, 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 Pu isotope doses are about seven orders of magnitude higher than the base case and are major contributors to peak dose after about 150,000 yr. The peak dose from either Pu 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 (1D), uniform, time varying gas flow field was coupled to equilibrium aqueous speciation, CO_2 gas evolution and transport, and calcite precipitation in a transient thermal field. The model was used to explore possible mechanisms for ^{14}C retardation and gas phase release. An initial pulse of $^{14}CO_2$ was modeled to exit the mountain after less than 2,000 years of transport. Subsequently, ^{14}C releases to the atmosphere were small and some ^{14}C remained trapped in precipitated calcite. However, these results were not incorporated in analyses of CCDFs.

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

In sensitivity analyses conducted by 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 PA 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 PA code [Total-system Performance Assessment (TPA)] 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 CLST 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 WPs) may have on the performance of the repository. Conceptual models of WP degradation and radionuclide release were also evaluated as part of the ENFE KTI sensitivity analyses. The results of the preliminary efforts using the TPA 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 YM 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 YM. Process-level model calculations by Lichtner, et al., (1998) suggest that strong alteration of the tuff host rock at YM 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, et al., (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 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, and

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_ds of radionuclides, particularly actinides, in cementitious environments are high (Bradbury and Sarott, 1995; Campbell and Krupka, 1997). For example, a comparison of K_ds of different elements specified in the TPA code input file with K_ds of those elements in cement environments taken from Bradbury and Sarott (1995) indicate that, except for Tc, Ra, Cs, and Se, radionuclide K_ds are higher, typically orders of magnitude higher, in cement-affected environments compared to a YM 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., 1996a) 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 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 m) 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 is 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 (PDF) 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 (UZ) is less than 30 years, then the travel time for transport of radionuclides in the UZ is assumed to be zero. In that case, performance is independent of the flow and transport properties of the UZ below the emplacement horizon. Since 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 3.1 code are presented in Jarzemba, 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 WP 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_ds, 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 WP 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 Cl and 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 Tc, the effect of iron will be more significant in helping to reduce Tc(VII) to Tc(IV), with lower solubility and increased sorption, but there is a lack of data to estimate Tc sorption behavior. Similar to Na, Cs 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 ($0.9 \leq R_d \leq 1.0$). Since 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 U concentrations are associated with hematite in fractures, but U migration into the matrix was limited (Pearcy, Prikryl, 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, Cm was assigned the same K_d distribution as Am and Nb. 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 yr at five km. 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 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 WP 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, potentially, 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. WP corrosion depends on the corrosion potential (E_{corr}) and the critical potential required to initiate localized corrosion. The value of E_{corr} is strongly dependent on oxygen concentration. In EBSFAIL, the critical potential, conservatively represented by the repassivation potential (E_{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_{rp} decreases with increasing chloride concentration while E_{corr} increases with increasing oxygen concentration. High chloride and oxygen concentrations are expected to induce earlier failures of WPs 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 WPs. The model takes into consideration the evaporative effects produced in a partially saturated environment by the heat released from the WP due to radioactive decay. When evaporation takes place, an increase in salinity occurs and pH increases as 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 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 WPs, 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 WP. Evaporative concentration of solutes could occur, leading to saturated salt solutions on the WP (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 atm is used in the TPA 3.1 input file assuming the maximum value expected in the near-field environment when the temperature of the WP 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 2.1×10^{-5} atm, reflecting carefully de-aerated water, and a maximum and peak values of 0.21 atm were adopted, corresponding to a mean value of 0.14 atm.

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 km or 20 km 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 3.0×10^{-2} atm. 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, 1996) 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 3.1 code is presented in Jarzemba, et al., (1999).

3.4.4 Conceptual Model of Oxidation Rate Controlled Limits on Radionuclide Release

Radionuclide release rates in the TPA code base case 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 proposed repository at YM. An alternate approach is to use data from natural analog systems to constrain release rates on a geologic time scale. The Nopal I U deposit at Peña Blanca, Chihuahua, Mexico, has been studied in detail as an analog of the proposed repository at YM. 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 Pearcy, 1992; Murphy, Pearcy, and Pickett, 1997). The rate is a maximum because a maximum limit was used for the infiltration that removed U 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 proposed YM 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 U removed from the site. Peña Blanca site-specific characteristics were used to determine this quantity, based on conservative estimates of the U solubility and the groundwater flux through the system. The maximum average U oxidation rate for the YM repository was determined by scaling for the masses of U at each site. For radionuclides incorporated in the matrix of spent fuel, the oxidation rate was assumed to limit their release rate.

Murphy, Pearcy, 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 Pearcy (1992) and a revised minimum time for oxidation based on radiometric ages of late forming uranophane (an oxidized U mineral) of 3 million yr (Pickett and Murphy, 1997). Calculation of the bounding rate depends on the amount of U that has been removed from the site, which is given by the product of the water flux and U concentration yielding the relation (Murphy and Pearcy, 1992)

$$R_0 = (U_e/t) + FC \tag{1}$$

where R_0 stands for the oxidation rate (e.g., grams year^{-1}), U_e represents the amount of oxidized U remaining at the deposit, t represents the period of time of oxidation, F stands for the water infiltration rate (e.g., liters year^{-1}), and C denotes the concentration of U in solution exiting the system. Peña Blanca site-specific characteristics were used to determine this quantity based on estimates of the U solubility and conservative limits on the groundwater flux through the system. The calculated maximum average rate is 140 g yr^{-1} for 320 metric tons of oxidized U remaining at the site and an estimated upper limit of 86 metric tons of U removed from the site over the 3 million yr 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 YM, although hydrologic transport rates may differ between the sites.

Scaling the YM repository to Peña Blanca, 70,000 tons of U at a hypothetical YM repository is 70,000/408 = 172 times the initial estimated (maximum) quantity of U at Peña Blanca. Supposing the whole of the repository inventory of spent fuel oxidizes at a comparable rate as that at Peña Blanca, the total rate is given by 172 × 140 g yr^{-1} (kg/1,000 g) = 24 kg yr^{-1}. This value represents a maximum average U oxidation rate for the YM repository, based on the maximum average rate of oxidation estimated for the Peña Blanca deposit.

U solubilities [see Eq. (1)] were used in calculation of the maximum average oxidation rate at the Peña Blanca deposit to estimate the maximum amount of U that has been removed from the deposit. Determination of the U solubility was based on the premise that secondary, oxidized alteration products control U solubility, which is indicated by the Peña Blanca data. This approach differs from that adopted in the DOE TSPA-95 to determine U solubility. TSPA-95 adopted a log β distribution of solubilities for U 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 U would be a factor in release of U in YM even though the ground water contains dissolved silica which could cause U silicates to precipitate." In this sensitivity analysis, the alternate hypotheses were taken that secondary uranyl minerals control U solubility and that oxidation rates are fast relative to transport of U 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 Pearcy (1992) adopted a U solubility of 10^{-7} mole liter^{-1}. 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 YM site. A variety of possible water chemistries and solubility controlling minerals was examined. The mean of the base 10 logarithm of the U solubility (in moles liter^{-1}) in these analyses was −7, corresponding to the value calculated previously by Murphy and Pearcy (1992). The standard deviation of the 32 computed values of the logarithm of U solubility was 1.8 units; 24 kg yr^{-1} corresponds to UserLeachRate[kg/yr/m^2] = 5×10^{-6} kg yr^{-1} m^{-2}. The release rate for each individual radionuclide is generally given by the U oxidation rate times its fractional inventory of the radionuclide in spent fuel.

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 km distance from the repository for various times up to 100,000 yr. 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 base case release model. The results from the natural analog study are not necessarily conservative, because we assume the release of all other radionuclides is directly related to release of solubilized uranium. This direct relationship between uranium and all other radionuclides is only possible if all radionuclides 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 Jarzemba 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 km from the waste emplacement at 10,000 and 50,000 years after repository closure. Both alternate models yield lower doses than the base case model in TPA 3.2 analyses.

4.0 REVIEW METHODS AND ACCEPTANCE CRITERIA

This IRSR and its acceptance criteria focus on providing a path to resolution. The acceptance criteria will be revised to become more specific once performance assessment studies are completed, other analyses and testing progress, the license application design of the repository is specified, and the NRC regulations for the YM repository (10 CFR 63; U.S. Nuclear Regulatory Commission, 1999a) are finalized. A systematic approach to resolving subissues is necessary. In the past, there has been little concerted effort to evaluate within a performance assessment framework how the evolution of the near-field geochemical environment influences repository performance. In addition, the lack of specificity of the acceptance criteria for this IRSR is a consequence, in part, of the instability in the repository design. 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 pre-closure period). As a result, the acceptance criteria are 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. The increased specificity of the acceptance criteria in the future will reflect only those processes that need to be addressed and, potentially, the bounds of the effects of coupled THC processes that need to be used for an acceptable performance assessment.

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. Acceptance criteria, upon which a more broad staff review of a DOE TSPA, that focus on key elements of subsystem abstraction (KESA) can be found in the TSPA Methodology IRSR (U.S. Nuclear Regulatory Commission, 1998c; see also Appendix A). The acceptance criteria for this KTI and each subissue are subsidiary to and designed to complement the broader-level acceptance criteria of the TSPA Methodology IRSR. In Revision 1 of the ENFE IRSR, the acceptance criteria and review methods have been grouped together to reflect the five technical and two programmatic acceptance criteria, and their associated review methods, found in the TSPA Methodology IRSR (U.S. Nuclear Regulatory Commission, 1998c).

Several relatively comprehensive review documents have been published that are relevant to coupled THC processes in the near field of the proposed repository at YM, 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

DOE has identified seepage of water into waste emplacement drifts and flow in the unsaturated zone as factors that are important to repository performance. Important factors affecting seepage and flow are: groundwater flux, fracture density and physical properties, presence or absence of fracture coatings, rock heterogeneity, moisture content, existence of fast pathways, fluid density gradients, and others. Several aspects of THC effects on seepage and flow, particularly the quantity and chemistry of water contacting WPs; distribution of mass flux between fracture and matrix; and the spatial and temporal distribution of flow, are included as key elements of the NRC performance assessment subsystem abstraction. Contributions will be required from several KTI groups to resolve the issue of coupled THC effects on seepage and flow, including the Unsaturated and Saturated Flow Under Isothermal Conditions (USFIC) KTI, TEF KTI, and RDTME KTI, as well as the ENFE KTI (U.S. Nuclear Regulatory Commission, 1998c). DOE must adequately estimate coupled THC effects on the quantity and chemistry of seepage into the drifts and unsaturated flow, and appropriately consider this subissue in its assessments of repository performance.

4.1.1 Review Methods and Acceptance Criteria

DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM is satisfactory, if the following acceptance criteria are met.

Review Method:

- Staff should ascertain whether DOE demonstrated that sufficient data exist to support the conceptual models and to define relevant parameters in DOE's abstractions. Staff should verify whether DOE provided sound bases for the inclusion or exclusion of certain observed phenomena in its conceptual models. The staff will evaluate the potential for DOE estimates of performance to be over-optimistic, given the excluded set of phenomena and the implementation of coupled geochemical processes in the PA. In its review, staff should determine whether DOE has performed sensitivity and uncertainty analyses to test for the possible need for additional data. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Data and Model Justification Acceptance Criteria

(1) Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow were considered.

(2) DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect seepage and flow.

(3) 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 for conceptual models and simulations of THC coupled processes that affect seepage and flow.

(4) Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.

(5) If the testing program for coupled THC processes on seepage and flow is not complete at the time of license application, or if sensitivity and uncertainty

analyses indicate additional data are needed, DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program.

Review Method:

- Staff should ascertain whether input values used in the quantity and chemistry of water affecting the engineered barrier system, spatial and temporal distribution of flow, and distribution of mass flux between fracture and matrix calculations within TSPA are reasonable, based on data from the YM region (e.g., drift-scale heater and niche test results) and other applicable laboratory tests and natural analogs. Staff should verify whether these values are consistent with the initial and boundary conditions and the assumptions of the conceptual models and design concepts for the YM site (e.g., estimations used in these abstractions should be based on the thermal loading strategy, including effects of ventilation; EBS design, including drift liner, and backfill and drip-shield, if present; and natural system masses and fluxes). In addition, the staff should verify that if any correlations between the input values exist, they have been appropriately established in DOE's TSPA. Finally, the staff should, to the extent feasible, evaluate DOE's input values by comparison to corresponding input values in the staff data set and use the NRC TPA code to test sensitivity of system performance to the input values and correlations used by DOE.

Data Uncertainty and Verification Acceptance Criteria

(1) Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on seepage and flow. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.

(2) Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow were considered.

(3) DOE's evaluation of coupled THC processes properly considered 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 seepage and flow.

(4) The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on seepage and flow were consistent with available data.

(5) DOE's performance confirmation program should assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on seepage and flow.

Review Method:

- Staff should ascertain whether DOE considered plausible alternative models and justified approaches used in the quantity and chemistry affecting the EBS, spatial and temporal distribution of flow, and distribution of mass flux between fracture and matrix abstractions. Staff should use the NRC TPA code to assist in verifying whether the intermediate outputs provided by DOE's approach reflects or bounds the range of uncertainties owing to alternative modeling approaches. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Model Uncertainty Acceptance Criteria

(1) Appropriate models, tests, and analyses were used that are sensitive to the THC couplings under consideration for both natural and engineering systems, as described in the following examples. The natural-setting data indicate processes that should be evaluated include: (i) zeolitization of volcanic glass, which could affect flow pathways; (ii) precipitation of calcite and opal on the footwall of fracture surfaces and the bottoms of lithophysal cavities, which indicates gravity-driven flow in open fractures that could affect permeability and porosity; and (iii) potential dehydration of zeolites and vitrophyre glass, which could release water affecting heat and fluid flow. The effects of THC coupled processes that may occur due to interactions with engineered materials or their alteration products include: (i) changes in water chemistry that may result from interactions between cementitious materials and groundwater, which, in turn, may affect seepage and flow; (ii) dissolution of the geologic barrier (e.g., tuff) by a hyperalkaline fluid that could lead to changes in the hydraulic properties of the geologic barrier; and (iii) precipitation of calcite or calcium-silica-hydrate phases along fracture surfaces as a result of migration of a hyperalkaline fluid that could affect hydraulic properties.

(2) Given the current design of the repository, it will be acceptable to ignore the potential effects of microbial processes on seepage and flow.

(3) Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

(4) DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC 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.

Review Method:

- Staff should ascertain whether DOE verified that the outputs of quantity and chemistry of water affecting the engineered barrier system, spatial and temporal distribution of flow, and distribution of mass flux between fracture and matrix abstractions reasonably reproduce or bound the results of corresponding process-level models or empirical observations. Staff should, to the extent feasible, evaluate the output of DOE's abstractions against results produced by process-level models developed by the staff.

Model Verification Acceptance Criteria

(1) The mathematical models for coupled THC effects on seepage and flow 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.

(2) DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on seepage and flow.

(3) Abstracted models for coupled THC effects on seepage and flow were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different *mathematical models to judge robustness of results.*

Review Method:

- Staff should ascertain whether consistent and appropriate assumptions and initial and boundary conditions have been propagated through DOE's abstraction approaches. For example, staff should determine if the conditions and assumptions used to generate the look-up tables or regression equations are consistent with all other conditions and assumptions in the TSPA for abstracting the quantity and chemistry of water affecting the engineered barrier system, spatial and temporal distribution of flow, and distribution of mass flux between fracture and matrix. The important design features that will set the initial and boundary conditions for these abstractions include drift lining, thermal loading strategy, including ventilation, etc. If DOE decides not to take credit for certain design features that have been demonstrated in NRC's or DOE's, or both analyses to provide only benefits and no deleterious effects, staff does not need to include such features in its review. Staff should verify whether DOE's dimensionality abstractions appropriately account for the various design features, site characteristics, and alternative conceptual approaches. In addition, staff should verify whether DOE's domain-based and temporal abstractions appropriately handled the THC couplings or sufficient justification has been provided to exclude these couplings. Staff should, to the extent feasible, use the NRC TPA code to selectively probe DOE's approach for these three abstractions for potential inconsistency in the analysis and non-defensible predictions.

Integration Acceptance Criteria

(1) DOE has considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, physical phenomena, and couplings, and used consistent and appropriate assumptions throughout.

(2) Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow.

(3) Not all THC couplings may be determined to be important to performance, and DOE may adopt assumptions to simplify PA analyses. If potentially important couplings are neglected, DOE should provide a technical basis for doing so. The technical basis can include activities such as independent modeling, laboratory or field data, or sensitivity studies.

(4) Where simplifications for modeling coupled THC effects on seepage and flow were used for PA analyses instead of detailed process models, the bases used for modeling assumptions and approximations were documented and justified.

Review Method:

- NRC will attend, as observers, activities conducted by DOE related to model abstractions, and track the progress made in resolving quality assurance deficiencies in the abstraction activities. If DOE uses peer reviews, staff should review DOE's implementation to ensure that the peer reviews followed the guidance in NUREG-1297 and NUREG-1298 (Altman, Donnelly, and Kennedy, 1988a,b), or other acceptable approaches. If staff has concerns, they will be noted at the time of staff's attendance and formally communicated to DOE. If DOE uses expert elicitations, NRC will attend, as observers, the elicitation workshops and review the documentation to ensure that the expert elicitations followed the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches. If staff has concerns, they will be noted at the time of the elicitation and formally communicated to DOE. Progress made in resolving these concerns will be tracked by the staff.

Programmatic Acceptance Criteria

(1) Data and models were collected, developed, and documented under acceptable quality assurance (QA) procedures.

(2) Deficiency reports concerning data quality on issues related to coupled THC effects on seepage and flow were closed.

(3) If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

4.1.2 Technical Bases for Review Methods and Acceptance Criteria for Effects of Coupled Thermal-Hydrologic-Chemical Processes on Seepage and Flow

The technical bases for the acceptance criteria are given in this section. These bases are primarily focused on explaining why the results of THC coupled processes may be important to seepage and flow. As mentioned previously in Section 3, limited analyses in past PAs on the effects of coupled THC processes on the potential repository seepage behavior and groundwater flow 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, materials used, etc.). This has resulted in acceptance criteria that primarily focus on ensuring that analysis of the 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, 1996, 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 these changes in porosity are within the range of values sampled for a particular modeled hydrologic unit in the base case PA, then the effect of that coupled process on porosity need not be considered further.

4.1.2.1 Coupled Thermal-Hydrologic-Chemical Processes Affecting Flow of Water

At YM, host rocks are silicic tuffs [70 to 80 percent SiO_2; (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 (SZ) 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 mV $\leq$ Eh $<$ 0 mV). The unsaturated zone (UZ) groundwaters differ substantially from SZ 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, Rattray, and Yu, 1996; Yang, et al., 1996). Aqueous silica concentrations in excess of cristobalite saturation are observed in tuffaceous aquifers at YM (Kerrisk, 1987), and higher concentrations are observed in the UZ (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 YM, 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 unit at YM, primary glass has been altered to analcime ± 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 YM is primarily air, approximately saturated with liquid water, and enriched in CO_2 relative to the atmosphere. Gas chemistry analyses show limited variability in the CO_2 content in space and time (Thorstenson, et al., 1990). The partial pressure of CO_2 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 uncompacted 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 YM 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 er of YM (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 YM 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_2 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 YM must be judicious, because the high temperatures, high pressures, saturated conditions, and short time scales of the experiments are unrepresentative of expected conditions at YM.

Mass transfer calculations that account for partial equilibrium and reaction kinetics in gas-water-rock interactions have provided geochemical models related to the YM 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_2 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, $p(CO_2)$, $p(O_2)$, 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_2 from the near-field environment. The vapor pressure of water at temperatures above 95 °C exceeds the hydrostatic pressure of less than 0.1 MPa at YM. Thus, the gas phase in the near field would tend to be dominated by H_2O. 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_2, silica, and calcium.

Silica redistribution in the YM 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 YM tuff under unsaturated conditions (Rimstidt, Newcomb, and Shettel, 1989). Silica and Fe, dissolved near the heater, were transported in solution and precipitated as amorphous silica, Fe 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 1.3×10^{-14} m^2 to about 10^{-17} m^2 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 yr.

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 TH simulations. Extensive development of heat pipe effects and refluxing at elevated temperatures could 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 effect 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).

Major geochemical changes in the near field are likely to depend primarily on the availability of water. Although unsaturated, the rocks at YM 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. Water is drawn by capillarity into the finest pores of the rock. Evaporation and mineral precipitation will occur dominantly 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 acidic water, due to dissolution of gaseous CO_2 into the condensate, and mineral dissolution are likely to occur on fracture surfaces. Together, these processes could lead to an increase of fracture permeability and a decrease of matrix permeability. 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 hydro-chemical modeling.

4.1.2.2 Effects of Engineered Materials on Seepage and Flow

The use of cementitious materials, in the form of concrete inverts and linings, is being considered for the estimated 179 km of emplacement drifts, roadways for construction, and emplacement ramps and service mains. Although cement is used primarily for its structural (e.g., high compressive strength) and physical (e.g., low permeability) properties, its effect on the near-field chemical environment of a repository could be pronounced. Our analysis of the effects of engineered materials on seepage assumes the reference case design for the proposed repository. The base case design assumes precast concrete liners for drift support and no backfill. For this design, the effects of engineered materials other than cementitious materials on seepage are assumed to be negligible. The following discussion of cementitious materials reflects only a limited effort by the ENFE KTI to address this topic during FY 1998.

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 'hermodynamically incompatible with silica, a major component of the proposed YM repository ,ost 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 analogue site for studying cement interaction in northern Jordan suggests that rapid interaction between cement-equilibrated, pore waters, and silicate-bearing rocks can be expected (Cowan, 1975). This may result in sealing of fractures and alteration of the host rock minerals. Calculations by Lichtner, et al., (1997, 1998) suggest that strong alteration of the tuff host rock at YM 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, CSH phases would precipitate and clog the pore spaces. Alternatively, precipitation of calcite and CSH 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 diminished seepage and groundwater flow through the repository. Results of Lichtner, et al., (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_2-high Ca cement pore fluid *mixes with the ambient groundwater containing high CO_2 concentrations* (Lichtner and Eikenberg, 1995; Steefel and Lichtner, 1994).

The results of Lichtner, et al., (1998) indicate that calcification of the cement could occur as vould be expected under ambient conditions. In the case of counter-diffusive transport, ,alcification 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. Joints in the inverts may provide fast pathways for water to move downward with short residence times for contact with cement. In such cases, there would be little change in pH, for example, due to cement interactions.

4.1.2.3 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 a potential repository's performance. These processes include canister corrosion and leaching of the glass waste form. Other potentially important microbial processes are supplying of organic by-products 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 sub-surface 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, 199 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 HLW repositories, several countries have assessed the potential importance of microbial processes in proposed HLW repositories (Christofi and Philp, 1997).

In contrast, microbiological study in the United States (US) HLW program has not progressed as far in determining the importance of microbial processes to repository performance. The efforts have focused on identifying microbial processes relevant to the YM 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 WP, modify water and gas chemistry outside the bounds predicted by abiotic chemical calculations, and alter the rate of radionuclide transport from breached WPs (Horn and Meike, 1995). Research efforts on the effect of microorganisms on radionuclide transport at YM were summarized and evaluated (Hersman, 1996). One conclusion from this study was that the elevated subsurface concentrations of CO_2, 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 has been proposed as part of the TSPA-VA (TRW Environmental Services, Inc., 1997b). This effort will attempt to address potential changes in gas chemistry (CO_2) from microbial processes. The effort will rely on models that have been developed in Europe to assess the potential importance of microbial processes on HLW repositories (TRW Environmental Services, Inc., 1997b; Grogan and McKinley, 1990).

Horn and Meike (1995) reviewed the conditions necessary for microbial activity in relation to deep sub-surface repositories, such as YM. For metabolic activity, microorganisms require an energy source (usually organic C); water; a source of N, S, and P; and suitable environmental conditions, such as temperature and pH (Horn and Meike, 1995). Most microorganisms are heterotrophic, using organic C compounds as their energy source. Oligotrophic bacteria (those that can grow in nutrient-poor water) require a minimum organic C content between 1–15 mg L^{-1} of water (Ehrlich, 1996). The organic C content of the groundwater in the vicinity of the repository (J-13 well water) is in the range of 0.15-0.55 mg L^{-1}, comprising principally humic acids and low molecular weight fatty acids (Means, Maest, and Crear, 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_2 concentration in the gas phase (Hersman, 1996; Thorstenson, et al., 1998).

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

Using samples collected from the ESF, Kieft, et al., (1997) conclusively demonstrated that water is the major limiting factor to growth and microbial activity at YM. Microbial abundance was low, and, in general, indicated microbes were most abundant in locations that were from less consolidated, non-welded, 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 simulation 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 YM 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 YM is unsaturated, pore fluids are oligotrophic, and microbial activity is limited under ambient conditions. Microbial activity in the near field during the post-closure 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 is directly controlled by the partial pressure of CO_2 in equilibrium with pore fluids in the UZ. Volatilization of CO_2 as water evaporates is expected to increase gas phase CO_2 concentration to many times that of the ambient conditions (Lichtner, 1997). This has been observed in the drift scale test at YM (unpublished results of Lawrence Berkeley Laboratory and Lawrence Livermore Laboratory). Ambient gas chemistry appears to be controlled by near surface microbial processes (Thorstenson, et al., 1998). The CO_2 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 YM.

Based on the groundwater studies, organic C is expected to be limiting for microbial growth and activity. Nevertheless; it needs to be recognized that unknown, but limited, quantities of xenobiotic C will be introduced into the emplacement drifts (Haldeman, et al., 1996; Horn and Meike, 1995) during the pre-closure 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 ESF (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 C source, the limited quantity of water in the drifts (due to ventilation during the pre-closure and radioactive decay heating in the post-closure) and the limited quantities of organic C 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 the repository's 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, then the potential for microbial activity that could affect seepage and flow would need to be reevaluated.

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

DOE identified long WP lifetime as a key factor in its Repository Safety Strategy (U.S. Department of Energy, 1998a). WP lifetime is embodied in a performance assessment framework by the NRC's KESA as mechanical disruption, WP corrosion, and quantity and chemistry of water contacting WPs and waste forms. The effects of coupled THC processes only influence the abstraction of the latter two KESA (see Appendix A). WP corrosion, which is addressed explicitly as a subissue in the CLST IRSR(U.S. Nuclear Regulatory Commission, 1998d), depends strongly on the near-field chemical environment. As discussed in Section 4.1, the ENFE KTI will help to determine the quantity and chemistry of water contacting WPs. Contributions from the CLST KTI, RDTME KTI, USFIC KTI, and TEF KTI will be required, in addition to those of the ENFE KTI to successfully abstract the two KESA that are necessary to assess the corrosion of waste packages. DOE must adequately consider coupled THC processes affecting the WP chemical environment in its assessments of repository performance.

4.2.1 Review Methods and Acceptance Criteria

DOE's approach to evaluate and abstract coupled THC effects on WP chemical environment in a TSPA for the proposed repository at YM is satisfactory, if the following acceptance criteria are met.

Review Method:

- Staff should ascertain whether DOE demonstrated that sufficient data exist to support the conceptual models and to define relevant parameters in DOE's abstractions. Staff should verify whether DOE provided sound bases for the inclusion or exclusion of certain observed phenomena in its conceptual models. The staff will evaluate the potential for DOE estimates of performance to be over-optimistic, given the excluded set of phenomena and the implementation of coupled geochemical processes in the PA. In its review, staff should determine whether DOE has performed sensitivity and uncertainty analyses to test for the possible need for additional data. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Data and Model Justification Acceptance Criteria

(1) Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment were considered.

(2) DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect the WP chemical environment.

(3) 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 for conceptual models and simulations of THC coupled processes that affect the WP chemical environment.

(4) A nutrient and energy inventory calculation (e.g., McKinley, West, and Grogan, 1985; Grogan and McKinley, 1990; Noy, et al., 1996) should be used to determine the potential for microbial activity that could impact the WP chemical environment.

(5) Should microbial activity be sufficient to allow microbial influenced corrosion (MIC) of the WP, then the time-history of temperature, humidity, and dripping should be used to constrain the probability for MIC (CRWMS M&O, 1997b).

(6) Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.

(7) If the testing program for coupled THC processes on WP chemical environment is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed, DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program.

Review Method:

- Staff should ascertain whether input values used in the quantity and chemistry of water affecting the WPs, and WP corrosion calculations within TSPA, are reasonable based on data from the YM region (e.g., heater test's results) and other applicable laboratory tests and natural analogs. Staff should verify whether these values are consistent with the initial and boundary conditions (design features) and the assumptions of the conceptual models and design concepts for the YM site (e.g., estimations used in these abstractions should be based on the thermal loading strategy, including effects of ventilation; engineered barrier system design, including drift liner, and backfill and drip-shield, if present; and natural system masses and fluxes). In addition, the staff should verify that if any correlations between the input values exist, they have been appropriately established in DOE's TSPA. Finally, the staff should, to the extent feasible, evaluate DOE's input values by comparison to corresponding input values in the staff data set and use the NRC TPA code to test sensitivity of system performance to the input values and correlations used by DOE.

Data Uncertainty and Verification Acceptance Criteria

(1) Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on the WP chemical environment. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.

(2) Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment were considered.

(3) DOE's evaluation of coupled THC processes properly considered 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 the WP chemical environment.

(4) The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on WP chemical environment were consistent with available data.

(5) DOE's performance confirmation program should assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on WP chemical environment.

Review Method:

- Staff should ascertain whether DOE considered plausible alternative models and justified approaches used in the quantity and chemistry affecting the WPs, and WP corrosion abstractions. Staff should use the NRC TPA code to assist in verifying whether the intermediate outputs provided by DOE's approach reflect or bounds the range of uncertainties owing to alternative modeling approaches. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Model Uncertainty Acceptance Criteria

(1) Appropriate models, tests, and analyses were used 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) TH effects on gas and water chemistry; (ii) hydrothermally driven geochemical reactions such as zeolitization of volcanic glass, which could affect water chemistry and WP environmental conditions; (iii) dehydration of hydrous phases liberating moisture that may affect the WP environment; (iv) effects of microbial process on the WP environment; and (v) changes in water chemistry that may result from the release of corrosion products from the WP and interactions between cementitious materials and groundwater, which, in turn, may affect the WP chemical environment.

(2) Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

(3) DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on WP chemical environment. 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.

Review Method:

- Staff should ascertain whether DOE verified that the outputs of quantity and chemistry of water affecting the WPs, and WP corrosion abstractions reasonably reproduce or bound the results of corresponding process-level models or empirical observations. Staff should, to the extent feasible, evaluate the output of DOE's abstractions against results produced by process-level models developed by the staff.

Model Verification Acceptance Criteria

(1) The mathematical models for WP chemical environment 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.

(2) DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate the WP chemical environment.

(3) Abstracted models for coupled THC effects on WP chemical environment were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different mathematical models to judge robustness of results.

Review Method:

- Staff should ascertain whether consistent and appropriate assumptions and initial and boundary conditions have been propagated through DOE's abstraction approaches. For example, staff should determine if the conditions and assumptions used to generate the look-up tables or regression equations are consistent with all other conditions and assumptions in the TSPA for abstracting the quantity and chemistry of water affecting the WPs, and WP corrosion. The important design features that will set the initial and boundary conditions for these abstractions include WP design and materials selection, drift lining, thermal loading strategy, including ventilation, etc. If DOE decides not to take credit for certain design features that have been demonstrated in NRC's or DOE's, or both analyses to provide only benefits and no deleterious effects, staff does not need to include such features in its review. Staff should verify whether DOE's dimensionality abstractions appropriately account for the various design features, site characteristics, and alternative conceptual approaches. In addition, staff should verify whether DOE's domain-based and temporal abstractions appropriately handled the THC couplings or sufficient justification has been provided to exclude these couplings. Staff should, to the extent feasible, use the NRC TPA code to selectively probe DOE's approach for these three abstractions for potential inconsistency in the analysis and non-defensible predictions.

Integration Acceptance Criteria

(1) DOE has considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, physical phenomena, and couplings, and used consistent and appropriate assumptions throughout.

(2) Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment.

(3) Not all THC couplings may be determined to be important to performance, and DOE may adopt assumptions to simplify performance assessment analyses. If potentially important couplings are neglected, DOE should provide a technical basis for doing so. The technical basis can include activities such as independent modeling, laboratory or field data, or sensitivity studies.

(4) Where simplifications for modeling coupled THC effects on WP chemical environment were used for performance assessment analyses instead of

detailed process models, the bases used for modeling assumptions and approximations were documented and justified.

Review Method:

- NRC will attend, as observers, activities conducted by DOE related to model abstractions, and track the progress made in resolving quality assurance deficiencies in the abstraction activities. If DOE uses peer reviews, staff should review DOE's implementation to ensure that the peer reviews followed the guidance in NUREG-1297 and NUREG-1298 (Altman, Donnelly, and Kennedy, 1988a,1988b) or other acceptable approaches. If staff has concerns, they will be noted at the time of staff's attendance and formally communicated to DOE. If DOE uses expert elicitations, NRC will attend, as observers, the elicitation workshops and review the documentation to insure that the expert elicitations followed the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches. If staff has concerns, they will be noted at the time of the elicitation and formally communicated to DOE. Progress made in resolving these concerns will be tracked by the staff.

Programmatic Acceptance Criteria

(1) Data and models were collected, developed, and documented under acceptable QA procedures.

(2) Deficiency reports concerning data quality on issues related to coupled THC effects on WP chemical environment were closed.

(3) If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

4.2.2 Technical Bases for Review Methods and Acceptance Criteria for Effects of Thermal-Hydrologic-Chemical Processes on Waste Package Chemical Environment

The technical bases for the acceptance criteria for coupled THC effects on WP 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 WP chemical environment. As mentioned previously in Section 3, limited analyses in past performance assessments on the effects of coupled geochemical processes on the potential repository WP 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, WP material, etc.). This has resulted in acceptance criteria that primarily focus on ensuring that analyses of the effects are completed. The sophistication of the analysis of the effect of a coupled process on the WP chemical environment that could be conducted and 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.

4.2.2.1 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, 1996). 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. It also is dependent on the composition and surface conditions of the container material. Evaluation of effects of corrosion potentials on WP lifetime are treated in detail in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d). 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 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 due to rearrangement of electronic structure within the solid and a streaming current results.

2. Thermoelectric potential: In metallic corrosion literature, the thermoelectric potentials have been 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 mV) have been measured during the single heater test within the ESF at YM. The measurements were reported to have been made using the same electrodes that were used for conductivity measurements and that were spaced approximately 1 m apart. However, details regarding the input impedance of the voltmeter and the electrode materials are not available at this time. The potential difference, thus, measured above the heater element was about 100 mV and that below the heater was about 800 mV. 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 that were 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) presence of conductive materials; (iv) discontinuity in the electrolyte layer between measuring points especially in a variably saturated medium; (v) 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 'iscussed in geophysical literature (e.g., Corwin and Hoover, 1979) as well as electrochemical literature (e.g., Bard and Faulkner, 1980). For example, assuming that the electrode materials were made of platinum that is 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 mV (Hoare, 1967). Further information is needed regarding the details of the measurement to assess the accuracy of self potential measurements made at the ESF. An accurate measurement of self potentials would entail using non-polarizable 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 WP performance. However, if during the performance confirmation period, 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 above, apply to this survey.

The ability to calculate moisture redistribution at the drift scale is essential to determine how fast the WP will corrode. The time period at which the RH exceeds a critical value, RH_c, is an important factor in determining the container performance. The corrosion rates in humid air at $RH > RH_c$, as well as in liquid, aqueous environments, are higher than those in dry air. The time at which rewetting of the containers occurs depends on the near-field environment. At this scale, the geometry of the individual WP 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 WPs. Thermohydrologic calculation of moisture redistribution is treated specifically in the TEF IRSR (U.S. Nuclear Regulatory Commission, 1998a). However, 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 YM performance calculations. Also, THC processes will affect the chemistry of gas and water that would interact with the WP.

Elevated temperatures in the near field at YM are expected to lead to important geochemical changes. Container oxidation under dry conditions is dictated by the WP temperature and the composition of the gaseous phase. In particular, the oxygen partial pressure is important for container oxidation. Localized reducing conditions could be promoted by near-field hydrologic effects and phase variations. Initial 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_2 and CO_2 from the near-field environment. The vapor pressure of water at temperatures above 95 °C exceeds the hydrostatic pressure of less than 0.1 MPa at YM. 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, 1997).

Aqueous corrosion of steel containers in the repository near field can create a local decrease in Eh, and 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 Fe is irreversible (far from equilibrium) and is diffusion-limited in the aqueous phase. Second, corrosion of Fe in an oxidizing environment leads to the formation of Fe oxides and oxyhydroxides that can further decrease the rate of electrochemical reduction of O_2. Finally, the initial formation of α-FeOOH can lead to a secondary reduction reaction to 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 α-FeOOH, and the cyclic process proceeds due to the electronic conductivity of the inner layer of Fe_3O_4.

4.2.2.2 Effects of Waste Package Corrosion Processes on Waste Package Chemical Environment

As discussed in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d), the outer steel overpack of the WP is included in the design as a corrosion allowance material that is expected to undergo slow, uniform corrosion. However, under some circumstances, dictated by the chemical composition of the WP environment, the steel can undergo localized corrosion or environmentally assisted cracking. The near-field environment may be conditioned by the presence of corrosion products or by modified conditions generated by localized corrosion processes. 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).

Geochemical parameters that affect container performance via uniform or localized corrosion, stress corrosion cracking (SCC), and hydrogen embrittlement include pH, Eh, temperature, Cl^-, NO_3^-, and HCO_3^-/CO_3^{2-}. 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 γ-Fe_2O_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 containers be neutral (pH = 7) or acidic, then active uniform corrosion will be important to WP lifetime calculations. SCC can occur in a HCO_3^-/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.

Under circumstances of localized corrosion, the environment experienced by the inner overpack and other WP materials is the environment inside occluded corrosion areas or cracks in the steel. The evolution of this local environment is affected by a combination of: (i) corrosion of steel that releases Fe^{2+} ions; (ii) reduction of oxygen that diffuses from the outside; (iii) reduction of water or H^+ inside the crevice created by the crack or localized corrosion front; (iv) the hydrolysis of Fe^{2+}; and (v) the electromigration of anionic species, such as Cl^- and SO_4^{2-}, into the crevice and transport of various ionic species out of the localized corrosion or cracked region. It has been predicted that, depending upon the rates of Fe oxidation to Fe^{2+}

and H_2O or H^+ reduction, the pH inside the crevice can be mildly acidic or alkaline (Sridhar, Lichtner, and Dunn, 1996). If Fe^{2+} is oxidized to Fe^{3+} by O_2 dissolved in water, lower pH conditions could be generated locally due to Fe^{3+} hydrolysis.

Changes in the crevice environment will also depend on the external environment, the presence of air, moisture film, and oxide scale on the steel surface. The increase in acidity and Cl^- concentration inside fissures has been observed in the case of meteoritic irons (Buchwald and Clarke, 1989). Increased Cl^- concentration, up to 1 weight percent of corrosion products, has been reported on land-based Fe archeological objects, and up to 13 weight percent Cl has been found in marine-based artifacts (Turgoose, 1982, 1989). The increase in Cl^- concentration in akaganeite (ß-FeOOH) formed in crevices is attributed to the disintegration of meteoritic objects over long time periods (Organ, 1977).

If penetration of the Alloy C-22 (or other Ni-base alloys) inner overpack occurs by localized dissolution, further local acidification may result from hydrolysis of Cr^{3+}. Experimental evidence for acidification within crevices of Ni-Cr-Mo alloys has been documented (Cavanaugh, et al., 1983; Sridhar and Dunn, 1994). At higher temperatures, crevice pH lower than one may be found in cracks or pits. It has been shown that the presence of Mo, which is added to increase the corrosion resistance of these alloys, can decrease the pH further, depending upon the potential inside the crevice.

Thus, depending upon the rate of movement and dripping of water into the container, the water in immediate contact with the containers may become acidic. However, several factors may decrease the corrosion potential established at the inner container surface below the repassivation potential for localized corrosion, precluding the occurrence of this phenomenon and, therefore, the generation of acidic conditions.

4.2.2.3 Effects of Cementitious Materials on Waste Package Chemical Environment

Our analysis of the effects of engineered materials on the chemical environment assumes the reference case design for the proposed repository. This design assumes pre-cast concrete liners, rather than carbon steel ribbing, for drift support. The effects of engineered materials other than cementitious material are likely to be important to containment; however, only a preliminary discussion is presented in the following.

Predicting the geochemical effects of the introduction of engineered materials in the near-field environment spans the engineering and geoscience disciplines. Stability in the repository can be enhanced by the use of engineering materials that are stable in the natural environment, such as copper in a low-sulfur, reducing environment, or by introduction of materials in the near field that react geochemically to improve isolation (Langmuir, 1987). A variety of metal alloys that are thermodynamically unstable in contact with oxidizing water, although kinetically passive, are being considered as container materials for the YM repository. In addition, if carbon steel is used as structural support for the drifts, then reactions affecting the containers could also affect the structural supports. Although corrosion of these materials may be slow, it would consume oxidants in the near-field environment.

It is important to predict changes in near-field chemistry as affected by cementitious materials over the time period of regulatory interest, particularly with respect to solution pH, which is a key parameter controlling container corrosion. However, the presence of cementitious materials will tend to keep HCO_3^-/CO_3^{2-} 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 time period on the order of a few hundred thousand years, under the low flow rates assumed in that study. However, 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-silica-hydrate (CSH) 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 yr), even in a closed system (Atkins, Glasser, and Kindness, 1991). For example, Atkinson, Everitt, and Guppy (1989) indicated that, if recrystallization of the CSH gel occurred in the long term, lower pH could result due to the lower solubility of the crystalline CSH phases. Formation of crystalline CSH phases by recrystallization of pre-existing CSH gel is likely in a HLW repository, due to 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 CSH 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 WP 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 HLW repository.

4.2.2.4 Microbial Effects on Waste Package Chemical Environment

The potential importance of microbial processes to nuclear waste repositories has been recognized internationally for over ten years, and microbial influenced corrosion (MIC) of the WP 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 US HLW program has also realized the potential importance of microbial processes on corrosion of WPs (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).

Horn and Meike (1995) concluded that microbial activity could potentially compromise the integrity of WPs at YM. Subsequent to that study, research has focused on MIC on carbon steel and Alloy 400 (Horn, et al., 1998). Microbial activities increased rates of carbon steel corrosion by a factor of 5-6-fold compared to abiotic corrosion rates (Horn, et al., 1998). DOE has recognized the potential importance of MIC on repository performance and is attempting to ıddress this process in its performance assessments (TRW Environmental Safety Systems, Inc., 1997b). To provide input for its TSPA-VA, DOE conducted an expert elicitation that addressed the potential for MIC (CRWMS M&O, 1997b). The potential for MIC was determined by evaluating the container material, environmental conditions, and the mechanisms that would lead to MIC (CRWMS M&O, 1997b). The TSPA-VA Methods and Assumptions report outlines an approach to address the potential for microbial activity in the near field that will be conducted as part of the near field geochemical environment abstraction activities (TRW Environmental Safety Systems, Inc., 1997b). Some factors that need to be considered in their evaluation and in future performance assessments are described next.

The conditions necessary for microbial activity in the near field have been discussed in Section 4.1.2.3. Only those aspects that differ and are pertinent to the chemical environment of the WP 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 greatly increased 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, then 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 humidity (Stroes-Gascoyne, et al., 1996; CRWMS M&O, 1997b). For instance, as long as the temperature is above 100 °C or the humidity is less than about 60 - 70%, then microbial activity will not occur (Stroes-Gascoyne, et al., 1996; Geesey, 1993). It is likely that the probability of growth of microbes will vary as a function of temperature (CRWMS M&O, 1997b).

To properly address the potential importance to performance of MIC and, if necessary, abstract it into a performance assessment will require two separate activities. First, a mass balance type of model should 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 MIC (CRWMS M&O, 1997b). A material inventory, identifying naturally occurring nutrients as well as those introduced during the pre-closure period of the repository (e.g., backfill derived from the muck pile if used), should be completed. 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). Conservatism in the analysis is introduced because most of the organic inventory will be refractory (Wilder, 1996). The refractory materials will not be assimilated by microbes. 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 HLW 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 YM, then the potential for MIC would need to be assessed. An assessment o MIC was attempted in the DOE WP expert elicitation (CRWMS M&O, 1997b). If abstraction of MIC was necessary, then a time-history for temperature and relative humidity, as well as the assessed potential for water dripping on the WPs, could be used to determine the potential for MIC at YM (CRWMS M&O, 1997b). Evaluation of this abstraction would be documented in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d).

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

DOE has identified slow rate of release of radionuclides from the waste form as a key factor for meeting its objective of acceptably low annual doses to a person living near the site (U.S. Department of Energy, 1998a). Radionuclide release rates and solubility limits constitute one of the NRC key element of system abstraction (KESA) for a TSPA. 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. Both the WP corrosion, and the quantity and chemistry of water contacting WPs and waste form are the two abstractions (see Appendix A) that incorporate THC processes that will determine the chemical environment for radionuclide release. Contributions from the CLST, TEF, and USFIC KTIs will also be required to resolve this 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 Review Methods and Acceptance Criteria

DOE's approach to evaluate and abstract coupled THC processes affecting the chemical environment for radionuclide release in a TSPA for the proposed repository at YM is satisfactory if the following acceptance criteria are met.

<u>Review Method:</u>

- Staff should ascertain whether DOE demonstrated that sufficient data exist to support the conceptual models and to define relevant parameters in DOE's abstractions. Staff should verify whether DOE provided sound bases for the inclusion or exclusion of certain observed phenomena in its conceptual models. The staff will evaluate the potential for DOE estimates of performance to be over-optimistic, given the excluded set of phenomena and the implementation of coupled geochemical processes in the PA. In its review, staff should determine whether DOE has performed sensitivity and uncertainty analyses to test for the possible need for additional data. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Data and Model Justification Acceptance Criteria

(1) Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release were considered.

(2) DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect the chemical environment for radionuclide release.

(3) Sufficient data were collected on 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 the chemical environment for radionuclide release.

(4) A nutrient and energy inventory calculation (e.g., McKinley, West, and Grogan, 1985; Grogan and McKinley, 1990; Noy, et al., 1996) should be used to determine the potential for microbial activity that could impact radionuclide release.

(5) Should microbial activity be sufficient to potentially affect the chemical environment for radionuclide release, then the time-history of temperature, humidity, and dripping (CRWMS M&O, 1997b) should be used 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) and microbial- enhanced dissolution of the HLW glass form (Staudigel, et al., 1995).

(6) Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.

(7) If the testing program for coupled THC processes on the chemical environment for radionuclide release from the engineered barrier system is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed, DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program.

Review Method:

- *Staff should ascertain whether input values used in the quantity and chemistry of water affecting the WPs and waste forms, WP corrosion, and radionuclide release rates and solubility limits calculations within TSPA are reasonable based on data from the YM region (e.g., heater test's results) and other applicable laboratory tests and natural analogs. Staff should verify whether these values are consistent with the initial and boundary conditions and the assumptions of the conceptual models and design concepts for the YM site. For instance, estimations used in each abstraction should be based on the thermal loading strategy, including effects of ventilation; engineered barrier system design, including drift liner, and backfill and drip-shield, if present; and natural system masses and fluxes. Estimation of the amount of the radionuclides released from breached WPs should be based on the initial inventory, chemical form of the radionuclides, and WP degradation model (i.e., how water flows in and out of the failed WPs). In addition, the staff should verify that if any correlations between* the input values exist, they have been appropriately established in DOE's TSPA Finally, the staff should, to the extent feasible, evaluate DOE's input values by comparison to corresponding input values in the staff data set and use the NRC TPA code to test sensitivity of system performance to the input values and correlations used by DOE.

Data Uncertainty and Verification Acceptance Criteria

(1) Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on the chemical environment for radionuclide release. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.

(2) Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release were considered.

(3) DOE's evaluation of coupled THC processes properly considered 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 the chemical environment for radionuclide release.

(4) The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on the chemical environment for radionuclide release were consistent with available data.

(5) DOE's performance confirmation program should assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on the chemical environment for radionuclide release form the EBS.

<u>Review Method:</u>

- Staff should ascertain whether DOE considered plausible alternative models and justified approaches used in the quantity and chemistry affecting the WPs and waste forms, WP corrosion, and radionuclide release rates and solubility limits abstractions. Staff should use the NRC TPA code to assist in verifying whether the intermediate outputs provided by DOE's approach reflects or bounds the range of uncertainties owing to alternative modeling approaches. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Model Uncertainty Acceptance Criteria

(1) Appropriate models, tests, and analyses were used 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) TH effects on gas and water chemistry; (ii) hydrothermally driven geochemical reactions, such as zeolitization of volcanic glass; (iii) dehydration of hydrous phases liberating moisture; (iv) effects of microbial processes; and (v) changes in water chemistry that may result from interactions between cementitious, or WP, materials and groundwater, which, in turn, may affect the chemical environment for radionuclide release.

(2) Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

(3) DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on the chemical environment for radionuclide release. 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.

Review Method:

- Staff should ascertain whether DOE verified that the outputs of quantity and chemistry of water affecting the WPs and waste forms, WP corrosion, and radionuclide release rates and solubility limits abstractions reasonably reproduce or bound the results of corresponding process-level models or empirical observations. Staff should, to the extent feasible, evaluate the output of DOE's abstractions against results produced by process-level models developed by the staff.

Model Verification Acceptance Criteria

(1) The mathematical models for coupled THC effects on the chemical environment for radionuclide release 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.

(2) DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on the chemical environment for radionuclide release.

(3) Abstracted models for coupled THC effects on the chemical environment for radionuclide release were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different mathematical models to judge robustness of results.

Review Method:

- Staff should ascertain whether consistent and appropriate assumptions and initial and boundary conditions have been propagated through DOE's abstraction approaches. For example, staff should determine if the conditions and assumptions used to generate the look-up tables or regression equations are consistent with all other conditions and assumptions in the TSPA for abstracting the quantity and chemistry of water affecting the WPs and waste forms, WP corrosion, and radionuclide release rates and solubility limits. The important design features that will set the initial and boundary conditions for these abstractions include WP design and materials selection, type of spent fuel, waste forms, drift lining, thermal loading strategy, including ventilation, etc. If DOE decides not to take credit for certain design features that have been demonstrated in NRC's or DOE's, or both analyses to provide only benefits and no deleterious effects, staff does not need to include such features in its review. Staff should verify whether DOE's dimensionality abstractions appropriately account for the various design features, site characteristics, and alternative conceptual approaches. In addition, staff should verify whether DOE's domain-based and temporal abstractions appropriately handled the THC couplings or sufficient justification has been provided to exclude these couplings. Staff should, to the extent feasible, use the NRC TPA code to selectively probe DOE's approach for these three abstractions for potential inconsistency in the analysis and non-defensible predictions.

Integration Acceptance Criteria

(1) DOE has considered the relevant features, events, and processes. The abstracted models adequately incorporated important design features; physical phenomena and couplings; and used consistent and appropriate assumptions throughout.

(2) Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release.

(3) Not all THC couplings may be determined to be important to performance, and DOE may adopt assumptions to simplify PA analyses. If potentially important couplings are neglected, DOE should provide a technical basis for doing so. The technical basis can include activities, such as independent modeling, laboratory or field data, or sensitivity studies.

(4) Where simplifications for modeling coupled THC effects on the chemical environment for radionuclide release were used for PA analyses instead of detailed process models, the bases used for modeling assumptions and approximations were documented and justified.

Review Method:

- NRC will attend, as observers, activities conducted by DOE related to model abstractions and track the progress made in resolving quality assurance deficiencies in the abstraction activities. If DOE uses peer reviews, staff should review DOE's implementation to ensure that the peer reviews followed the guidance in NUREG-1297 and NUREG-1298 (Altman, Donnelly, and Kennedy, 1988a,1988b) or other acceptable approaches. If staff has concerns, they will be noted at the time of staff's attendance and formally communicated to DOE. If DOE uses expert elicitations, NRC will attend, as observers, the elicitation workshops and review the documentation to insure that the expert elicitations followed the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches. If staff has concerns, they will be noted at the time of the elicitation and formally communicated to DOE. Progress made in resolving these concerns will be tracked by the staff.

Programmatic Acceptance Criteria

(1) Data and models were collected, developed, and documented under acceptable QA procedures.

(2) Deficiency reports concerning data quality on issues related to coupled THC effects on the chemical environment for radionuclide release were closed.

(3) If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

4.3.2 Technical Bases for Review Methods and Acceptance Criteria for Effects of Coupled Thermal-Hydrologic-Chemical Processes on the Chemical Environment for Radionuclide Release

The technical bases for the acceptance criteria for rate of radionuclide release are given in this section. These bases are primarily focused on explaining how 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, WP 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 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, high pH solutions may be generated due to interaction of water with the concrete and this may significantly alter the solubility of radionuclides. DOE's assessment of this impact would be acceptable if it demonstrated using geochemical equilibrium modeling codes, such a 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.

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

Near-field environmental factors, including Eh, pH, temperature, Cl^-, NO_3^-, and HCO_3^-, and F^-, are governed by near-field THC processes. These factors affect degradation modes of spent fuel (SF) 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 (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 (Cragnolino and Galvele, 1978). Such a potential can be attained naturally under slightly oxidizing conditions (i.e., in the presence of Fe^{3+}). Under the environmental and potential conditions leading to pitting, SCC 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 SCC, 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 ZrF_6^{2-} complexes compared to that of the passive ZrO_2 film.

Corrosion of SF (predominantly UO_2) by contact with the 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 SF is determined by the corrosion potential E_{corr}. UO_{2+x} is a relatively good electronic conductor because of its deviation from stoichiometry. As a result, E_{corr} is a well-defined electrochemical parameter for SF immersed in an aqueous environment. The rates of reduction of species, such as O_2 and H_2O_2, are coupled to the rate of oxidation of UO_{2+x}, establishing E_{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 UO_2.

The pH has an effect on the rate of dissolution of SF 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 U-H_2O 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 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 SF. Anions such as CO_3^{2-}, that form stable soluble complexes with U^{6+} cations, substantially increase the rate of oxidative dissolution (Blesa, Morando, and Regazzoni, 1994). At low CO_3^{2-} concentrations (0.001 M), the rate of dissolution is proportional to the total concentration (Blesa, Morando, and Regazzoni, 1994). At intermediate concentrations of CO_3^{2-} (0.5 M), the dissolution rate depends on the square root of the total concentration (Grambow, 1989). At a higher CO_3^{2-} concentration (1.0 M 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 $Cl^- < PO_4^{3-} < SO_4^{2-} < F^- < CO_3^{2-}$. In the case of PO_4^{3-} and SO_4^{2-}, a maximum in the corrosion rate is observed at intermediate concentrations (about 1.5×10^{-2} M) (Blesa, Morando, and Regazzoni, 1994).

Other species, such as $SiO_2(aq)$, $H_3SiO_4^-$, and $H_2SiO_4^{2-}$, can react with U(VI) 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 SF. Under certain circumstances, acceleration of SF dissolution can occur as a result of spallation of the alteration layers.

Rapid increases in the concentration of SF 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 SF led to precipitation of most of the transuranic elements (Am, Cm, and Pu). Np was not precipitated and remained in solution. Wilson (1990) used semi-static experiments (i.e., involving periodic removal of a leachant aliquot and replacement with fresh solution). He observed that actinide (U, Pu, Am, Cm, and Np) concentrations reached constant values rapidly. This observation suggests that steady-state conditions between SF dissolution and secondary-phase formation are established. Formation of U^{6+} secondary phases, such as uranophane, was confirmed. Actinide concentrations, with the exception of Np, 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 Pu, Am, and Cm 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 UO_2 matrix. Soluble radionuclides, such as ^{137}Cs, ^{90}Sr, and ^{125}Sb, exhibited congruent dissolution from SF 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 U from the UO_2 matrix (Gray, Leider, and Steward, 1992). In semi-static tests, the fractional release of ^{90}Sr, ^{137}Cs, ^{129}I, and ^{99}Tc increased with temperature, and almost linearly with time. Species such as Ca^{2+}, Mg^{2+}, $H_3SiO_4^-$, $H_2SiO_4^{2-}$, and 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 SF (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_3 by alpha-radiolysis of humid air, as well as the generation of formate and oxalate from inorganic C, 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 YM. However, acid generating processes may be counteracted by alkalinity deriving from cement-water interactions.

Through interactions with oxidizing components, including radiolytic products, SF will eventually oxidize and form a large quantity of UO_2^{2+}-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 SF) are rapid relative to transport of U away from the natural geologic setting or the experimentally simulated WP, respectively. Therefore, secondary oxidation products will accumulate and uranyl minerals will have a large effect on near-field physical and chemical conditions.

Secondary U 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 because of the volume expansion. Third, Np, Pu, and other radioactive waste species may be incorporated into secondary U phases by coprecipitation. Fourth, the secondary U phases may limit ingress of water and oxidants to unaltered wastes. Finally, the secondary U phases may control the solubility or dissolution rate of spent fuel. Thus, secondary U phases may control the source term for radionuclide (not just U) releases from the breached WPs. 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 SF will tend to increase with time. In contrast, through a process of Ostwald ripening, increasingly stable secondary phases, with lower solubilities, will crystallize.

The second main waste form planned for the proposed repository at YM is borosilicate glass. Environmental factors affecting the general or localized dissolution rate of borosilicate glasses include Eh, pH, temperature, Cl^-, HCO_3^-, $H_3SiO_4^-$, F^-, and 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, 'he 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 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, Rattray, and Yu, 1996). A glass waste form would be expected to be fairly unreactive for these conditions. Other components of the glass (e.g., B) 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 YM. Both the clay and zeolite minerals are likely to incorporate augmented quantities of components of the engineered barrier system, such as Fe and Ca. 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.

4.3.2.2 Effects of Engineered Materials on the Chemical Environment for Radionuclide Release

Our analysis of the effects of engineered materials on the chemical environment for radionuclide release assumes the reference case design for the proposed repository. This design assumes pre-cast concrete liners, rather than carbon steel ribbing, for drift support. The effects of engineered materials other that cementitious material are likely to be important to the chemical environment for radionuclide release; however, only a preliminary discussion is presented below.

A variety of metal alloys that are thermodynamically unstable in contact with oxidizing water are being considered as container materials for the YM 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 SF. 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 affect 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 Fe^{3+}. The pH can decrease by the hydrolysis of highly-charged cations, such as Cr^{3+} and Fe^{2+}, 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 SF due to the formation of complexing species and chelating species.

The use of cementitious materials, in the form of concrete inverts and linings, is being considered for the estimated 179 km of emplacement drifts of the proposed YM HLW repository. The use of these materials is in addition to the planned use of cement in roadways for construction, and emplacement ramps and service mains (TRW Environmental Safety Systems, Inc., 1996b). 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 CSH because it contains Ca, Si, and H_2O) 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 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.

4.3.2.3 Radiolysis Effects on Radionuclide Release

Radiolysis has complex effects on aqueous oxidation-reduction conditions. According to Dubessy, et al., (1988), the dose of absorbed γ rays is only 0.02 times the dose of absorbed α particles in a given time. Also, according to Spinks and Woods (1976) (cited in Dubessy, et al., 1988), a single 1 MeV α particle can ionize 10^5 molecules as it loses energy. Therefore, the primary cause of water radiolysis is α-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 $OH^\bullet$, where "$\bullet$" denotes a free radical, H_2O_2, $HO_2^\bullet$, and O^{2-} (Spinks and Woods, 1976), could oxidize reduced species (e.g., Fe^0 in the WP to Fe^{2+} and Fe^{3+}, N_2(aq) to NO_2^- or NO_3^-, and U^{4+} to U^{6+}). Molecular hydrogen (H_2) produced as the result of the combination of two $H^\bullet$, in contrast, is relatively non-reactive. 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 SF and glass) instability and radionuclide mobility through both enhancement of oxidative processes and lowering of pH (Wronkiewicz, Young, nd 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 YM repository. On the other hand, bicarbonate could limit the radiolytic pH lowering (Van Konynenburg, 1986), as could cement-water interactions.

4.3.2.4 Microbial Effects on the Chemical Environment for Radionuclide Release

The potential importance of microbial processes to affect 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 HLW glass dissolution and the production of chelating and complexing ligands as result of microbial activity has also been recognized by DOE as being potentially important to radionuclide release. These topics are addressed in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d). 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 YM indigenous microbes, documented in Hersman (1996), suggested that they are capable of producing enough chelating agents (e.g., siderophores) to chelate 0.2 g L^{-1} of ^{239}Pu 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 is not considering microbial effects on radionuclide release in its TSPA-VA (TRW Environmental Safety System, 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), that microbial activity within the engineered barrier system is unlikely to be of significance. This approach has been discussed in detail in Section 4.2.2.3 and a detailed discussion of the limitations for microbial activity has been presented in Section 4.1.2.3. 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, should be conducted. DOE stated that they will 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

DOE considers radionuclide concentration reduction during transport through engineered and natural barriers a key performance attribute of the proposed repository (U.S. Department of Energy, 1998a). Distribution of mass flux between fracture and matrix and retardation of radionuclides in fractures in the unsaturated zone and in the saturated zone constitute three of the NRC key element of system abstraction (KESA) for TSPA. Each of these abstractions (se Appendix A) will be affected by coupled THC processes and are necessary to appropriately describe transport of radionuclides through engineered and natural barriers. Contributions from USFIC and RT KTIs will also be required to resolve this 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 Review Methods and Acceptance Criteria

DOE's approach to evaluate and abstract coupled THC effects on radionuclide transport through engineered and natural barriers in a TSPA is satisfactory if the following acceptance criteria are met.

Review Method:

- Staff should ascertain whether DOE demonstrated that sufficient data exist to support the conceptual models and to define relevant parameters in DOE's abstractions. Staff should verify whether DOE provided sound bases for the inclusion or exclusion of certain observed phenomena in its conceptual models. The staff will evaluate the potential for DOE estimates of performance to be over-optimistic, given the excluded set of phenomena and the implementation of coupled geochemical processes in the PA. In its review, staff should determine whether DOE has performed sensitivity and uncertainty analyses to test for the possible need for additional data. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Data and Model Justification Acceptance Criteria

(1) Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on transport of radionuclides in the near field were considered.

(2) DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect radionuclide transport in the near field.

(3) Sufficient data were collected on 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 transport of radionuclides in the near field.

(4) A nutrient and energy inventory calculation (e.g., McKinley, West, and Grogan, 1985; Grogan and McKinley, 1990; Noy, et al., 1996) should be used to determine the potential for microbial activity that could adversely affect radionuclide transport through engineered and natural barriers.

(5) Should microbial activity be sufficient to potentially cause adverse microbial effects on transport of radionuclides through engineered and natural barriers, then the time-history of temperature, humidity, and water saturation in engineered and natural materials should be used to constrain the probability for these effects.

(6) Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine if additional new data are needed to better define ranges of input parameters.

(7) If the testing program for the effects of coupled THC processes on radionuclide transport is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed, DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program.

Review Method:

- Staff should ascertain whether input values used in the distribution of mass flux between fracture and matrix, retardation in fractures in the unsaturated zone, and retardation in water production zone calculations within TSPA are reasonable, based on data from the YM region (e.g., niche, C-wells, and Busted Butte test's results) and other applicable laboratory tests and natural analogs. Staff should verify whether these values are consistent with the initial and boundary conditions and the assumptions of the conceptual models and design concepts for the YM site. For instance, estimations used in each of the abstractions should be based on the thermal loading strategy, including effects of ventilation; engineered barrier system design, including drift liner, and backfill and drip-shield, if present; and natural system masses and fluxes. Estimation of radionuclide retardation along the transport path from the repository to the water table should be based on the chemical properties of the radionuclide, properties of the various hydrogeologic units, and the affects of coupled THC processes on their properties. In addition, the staff should verify that if any correlations between the input values exist, they have been appropriately established in DOE's TSPA. Finally, the staff should, to the extent feasible, evaluate DOE's input values by comparison to corresponding input values in the staff data set and use the NRC TPA code to test sensitivity of system performance to the input values and correlations used by DOE.

Data Uncertainty and Verification Acceptance Criteria

(1) Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on transport of radionuclides in the near field. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.

(2) Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on radionuclide transport in the near field were considered.

(3) DOE's evaluation of coupled THC processes properly considered 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 transport of radionuclides in the near field.

(4) The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on radionuclide transport in the near field were consistent with available data.

(5) DOE's performance confirmation program should assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on transport of radionuclides in the near field.

Review Method:

- Staff should ascertain whether DOE considered plausible alternative models and justified approaches used in the distribution of mass flux between fracture and matrix, retardation in fractures in the unsaturated zone, and retardation in water production zones abstractions. Staff should use the NRC TPA code to assist in verifying whether the intermediate outputs provided by DOE's approach reflects or bounds the range of uncertainties owing to alternative modeling approaches. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Model Uncertainty Acceptance Criteria

(1) Appropriate models, tests, and analyses were used 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) TH effects on gas and water chemistry in the unsaturated zone and saturated zone; (ii) precipitation of calcite and opal on the footwall of fracture surfaces and the bottoms of lithophysal cavities, which indicates gravity-driven flow in open fractures, and isolation of transport pathways from sorption sites in the rock matrix; (iii) zeolitization of volcanic glass, that could affect transport pathways; (iv) precipitation and dissolution of oxides and hydroxides on fracture surfaces, illitization of smectite, and recrystallization of zeolites to analcime, which could affect sorption characteristics; (v) effects of microbial processes; (vi) effects of corrosion products of container materials and waste forms on transport of radionuclides in the near field; and (vii) changes in hydraulic and sorptive properties of the natural system resulting from interactions between cementitious materials and groundwater.

(2) Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

(3) DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on radionculide transport in the near field. 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.

Review Method:

- Staff should ascertain whether DOE verified that the outputs of the distribution of mass flux between fracture and matrix, retardation in fractures in the unsaturated zone, and retardation in water production zones abstractions reasonably reproduce or bound the results of corresponding process-level models or empirical observations. Staff should, to the extent feasible, evaluate the output of DOE's abstractions against results produced by process-level models developed by the staff.

Model Verification Acceptance Criteria

(1) The mathematical models for coupled THC effects on radionculide transport in the near field 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.

(2) DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on transport of radionuclides in the near field.

(3) Abstracted models for coupled THC effects on seepage and flow were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different mathematical models to judge robustness of results.

Review Method:

- Staff should ascertain whether consistent and appropriate assumptions and initial and boundary conditions have been propagated through DOE's abstraction approaches. For example, staff should determine if the conditions and assumptions used to generate the look-up tables or regression equations are consistent with all other conditions and assumptions in the TSPA for abstracting the distribution of mass flux between fracture and matrix, retardation in fractures in the unsaturated zone, and retardation in water production zones. The important design features that will set the initial and boundary conditions for these abstractions include WP design and materials selection, waste forms, drift lining, thermal loading strategy, including ventilation, etc. If DOE decides not t take credit for certain design features that have been demonstrated in NRC's o DOE's, or both analyses to provide only benefits and no deleterious effects, staff does not need to include such features in its review. Staff should verify whether DOE's dimensionality abstractions appropriately account for the various design features, site characteristics, and alternative conceptual approaches. In addition, staff should verify whether DOE's domain-based and temporal abstractions appropriately handled the THC couplings or sufficient justification has been provided to exclude these couplings. Staff should, to the extent feasible, use the NRC TPA code to selectively probe DOE's approach for these three abstractions for potential inconsistency in the analysis and non-defensible predictions.

Integration Acceptance Criteria

(1) DOE has considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, physical phenomena, and couplings, and used consistent and appropriate assumptions throughout.

(2) Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on transport of radionuclides in the near field.

(3) Not all THC couplings may be determined to be important to performance, and DOE may adopt assumptions to simplify performance assessment analyses. If potentially important couplings are neglected, DOE should provide a technical basis for doing so. The technical basis could include activities, such as independent modeling, laboratory or field data, or sensitivity studies.

(4) Where simplifications for modeling coupled THC effects on radionuclide transport in the near field were used for performance assessment analyses instead of detailed process models, the bases used for modeling assumptions and approximations were documented and justified.

<u>Review Method:</u>

- NRC will attend, as observers, activities conducted by DOE related to model abstractions, and track the progress made in resolving quality assurance deficiencies in the abstraction activities. If DOE uses peer reviews, staff should review DOE's implementation to ensure that the peer reviews followed the guidance in NUREG-1297 and NUREG-1298 (Altman, Donnelly, and Kennedy, 1988a,b) or other acceptable approaches. If staff has concerns, they will be noted at the time of staff's attendance and formally communicated to DOE. If DOE uses expert elicitations, NRC will attend, as observers, the elicitation workshops and review the documentation to ensure that the expert elicitations followed the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches. If staff has concerns, they will be noted at the time of the elicitation and formally communicated to DOE. Progress made in resolving these concerns will be tracked by the staff.

Programmatic Acceptance Criteria

(1) Data and models were collected, developed, and documented under acceptable QA procedures.

(2) Deficiency reports concerning data quality on issues related to coupled THC effects on transport of radionuclides in the near field were closed.

(3) If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

4.4.2 Technical Bases for Review Methods and Acceptance Criteria for 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 previously 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 acceptance criteria that primarily focus on ensuring that some type of analysis of the effects is 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 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, as a result of the water reacting with concrete as it leaves the drift, fluids along flow paths beneath the repository horizon might have a high pH. This condition could substantially affect the values assumed for retardation in the unsaturated zone. One acceptable approach to evaluate this effect would be to conduct sensitivity studies using the DOE PA code, which could be completed using values of K_ds associated with cementitious repositories. If substantial negative changes in the performance of the repository resulted from the use of these alternative K_ds, then it would be expected that these alternative values be used to assess performance of the repository.

4.4.2.1 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 the 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 U in solution through precipitation of reduced U 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 U, but also other actinides through coprecipitation (Murphy and Prikryl, 1996).

The 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. 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.

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 Fe, Mn, and Si, are common fracture lining minerals in the YM 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., teel 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 (TcO_4^-) and does not sorb strongly. Under reducing conditions, Tc^{4+} is predominant species of TC 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). Cations, such as UO_2^{2+}, NpO_2^+, and Am^{3+}, 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 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 TcO_4^- and SeO_4^{2-}, the reverse is true. For these ion 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 sorbant 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 YM 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).

Zeolites, such as clinoptilolite, heulandite, and analcime, may also be important for retarding transport of radionuclides in the near field at YM. Zeolites exhibit a fixed charge developed by substitution of Al^{3+} for Si^{4+} in the zeolite structure. The charge imbalance is compensated by Na^+, K^+, Ca^{2+}, and 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 Cs^+ and 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 (Ca^{2+} or Mg^{2+}) and the carbonate anion (CO_3^{2-}). For this reason, sorption on carbonates is a complex function of pH, solution chemistry, and p(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 (^{125}Sb, ^{60}Co, and ^{54}Mn) were concentrated at the hot side of the experiment. This was probably due to sorption on Fe oxyhydroxides. ^{144}Ce and ^{99}Tc 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 (pH_{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 Am, Ba, Ce, Cs, Eu, Pu, Sr, and U on crushed tuff materials either remain constant or increase with increasing temperature (Meijer, 1990). This assumption is made in current DOE TSPA 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.

Diffusion

Diffusion is a retardation mechanism that is potentially important in the near field at YM. Diffusion is the migration of water from fractures, where transport may be relatively rapid, into the matrix where flow and transport are slow. Field studies at the Nopal I U deposit in the Peña Blanca mining district, Chihuahua, Mexico, suggest that diffusion into the matrix is of limited importance in U retardation (Pearcy, Prikryl, and Leslie, 1995). Instead, U 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, U appears to be coprecipitated with Fe oxyhydroxides and clays. Similar paragenesis have been observed in long-term drip experiments using water, related to that from the J-13 well at YM, and unirradiated UO_2 (Wronkiewicz, et al., 1992).

Gas Transport

Vaporization would partition $^{14}CO_2$ 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 $^{14}CO_2$ into the liquid phase.

Codell and Murphy (1992) performed 1D simulations of ^{14}C transport in unsaturated rock. The results indicated an early initial release of ^{14}C to the gas phase. CO_2 was predicted then to dissolve into the aqueous phase and calcite precipitation served to sequester ^{14}C 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.

Current DOE TSPA models do not explicitly include $^{14}CO_2$ gas transport (TRW Environmental Safety Systems, Inc., 1995). The decision by DOE to not include this mode of transport is consistent with recent recommendations of the National Academy of Sciences (National Research Council, 1995). The Academy considered that $^{14}CO_2$ 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 ^{129}I and ^{36}Cl, 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 are typically called radiocolloids and have been divided into two types (Maiti, Smith, and Laul, 1989; Manaktala, et al., 1995). "True" or "real" colloids are generally 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 C, calcite, silica, clay particles, and oxyhydroxide compounds of metals, such as Fe, Mn, and Al. 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 $p(CO_2)$ 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 has also demonstrated that colloids can be formed as secondary precipitates and clay alteration products. These mineral phases can be released from HLW 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 HLW repository is the most likely source of colloidal materials in the near field. Organics associated with dissolution of vitrified waste forms; secondary alteration products from spent fuel, glass, WPs, 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 HCO_3^- and 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. These radionuclides are then free to sorb 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 pH_{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 (0.3–1 nm) 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.

4.4.2.2 Effects of Engineered Materials on Radionuclide Transport Through Engineered and Natural Barriers

Our analysis of the effects of engineered and man-introduced materials on transport of radionuclides in the near field assumes the reference case design for the proposed repository. This design assumes pre-cast concrete liners, rather than carbon steel ribbing, for drift support. The effects of engineered and man-introduced materials other than cementitious material are likely to be important to radionuclide release.

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} M) can significantly reduce the amount of Np(V) sorbed on kaolinite. EDTA concentrations of the order 10^{-6} M, 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).

Transport of radionuclides potentially can 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 favor 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-silica-hydrate (CSH) 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 CSH 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, et al., (1997) suggest that strong alteration of the YM 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 Fe, Mn, and Si, may be created by oxidation of materials introduced during the construction and operation of the repository (e.g., steel containers and rock bolts). These highly sorbant minerals may form pseudocolloids and facilitate radionuclide transport within the near field. Recent work suggests that colloids that could be formed within the WP or from the spent fuel would most likely be agglomerated as a result of interaction with alkaline fluids associated with cementitious materials (Savage, 1997). Since a concrete invert is envisioned for the drifts, any colloids generated hydrologically up gradient (i.e., in the WP) would be subject to alkaline pore fluids associated with the invert.

4.2.3 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. U 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 U 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) follows. It appears, however, that the organic material hosting the U 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 UO_2^{2+} carbonate complexes would be set up that would diffuse toward the precipitated UO_2, thereby, increasing the probability of U adsorption, reduction, and precipitation. Further study of U 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. Nevertheless, the potential for radiolytic reduction needs to be considered and its effect on mobility understood. Notably, DOE's TSPA completed in 1995 (TRW Environmental Safety Systems, Inc., 1995) does not consider potential radiolytic effects on the source term or radionculide 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, perhaps radiolysis can 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.

4.4.2.4 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 have also 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 YM, 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, the DOE program has argued that potentially deleterious impacts of microbial activity on the radioactive waste environment at YM include the increased rate of transport of radionuclides from breached WPs (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 is not planning to address microbial effects on radionuclide transport in its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b).

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 4.2.2.3 and a detailed discussion of the limitations for microbial activity has been presented in Section 4.1.2.3. DOE has stated that it will use this type of approach in its TSPA-VA to evaluate the potential for microbiological processes in the near field geochemical environment (TRW Environmental Safety Systems, Inc., 1997b).

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 HLW creates a potential for sustained neutron chain reaction (criticality event). Such an event could arise if there is failure of the WP, dissolution of the fissile material, and redeposition outside the WP in the near-field environment. The acceptance criteria developed here are limited to consideration of criticality resulting from coupled THC processes in the near-field environment of the proposed YM repository. Criticality issues related to canister and waste form design issues are considered in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d). Criticality issues in the far field of the proposed repository are addressed in the RT IRSR (U.S. Nuclear Regulatory Commission, 1998e). The review process will determine whether nuclear criticality in the near-field environment due to coupled THC processes has been adequately considered by 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 Review Methods and Acceptance Criteria

DOE's approach to abstract the affects of coupled THC processes on potential nuclear criticality in the near field in a TSPA is satisfactory if the following acceptance criteria are met. Note that acceptance criteria for scenario analysis, contained in the TSPA IRSR (U.S. Nuclear Regulatory Commission, 1998f), were not yet available when Revision 1 of the ENFE IRSR was completed (U.S. Nuclear Regulatory Commission, 1998g). As a consequence, all the acceptance criteria necessary to evaluate scenarios that can affect the performance of the proposed repository over the compliance period, such as criticality in the near field, were not included in Revision 1 of the ENFE IRSR. Subsequently, acceptance criteria for scenario analysis were developed (U.S. Nuclear Regulatory Commission, 1998c). The scenario analysis acceptance criteria allow features, events, or processes to be excluded from a performance assessment based on regulatory, probability, or consequence arguments (U.S. Nuclear Regulatory Commission, 1998c). The approach developed in Revision 1 of the ENFE IRSR was based on a consequence approach. This approach examines the consequences of criticality (steady state or transient) without evaluating the probability of processes that could lead to criticality.

Review Method:

- Staff should verify whether DOE provided adequate technical justification or a conservative basis for the inclusion or exclusion of certain observed phenomena in its conceptual models. The staff will evaluate the potential for DOE estimates of performance to be over-optimistic, given the excluded set of phenomena and the implementation of coupled geochemical processes in the PA. In its review, staff should determine whether DOE has performed sensitivity and uncertainty analyses to test for the potential consequences on repository performance and for the possible need for additional data.

Scenario Screening Acceptance Criterion

(1) Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were completed to determine whether criticality will impact repository performance, and whether additional new data are needed to better define ranges of input parameters.

Should sensitivity studies indicate that the consequences of criticality in the near field would affect repository performance during the compliance period, then the following review methods and acceptance criteria would apply to the abstraction of criticality into a TSPA.

Review Method:

- Staff should ascertain whether DOE demonstrated that sufficient data exist to support the conceptual models and to define relevant parameters in DOE's abstractions. Staff should verify whether DOE provided sound bases for the inclusion or exclusion of certain observed phenomena in its conceptual models. In its review, staff should determine whether DOE has performed sensitivity and uncertainty analyses to test for the possible need for additional data. The description of the performance confirmation program should be reviewed to ascertain whether necessary information will be collected.

Data and Model Justification Acceptance Criteria

(1) Available 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 were considered.

(2) DOE's evaluation of coupled THC processes properly considered 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.

(3) Sufficient data were collected on 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 may affect nuclear criticality in the near-field environment.

Review Method:

- Staff should ascertain whether input values used in criticality calculations for the near field within TSPA are reasonable, based on data from the YM region and other applicable laboratory tests and natural analogs. Staff should verify whether these values are consistent with the initial and boundary conditions and the assumptions of the conceptual models and design concepts for the YM site. In addition, the staff should verify that if any correlations between the input values exist, they have been appropriately established in DOE's TSPA. Finally, the staff should, to the extent feasible, evaluate DOE's input values by comparison to corresponding input values in the staff data set and use the NRC TPA code to test sensitivity of system performance to the input values and correlations used by DOE.

'ata Uncertainty Verification Acceptance Criteria

(1) Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on potential nuclear criticality in the near-field environment. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.

(2) Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality were considered.

(3) DOE's evaluation of coupled THC processes properly considered 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.

(4) The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on potential nuclear criticality in the near-field environment were consistent with available data.

Review Method:

- Staff should ascertain whether DOE considered plausible alternative models and justified approaches used in the abstractions of criticality in the near field. Staff should use the NRC TPA code to assist in verifying whether the intermediate outputs provided by DOE's approach reflects or bounds the range of uncertainties owing to alternative modeling approaches.

Model Uncertainty Acceptance Criteria

(1) Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

(2) DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on potential nuclear criticality. 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.

Review Method:

- Staff should ascertain whether DOE verified that the outputs of the criticality abstractions reasonably reproduce or bound the results of corresponding process-level models or empirical observations. Staff should, to the extent feasible, evaluate the output of DOE's abstractions against results produced by process-level models developed by the staff.

Model Verification Acceptance Criteria

(1) The mathematical models for coupled THC effects on potential nuclear criticality 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.

(2) DOE appropriately adopted accepted, and well-documented, procedures to construct and test the numerical models used to simulate coupled THC effects on potential nuclear criticality.

(3) Abstracted models for coupled THC effects on potential nuclear criticality were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different mathematical models to judge robustness of results.

Review Method:

- Staff should ascertain whether consistent and appropriate assumptions and initial and boundary conditions have been propagated through DOE's abstraction approaches. For example, staff should determine if the conditions and assumptions used to generate the look-up tables or regression equations are consistent. The important design features that will set the initial and boundary conditions for the abstraction include WP design and materials selection, waste forms, drift lining, thermal loading strategy, including ventilation, etc. If DOE decides not to take credit for certain design features that have been demonstrated in NRC's or DOE's, or both analyses to provide only benefits and no deleterious effects, staff does not need to include such features in its review. Staff should verify whether DOE's dimensionality abstractions appropriately account for the various design features, site characteristics, and alternative conceptual approaches. In addition, staff should verify whether DOE's domain-based and temporal abstractions appropriately handled the THC couplings or sufficient justification has been provided to exclude these couplings. Staff should, to the extent feasible, use the NRC TPA code to selectively probe DOE's approach for these three abstractions for potential inconsistency in the analysis and non-defensible predictions.

Integration Acceptance Criteria

(1) DOE has considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, including criticality safety features; physical phenomena and couplings, including neutron absorbers; and used consistent and appropriate assumptions throughout.

(2) Important mass transfer and mass transport processes and mechanisms considered for formation of both a critical mass and configuration are plausible for the YM near-field environment.

(3) Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality.

(4) Criticality in the near field, and not all THC couplings, may be determined to be important to performance, and DOE may adopt assumptions to simplify PA analyses. If potentially important couplings and criticality in the near field are neglected, DOE should provide a technical basis for doing so. The technical basis could include activities, such as independent modeling, laboratory or field data, or sensitivity studies.

(5) Where simplifications for modeling coupled THC effects on potential nuclear criticality were used for PA analyses instead of detailed process models, the bases used for modeling assumptions and approximations were documented and justified.

Review Method:

- NRC will attend, as observers, activities conducted by DOE related to mode abstractions, and track the progress made in resolving quality assurance deficiencies in the abstraction activities. If DOE uses peer reviews, staff should review DOE's implementation to ensure that the peer reviews followed the guidance in NUREG-1297 and NUREG-1298 (Altman, Donnelly, and Kennedy, 1988a, b) or other acceptable approaches. If the staff has concerns, they will be noted at the time of staff's attendance and formally communicated to DOE. If DOE uses expert elicitations, NRC will attend, as observers, the elicitation workshops and review the documentation to ensure that the expert elicitations followed the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches. If staff has concerns, they will be noted at the time of the elicitation and formally communicated to DOE. Progress made in resolving these concerns will be tracked by the staff.

Programmatic Acceptance Criteria

(1) Data and models were collected, developed, and documented under acceptable QA procedures.

(2) Deficiency reports concerning data quality on issues related to coupled THC effects on the potential for nuclear criticality were closed.

(3) If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

4.5.2 Technical Bases for Review Methods and Acceptance Criteria for 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 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 HLW repository at YM can theoretically reach criticality in a geologic medium. These materials may include weapons-grade Pu, highly enriched uranium from naval and research reactors, and small amounts of plutonium and enriched U from commercial and DOE-owned spent fuel.

4.5.2.1 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, then the system is subcritical. If an equal number of neutrons are produced as are lost or absorbed, then the system is critical. If more neutrons are produced than are lost or absorbed, then the system is supercritical.

The critical state is mathematically represented by a parameter called k_{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_{eff} equal to one. A subcritical system has a neutron population that decreases in time, k_{eff} less than one. A supercritical system has a neutron population that increases in time, k_{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_{∞}. The value of k_{eff} is always less than or equal to the value of k_{∞}. Typically, an upper bound for subcritical conditions is to have a calculated k_{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 alled 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 HLW disposal operations, the factors most important to criticality include: (i) concentration of fissile material, principally ^{239}Pu and ^{235}U; (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 ^{239}Pu concentration of 7.2 g/L is generally reported as the minimum critical concentration of Pu (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.*, in 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 HLW 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 Pu 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.

4.5.2.2 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 ^{239}Pu, ^{235}U and other fissile materials are mobilized from waste forms in the YM 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 Pu into a greater volume of rubbilized 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 ^{239}Pu with water and SiO_2 as a proxy for rock. For the idealized spherical Pu-H_2O-SiO_2 geometry considered, Bowman and Venneri (1995) noted that a sphere of about 25 cm 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 kg of ^{239}Pu 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 ^{235}U would require an even larger critical mass than ^{239}Pu. 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. Several critiques of the hypothesis proposed by Bowman and Venneri (1995) have been prepared (Murphy, Jarzemba, 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 YM 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 whether colloid transport through the unsaturated zone can move and concentrate the amounts of Pu that are necessary. At present, experiments suggest that glass does not alter to form pure Pu phases, but instead alters to a suite of clays and secondary minerals (Bates, et al., 1992). In addition, if Pu is bound to existing particles and transported as pseudocolloids, the mass of Pu required for criticality will be larger than that required for a pure Pu phase.

- Low Pu solubility limits (10^{-6} to 10^{-12} molar) that potentially require large volumes of water to provide the mass of ^{239}Pu 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 cm radius sphere to transport the potentially critical 15 kg of ^{239}Pu by means of dissolution and redeposition (Murphy, Jarzemba, and Lichtner, 1995).

- The poor analogy between SiO_2 and the host rock at YM. Other constituents in the waste (e.g., ^{238}U), waste canister (e.g., Fe), 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 ^{239}Pu) from consideration in the YM system. Also, the heterogeneity of the YM 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 YM system would be necessary to evaluate the masses of ^{239}Pu or ^{235}U 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 analyses 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).

4.5.2.3 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, for criticality to occur would require sufficient neutron moderators and with sufficiently few poisons. 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 Pu 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.

It would be a difficult task to determine in detail how these processes would work over long time frames in a repository at YM and to determine the probability of generation of a critical configuration. Considering the very restricted configurational, chemical, and isotopic conditions under which criticality can occur, it appears to be an unlikely possibility in the near field, particularly relative to criticality in waste containers where fissile materials are already concentrated.

.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 evolution of the near-field environment. The site characterization concerns were documented in the Staff Site Characterization Analysis (SCA) of DOE's Site Characterization Plan (SCP), YM Site, Nevada (U.S. Nuclear Regulatory Commission, 1989). Additionally, a letter from NRC to DOE with results of our review of DOE's thermohydrology testing and modeling program (U.S. Nuclear Regulatory Commission, 1997a) described concerns on DOE's coupled THC modeling efforts. Our concerns on near field topics within DOE's performance assessment program have been documented (U.S. Nuclear Regulatory Commission, 1996; U.S. Nuclear Regulatory Commission, 1998h). 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 our application of review methods and acceptance criteria, presented in Section 4, to the DOE Viability Assessment (VA). In Section 5.5 a summary is provided that updates the status of subissue resolution.

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 SCP (U.S. Nuclear Regulatory Commission, 1989) and Study Plan (SP) 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 SCA, DOE has adopted a revised program plan (U.S. Department of Energy, 1996b) and the Repository Safety Strategy (U.S. Department of Energy, 998a). The safety strategy included a number of hypotheses concerning safety attributes of he proposed HLW repository (U.S. Department of Energy, 1998a). DOE's refocused program, a result of Congressional direction (U.S. Department of Energy, 1996b; page 11), incorporates the predecessor to the Repository Safety Strategy (U.S. Department of Energy, 1996a). Now DOE's 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 SCP have changed in scope, been deferred, or canceled (U.S. Department of Energy, 1997a; Appendices A and G). NRC has refocused its pre-licensing program to address those issues most significant to repository performance (Sagar, 1996). In addition, the proposed NRC site-specific regulations for the proposed repository are performance based (U.S. Nuclear Regulatory Commission, 1999a). Some of the SCA and SP comments are no longer valid as the result of the changes in the overall DOE program and NRC's refocused program. Additionally, information from both DOE and ongoing work by NRC and Center staff has become available to close open items. As a result, two of the SCA 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 SCA and SP. However, the technical concerns of some of these issues remain with the staff. NRC's disposition of the SCA and SP comments and questions is provided below and is summarized in Table 1.

SCA Comment 25: The SCP 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, page 4-29).

NRC DISPOSITION OF COMMENT: 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 CLST KTI has lead responsibility for this comment, the ENFE KTI also considers 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 (WP) 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 DOE's Repository Safety Strategy Hypotheses 13 and 14. DOE intends to submit a study plan (8.3.4.2.4.1) to NRC to address this issue and to submit a supplemental response to this comment to NRC (U.S. Department of Energy, 1997a).

DOE is addressing resolution of this comment in a site characterization framework through submission of a study plan. In addition, DOE is also evaluating the potential impact to performance of WP degradation products within its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). We still consider the effects of degradation products on repository performance important. To resolve this comment, we will apply the acceptance criteria in this IRSR to DOE's approach to evaluate the effects of WP degradation products on transport of radionuclides in the TSPA-VA (see Section 5.4.3 and 5..4.4 for detailed analysis).

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 conditions within the waste packages were omitted. 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 WP degradation products on radionuclide transport.

SCA 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; page 4-32).

Comment 29 relates to subissue 2 (WP lifetime), subissue 3 (rate of release of radionuclides from breached WPs), and subissue 4 (transport of radionuclides through engineered barriers and natural barriers). We considered this comment resolved based on DOE's 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 the Study 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 DOE's characterization of microbial effects as part of the issue resolution process for the ENFE KTI.

SCA Comment 79: It has not been demonstrated that the test environment in WP corrosion tests is fully representative of the repository environment (U.S. Nuclear Regulatory Commission, 1989; page 4-66).

NRC DISPOSITION OF COMMENT: Comment 79 relates to Subissue 2 (chemical environment of WP). The ENFE KTI has lead responsibility for this comment. We considered this comment open because DOE indicated that the test environments for WP corrosion tests will evolve as site data and detailed designs become available (U.S. Nuclear Regulatory Commission, 1991). NRC technical concerns are embodied in DOE's Repository Safety Strategy Hypothesis 7. DOE intends to submit a study plan (8.3.4.2.4.1) to NRC to address this issue and to submit a supplemental response to this comment to NRC (U.S. Department of Energy, 1997a).

DOE is addressing resolution of this comment in a site characterization framework through submission of a study plan. In addition, DOE is also evaluating the potential impact to performance of the WP chemical environment within the WP degradation module of its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). The DOE model is expected to include the interaction between inner corrosion resistant alloy and outer corrosion allowance material. Crevice formation between the outer barrier, and the outer barrier corrosion product precipitates 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 will be considered in the TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b, page 6-60).

The chemistry within pits and crevices may vary substantially from the bulk fluid chemistry in contact with the WP 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 $FeCl_3$ solutions that may occur in crevices (Sridhar, Lichtner, and Dunn, 1996). Resolution of the chemistry in this environment will require input from the CLST KTI. Thus, we still consider the test environment in WP corrosion tests important. To resolve this comment, we will apply the acceptance criteria in this IRSR, in combination with review by CLST staff, according to the acceptance criteria in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d).

The CLST KTI resolved this comment within their IRSR as a result of an Appendix 7 meeting on the DOE 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 CLST KTI. This comment is resolved.

SCA Comment 81: Investigation into the stress corrosion behavior of the container alloys assume 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: The CLST KTI has lead responsibility for Comment 81. This comment was open because DOE indicated that the test environments for WP corrosion tests will evolve as site data and detailed designs become available (U.S. Nuclear Regulatory

Commission, 1991). In Revision 0 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1997b), the ENFE KTI considered that Comment 81 was related to Subissue 2 (chemical environment of the waste package), partly because the CLST KTI was not fully funded, and detailed discussions with cognizant staff were not possible. Discussions with the CLST KTI team, which has been funded at the Center during FY 1998, 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 and will not be considered in future revisions of the ENFE IRSR.

The CLST KTI resolved this comment within their IRSR as a result of an Appendix 7 meeting on the DOE 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 CLST KTI. This comment is resolved.

SCA 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; page 4-68).

NRC DISPOSITION OF COMMENT: The CLST 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). In Revision 0 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1997b), the ENFE KTI considered that Comment 84 was related 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), partly because the CLST KTI was not fully funded, and detailed discussions with cognizant staff were not possible. Discussions with the CLST KTI team, which has been funded at the Center during FY 1988, 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 and will not be considered in future revisions of the ENFE IRSR.

The CLST KTI resolved this comment within their IRSR as a result of an Appendix 7 meeting on the DOE 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 CLST KTI. This comment is resolved.

SCA 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; page 4-71).

NRC DISPOSITION OF COMMENT: 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 CLST KTI also considers the comment important to issue resolution. We considered Comment 89 open. DOE's response indicated that testing programs will investigate how water chemistry is changed by the WP 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 NRC to address this issue and to submit a supplemental response to this comment to NRC (U.S. Department of Energy, 1997a).

DOE is addressing resolution of this comment in a site characterization framework through submission of a study plan. In addition, DOE is also evaluating the potential importance of variation in the pH on the performance of the WP 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. To resolve this comment, we will apply the acceptance criteria in this IRSR to DOE's approach in the TSPA-VA. We will evaluate DOE's analysis of the effects of pH variation, resulting from the use of grouts, cements, and organic materials, on WP and glass form degradation (see Section 5.4.3 for detailed analysis).

The CLST KTI partially resolved this comment within their IRSR as a result of an Appendix 7 meeting on the DOE 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 CLST IRSR indicated that a full range of the evolving chemical environments at the proposed YM repository should be considered in the long-tem glass corrosion studies (U.S. Nuclear Regulatory Commission, 1998d, page 55). The ENFE staff concurs with the conclusion of the CLST KTI. Thus the technical concerns of this comment remain with the staff.

SCA Comment 90: The effects of varying oxygen concentrations on corrosion of the metal canisters are not considered (U.S. Nuclear Regulatory Commission, 1989; page 4-71).

NRC DISPOSITION OF COMMENT: Comment 90 is related to Subissue 2 (chemical environment of waste package). The CLST 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 RH chambers are being performed at Lawrence Livermore National Laboratory in support of DOE's WP 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 CLST KTI also resolved this comment within their IRSR as a result of an Appendix 7 meeting on the DOE 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 CLST KTI. This comment is resolved.

SCA 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; page 4-72). .

Comment 92 relates to all of the subissues of the ENFE KTI. We considered this comment resolved based on the DOE 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 NRC guidance and in DOE 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 evolution of the near-field environment and its impact on performance as part of the issue resolution process within the ENFE KTI.

SCA 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; page 4-115)?

NRC DISPOSITION OF COMMENT: 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 CLST 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. DOE intends to submit the study plan (8.3.4.2.4.1) to NRC to address this issue and to submit a supplemental response to this comment to NRC (U.S. Department of Energy, 1997a).

DOE is addressing resolution of this comment in a site characterization framework (Wang et al., 1998). In addition, DOE is also evaluating the potential impact to performance of the WP chemical environment within the WP degradation module in its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b). We still consider the chemical environment (water quality) of the WP to be important to repository performance. To resolve this comment, we will apply the acceptance criteria in this IRSR to DOE's approach to evaluate the effects of the chemical environment on WP degradation in the TSPA-VA (see Section 5.4.2 for a detailed analysis).

Our review of the TSPA-VA confirms that the technical concerns of this comment remains with the staff. For instance, the effects of salt formation and increased pH as a result of interaction with concrete on WP corrosion rates were omitted. The impact of water reacting with the concrete drift liner could lead to early failure of the outer carbon steel layer of the waste package. This could lead to rapid failure of the waste package because of the greater likelihood of localized corrosion of the inner barrier.

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. NRC staff recommended that the study include petrographic analysis for determining textural relationships of minerals along the transport pathways between the Topopah Spring and the accessible environment.

NRC DISPOSITION OF COMMENT: This comment is new to this revision of the ENFE IRSR. The comment was incorporated from the Open Item Tracking System (OITS). The OITS 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: This question is new to this revision of the ENFE IRSR. The question was incorporated from the OITS. 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: This question is new to this revision of the ENFE IRSR. The question was incorporated from the OITS. 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: This question is new to this revision of the ENFE IRSR. The question was incorporated from the OITS. 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).

Table 1. Summary of ENFE KTI open Item status

Item ID	Status	Title	Comment
OSC0000001347C025	Closed	No rationale for additional testing needs to determine the influence of package degradation products and radionuclide interactions on sorption	
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 EBS 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 if varying oxygen concentrations on waste package corrosion is 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 generated heat	U.S. Nuclear Regulatory Commission, 1997b

Table 1. Summary of ENFE KTI open item status (cont'd)

Item ID	Status	Title	Comment
OSC0000001347Q030	Closed	Expected quality if water which may contact waste packages and changes in quality over time	
OSP0000831321C001	Closed	Inadequate information collected to determine textural relationships of minerals along groundwater pathways	Revision 2 of ENFE IRSR
OSP0000831321Q002	Closed	Potential for bias in retardation calculations from use of vertical core thin sections to obtain groundwater pathway mineral data	Revision 2 of ENFE IRSR
OSP0000831321Q004	Closed	Methods for determining changes in lithology for mineral studies	Revision 2 of ENFE IRSR
OSP0000831321Q005	Closed	The difference between software and model verification/validation for mineralogy studies	Revision 2 of ENFE IRSR

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 three 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 three addressed whether the DOE testing and modeling strategy included means for bounding the effects of thermal-hydrologic-chemical coupled processes on repository performance.

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 yet fully evaluated the potential impacts on repository performance. 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). DOE has stated that "simplifications that patch thermal-mechanical and/or thermal-chemical influences into an UZ-TH 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-VA to ensure that conservative bounds on the effects of coupled THC processes on repository performance have been demonstrated and used in 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 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; 1998h). Many conservatisms were considered in TSPA-95. However, several potential WP 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 the DOE 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 ar 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 overpack material. We will evaluate how DOE accounts for these effects in its evaluation of WP and total system performance in TSPA-VA.

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 Np, Pu, and Am 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-VA (TRW Environmental Safety System, Inc., 1997b).

NRC has stated its continuing concerns with some aspects of DOE's performance assessment as they relate to an acceptable license application (U.S. Nuclear Regulatory Commission, 1998h). Some of the concerns address issues contained in this IRSR. For example, there is an apparent inconsistency in how the cementitious liner is included in the analysis of performance in different models. The liner is accounted for near-field chemistry calculations, but is apparently neglected in hydrologic models. The cementitious liner will affect seepage while intact and may affect flow within and out of the drift after it has collapsed. We will review the TSPA-VA information to ensure that all significant features and processes associated with the evolution of the near field environment were considered by DOE in its PA.

Uncertainty in near-field environmental conditions, as it impacts WP performance, does not appear to be fully captured by the performance assessment modeling (U.S. Nuclear Regulatory Commission, 1998h). There are two concerns arising from the expert elicitations that 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 were unable to be used to develop the initial and boundary conditions for the WP corrosion panel. This situation arose because the WP elicitation was completed prior to the initiation of the near field elicitation. Our concerns on DOE's PAs will be evaluated by applying the acceptance criteria in this IRSR to the TSPA-VA.

5.4 STATUS OF SUBISSUE RESOLUTION AT THE STAFF LEVEL

Revision 1 of the ENFE IRSR has focused on developing somewhat general acceptance criteria for all subissues, including the new subissue of criticality with the near field environment. This systematic approach has been taken to issue resolution within the ENFE IRSR, as limited PA calculations are available to refine the acceptance criteria to be repository and design specific, and performance-based. A technical approach, and its associated acceptance criteria, has been presented to evaluate the potential for microbial activity in the near field environment. Revision 2 will describe the status of issue resolution as a result of the application of the acceptance criteria to DOE's TSPA-VA. The coupled THC processes that may impact performance and the extent of subissue resolution are summarized below for each subissue.

Revision 2 describes the status of issue resolution as a result of application of ENFE IRSR acceptance criteria to DOE's TSPA-VA. At this time, the repository concept of operations (i.e., thermal load and ventilation) and the design of the engineered barrier system remain in flux. In the next revision of the IRSR, acceptance criteria in Section 4 of the ENFE IRSR will be applied to whatever repository concept and design is chosen by DOE for its TSPA-Site Recommendation.

NRC has completed its review of the Viability Assessment (VA). We provided DOE our evaluation of the VA (U.S. Nuclear Regulatory Commission, 1999b). The staff identified several challenges for DOE to assemble a complete a high-quality LA within the time frame envisioned in the LA Plan. We reviewed the preliminary design concept, total system performance assessment, and LA Plan (Table 2). In revising this IRSR, supporting documents such as the TSPA-VA Technical Basis Document and the Near-Field/Altered-Zone Models Report (Table 2) were also examined. The comments on TSPA-VA provided to the DOE addressed key elements of DOE's PA (U.S. Nuclear Regulatory Commission, 1999b). This review approach is described in the Total System Performance Assessment and Integration (TSPAI) IRSR (U.S. Nuclear Regulatory Commission, 1998c). Review of each performance assessment abstractions is based on five technical acceptance criteria (U.S. Nuclear Regulatory Commission, 1998c): (i) data and model justification, (ii) data uncertainty and verification, (iii) model uncertainty, (iv) model verification, and (v) integration.

The review presented here focuses on the ENFE Key Technical Issues and its subissues. Each subissue is pertinent to more than one abstraction within the PA. The abstractions influenced by the evolution of the near-field environment in each subissue are reiterated in introductory comments for each subissue. The coupled processes that need to be considered in the evaluation of each abstraction are also presented in introductory remarks for each subissue. The review in the following sections provides more detailed comments than in our previous comments to the DOE (U.S. Nuclear Regulatory Commission, 1999b). However, particular challenges for DOE, presented in our review of the VA (U.S. Nuclear Regulatory Commission, 1999b), are also summarized in the introductions to each subissue. These comments are followed by detailed comments on the TSPA-VA, developed as a result of the application of the acceptance criteria of this IRSR to the TSPA-VA.

Acceptance criteria for each ENFE subissue are grouped according to the technical acceptance criteria used in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 1998c). Review methods used by us to evaluate the TSPA-VA in this IRSR are presented in Section 4. The DOE Viability Assessment (e.g., U.S. Department of Energy, 1998b) was responsive to subissues identified in Revision 0 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1997b). This responsiveness helped to facilitate our review of the VA.

Table 2. Primary references reviewed to assess the status of resolution

Document Title	Citation
Viability Assessment of a Repository at Yucca Mountain, Volume 2: Preliminary Design Concept for the Repository and Waste Package	U.S. Department of Energy, 1998c
Viability Assessment of a Repository at Yucca Mountain, Volume 3: Total System Performance Assessment	U.S. Department of Energy, 1998b
Viability Assessment of a Repository at Yucca Mountain, Volume 4: License Application Plan and Costs	U.S. Department of Energy, 1998d
Total System Performance Assessment–Viability Assessment (TSPA-VA) Analyses Technical Basis Document	Civilian Radioactive Waste Management System Management and Operating Contractor (CRWMS M&O), 1998d
Near-Field/Altered-Zone Models Report	Hardin, 1998

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

Processes that should be evaluated to determine their potential importance to repository performance include: (i) zeolitization of volcanic glass, which could affect flow pathways; (ii) precipitation of calcite and opal on the footwall of fracture surfaces and the bottoms of lithophysal cavities, which indicates gravity driven flow in open fractures that could affect permeability and porosity; and (iii) potential dehydration of zeolites and vitrophyre glass, which could release water affecting heat and fluid flow. The effects of THC coupled processes that may occur due to interactions with engineered materials or their alteration products include: (i) changes in water chemistry that may result from interactions between cementitious materials and groundwater, which, in turn, may affect seepage and flow; (ii) dissolution of the geologic barrier (e.g., tuff) by a hyperalkaline fluid that could lead to changes in the hydraulic properties of the geologic barrier; and (iii) precipitation of calcite or CSH phases along fracture surfaces as a result of migration of a hyperalkaline fluid that could affect hydraulic properties.

Changes in pore-water and gas chemistry due to microbial activity that could affect seepage and flow, and the repository's performance, will be insignificant relative to chemical changes expected as the result of THC process 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 PA.

Three abstractions are influenced by the evolution of the near-field environment within the scope of the seepage and flow subissue (see Appendix A). The abstractions are distribution of mass flux between fracture and matrix; spatial and temporal distribution of flow; and quantity and chemistry of water contacting the waste packages and waste forms. In the near-field environment, three main coupled thermal-hydrologic-chemical (THC) processes will occur that have the potential to affect seepage and flow and, hence, repository performance. These processes need to be considered in the evaluation of each abstraction. The processes are dehydration of zeolitic horizons, coupled THC processes that affect the porosity and permeability of the natural system, and coupled THC processes that occur at the interface of the natural system and the engineered components.

Both the distribution of mass flux between fracture and matrix abstraction and the spatial and temporal distribution of flow abstraction were evaluated (U.S. Nuclear Regulatory Commission, 1999b). Based on the status of DOE efforts in these technical areas, as reflected in the TSPA-VA and License Application Plan (U.S. Department of Energy, 1998b,d), there is general staff agreement that DOE's planned work appears adequate (U.S. Nuclear Regulatory Commission, 1999b). Nonetheless, review of the TSPA-VA is presented below to document the status of subissue resolution and to outline the path forward to resolution.

However, data and models used in the TSPA-VA to calculate the quantity and chemistry of water dripping on waste packages were determined to be inadequate to describe the process and extent of potential dripping under thermally-altered conditions (U.S. Nuclear Regulatory Commission, 1999b). This is an important issue because both DOE and NRC PA analyses indicate that the fraction of WPs contacted by water is the most important factor affecting dose for the groundwater pathway. In addition, we determined that current DOE testing and modeling plans are not sufficient to resolve the issue prior to any license application (LA). There are, however, activities that DOE could complete prior to LA submission that would provide additional support for addressing this issue. Systematic air permeability measurements conducted in horizontal boreholes in the three repository host rock units could provide data on the scales of variability and heterogeneity in rock properties that are necessary to describe seepage of water into the drifts. In addition, additional model development efforts should focus on explaining the observed patterns of seepage in the niche experiments.

Coupled THC processes that affect seepage and flow were not considered explicitly in the TSPA-VA (U.S. Department of Energy, 1998b). The acceptance criteria are based on the concept that features, events, or processes necessary to describe repository performance are abstracted into a performance assessment. Because DOE did not abstract coupled THC processes, that may affect seepage and flow, into the TSPA-VA, the applicability of the acceptance criteria is severely limited. Nonetheless, the acceptance criteria are applied to the topic to indicate to DOE what needs to be completed for a high-quality license application.

Although DOE did not abstract the effects of coupled THC processes on seepage and flow into TSPA-VA, they appear to be planning to address this topic in their future TSPAs. Presentations at the Thermal-Hydrology and Coupled Processes (THCP) Workshop and the In-Drift Geochemical Environment (IDGE) and Engineered Barrier System Transport Workshops indicate that DOE is attempting to address all the acceptance criteria for this subissue, with some exceptions noted below. These workshops served as the beginning of the model abstraction process for the DOE's TSPAs to be used for DOE's Site Recommendation and a potential License Application. We will continue to attend DOE-sponsored meetings to ensure that deficiencies identified below are being addressed as part of the DOE program to develop a license application.

In general, DOE's approach to assess the effects of coupled THC processes on seepage and flow must meet the following generic acceptance criteria: (i) data and model justification; (ii) data uncertainty and verification; (iii) model uncertainty; (iv) model verification; (v) integration; (vi) quality assurance; and (vii) expert elicitation (U.S. Nuclear Regulatory Commission, 1998c).

5.4.1.1 Data and Model Justification for Subissue 1

The following five acceptance criteria address data and model justification. DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow were considered.

There are sufficient data from the DOE thermal testing program; observations from natural analogs (*e.g.*, Matyskiela, 1997); and interpretations from modeling (*e.g.*, Hardin, 1998) to suggest that the effects of the coupled THC processes are likely important to repository performance. "The assumptions made in the TSPA-VA that the effects of THC activities will be short-lived and can be omitted are not warranted; the analysis by Hardin (1998) indicates these processes will persist over the life of the repository" (Whipple, et al., 1999, page 53). Coupled THC processes will affect the permeability field as a function of time and will alter the chemical composition of water contacting the WP and waste forms. Effects of coupled THC processes on the permeability and capillary structure of the fracture network will alter seepage patterns as a function of time (Whipple, et al., 1999). DOE should use available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow in their abstractions.

Criterion 2 DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect seepage and flow.

Potential episodic seepage events during the thermal period cannot be excluded (see Large Block Test results in Hardin, 1998). Using homogeneous hydrologic properties in modeling coupled thermal-hydrologic-chemical processes does not adequately account for spatial and temporal variability in fracture flow. This conclusion is apparent from results of the drift scale heater test at Yucca Mountain. Thus, DOE should use results from their thermal testing program to establish initial and boundary conditions for their conceptual models in their abstractions.

Criterion 3 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 for conceptual models and simulations of THC coupled processes that affect seepage and flow.

The VA reference design of the repository includes use of a concrete liner in emplacement drifts. The liner will promote shedding of ambient percolation flux and reflux as long as the liner remains substantially intact, delaying or reducing contact of water with waste packages during early time periods. DOE analyses assume that the concrete liner will collapse within the first few hundred years, thus the liner is not considered for seepage calculations described in the TSPA-VA. To the extent that it is conservative to assume that no concrete liner remains intact to prevent early contact of water with the WP, we agree that the concrete liner need not be accounted for in seepage calculations. However, DOE did not consider the potential adverse consequences to repository performance from their assumption that the concrete liner collapses early in the post-closure period. In particular, the performance of the waste package may be substantially degraded by high-pH waters that traversed the concrete liner (Section 5.2.2). In addition, DOE did not collect sufficient data on the quantity and reactivity of concrete used in the drift that may affect flow in the unsaturated zone.

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. For instance, DOE should continue with studies of mineral precipitation and dissolution, focusing on the potential formation of durable permeability heterogeneities and the resulting effects on flow and seepage into and from the drifts. These properties should be used in their abstractions.

Proposed changes in repository design may not include concrete in the emplacement drifts. Thus, the potential interactions between concrete and tuff which affect seepage and flow may no longer be pertinent. However, regardless of the design, sufficient data are necessary to establish initial and boundary conditions for conceptual models. There is general agreement that there is a general lack of data to support assumptions in the models (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999).

Criterion 4 Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.

No abstraction of the effects of coupled THC on seepage and flow was completed by DOE. Thus, no sensitivity and uncertainty analyses were conducted using the abstracted model to determine whether additional new data are needed. DOE should use sensitivity and uncertainty analyses of their abstracted model to determine whether additional new data are needed to support the abstracted model.

Criterion 5 DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program, if the testing program for coupled THC processes on seepage and flow is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed.

The performance confirmation program was not a focus of our VA review. In addition, sensitivity and uncertainty studies were not conducted by DOE. The testing program for coupled THC processes on seepage and flow is not expected to be complete at the time of license application. Thus, DOE should identify specific plans to acquire the necessary information as part of the performance confirmation program.

5.4.1.2 Data Uncertainty and Verification for Subissue 1

The following five acceptance criteria address data uncertainty and verification. DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on seepage and flow. Parameter values, assumed ranges, probability distributions, and bounding assumptions were technically defensible and reasonably accounted for uncertainties.

Simulations of the process of silica redistribution near the emplacement drifts are described in the Near-Field/Altered-Zone Models Report (Hardin, 1998, Section 5.6). A summary also appears in Chapter 3 of the TSPA-VA Technical Basis Document (CRWMS M&O, 1998d). The modeling studies used a dual permeability formulation for coupled reactive transport and multiphase flow. Silica dissolution and precipitation of quartz, cristobalite, and amorphous silica were considered. Dissolution and precipitation of all other mineral phases were ignored. Detailed dependencies of the reaction rate on solution composition (e.g., pH and ionic strength) were ignored also. Precipitation and dissolution altered the porosity in both the fracture and matrix flow systems, which in turn modified the permeability. The modeling study predicted precipitation of silica minerals near the emplacement drifts, with dissolution and silica removal at greater distances. Although models for silica redistribution considered the effect of permeability reduction on the evolution of the flow system under nonisothermal conditions, the effect of the durable changes on drift seepage after the thermal pulse was not considered.

The main effect of the mineral redistribution process is to decrease the fracture permeability near the emplacement drifts. In some scenarios considered, the magnitude of this reduction is as large as several factors of ten. Formation of the mineral cap appears to be a robust process due to boiling mineral-laden water at the bottom of the heat pipe, a process independent of details of the chemistry. Effects of mineral cap precipitation depend on its location. For example, reduction of permeability in the fracture continuum in the vicinity of the drift wall could result in dripping at lower infiltration and percolation rates. Provided DOE does not take credit for an associated reduction in seepage, then the use of simplified chemistry in the mineral cap formation modeling is justified.

DOE should use reasonable or conservative ranges of parameters to determine effects of coupled THC processes on seepage and flow in their abstracted models. Continuation with studies of mineral precipitation in fractures based on more realistic chemistry should be considered. DOE should use technically defensible parameter values, assumed ranges, and probability distributions. If DOE uses bounding assumptions, they should be technically defensible and reasonably account for uncertainties.

Criterion 2 Uncertainties in data due to both temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow were considered.

Primary mineral redistribution below the drifts and in the pillar regions is sensitive to modeling details (Hardin, 1998, Section 5.6). For these processes, uncertainties in water-rock interactions and solution composition dependencies in the chemical rate parameters may be more important. Of particular concern is the reactive surface area, which is highly uncertain and subject to spatial and temporal variability. DOE should consider contributions to data uncertainty from both temporal and spatial variability. These contributions to uncertainty should be implemented in their abstracted models of the effects of coupled THC processes on seepage and flow. Presentations at THCP and IDGE workshops indicate that spatial and temporal variations in results from the drift scale heater test are now being considered in the process-level modeling studies. These process-level modeling results would form the basis for an abstracted model in a performance assessment. Thus, it appears that DOE is attempting to address this acceptance criterion in its abstraction process.

Criterion 3 DOE's evaluation of coupled THC processes properly considered 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 seepage and flow.

For coupled THC processes that are sensitive to modeling details, such as primary mineral redistribution below the drifts and in the pillar regions (Hardin, 1998, Section 5.6), spatial and temporal variation may introduce significant uncertainties.

DOE did not attempt to abstract the effects of concrete-tuff interactions on seepage and flow. Thus, they also did not consider uncertainties in the characteristics of the engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for their conceptual models. Proposed changes in repository design could also remove concrete from the emplacement drifts. Thus, the potential interactions between concrete and tuff which affect seepage and flow may no longer be pertinent. Regardless of the repository design, DOE should consider uncertainties in the characteristics of the natural and engineered materials in their abstraction of the effects of coupled THC processes on seepage and flow.

Criterion 4 The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on seepage and flow were consistent with available data.

No abstraction of the effects of coupled THC on seepage and flow was completed by DOE. Thus, no sensitivity analyses were conducted. DOE should use initial conditions, boundary conditions, and computational domains in their sensitivity analyses involving coupled THC effects on seepage and flow that are consistent with available data.

Criterion 5 DOE's performance confirmation program will assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on seepage and flow.

The performance confirmation program was not a focus of our VA review. DOE should address this objective in their performance confirmation plan.

5.4.1.3 Model Uncertainty for Subissue 1

The following four acceptance criteria address model uncertainty. DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Appropriate models, tests, and analyses were used that are sensitive to the THC couplings under consideration for both natural and engineering systems.

The natural-setting data indicate processes that should be evaluated include: (i) zeolitization of volcanic glass, which could affect flow pathways; (ii) precipitation of calcite and opal on the footwall of fracture surfaces and the bottoms of lithophysal cavities, which indicates gravity-driven flow in open fractures that could affect permeability and porosity; and (iii) potential dehydration of zeolites and vitrophyre glass, which could release water affecting heat and fluid flow. The effects of THC coupled processes that may occur due to interactions with engineered materials or their alteration products include: (i) changes in water chemistry that may result from interactions between cementitious materials and groundwater, which, in turn, may affect seepage and flow; (ii) dissolution of the geologic barrier (e.g., tuff) by a hyperalkaline fluid that could lead to changes in the hydraulic properties of the geologic barrier; and (iii) precipitation of calcite or calcium-silica-hydrate phases along fracture surfaces as a result of migration of a hyperalkaline fluid that could affect hydraulic properties. These processes were not considered in the TSPA-VA.

For some processes considered in the Near-Field/Altered Zone Models Report (Hardin, 1998), the supporting analyses were preliminary and did not adequately address significant uncertainties with process descriptions and modeling methodology [e.g., the analysis of prima mineral redistribution and secondary mineral formation in fractures (Hardin, 1998; Sections 5. and 5.7)].

The main concern with the effects of primary mineral redistribution on flow is persistent THC-induced permeability heterogeneities. Durable THC-induced heterogeneities in permeability below the repository could focus flow in these areas, thereby reducing radionuclide travel time and retention processes such as sorption. These changes are sensitive to details in the modeling studies and are thus subject to greater uncertainty than the mineral cap formation (Hardin, 1998). Permeability heterogeneities in the mineral cap could also focus flow, even if the overall effect of the mineral cap is to reduce permeability. Flow could be diverted toward the repository edges where the mineral cap is expected to be smaller, for example. DOE should use appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems.

Criterion 2 The potential effects of microbial processes on seepage and flow were addressed.

A model for bounding the magnitude of development of microbial communities was developed. The model was not used in the base case, but provided first-order limits on potential microbial effects (CRWMS M&O, 1998d). The DOE approach is acceptable and we have no current concerns regarding the potential effects of microbial processes on seepage and flow.

Criterion 3 Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

Reactive transport modeling of secondary mineral formation in fractures has also been undertaken (Hardin, 1998, Section 5.7). Secondary mineral formation in fractures is a process that could potentially alter fracture permeability and flow between fractures and matrix. This study makes significant progress at representing more detailed chemistry in the rock alteration models. However, important processes associated with the precipitation and dissolution of calcite and other minerals in fractures are omitted. For example, the GIMRT code used in this study is unable to treat phenomena associated with boiling, which is expected to be a dominant process controlling mineral precipitation. These issues should be examined using multiphase coupled models, which can handle the effects of boiling.

Criterion 4 DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on seepage and flow. The description included a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model.

DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on seepage and flow. DOE did discuss some of the limitations and uncertainties of their process-level model. However, their description did not include a discussion of alternative modeling approaches not considered in its final analysis. For example, the GIMRT code used in this study is unable to treat phenomena associated with boiling, which is expected to be a dominant process controlling mineral precipitation. These issues should be examined using multiphase coupled models, which can handle the effects of boiling. Presentations at the IDGE workshop on the TOUGH-REACT code modeling efforts indicate that DOE is attempting to address this acceptance criterion.

5.4.1.4 Model Verification for Subissue 1

The following three acceptance criteria address model verification. DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 The mathematical models for coupled THC effects on seepage and flow 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.

The effects of coupled THC processes were not abstracted in the TSPA-VA. Thus, it is not possible to compare abstracted model results to detailed process models or empirical observations. The general lack of data to support and test critical assumptions in mathematical models has been noted by others (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, 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. We recommend 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.

Criterion 2 DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on seepage and flow.

The effects of coupled THC processes were not abstracted in the TSPA-VA. Thus, it is not possible to evaluate whether DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on seepage and flow. DOE used well-documented procedures to construct and test process level numerical models such as TOUGH. This code only supports thermal-hydrologic calculations and may be used to support future abstractions. DOE should continue to apply accepted testing procedures in the development of TOUGH-REACT and other numerical codes that will be used to generate models providing the basis for abstraction of the effects of coupled THC processes on seepage and flow.

Criterion 3 Abstracted models for coupled THC effects on seepage and flow were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results were compared with different mathematical models to judge robustness of results.

The effects of coupled THC processes were not abstracted in the TSPA-VA. Thus, it is not possible to compare abstracted model results to detailed process models or empirical observations. DOE should use the results from both their laboratory and field heater test program to test their abstracted models for consistency with observations. We recommend 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.

5.4.1.5 Integration for Subissue 1

The following four acceptance criteria address integration. DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 DOE considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, physical phenomena, and couplings, and used consistent and appropriate assumptions throughout.

The general approach taken in the TSPA-VA is to decouple system behavior according to type of process. This assumption of weak feedback among processes is considered inadequate when applied to THC-induced changes in the repository flow system, which are characterized by strong coupling between thermal-hydrology effects, multicomponent chemistry, and rock-water interaction (CRWMS M&O, 1998d, Section 4.1.2.1). DOE staff recognized the complexity of these issues and summarized recent progress in the Near-Field/Altered-Zone Models Report (Hardin, 1998). The approach that DOE used for the VA did not include a formal screening process for features, events, and processes (FEP). As a result many important design features, physical phenomena, and couplings were not evaluated in a performance assessment framework. Presentations at the THCP Workshop and the IDGE Workshop indicate that DOE will complete a FEP analysis for the TSPA that could be used for a Site Recommendation and a potential License Application.

Criterion 2 Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow.

The general approach taken in the TSPA-VA is to decouple system behavior according to type of process. This assumption of weak feedback among processes is considered inadequate when applied to THC-induced changes in the repository flow system, which are characterized by strong coupling between thermal-hydrology effects, multicomponent chemistry, and rock-water interaction (CRWMS M&O, 1998d, Section 4.1.2.1). DOE did not include important design features, physical phenomena, and couplings that cause temporal and spatial variations in conditions. Thus the TSPA-VA models do not account for known temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow.

Criterion 3 If potentially important couplings were neglected, DOE provided a technical basis for doing so.

There must be a technical basis for omitting potentially important couplings that may significantly affect seepage and flow. The TSPA-VA omitted several important THC processes that could significantly affect seepage and flow. The TSPA-VA and supporting documents did not provide a technical basis for omitting the following processes: coupling the chemistries of the gas, aqueous, and solid phases with fluid flow through fractures and matrix; zeolitization of volcanic glass; dehydration of zeolitic or vitrophyre units; dissolution of tuff by a hyperalkaline fluid produced by water interaction with cementitious material; precipitation of calcite from cement-altered fluids onto fracture surfaces; and precipitation of calcite due to elevated temperatures in the saturated zone (SZ). DOE should provide a technical basis for omitting these processes or include these effects in the performance analyses via abstracted models or bounding assumptions.

Presentations at the THCP Workshop and the IDGE Workshop indicate that DOE will complete a FEP analysis for the TSPA that will be used for DOE's Site Recommendation and a potential License Application. DOE should include the processes listed above in their FEP analysis. The FEP analysis should meet the acceptance criteria in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 1998c) for scenario analysis. This FEP analysis will be the single most important action for DOE to take to facilitate resolution on this subissue.

Criterion 4 The bases used for modeling assumptions and approximations were documented and justified where simplifications for modeling coupled THC effects on seepage and flow were used for PA analyses instead of detailed process models.

DOE analyses of seepage described in the TSPA-VA did not consider the presence of a concrete liner in the emplacement drifts. The drift liner was neglected based on the assumption that the liner will collapse within the first few hundred years. However, DOE recognized that the collapsed concrete mass will still be available to react with water on the lower portion of the WPs for a long time. DOE did not consider the potential effect on flow of changes in unsaturated zone (UZ) porosity and permeability caused by alkaline plumes. These plumes, caused by interaction of water with cementitious materials, could enhance flow in the UZ by dissolving the host rock and increasing the porosity and permeability of the fractures, matrix, or both (e.g., Lichtner, et al., 1998). DOE should document the bases for simplifications used in modeling coupled THC effects on seepage and flow.

5.4.1.6 Quality Assurance and Expert Elicitation Concerns for Subissue 1

The following three acceptance criteria address quality assurance and expert elicitation. DOE's approach to evaluate and abstract coupled THC effects on seepage and flow in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Data and models were collected, developed, and documented under acceptable quality assurance (QA) procedures.

It was acknowledged that "analyses presented within were not performed with qualified data sets, all results within this report are considered unqualified" (CRWMS M&O, 1998d; Section 4.4.1). We have already noted our concerns with DOE's QA program (U.S. Nuclear Regulatory Commission, 1999b). These concerns are applicable to all subissues of this key technical issue (KTI). Although the VA was not designed to be a Quality Controlled document, we are concerned that limited time remains for DOE to qualify these data. We have formed an NRC QA Task Force to conduct an independent and objective review of the DOE HLW QA program and its implementation (U.S. Nuclear Regulatory Commission, 1999b). Presentations at the recent THCP Workshop and the IDGE Workshop indicate that DOE will focus on qualifying the data and models that could be used in the TSPA used for DOE's Site Recommendation and a potential License Application.

Criterion 2 Deficiency reports concerning data quality on issues related to coupled THC effects on seepage and flow were closed.

We have already noted our concerns with DOE's QA program (U.S. Nuclear Regulatory Commission, 1999b). As of May 18, 1999, three QA deficiency reports concerning issues related to coupled THC effects on seepage and flow remain open. Each of these reports has remained open for over one year. The three reports are LLNL-98-D-007, LVMO-98-D-064, and LLNL-98-D-065. Different aspects of the Drift Scale Heater Test (DST) program are covered by these deficiencies reports. The DST is one of the main bases for abstracting the effects of coupled THC processes on seepage and flow. These deficiency reports also cover topics within the other subissues of this KTI. Thus, DOE should vigorously pursue closing these deficiency reports.

Criterion 3 Expert elicitations, if used, were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

Concerns arising from the expert elicitations that DOE has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c) have already been documented (U.S. Nuclear Regulatory Commission, 1998h). These concerns were not directly associated with seepage and flow ubissue. However, one of the concerns is still relevant, and pertains to each subissue of this KTI. DOE needs to understand the expected use of the elicited expert judgements prior to convening the expert panel. For instance, the results from the near-field/altered-zone expert elicitation were not used in the near-field seepage abstraction. A positive aspect of the near-field/altered-zone expert elicitation was that experts were elicited for recommendations on what actions should be taken to reduce uncertainties (CRWMS M&O, 1998c). Based on presentations at the THCP Workshop and the IDGE Workshop, it is not clear that these recommendations are being considered.

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

The consequences of THC processes not only affect the potential chemical environment under which corrosion could occur, but they also determine the extent and type of corrosion products that will form. These corrosion products could have substantial impacts on repository performance, especially on radionuclide release and transport of radionuclides. Processes that should be evaluated to determine their potential importance to repository performance include: (i) thermal-hydrology 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 (WP) environmental conditions; (iii) dehydration of hydrous phases liberating moisture that may affect the WP environment; (iv) effects of microbial process on the WP environment; and (v) changes in water chemistry that may result from interactions between cementitious materials and groundwater, which, in turn, may affect the WP chemical environment. The large self potentials measured in certain DOE field tests have no impact on WP performance and need not be considered in a repository PA.

Two abstractions are influenced by the evolution of the near-field environment within the scope of the WP chemical environment subissue (see Appendix A). The abstractions are the quantity and chemistry of water contacting the WPs and waste forms, and WP corrosion (temperature, humidity, and chemistry). The processes listed above need to be considered in the evaluation of each abstraction. We evaluated DOE's TSPA-VA abstractions of the quantity and chemistry of water contacting the WPs and waste forms, and the waste package corrosion (U.S. Nuclear Regulatory Commission, 1999b).

Data and models used in the TSPA-VA to calculate the quantity and chemistry of water dripping on WPs were determined to be inadequate to describe the process and extent of potential dripping under thermally-altered conditions (U.S. Nuclear Regulatory Commission, 1999b). This is an important issue because both DOE and NRC PA analyses indicate that the fraction of WPs contacted by water is the most important factor affecting dose for the groundwater pathway. In addition, we determined that the current DOE testing and modeling plans are not sufficient to resolve the issue prior to LA submission. There are, however, activities that DOE could complete prior to LA that would provide additional support for addressing this issue. Systematic air permeability measurements conducted in horizontal boreholes in the three repository host rock units could provide data on the scales of variability and heterogeneity in rock properties that are necessary to describe coupled THC processes affecting seepage of water into the drifts. Additional model development efforts should focus on explaining the observed patterns of seepage in the niche experiments.

In addition, we concluded that it is unclear whether DOE will be able to acquire sufficient data—applicable to conditions at the proposed repository—in time to demonstrate compliance with NRC requirements (U.S. Nuclear Regulatory Commission, 1999b). This conclusion was based on our analysis of DOE's waste package corrosion abstraction. Sensitivity analyses indicate that the lifetime of WPs has a significant effect on dose. Corrosion performance of the WPs is a critical factor that may be affected by detrimental interactions between different materials and is influenced by the chemistry of the water contacting the WP. In addition, we found that the bulk of the long-term data used in the TSPA-VA may not be applicable to the environmental conditions at YM, particularly with regard to water chemistry (U.S. Nuclear Regulatory Commission, 1999b). The data that DOE used were gathered from expert elicitations and literature reviews.

Coupled THC processes that affect seepage and flow were not considered explicitly in the TSPA-VA (U.S. Department of Energy, 1998b). The effects of coupled THC processes that affect seepage and flow are also important in characterization of the WP chemical environment. In addition, models for the waste package chemical environment were developed but not used in the abstraction of waste package corrosion. The acceptance criteria are based on the concept that features, events, or processes necessary to describe repository performance are abstracted into a performance assessment. Because DOE did not abstract coupled THC processes, that may affect seepage and flow and the waste package chemical environment, into the TSPA-VA, the applicability of the acceptance criteria is somewhat limited. Nonetheless, the acceptance criteria are applied to the topic to indicate to DOE what needs to be completed for a high-quality license application.

Although DOE did not abstract the effects of coupled THC processes on seepage and flow and the waste package chemical environment into TSPA-VA, they appear to be planning to address this topic in their future TSPAs. Presentations at the Thermal-Hydrology and Coupled Processes (THCP) Workshop and the In-Drift Geochemical Environment (IDGE) and Engineered Barrier System Transport Workshops indicate that DOE is attempting to address all the acceptance criteria for this subissue, with some exceptions noted below. We will continue to attend DOE-sponsored meetings to ensure that deficiencies identified below are being addressed as part of the DOE program to develop a potential license application.

In general, DOE's approach to assess the effects of coupled THC processes on the waste package chemical environment must meet the following generic acceptance criteria: (i) data and model justification; (ii) data uncertainty and verification; (iii) model uncertainty; (iv) model verification; (v) integration; (vi) quality assurance; and (vii) expert elicitation (U.S. Nuclear Regulatory Commission, 1998c).

5.4.2.1 Data and Model Justification for Subissue 2

The following seven acceptance criteria address data and model justification. DOE's approach to evaluate and abstract coupled THC effects on the waste package chemical environment in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment were considered.

There are sufficient data from the DOE thermal testing program; observations from natural analogs; and interpretations from modeling (*e.g.*, Hardin, 1998) to suggest that the effects of the coupled THC processes are likely important to repository performance. "The assumptions made in the TSPA-VA that the effects of THC activities will be short-lived and can be omitted are not warranted; the analysis by Hardin (1998) indicates these processes will persist over the life of the repository" (Whipple, et al., 1999, page 53). In addition, we found that the bulk of the long-term data used in the TSPA-VA may not be applicable to the environmental conditions at YM, particularly with regard to water chemistry (U.S. Nuclear Regulatory Commission, 1999b). The data that DOE used was gathered from expert elicitations and literature reviews.

DOE should use available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment in their abstractions. For instance, presentations at the IDGE workshop on laboratory work at Lawrence Livermore National Laboratory (LLNL) on the formation of evaporative deposits of salt on the waste package was described. If waste package materials chosen for the SR and LA are sensitive to the salt concentration, then this work should be considered in the abstraction of the waste package corrosion process.

Criterion 2 DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect the WP chemical environment.

DOE's Near Field Geochemical Environment model focused on the central part of the repository, which was rationalized on the basis that the magnitude of effects is likely to be largest there (CRWMS M&O, 1998d; Section 4.4.2.1). However, effects of certain coupled THC effects may be most important on the margins of the emplacement zone. Refluxing water may be shed to the margins of the emplacement zone and the ingress of ambient oxygen may be greatest there. These processes will be adverse to waste package performance. Thus, 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.

Potential episodic seepage events during the thermal period cannot be excluded (see Large Block Test results in Hardin, 1998). Thus, DOE should use results from their thermal testing program to establish initial and boundary conditions for their conceptual models in their abstractions. Presentations on modeling efforts to determine edge effects and the effect of heterogeneities on dripping at the recent THCP Workshop and the IDGE Workshop indicate that DOE is attempting to address this acceptance criterion.

Criterion 3 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 for conceptual models and simulations of THC coupled processes that affect the WP chemical environment.

The DOE base case analysis does not include the effect of seepage water interacting with the concrete drift lining. In the base case DOE assumed that the drift lining collapses early in the simulation (U.S. Department of Energy, 1998b, Section 3.4.2). However, the effect of concrete-modified water on the WP has been evaluated by DOE in sensitivity analyses using a submodel for aqueous corrosion of the outer barrier (U.S. Department of Energy, 1998b, Section 5.3.2.1). Results of the sensitivity analyses indicate that when seepage water has a pH greater than 10, the carbon-steel outer barrier undergoes high aspect ratio pitting corrosion. This causes failure of the outer barrier shortly after corrosion starts (U.S. Department of Energy, 1998b, Section 5.4.4). Early failure of the outer barrier leads to early, rapid failure of the WP. The rapid failure of the WPs is the result of a higher probability of localized corrosion of the inner barrier. The early failure of WPs also leads to early release of radionuclides. For the concrete-modified water case, the peak total release rate from the engineered barrier system is increased more than three orders of magnitude compared to the base case. DOE cited 1 yr experiments at LLNL indicating that carbon steel in concentrated J-13 water with pH 9.7 does not show high aspect ratio pitting corrosion (McCright, 1998). DOE used this observation to suggest that the concrete-modified water may not impact repository performance. We consider the LLNL results inadequate to disprove the potential for enhanced pitting at alkaline pH because many of the LLNL experiments were conducted at a lower pH and none at pH 10 or higher.

Should DOE continue to have a repository design with large amounts of cementitious materials, then we will expect the concrete-modified water case to be part of the base case in the DOE PA analyses for any potential LA. Proposed changes in repository design could also remove concrete from the emplacement drifts. Thus, the potential interactions between concrete and water which affect WP chemical environment may no longer be pertinent. However, regardless of the design, sufficient data are necessary to establish initial and boundary conditions for conceptual models. There is general agreement that there is a general lack of data to support assumptions in the models (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999). We will evaluate experimental data presented by the DOE to support its conclusion that the corrosion pitting model used in the TSPA-VA sensitivity analyses is unrealistically conservative. This analysis will be documented in the next revision of the Container Life and Source Term IRSR.

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. These properties should be used in their abstractions.

Criterion 4 A nutrient and energy inventory calculation (e.g., McKinley, West, and Grogan, 1985; Grogan and McKinley, 1990; Noy, et al., 1996) was used to determine the potential for microbial activity that could impact the WP chemical environment.

A model for bounding the magnitude of development of microbial communities was developed. The in-drift microbial communities model has been judged by others to be at a preliminary stage (Whipple, et al., 1999). Two aspects of the DOE effort to constrain the potential for microbial activity were not considered. First, the potential for the release of organic carbon resulting from tuff-water interaction under thermal conditions was not considered. For instance, organic carbon in excess to that observed of local groundwaters has been measured in the EJ-13 water used in waste form experiments (Wronkiewicz, et al., 1992). The EJ-13 water is the esult of tuff-groundwater interactions at 90 °C. The potential for organic carbon release resulting from thermal reactions could be determined from observations in the Drift Scale Heater Test (*i.e.*, organic carbon measurements from water collected in boreholes). Second, high inorganic carbon concentrations, relative to the organic concentrations, would not limit autotrophic metabolism (Davis, et al., 1998). While both of these processes were not considered in the nutrient and energy inventory calculation, the uncertainties in the modeled results are large (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 criteria was used by DOE. We have 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 WP chemical environment.

Criterion 5 If microbial activity was predicted to be sufficient to allow microbial influenced corrosion (MIC) of the WP, then the time-history of temperature, humidity, and dripping was used to constrain the probability for MIC (CRWMS M&O, 1997b).

The microbial activity model was not used in the base case, but provided first-order limits on potential microbial effects (CRWMS M&O, 1998d). Rather than using the approach provided in this acceptance criterion, the DOE assigned a multiplying factor of five to the corrosion rates in a sensitivity study (CRWMS M&O, 1998d, Chapter 5). The TSPA-VA Peer Review Panel has concluded that this approach is highly conservative (Whipple, et al., 1999). Our analysis of MIC will be documented in the next revision of the Container Life and Source Term IRSR.

Criterion 6 Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.

The effect of concrete-modified water on the WP has been evaluated by DOE in sensitivity analyses using a submodel for aqueous corrosion of the outer barrier (U.S. Department of Energy, 1998b, Section 5.3.2.1). The peak total release rate from the engineered barrier was increased more than three orders of magnitude compared to the base case. Rather than concluding additional data are needed to better define ranges of input parameters, DOE cited 1 yr experiments at LLNL indicating that carbon steel in concentrated J-13 water with pH 9.7 does not show high aspect ratio pitting corrosion (McCright, 1998). DOE used this observation to suggest that the concrete-modified water may not impact repository performance. We consider the LLNL results inadequate to disprove the potential for enhanced pitting at alkaline pH because many of the LLNL experiments were conducted at a lower pH and none at pH 10 or higher. DOE should use sensitivity and uncertainty analyses of their abstracted model to determine whether additional new data are needed to support the abstracted model.

Criterion 7 DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program if the testing program for coupled THC processes on WP chemical environment is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed.

The performance confirmation program was not a focus of our VA review. Sensitivity and uncertainty studies were conducted by DOE. These analyses indicated that for a repository system that contained large quantities of cementitious materials additional data are needed. These data would address the potential for enhanced pitting at alkaline pH. Additional data were needed because many of the LLNL experiments were conducted at a lower pH and none at pH 10 or higher. Thus, DOE should have identified specific plans to acquire the necessary information. For future TSPAs, DOE should identify specific plans to acquire the necessary information as part of the performance confirmation program, if sensitivity studies indicate additional data are needed.

5.4.2.2 Data Uncertainty and Verification for Subissue 2

The following five acceptance criteria address data uncertainty and verification. DOE's approach to evaluate and abstract coupled THC effects on the waste package chemical environment in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on the WP chemical environment. Parameter values, assumed ranges, probability distributions, and bounding assumptions were technically defensible and reasonably accounted for uncertainties.

DOE has not used reasonable or conservative range of parameters to determine the effects of coupled thermal-hydrologic-chemical processes on the WP chemical environment. Some unrealistic model results indicate 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. High sanidine, a high-temperature volcanic phase, and the Mg-rich phase celadonite are all unrealistic members of the secondary mineral assemblage modeled for evaporative precipitation (CRWMS M&O, 1998d; figs. 4-71, 72, 73). 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, 5-49). In addition, only local equilibrium models were developed for in-drift water chemistry evaluations (CRWMS M&O, 1998d; Section 4.5.3). This approach did not consider reaction kinetics, except in the case of spent fuel dissolution and kinetic suppression of precipitation of certain minerals. We recognize the challenges associated with compilation of an internally consistent and validated thermodynamic and kinetic database for relevant mineral phases.

DOE should use reasonable or conservative ranges of parameters to determine effects of coupled THC processes on WP chemical environment in their abstracted models. DOE should continue with studies of evaporative chemistry at LLNL if the materials chosen for the waste package are sensitive to the salt content of the water contacting them. We also recommend exercises using experimental, site, and natural analog data to aid verification of 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, they should be technically defensible and reasonably account for uncertainties.

Criterion 2 Uncertainties in data due to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment were considered.

DOE did not consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment. DOE should consider contributions to data uncertainty from both temporal and spatial variability. These contributions to uncertainty should be implemented in their abstracted models of the effects of coupled THC processes on waste package chemical environment. Presentations at the IDGE workshop indicate that spatial and temporal variations in results from the drift scale heater test are now being considered in the process-level modeling studies. These process-level modeling results would form the basis for an abstracted model in a performance assessment for the incoming water chemistry. However, it is not clear that uncertainty in data due to both temporal and spatial variations in conditions affecting evaporative effects of water on the waste packages will be considered. Thus, it appears that DOE will only partially address this acceptance criterion in its abstraction process.

Criterion 3 DOE's evaluation of coupled THC processes properly considered 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 the WP chemical environment.

Some mineral phases, anticipated on the basis of alteration in the vicinity of YM (e.g., smectite, illite, and zeolites), are absent from the models. These limitations are generally recognized by DOE. Regardless of the repository design, 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 WP chemical environment.

Criterion 4 The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on WP chemical environment were consistent with available data.

The effect of concrete-modified water on the WP has been evaluated by DOE in sensitivity analyses using a sub-model for aqueous corrosion of the outer barrier (U.S. Department of Energy, 1998b, Section 5.3.2.1). DOE cited 1 yr experiments at LLNL that indicated carbon steel in concentrated J-13 water with pH 9.7 does not show high aspect ratio pitting corrosion (McCright, 1998). DOE used this observation to suggest that the concrete-modified water may not impact repository performance. We consider the LLNL results inadequate to disprove the potential for enhanced pitting at alkaline pH because many of the LLNL experiments were conducted at a lower pH and none at pH 10 or higher. These higher pH fluids are expected for waters that interact with cementitious materials (Atkins, et al., 1990). DOE should use initial conditions, boundary conditions, and computational domains in their sensitivity analyses involving coupled THC effects on WP chemical environment that are consistent with available data.

Criterion 5 DOE's performance confirmation program will assess whether the natural syste and engineered materials are functioning as intended and anticipated with rega to coupled THC effects on WP chemical environment.

The performance confirmation program was not a focus of our VA review. DOE should address this objective in their performance confirmation plan.

5.4.2.3 Model Uncertainty for Subissue 2

The following three acceptance criteria address model uncertainty. DOE's approach to evaluate and abstract coupled THC effects on the waste package chemical environment in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Appropriate models, tests, and analyses were used that are sensitive to the THC couplings under consideration for both natural and engineering systems.

The major limitation recognized by the 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. For example, salt formation was considered in the DOE near field geochemical environment (NFGE) model, but not used in the TSPA-VA analyses. "Although the amount of salt precipitation was assessed in the NFGE component (see Sections 4.4 and 4.6), it was not directly used in the TSPA-VA analyses" (CRWMS M&O, 1998d; Section 4.2.3.2.2.4). In addition, "the local exposure environment on the corrosion resistant material and its evolution with time and the bulk condition are poorly defined, and the current (stochastic) [WP degradation model] is not yet capable of modeling th specific chemistry of the corrosion resistant material localized corrosion" (CRWMS M&O, 1998d; Section 5.9.1). DOE should use appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems.

Criterion 2 Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

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, 5-48). We concur with this observation. Present models are recognized to be inadequate. Presentations at the IDGE Workshop on the TOUGH-REACT code modeling efforts indicate that DOE is attempting to address this acceptance criterion.

Criterion 3 DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on WP chemical environment. The description included a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model.

DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on WP chemical environment. DOE did discuss some of the limitations and uncertainties of their process-level model. A reasonable suite of gas and aqueous phase components was considered in these models. Generally these models and analyses are considered to be preliminary and inadequate. Modeled chemical conditions are generally omitted in evaluations of container corrosion processes. Instead, chemical conditions provided to an expert elicitation panel were used for abstraction of the WP corrosion. See the Container Life and Source Term Issue Resolution Status Report for a detailed discussion of corrosion processes (U.S. Nuclear Regulatory Commission, 1998d). DOE did not clearly distinguish between the different approaches they used and did not describe how the modeling approach they did not use in their final analysis could have affected the results.

5.4.2.4 Model Verification for Subissue 2

The following three acceptance criteria address model verification. DOE's approach to evaluate and abstract coupled THC effects on the waste package chemical environment in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 The mathematical models for WP chemical environment 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.

To the extent that models are generally regarded as preliminary, model verification for the effects of coupled THC processes on the WP chemical environment is premature. The abstraction of the effects of coupled THC processes was not used in the TSPA-VA. Thus, it is not possible to compare abstracted model results to detailed process models or empirical observations. The general lack of data to support and test critical assumptions in mathematical models has been noted by others (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, 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. We recommend 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.

Criterion 2 DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate the WP chemical environment.

To the extent that models are generally regarded as preliminary and were not used in the abstraction of waste package corrosion, model verification for the effects of coupled THC processes on the WP chemical environment is premature. The efforts to abstract the effects of coupled THC processes on the waste package chemical environment were not used in the TSPA-VA. Thus, it is not possible to evaluate whether DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on waste package chemical environment. As part of DOE's effort to verify their abstracted model of the effects of coupled THC effects on waste package chemical environment, DOE should use well-documented procedures to construct and test the numerical models.

Criterion 3 Abstracted models for coupled THC effects on WP chemical environment were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results were compared with different mathematical models to judge robustness of results.

To the extent that models are generally regarded as preliminary, model verification for the effects of coupled THC processes on the WP chemical environment is premature. The abstraction of the effects of coupled THC processes on the waste package chemical environment was not used in the TSPA-VA. DOE should use the results from both their laboratory and field heater test program to test their abstracted models for consistency with observations. We recommend 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.

5.4.2.5 Integration for Subissue 2

The following four acceptance criteria address integration. DOE's approach to evaluate and abstract coupled THC effects on the waste package chemical environment in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 DOE considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, physical phenomena, and couplings, and used consistent and appropriate assumptions throughout.

The major limitation recognized by the NRC and DOE staffs is that although conceptually the complexity of coupled THC processes is recognized, many aspects of these processes are omitted in the TSPA-VA analyses. For example, salt formation was considered in the DOE near field geochemical environment (NFGE) model, but not used in the TSPA-VA analyses. "Although the amount of salt precipitation was assessed in the NFGE component (see Sections 4.4 and 4.6), it was not directly used in the TSPA-VA analyses" (CRWMS M&O, 1998d; Section 4.2.3.2.2.4).

Models for the WP chemical environment were developed through analyses of successive changes to water entering and passing through the drift to define specific characteristics of pH, total dissolved solids, total carbonate concentration, and ionic strength (CRWMS M&O, 1998d, Section 4.5.1.2). These models appear to have little or no relation to water chemistries adopted 'y the DOE expert elicitation team charged with estimating corrosion rates as described in Section 5.8.2.1 of CRWMS M&O, 1998a. This is a major inconsistency and points to a lack of integration. The approach that DOE used for the VA 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.

Criterion 2 Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment.

DOE did not include some important design features, physical phenomena, and couplings that cause temporal and spatial variations in conditions. For instance, spatial variation in the thermal reflux is expected and DOE did not evaluate the potential variation. Thus the TSPA-VA models do not account for known temporal and spatial variations in conditions affecting coupled THC effects on the WP chemical environment.

Nevertheless, we consider that DOE made considerable progress in addressing effects of coupled THC processes on the chemical environment of the WPs in the TSPA-VA compared to prior TSPA activities. Consistently, in technical documents such as the TSPA-VA Plan (TRW Environmental Safety Systems, Inc., 1996c) and the Near-Field/Altered-Zone Models Report (Hardin, 1998), DOE has acknowledged the potential significance of coupled THC processes on repository performance. For the TSPA-VA, stepped temporal changes in the chemical composition of water entering and traversing the emplacement drift were modeled. This is a significant advance.

Criterion 3 If potentially important couplings were neglected, DOE provided a technical basis for doing so.

There must be a technical basis for omitting potentially important couplings that may significantly affect WP chemical environment. While DOE did consider many pertinent important couplings in their near field geochemical environment model, these abstractions were not used to evaluate the WP corrosion process. In addition some processes, such as evaporative concentration of solutes on the WP and the impact of high pH from cement-modified waters interacting with the WP, were not incorporated into the TSPA-VA. Instead, the TSPA-VA relied on an expert elicitation to determine parameters used in the WP corrosion abstraction. DOE did not provide an adequate technical basis to determine whether the parameters used in the expert elicitation would be bounded by the results from the near field geochemical environment model. DOE should provide a technical basis for omitting these processes or include these effects in the performance analyses via abstracted models or bounding assumptions.

The FEP analysis should include the processes described above and those listed in Section 5.4.2. The FEP analysis should meet the acceptance criteria in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 1998c) for scenario analysis. This FEP analysis will be an important action for DOE to take to facilitate resolution on this subissue. Additionally, integration between the major abstraction efforts needs to improve.

Criterion 4 The bases used for modeling assumptions and approximations were documented and justified where simplifications for modeling coupled THC effe on WP chemical environment were used for performance assessment analyses instead of detailed process models.

The simplified chemical environment for WP corrosion was given to the WP corrosion expert elicitation panel and did correspond to detailed process-level models. The bases used and the simplifications of the chemical environment used for the expert elicitation were not justified. Nevertheless, we acknowledge that DOE made considerable progress in addressing effects of coupled THC processes on the chemical environment of the waste packages in the TSPA-VA compared to prior TSPA activities. Consistently, in technical documents such as the TSPA-VA Plan (TRW Environmental Safety Systems, Inc., 1996c) and the Near- Field/Altered-Zone Models Report (Hardin, 1998), DOE has acknowledged the potential significance of coupled THC processes on repository performance. The stepped temporal changes in the chemical composition of water entering and traversing the emplacement drift used in TSPA-VA is a significant advance. DOE should continue to document the bases for simplifications used in modeling coupled THC effects on the waste package chemical environment.

5.4.2.6 Quality Assurance and Expert Elicitation Concerns for Subissue 2

The following three acceptance criteria address quality assurance and expert elicitation. DOE's approach to evaluate and abstract coupled THC effects on the waste package chemical environment in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Data and models were collected, developed, and documented under acceptabl quality assurance (QA) procedures.

The assessment of DOE's QA program in relation to this criterion is presented in Section 5.4.16 and is not repeated here.

Criterion 2 Deficiency reports concerning data quality on issues related to coupled THC effects on waste package chemical environment were closed.

As of May 18, 1999, three QA deficiency reports concerning issues related to coupled THC effects on waste package chemical environment remain open. Two of three reports have remained open for greater than 6 months. The three reports are LVMO-99-D-044, K/PB-98-D-105, and LVMO -98-D-139. Different aspects of the Tracers, Fluids and Material (TFM) Control program are covered by these deficiency reports. The TFM program is one of the main bases for abstracting the effects of microbial processes on the waste package chemical environment and the potential for microbial influenced corrosion. Thus, DOE should pursue closing these deficiency reports.

Criterion 3 Expert elicitations, if used, were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

Concerns arising from the expert elicitations that DOE has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c) have already been documented (U.S. Nuclear Regulatory Commission, 1998h). These concerns are directly associated with the WP chemical environment. Two concerns arose from the expert elicitations that 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. The results from the near-field/altered-zone expert elicitation were unable to be used to develop the initial and boundary conditions for the WP corrosion panel. This situation arose because the WP elicitation was completed prior to the initiation of the near field elicitation. A positive aspect of the near-field/altered-zone expert elicitation was that experts were elicited for recommendations on what actions should be taken to reduce uncertainties (CRWMS M&O, 1998c). Based on presentations at the THCP Workshop and the IDGE Workshop, it is not clear that these recommendations are being considered. We will attend DOE-sponsored expert elicitations to ensure that the deficiencies identified above are addressed as part of the DOE program to develop a license application.

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

Processes that should be evaluated to determine their potential importance to repository performance include: (i) TH effects on gas and water chemistry; (ii) hydrothermally driven geochemical reactions, such as zeolitization of volcanic glass; (iii) dehydration of hydrous phases liberating moisture; (iv) effects of microbial processes; and (v) changes in water chemistry that may result from interactions between cementitious, or WP, materials and groundwater, which, in turn, may affect the chemical environment for radionuclide release.

Three abstractions are influenced by the ENFE within the scope of the chemical environment for radionuclide release subissue (see Appendix A). The abstractions are waste package (WP) corrosion (temperature, humidity, and chemistry), the quantity and chemistry of fluids contacting waste forms, and radionuclide release rates and solubility limits. Radionuclide release is addressed in more detail in the CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d). Abstraction of radionuclide release and solubility limits requires input from the other two abstractions in this subissue (U.S. Nuclear Regulatory Commission, 1998c). The processes listed above need to be considered in the evaluation of each abstraction. We evaluated DOE's TSPA-VA abstractions of the quantity and chemistry of water contacting the WPs and waste forms, waste package corrosion, and radionuclide release (U.S. Nuclear Regulatory Commission, 1999b).

The radionuclide release and solubility limit abstraction was reviewed. Based on the status of DOE efforts in this KESA, as reflected in the TSPA-VA and License Application Plan (U.S. Department of Energy, 1998b,d), the staff had no major concerns (U.S. Nuclear Regulatory Commission, 1999b). Nonetheless, a review of the TSPA-VA is presented below to document the status of subissue resolution and to outline the path forward to resolution.

Data and models presented in the TSPA-VA used to calculate the quantity and chemistry of water contacting the WPs and waste forms were determined to be inadequate to describe the process under thermally-altered conditions (U.S. Nuclear Regulatory Commission, 1999b). This is an important issue because both DOE and NRC PA sensitivity analyses (U.S. Department of Energy, 1998b; Murphy and Codell, 1999) indicate that alternate source term abstractions are important factors affecting dose for the groundwater pathway. These alternate abstractions attempt to take into account the potential effects of the chemistry of the water interacting with the waste form and lead, for certain sets of assumptions, to lower doses (Murphy and Codell, 1999). We determined that the current DOE testing and modeling plans are not sufficient to resolve the issue of the quantity and chemistry of water contacting the WPs and waste forms prior to any LA submission. There are, however, activities that DOE could complete prior to LA that would provide additional support for addressing this issue. Systematic air permeability measurements conducted in horizontal boreholes in the three repository host rock units could provide data on the scales of variability and heterogeneity in rock properties that are necessary to describe the effects of coupled THC processes on seepage of water into the drifts. In addition, additional model development efforts should focus on explaining the observed patterns of seepage in the niche experiments.

Based on our analysis of DOE's waste package corrosion abstraction, we concluded that it is unclear whether DOE will be able to acquire sufficient data, applicable to conditions at the proposed repository, in time to demonstrate compliance with NRC requirements (U.S. Nuclear Regulatory Commission, 1999b). Sensitivity analyses indicate that the lifetime of WPs has a significant effect on dose. Corrosion performance of the WPs and waste forms is a critical factor that may be affected by detrimental interactions between different materials and is influenced by the chemistry of the water contacting the WP. We found that the bulk of the long-term data used the abstraction of WP corrosion in the TSPA-VA may not be applicable to the environmental conditions at YM (U.S. Nuclear Regulatory Commission, 1999b). This conclusion is particularly relevant to the assumed water chemistry.

Coupled THC processes that affect seepage and flow were not considered explicitly in the TSPA-VA (U.S. Department of Energy, 1998b). The effects of coupled THC processes that affect seepage and flow are also important in characterization of the chemical environment for radionuclide release. Models for the chemical environment for radionuclide release were developed but not used in the abstraction of radionuclide release from the glass waste form. The acceptance criteria are based on the concept that features, events, or processes necessary to describe repository performance are abstracted into a performance assessment. Because DOE did not abstract coupled THC processes, that may affect seepage and flow and the chemical environment for radionuclide release, into the TSPA-VA, the applicability of the acceptance criteria is somewhat limited. Nonetheless, the acceptance criteria are applied to the topic to indicate to DOE what needs to be completed for a high-quality license application. Many of the uncertainties and concerns described in the discussion of the WP chemical environment (Section 5.4.2 of this IRSR) apply equally to the chemical environment for radionuclide release.

Although DOE did not abstract the effects of coupled THC processes on seepage and flow and the chemical environment for radionuclide release into TSPA-VA, they appear to be planning to address this topic in their future TSPAs. Presentations at the Thermal-Hydrology and Coupled Processes (THCP) Workshop and the In-Drift Geochemical Environment (IDGE) and Engineered Barrier System Transport Workshops indicate that DOE is attempting to address all the acceptance criteria for this subissue, with some exceptions noted below. We will continue attend DOE-sponsored meetings to ensure that deficiencies identified below are being addressed as part of the DOE program to develop a potential license application.

In general, DOE's approach to assess the effects of coupled THC processes on the chemical environment for radionuclide release must meet the following generic acceptance criteria: (i) data and model justification; (ii) data uncertainty and verification; (iii) model uncertainty; (iv) model verification; (v) integration; (vi) quality assurance; and (vii) expert elicitation (U.S. Nuclear Regulatory Commission, 1998c).

5.4.3.1 Data and Model Justification for Subissue 3

The following seven acceptance criteria address data and model justification. DOE's approach to evaluate and abstract coupled THC effects on the chemical environment for radionuclide release in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release were considered.

There are sufficient data from the DOE thermal testing program; observations from natural analogs; and interpretations from modeling (e.g., Hardin, 1998) to suggest that the effects of the coupled THC processes are likely important to repository performance. Coupled THC modeling in the TSPA-VA as applied to the chemical environment for radionuclide release represents a significant advance relative to previous TSPA evaluations. Nevertheless, many pproximations and assumptions were made, and their limitations are generally recognized. We ound that the bulk of the long-term data used in the TSPA-VA WP corrosion abstraction may not be applicable to the environmental conditions at YM, particularly with regard to water chemistry (U.S. Nuclear Regulatory Commission, 1999b). The WP corrosion process will affect the chemistry of water contacting waste forms. This will impact radionuclide release. The data that DOE used in the WP corrosion abstraction was gathered from expert elicitations and literature reviews. DOE should use available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment in their abstractions.

Criterion 2 DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect the chemical environment for radionuclide release.

DOE 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). However, effects of certain coupled THC processes may be most important on the margins of the emplacement zone. Refluxing water may be shed to the margins of the emplacement zone and the ingress of ambient oxygen may be greatest there. These processes will be adverse to waste form performance and could lead to earlier and larger radionuclide releases. Thus, neglecting processes at the edges of the emplacement zone may not be conservative. DOE should justif neglecting processes at the margin of the emplacement area.

Potential episodic seepage events during the thermal period cannot be excluded (see Large Block Test results in Hardin, 1998). Thus, DOE should use results from their thermal testing program to establish initial and boundary conditions for their conceptual models in their abstractions. Presentations at THCP and IDGE workshops on modeling efforts to determine edge effects and the effect of heterogeneities on dripping indicate that DOE is attempting to address this acceptance criterion.

Criterion 3 Sufficient data were collected on 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 the chemical environment for radionuclide release.

The DOE base case analysis does not include the effect of seepage water interacting with the concrete drift lining. In the base case DOE assumed that the drift lining collapses early in the simulation (U.S. Department of Energy, 1998b, Section 3.4.2). However, the effect of concrete-modified water on the WP and radionuclide release has been evaluated by DOE in sensitivity analyses using a sub-model for aqueous corrosion of the outer barrier (U.S. Department of Energy, 1998b, Section 5.3.2.1). Results of the sensitivity analyses indicate that when seepage water has a pH greater than 10, the carbon-steel outer barrier undergoes high aspect ratio pitting corrosion. This causes an early failure of WPs and leads to early release of radionuclides. For the concrete-modified water case, the peak total release rate from the engineered barrier system is increased more than three orders of magnitude compared to the base case. DOE cited 1 yr experiments at Lawrence Livermore National Laboratory (LLNL) indicating that carbon steel in concentrated J-13 water with pH 9.7 does not show high aspect ratio pitting corrosion (McCright, 1998). DOE used this observation to suggest that the concrete-modified water may not impact repository performance. Many of the LLNL experiments were conducted at a lower pH and none at pH 10 or higher. Additional insights from other studies suggest that concrete-modified fluids would have a pH greater than 10 (Atkins, et al., 1990). Thus, we consider the LLNL results inadequate to disprove the potential for enhanced pitting at alkaline pH and the enhanced early release of radionuclides.

We will expect the concrete-modified water case to be part of the base case in the DOE PA analyses for any LA should DOE continue to have a repository design with large amounts of cementitious materials. Regardless of the design, sufficient data are necessary to establish initial and boundary conditions for conceptual models. There is general agreement that there is a general lack of data to support assumptions in the models (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999).

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. These properties should be used in their abstractions. Given the rapid rate of spent fuel alteration predicted for the YM near-field environment, studies of environmental conditions affecting the stability of oxidized engineered and waste form materials, such as secondary uranyl phases, would provide an important step forward.

Criterion 4 A nutrient and energy inventory calculation (e.g., McKinley, West, and Grogan, 1985; Grogan and McKinley, 1990; Noy, et al., 1996) was used to determine the potential for microbial activity that could impact radionuclide release.

A model for bounding the magnitude of development of microbial communities was developed. 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 criteria was used by DOE. We have 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 chemical environment for radionuclide release.

Criterion 5 If microbial activity was predicted to be sufficient to potentially affect the chemical environment for radionuclide release, then the time-history of temperature, humidity, and dripping (CRWMS M&O, 1997b) was used 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) and microbially enhanced dissolution of the HLW glass form (Staudigel, et al., 1995).

The microbial activity model was not used in the base case, but provided first-order limits on potential microbial effects (CRWMS M&O, 1998d). DOE did not attempt to evaluate potential microbial effects on radionuclide release. DOE should use the time-history of temperature, humidity, and dripping (CRWMS M&O, 1997b) 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) and microbially enhanced dissolution of the HLW glass form (Staudigel, et al., 1995). From presentation at the IDGE workshop it is not clear that DOE will pursue any further work on microbial processes in the near-field geochemical environment. Neglecting the potential impact of microbial processes on radionuclide release will require justification (see Section 5.4.3.5, Criterion 3).

Criterion 6 Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine whether additional new data are needed to better define ranges of input parameters.

Both DOE and NRC PA sensitivity analyses (U.S. Department of Energy, 1998b; Murphy and Codell, 1999) indicate that alternate source term abstractions are important factors affecting dose for the groundwater pathway. These alternate abstractions attempt to take into account the potential effects of the chemistry of the water interacting with the waste form and lead, for certain sets of assumptions, to lower doses (Murphy and Codell, 1999). A sensitivity study was conducted of the evolution of water and mineral chemistry inside the emplacement drift using the numerical reactive transport model AREST-CT (CRWMS M&O, 1998d, Section 6.4.3.2).

Similarly to TSPA-VA model results, spent fuel dissolves rapidly (e.g., less than 500 years) in the AREST-CT model. The model indicates that schoepite is the dominant secondary phase and that uranophane (and to a lesser extent soddyite) is also produced limited by calcium (an silica) supply. This study represents an important, relatively mechanistic, advance over other treatments of the radionuclide source term in performance assessments, and one that is relatively well supported by laboratory, natural analog and thermodynamic considerations. Limitations as well as potential contributions of this modeling effort are well recognized in the documentation (CRWMS M&O, 1998d, Section 6.4.3.2). For instance, large uncertainties are recognized in the thermodynamic and kinetic data used for the sensitivity study. DOE should use sensitivity and uncertainty analyses of their abstracted model to determine whether additional new data are needed to support the abstracted model. Given the rapid rate of spent fuel alteration predicted for the YM near-field environment, studies of environmental conditions affecting the stability of oxidized engineered and waste form materials, such as secondary uranyl phases, would provide an important step forward.

Criterion 7 DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program if the testing program for coupled THC processes on the chemical environment for radionuclide release from the engineered barrier system is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed.

The performance confirmation program was not a focus of our VA review. Sensitivity and uncertainty studies were conducted by DOE. The waste form sensitivity studies demonstrated the effects of secondary uranium mineral formation on radionuclide release. Because the formation of secondary uranium minerals has the potential to substantially improve repository performance, it has been suggested (Whipple, et al., 1999) that substantial more study in this area is needed. Based on presentations at the IDGE workshop, it does not appear that DOE will change its spent fuel dissolution model approach. Thus, DOE did not have to identify specific plans to acquire the necessary information on the secondary uranium mineral release model. For future TSPAs, DOE should identify specific plans to acquire the necessary information as part of the performance confirmation program, if sensitivity studies indicate additional data are needed.

5.4.3.2 Subissue 3: Data Uncertainty and Verification Acceptance Criteria

The following five acceptance criteria address data uncertainty and verification. DOE's approach to evaluate and abstract coupled THC effects on the chemical environment for radionuclide release in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on the chemical environment for radionuclide release. Parameter values, assumed ranges, probability distributions, and bounding assumptions were technically defensible and reasonably accounted for uncertainties.

DOE has not used reasonable or conservative range of parameters to determine the effects of coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release. Some unrealistic model results indicate that a review of the parameters used in the process-level models is warranted. Andradite (a garnet mineral), diopside, and wollastonite are ypically 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, 5-49). In addition, only local equilibrium models were developed for in-drift water chemistry evaluations. This approach did not consider reaction kinetics, except in the case of spent fuel dissolution and kinetic suppression of precipitation of certain minerals. We recognize the challenges associated with compilation of an internally consistent and validated thermodynamic and kinetic database for relevant mineral phases.

DOE should use reasonable or conservative ranges of parameters to determine effects of coupled THC processes on WP chemical environment in their abstracted models. We recommend exercises using experimental, site, and natural analog data to aid verification of 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, probability distributions. If DOE uses bounding assumptions, they should be technically defensible and reasonably account for uncertainties.

Criterion 2 Uncertainties in data due to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release were considered.

DOE did not consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release. DOE should consider contributions to data uncertainty from both temporal and spatial variability. These contributions to uncertainty should be implemented in their abstracted models of the effects of coupled THC processes on the chemical environment for radionuclide release.

Criterion 3 DOE's evaluation of coupled THC processes properly considered 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 the chemical environment for radionuclide release.

Some mineral phases, anticipated on the basis of alteration in the vicinity of YM (e.g., smectite, illite, and zeolites), are absent from the models. These limitations are generally recognized by DOE. Regardless of the repository design, 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 chemical environment for radionuclide release.

Criterion 4 The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on the chemical environment for radionuclide release were consistent with available data.

The effect of concrete-modified water on the waste package and radionuclide release has been evaluated by DOE in sensitivity analyses using a sub-model for aqueous corrosion of the outer barrier (U.S. Department of Energy, 1998b, Section 5.3.2.1). DOE cited 1 yr experiments at

LLNL that indicated carbon steel in concentrated J-13 water with pH 9.7 does not show high aspect ratio pitting corrosion (McCright, 1998). DOE used this observation to suggest that the concrete-modified water may not impact repository performance. We consider the LLNL results inadequate to disprove the potential for enhanced pitting at alkaline pH and for earlier larger radionuclide releases. Many of the LLNL experiments were conducted at a lower pH and none at pH 10 or higher. These higher pH fluids are expected for waters that interact with cementitious materials (Atkins, et al., 1990).

A sensitivity study was conducted of the evolution of water and mineral chemistry inside the emplacement drift using the numerical reactive transport model AREST-CT (CRWMS M&O, 1998d, Section 6.4.3.2). Pore water velocities, temperature, incoming water and gas chemistry, and fraction (and surface area) of spent fuel exposed to water are inputs to the AREST-CT model from other near-field geochemical environment models. A kinetic rate law based on data from Gray et al. (1992) is implemented for spent fuel dissolution. This rate law differs from that used in the TSPA-VA analyses to be consistent with a transition state law kinetic expression. Four secondary uranyl minerals are considered in the model based on laboratory data, natural analog observations, and availability of thermodynamic and kinetic parameters. However, large uncertainties are recognized in the thermodynamic and kinetic data. This study represents an important, relatively mechanistic, advance over other treatments of the radionuclide source term in performance assessments, and one that is relatively well supported by laboratory, natural analog and thermodynamic considerations. Limitations as well as potential contributions of this modeling effort are well recognized in the documentation (CRWMS M&O, 1998d, Section 6.4.3.2). DOE should continue to use initial conditions, boundary conditions, and computational domains in their sensitivity analyses involving coupled THC effects on the chemical environment for radionuclide release that are consistent with available data.

Criterion 5 DOE's performance confirmation program will assess whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on the chemical environment for radionuclide release from the EBS.

The performance confirmation program was not a focus of our VA review. DOE should address this objective in their performance confirmation plan.

5.4.3.3 Model Uncertainty for Subissue 3

The following three acceptance criteria address model uncertainty. DOE's approach to evaluate and abstract coupled THC effects on the chemical environment for radionuclide release in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems were used.

The major limitation recognized by the 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 alteration of cladding and basket materials on the chemistry in the WPs controlling radionuclide releases were omitted

CRWMS M&O, 1998d, Section 4.2.3.2.2). These effects could be important because both basket and cladding materials may have different compositions than the waste packages. The additional materials could have a strong effect on corrosion products and associated water chemistry of the waste form environment. In addition, no process model exists for evolution of the gas phase chemistry within the drifts (CRWMS M&O, 1998d, Section 4.4.1). Neglect of these processes contributes to model uncertainty and should be justified or remedied.

Results of DOE modeling indicate that degradation of the high-level radioactive glass waste form occurs over a period of 8,000-10,000 yrs. and, thus, is not a key parameter in controlling repository performance during this period. However, this lack of importance of glass waste form degradation rate could change if the analysis evaluated the potential effect of alkaline pH (>10) on glass waste dissolution. For instance, experiments by Heimann (1988) indicated that cement and glass interaction leads to accelerated dissolution and alteration of nuclear waste glass compared to a system without cement. DOE near-field geochemical analyses for the composition of water reacting with concrete support components in the emplacement drifts indicate that water will have a pH near 11 for at least 10,000 yrs. Although DOE claims to have waste form dissolution models that directly incorporate rates that depend on pH (CRWMS M&O, 1998d, Section 6.3.3), the TSPA-VA analysis does not include a consideration of alkaline pH effects on waste glass dissolution. DOE should use appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems.

Criterion 2 Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

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-1 3 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, 5-48). We concur with this observation. Present models are recognized to be inadequate. Presentations at the IDGE Workshop on the TOUGH-REACT code modeling efforts indicate that DOE is attempting to address this acceptance criterion. This code will address the chemistry of water entering the emplacement drifts.

However, it is not clear that additional modeling work on the AREST-CT will be completed. This alternative approach is consistent with available evidence and current scientific understanding. While DOE did complete some analyses with this alternative approach, the results weren't fully considered.

Criterion 3 DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on the chemical environment for radionuclide release. The description included a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model.

DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on the chemical environment for radionuclide release. DOE did discuss some of the limitations and uncertainties of their process-level model. Generally these models and analyses are considered to be preliminary and inadequate. Modeled chemical conditions are generally omitted in evaluations of radionuclide release from the glass waste form. See the CLST IRSR for a detailed discussion of radionuclide release from the glass waste form (U.S. Nuclear Regulatory Commission, 1998d). DOE did not clearly distinguish between the different approaches that they used and did not describe how the modeling approach they did not use in its final analysis could have affected the results.

5.4.3.4 Model Verification for Subissue 3

The following three acceptance criteria address model verification. DOE's approach to evaluate and abstract coupled THC effects on the chemical environment for radionuclide release in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 The mathematical models for coupled THC effects on the chemical environment for radionuclide release 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.

Section 6.4.3.2.2.3 of the Technical Basis Document (CRWMS, M&O 1998d) draws comparisons between model results and laboratory and natural analog data in an effort at model verification. In general, good correspondence led to the conclusion that models provide a reasonably good representation. Nevertheless, qualitative similarities and some discrepancies are noted, and the validation is described as partial and preliminary. The general lack of data to support and test critical assumptions in mathematical models has been noted by others (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999). We recommend additional exercises using experimental, site, and natural analog data to aid verification of their models. DOE should use the results from both their ongoing waste form dissolution experiments 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.

Criterion 2 DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on the chemical environment for radionuclide release.

To the extent that models are generally regarded as preliminary, model verification for the effects of coupled THC processes on the chemical environment for radionuclide release is premature. As part of DOE's effort to verify their abstracted model of the effects of coupled THC effects on the chemical environment for radionuclide release, DOE should use well-documented procedures to construct and test the numerical models.

Criterion 3 Abstracted models for coupled THC effects on the chemical environment for radionuclide release were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results were compared with different mathematical models to judge robustness of results.

DOE should use the results from both their waste form laboratory experiments, field heater test program, and their natural analog program to test their abstracted models for consistency with observations. We recommend 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.

5.4.3.5 Integration for Subissue 3

The following four acceptance criteria address integration. DOE's approach to evaluate and abstract coupled THC effects on the chemical environment for radionuclide release in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 DOE considered the relevant features, events, and processes. The abstracted models adequately incorporated important design features; physical phenomena and couplings; and used consistent and appropriate assumptions throughout.

Many uncertainties associated with the chemical environment for radionuclide release are comparable to those identified in Section 5.4.2 concerning the WP chemical environment. In general, DOE recognizes the significance of the chemical environment to radionuclide release with regard to repository performance, and DOE recognizes the significance of coupled THC processes for prediction of the evolution of this environment. Efforts to model these phenomena represent an advance over prior TSPA efforts. Nevertheless, these models retain notable uncertainties and limitations such as neglect of concrete, cladding, basket, container, and waste form alteration products on water chemistry; and neglect of coupled gas and aqueous phase chemical evolution.

Interactions with corrosion products of the metallic WP are assumed represented by equilibrium with goethite. However, goethite is not identified as a corrosion product in the models for WP corrosion. Other phases could predominate, and the metals are not necessarily predominantly iron. Corrosion of metals such as nickel, molybdenum, and aluminum will likely result in minerals sufficiently different from goethite that the water chemistry contacting the waste form will be influenced strongly. In addition, the potential for local oxygen depletion of aqueous solutions and formation of reduced solutions due to reactions with metals or reduced waste form materials are not considered in the water chemistry models.

The approach that DOE used for the VA did not include a formal screening process for features, events, and processes (FEP). As a result many important design features, physical phenomena, and couplings were not evaluated in a performance assessment framework.

Criterion 2 Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release.

DOE did not include some important design features, physical phenomena, and couplings that cause temporal and spatial variations in conditions. For instance, spatial variation in the thermal reflux is expected and DOE did not evaluate the potential variation. Thus the TSPA-VA models do not account for known temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release.

Nevertheless, we conclude that DOE made considerable progress in addressing effects of coupled THC processes on the chemical environment for radionuclide release in the TSPA-VA compared to prior TSPA activities. Consistently, in technical documents such as the TSPA-VA Plan (TRW Environmental Safety Systems, Inc., 1996c) and the Near-Field/Altered-Zone Models Report (Hardin, 1998), DOE has acknowledged the potential significance of coupled THC processes on repository performance. For the TSPA-VA, stepped temporal changes in the chemical composition of water entering and traversing the emplacement drift were modeled. This is a significant advance.

Criterion 3 If potentially important couplings were neglected, DOE should provide a technical basis for doing so.

Interactions with corrosion products of the metallic WP are assumed represented by equilibrium with goethite. However, goethite is not identified as a corrosion product in the models for WP corrosion. Other phases could predominate, and the metals are not necessarily predominantly iron. Corrosion of metals such as nickel, molybdenum, and aluminum will likely result in minerals sufficiently different from goethite that the water chemistry contacting the waste form will be influenced strongly. In addition, the potential for local oxygen depletion of aqueous solutions and formation of reduced solutions due to reactions with metals or reduced waste form materials are not considered in the water chemistry models.

There must be a technical basis for omitting potentially important couplings that may significantly affect the chemical environment for radionuclide release. While DOE did consider many pertinent important couplings in their near field geochemical environment model, these abstractions were not used to evaluate the WP corrosion process. In addition some processes, such as evaporative concentration of solutes on the waste form, and the impact of high pH from cement-modified waters interacting with the glass waste form, were not incorporated into the TSPA-VA. DOE did not provide an adequate technical basis to determine whether the parameters used in the expert elicitation would be bounded by the results from the near field geochemical environment model. DOE should provide a technical basis for omitting these processes or include these effects in the performance analyses via abstracted models or bounding assumptions.

The FEP analysis should include the processes described above and those listed in Section 5.4.3. The FEP analysis should meet the acceptance criteria in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 1998c) for scenario analysis. This FEP analysis will an important action for DOE to take to facilitate resolution on this subissue.

Criterion 4 The bases used for modeling assumptions and approximations were documented and justified, where simplifications for modeling coupled THC effects on the chemical environment for radionuclide release were used for PA analyses instead of detailed process models.

DOE made considerable progress in addressing effects of coupled THC processes on the chemical environment for radionuclide release in the TSPA-VA compared to prior TSPA activities. Consistently, in technical documents such as the TSPA-VA Plan (TRW Environmental Safety Systems, Inc., 1996c) and the Near-Field/Altered-Zone Models Report (Hardin, 1998), DOE has acknowledged the potential significance of coupled THC processes on repository performance. The stepped temporal changes in the chemical composition of water entering and traversing the emplacement drift used in TSPA-VA is a significant advance. DOE should continue to document the bases for simplifications used in modeling coupled THC effects on the chemical environment for radionuclide release.

5.4.3.6 Quality Assurance and Expert Elicitation Concerns for Subissue 3

The following three acceptance criteria address quality assurance and expert elicitation. DOE's approach to evaluate and abstract coupled THC effects on the chemical environment for radionuclide release in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Data and models were collected, developed, and documented under acceptable quality assurance (QA) procedures.

The assessment of DOE's program in relation to this criterion is presented in Section 5.4.1.6 and is not repeated here.

Criterion 2 Deficiency reports concerning data quality on issues related to coupled THC effects on radionuclide release were closed.

As of May 18, 1999, three QA deficiency reports concerning issues related to coupled THC effects on the chemical environment for radionuclide release remain open. These are the same reports mentioned in Section 5.4.2.6. Different aspects of the Tracers, Fluids and Material (TFM) Control program are covered by these deficiency reports. The TFM program is one of the main bases for abstracting the effects of microbial processes on the chemical environment for radionuclide release. Thus, DOE should pursue closing these deficiency reports.

Criterion 3 Expert elicitations, if used, were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

Concerns arising from the expert elicitations that DOE has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c) have already been documented (U.S. Nuclear Regulatory Commission, 1998h). These concerns were not directly associated with the chemical environment for radionuclide release. However, the expected use of the elicited expert judgements needs to be understood prior to convening the expert panel. The results from the near-field/altered-zone expert elicitation were not used to develop the initial and boundary conditions for the radionuclide release abstraction. This situation arose because the waste form elicitation was completed prior to the initiation of the near field elicitation. A positive aspect of the near-field/altered-zone expert elicitation was that experts were elicited for recommendations on what actions should be taken to reduce uncertainties (CRWMS M&O, 1998c). Based on presentations at the THCP Workshop and the IDGE Workshop, it is not clear that these recommendations are being considered. We will attend DOE-sponsored expert elicitations to ensure that the deficiencies identified above are addressed as part of the DOE program to develop a license application.

5.4.4 Subissue 4: Effects of Coupled Thermal-Hydrologic-Chemical Processes on Radionuclide Transport

Processes that should be evaluated to determine their potential importance to repository performance include: (i) thermal-hydrologic effects on gas and water chemistry in the unsaturated zone (UZ) and saturated zone (SZ); (ii) precipitation of calcite and opal on the footwall of fracture surfaces and the bottoms of lithophysal cavities, which indicates gravity driven flow in open fractures, and isolation of transport pathways from sorption sites in the rock matrix; (iii) zeolitization of volcanic glass, that could affect transport pathways; (iv) precipitation and dissolution of oxides and hydroxides on fracture surfaces, illitization of smectite, and recrystallization of zeolites to analcime, which could affect sorption characteristics; (v) effects of microbial processes; (vi) effects of corrosion products of container materials and waste forms on transport of radionuclides in the near field; and (vii) changes in hydraulic and sorptive properties of the natural system resulting from interactions between cementitious materials and groundwater.

Three abstractions are influenced by the evolution of the near-field environment within the scope of the radionuclide transport subissue (see Appendix A). The abstractions are distribution of mass flux between fracture and matrix, retardation in fractures in the UZ, and retardation in water-production zones and alluvium. Radionuclide transport is addressed in more detail in the Radionuclide Transport IRSR (U.S. Nuclear Regulatory Commission, 1998e). The NRC's abstraction of radionuclide transport requires input from the three abstractions in this subissue (U.S. Nuclear Regulatory Commission, 1998c). The processes listed above need to be considered in the evaluation of each abstraction. We evaluated DOE's TSPA-VA abstractions of the distribution of mass flux between fracture and matrix, retardation in fractures in the UZ, and retardation in water-production zones and alluvium (U.S. Nuclear Regulatory Commission, 1999b).

Based on the status of DOE efforts in the distribution of mass flux between fracture and matrix, retardation in fractures in the UZ, and retardation in water-production zones and alluvium KESAs, as reflected in the TSPA-VA and License Application Plan (U.S. Department of Energy, 1998b,d), the staff had no major comments (U.S. Nuclear Regulatory Commission, 1999b). Nonetheless, review of the TSPA-VA is presented below to document the status of subissue resolution and to outline the path forward to resolution.

Coupled THC processes that affect seepage and flow were not considered explicitly in the TSPA-VA (U.S. Department of Energy, 1998b). The effects of coupled THC processes that affect seepage and flow are also important in characterization of radionuclide transport through engineered and natural barriers. The acceptance criteria are based on the concept that ıatures, events, or processes necessary to describe repository performance are abstracted into a performance assessment. Because DOE did not abstract coupled THC processes that may affect seepage and flow and the radionuclide transport into the TSPA-VA, the applicability of the acceptance criteria is limited. Nonetheless, the acceptance criteria are applied to the topic to indicate to DOE what needs to be completed for a high-quality license application.

Although DOE did not abstract the effects of coupled THC processes on radionuclide transport through engineered and natural barriers into TSPA-VA, they appear to be planning to address this topic in their future TSPAs. Presentations at the Thermal-Hydrology and Coupled Processes (THCP) Workshop and the In-Drift Geochemical Environment (IDGE) and Engineered Barrier System Transport Workshops indicate that DOE is attempting to address all the acceptance criteria for this subissue, with some exceptions noted below. These workshops served as the beginning of the model abstraction process for the DOE's TSPAs to be used for DOE's Site Recommendation and a potential License Application. We will continue to attend DOE-sponsored meetings to ensure that deficiencies identified below are being addressed as part of the DOE program to develop a license application.

In general, DOE's approach to assess the effects of coupled THC processes on radionuclide transport through engineered and natural barriers must meet the following generic acceptance criteria: (i) data and model justification; (ii) data uncertainty and verification; (iii) model uncertainty; (iv) model verification; (v) integration; (vi) quality assurance; and (vii) expert elicitation (U.S. Nuclear Regulatory Commission, 1998c).

5.4.4.1 Data and Model Justification for Subissue 4

The following seven acceptance criteria address data and model justification. DOE's approach to evaluate and abstract coupled THC effects on radionuclide release transport in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on transport of radionuclides in the near field were considered.

There are sufficient data from the DOE thermal testing program; observations from natural analogs; and interpretations from modeling (e.g., Hardin, 1998) to suggest that the effects of the coupled THC processes are likely important to repository performance. The DOE has made progress since TSPA-95 in abstracting detailed process models for radionuclide transport through the engineered and natural barriers in the near field (TRW Environmental Safety Systems, Inc., 1995). For the first time in the TSPA-VA, DOE implemented models of colloid transport and retardation by invert material. Nevertheless, many approximations and assumptions were made, and DOE recognizes the limitations. DOE should use available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on radionuclide transport in their abstractions.

Criterion 2 DOE's evaluation of coupled THC processes properly considered site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect radionuclide transport in the near field.

DOE 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). However, effects of certain coupled THC processes may be most important on the margins of the emplacement zone. Refluxing water may be shed to the margins of the emplacement. 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.

Potential episodic seepage events during the thermal period cannot be excluded (see Large Block Test results in Hardin, 1998). Thus, DOE should use results from their thermal testing program to establish initial and boundary conditions for their conceptual models in their abstractions.

Criterion 3 Sufficient data were collected on 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 transport of radionuclides in the near field.

The DOE base case analysis does not include the effect of seepage water interacting with the concrete drift lining. Interaction of water with the concrete liner and drift invert will affect radionuclide transport. We will expect the concrete-modified water case to be part of the base case in the DOE PA analyses for any potential LA should DOE continue to have a repository design with large amounts of cementitious materials. Regardless of the design, sufficient data are necessary to establish initial and boundary conditions for conceptual models. There is general agreement that there is a general lack of data to support assumptions in the models (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999).

Ongoing work described in Hardin (1998) is designed to evaluate the effects of system chemistry and mineralogy on transport through the near field, but results were not included in the VA analyses. Presumably, the work described in Hardin (1998) will continue and be used to constrain sorption parameters and provide a theoretical basis for abstractions for TSPA-LA.

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. These properties should be used in their abstractions.

Criterion 4 A nutrient and energy inventory calculation (e.g., McKinley, West, and Grogan, 1985; Grogan and McKinley, 1990; Noy, et al., 1996) was used to determine the potential for microbial activity that could adversely affect radionuclide transport through engineered and natural barriers.

model for bounding the magnitude of development of microbial communities was developed. 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 was used by DOE. We have 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.

Criterion 5 If microbial activity was predicted to be sufficient to potentially cause adverse microbial effects on transport of radionuclides through engineered and natural barriers, then the time-history of temperature, humidity, and water saturation in engineered and natural materials was used to constrain the probability for these effects.

The microbial activity model was not used in the base case, but provided first-order limits on potential microbial effects (CRWMS M&O, 1998d). DOE did not attempt to evaluate potential microbial effects on radionuclide transport. DOE should use the time-history of temperature, humidity, and dripping (CRWMS M&O, 1997b) 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). From presentation at the IDGE workshop it is not clear that DOE will pursue any further work on microbial processes in the near-field geochemical environment. Neglecting the potential impact of microbial processes on radionuclide transport will require justification (see Section 5.4.4.5, Criterion 3).

Criterion 6 Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were used to determine if additional new data are needed to better define ranges of input parameters.

We believe that the DOE approach of setting the K_Ds of the actinides to zero for sensitivity studies is appropriately conservative, and concur with the DOE use of sensitivity analyses to evaluate the relative importance of sorption in the unsaturated zone to repository performance and to bound the potentially nonconservative interactions. Nevertheless, placeholder K_Ds used in transport through the engineered barrier system should be replaced by more realistic limits on sorption as additional experimental data become available. DOE should use sensitivity and uncertainty analyses of their abstracted model to determine whether additional new data are needed to support the abstracted model.

Criterion 7 DOE has identified specific plans to acquire the necessary information as part of the performance confirmation program, if the testing program for the effects of coupled THC processes on radionuclide transport is not complete at the time of license application, or if sensitivity and uncertainty analyses indicate additional data are needed.

The performance confirmation program was not a focus of our VA review. For future TSPAs, DOE should identify specific plans to acquire the necessary information as part of the performance confirmation program, if sensitivity studies indicate additional data are needed.

5.4.4.2 Data Uncertainty and Verification for Subissue 4

The following five acceptance criteria address data uncertainty and verification. DOE's approach to evaluate and abstract coupled THC effects on radionuclide transport in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on transport of radionuclides in the near field. Parameter values, assumed ranges, probability distributions, and bounding assumptions were technically defensible and reasonably accounted for uncertainties.

DOE has not used reasonable or conservative range of parameters to determine the effects of coupled thermal-hydrologic-chemical processes on the transport of radionuclides. Model parameters, particularly K_c and the irreversibly sorbed fraction, are not well constrained and will require additional experiments or other analyses to establish reasonable bounding limits. In addition, the modeling approach has been limited to plutonium. This approach should be applied to other strongly sorbing radionuclides that may be subject to colloid transport such as americium and thorium, or DOE should clearly demonstrate that colloidal transport of these radionuclides is not important to repository performance.

Some unrealistic model results from the thermal-chemical evaluations indicate 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, 5-49). In addition, only local equilibrium models were developed for in-drift water chemistry evaluations. We recognize the challenges associated with compilation of an internally consistent and validated thermodynamic and kinetic database for relevant mineral phases.

DOE should use reasonable or conservative ranges of parameters to determine effects of coupled THC processes on radionuclide transport in their abstracted models. We recommend exercises using experimental, site, and natural analog data to aid verification of 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, probability distributions. If DOE uses bounding assumptions, they should be technically defensible and reasonably account for uncertainties.

Criterion 2 Uncertainties in data due to both temporal and spatial variations in conditions affecting coupled THC effects on radionuclide transport in the near field were considered.

DOE did not consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on radionuclide transport. DOE should consider contributions to data uncertainty from both temporal and spatial variability. These contributions uncertainty should be implemented in their abstracted models of the effects of coupled THC processes on the transport of radionuclides. Presentations at the IDGE workshop indicate that spatial and temporal variations in results from the drift scale heater test are now being considered in the process-level modeling studies.

Criterion 3 DOE's evaluation of coupled THC processes properly considered 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 transport of radionuclides in the near field.

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. There is also uncertainty in the parameters used in the TSPA-VA abstracted models. For example, the plutonium K_Ds used in modeling colloid transport are based on experimental data reported by Lu et al. (1998) and cover a range of about seven orders of magnitude (100–10^9 mL/g). The geochemical processes and conditions that result in this parameter variability are not explicitly incorporated in the TSPA-VA calculations.

Transport of radionuclides through the engineered barrier system is simulated using a K_D to represent interaction with the invert. Sorption characteristics of the invert are poorly understood (U.S. Department of Energy, 1998b, Section 3.5.3.6), and the sorption coefficients used in the TSPA-VA are referred to as "placeholder K_Ds" by DOE (CRWMS M&O, 1998d, Section 6.5.2). The ranges for all sorbing radionuclides (e.g., U, Np, Pa, and Pu) vary from a minimum value of zero to a maximum value that is typically greater than values used for unsaturated zone and saturated zone transport (U.S. Department of Energy, 1998b, Section 4.1.10).

Some mineral phases, anticipated on the basis of alteration in the vicinity of YM (e.g., smectite, illite, and zeolites), are absent from the models of coupled thermal-chemical effects. These limitations are generally recognized by DOE. Regardless of the repository design, 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.

Criterion 4 The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on radionuclide transport in the near field were consistent with available data.

DOE's completed sensitivity analyses for transport through the invert using the 5th and 95th percentile K_D values with all other parameters set at base case expected values. Their results suggest no influence on dose out to 10,000 yrs., 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 yrs., with the effect decreasing at times greater than about 250,000 yrs. DOE should continue to use initial conditions, boundary conditions, and computational domains in their sensitivity analyses involving coupled THC effects on the transport of radionuclides that are consistent with available data.

Criterion 5 DOE's performance confirmation program will assess whether the natural systr and engineered materials are functioning as intended and anticipated with rega to coupled THC effects on transport of radionuclides in the near field.

The performance confirmation program was not a focus of our VA review. DOE should address this objective in their performance confirmation plan.

5.4.4.3 Model Uncertainty for Subissue 4

The following three acceptance criteria address model uncertainty. DOE's approach to evaluate and abstract coupled THC effects on radionuclide transport in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems were used.

Transport of radionuclide through the engineered barrier system and near-field environment, including sorption onto engineered barrier system materials is new to DOE's TSPAs. Only sorption onto the invert supporting the waste package is considered (U.S. Department of Energy, 1998b, Section 6.5.1). The invert is assumed to be intact concrete, an assumption that DOE recognizes is possibly nonconservative (CRWMS M&O, 1998d, Section 6.5.1). Each invert cell is assumed to have a porosity of 10 percent, and both advective and diffusive transport are assumed between adjacent invert cells. Advective flow is set equal to the seepage through the drift. A maximum (conservative) concentration gradient driving diffusion is modeled for high solubility radionuclides by assuming zero concentration at the edge of the engineered barrier system, where radionuclides are "swept away" as soon as the boundary is reached. Low solubility radionuclides are presumed to precipitate at the engineered barrier system boundary and are removed slowly as a function of seepage flux. The approach to modeling radionuclide transport through the engineered barrier system is consistent with models used to simulate transport through the natural barrier system. DOE recognizes, however, that some modeling assumptions may not be conservative. The assumption of intact concrete needs to be refined to demonstrate the effects of changing chemical conditions on transport.

Colloid transport of plutonium is modeled by calculating an effective sorption coefficient based on colloid concentration and the degree to which plutonium sorption on a colloid phase is enhanced relative to sorption on the rock matrix. Most plutonium attachment onto colloids is assumed to be fast and reversible, but a small fraction is assumed to sorb irreversibly to colloids. The partitioning between the aqueous and colloid phase is represented by a single lumped parameter referred to as the aqueous-colloid partitioning coefficient, K_C. The mass of colloids in each of the waste package cell pathways is dependent on the volume of water in the cell and the colloid concentration per unit volume of water. Filtration is not incorporated into the model, and the only measure of colloid stability is an empirical relationship of colloid concentration to ionic strength. Four types of colloids are considered in the engineered barrier system: natural iron-oxide and clay and colloids derived from spent fuel and glass waste. Sorption on colloids is modeled using a linear sorption coefficient (K_D) that ranges from 10^2 to 10^9 mL/g. The irreversible fraction is assumed to be defined by a log-uniform distribution.

The major limitation recognized by the 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 and should be justified or remedied. DOE should use appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems. Presentations at the IDGE Workshop on corrosion products and the their impact on the chemistry of water reacting with waste forms indicate that DOE is attempting to address this acceptance criterion.

Criterion 2 Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

A more detailed simulation of colloidal transport was developed as an auxiliary analysis for colloid transport in the saturated zone (CRWMS M&O, 1998d, Section 8.5.2.5). This model includes sorption and desorption rates, colloid filtration, and effective porosities. DOE recommends this type of approach be used for the in-drift colloid model, but it requires additional laboratory, field, and site-scale observations to develop the necessary model parameters.

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, 5-48). We concur with this observation. Present models are recognized to be inadequate.

Criterion 3 DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on radionculide transport in the near field. The description included a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model.

The DOE TSPA-VA base case evaluation includes diffusion through and radionuclide retardation in the concrete invert for neptunium, plutonium, uranium, and protactinium. The results indicate that there is a transport delay of neptunium-237 in the invert. A K_D value of 100 mL/g for neptunium (600 mL/g for plutonium) makes travel time in the dry climate slow (on the order of 100,000 yrs. through the 1 m of invert), whereas in the long-term average climate the travel time is about 7,500 yrs. The DOE analysis omits the potential effect of degradation of the invert in its calculations of radionuclide transport through the engineered barrier system and assumes that its effect on overall system performance is insignificant because of the small transport length involved relative to the total transport length. DOE supported this conclusion with results of sensitivity analyses indicating that retardation in the invert has little significance to total dose. We agree 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, the delay caused by transport through the invert is a substantial portion of a 10,000 year regulatory period. Thus the model will need to be consistent with expected engineered materials. For instance, the model used by the DOE for the invert allows for substantial retardation, while the model chosen by NRC in the TPA 3.2 code shows negligible impact on radionuclide retardation.

DOE did not assess the effects of coupled THC processes on radionuclide transport. Thus, they did not provide a reasonable description of the mathematical models included in its analyses of coupled THC effects on the transport of radionuclides. DOE discussed some of the limitations and uncertainties in their approach to colloidal transport. Generally these models and analyses are considered to be preliminary and inadequate. DOE did distinguish between the different approaches that they used in the saturated zone and in-drift colloid transport. DOE described a more detailed simulation that was developed as an auxiliary analysis for colloid transport in the saturated zone (CRWMS M&O, 1998d, Section 8.5.2.5). This model includes sorption and desorption rates, colloid filtration, and effective porosities. DOE recommends this type of approach be used for the in-drift colloid model.

5.4.4.4 Model Verification for Subissue 4

The following three acceptance criteria address model verification. DOE's approach to evaluate and abstract coupled THC effects on radionuclide transport in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 The mathematical models for coupled THC effects on radionculide transport in the near field 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.

The general lack of data to support and test critical assumptions in mathematical models has been noted by others (U.S. Nuclear Waste Technical Review Board, 1999; Whipple, et al., 1999). We recommend additional exercises using experimental, site, and natural analog data to aid verification of 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. .

Criterion 2 DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on transport of radionuclides in the near field.

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 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.

Criterion 3 Abstracted models for coupled THC effects on radionuclide transport were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results were compared with different mathematical models to judge robustness of results.

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 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.

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-LA. These models appear to represent a reasonable first approach to colloid transport through the engineered barrier system, and they are consistent with models used to simulate colloid transport through the natural barrier system. The proposed work described in Hardin (1998) includes site-specific studies and laboratory and modeling work for other waste package materials, concretes, and cementitious materials. These tests, if carried out and properly documented, are likely to meet the acceptance criteria presented in this subissue.

Should DOE continue to take credit for retardation in the engineered barrier system, DOE should continue its work investigating more detailed process models of colloid transport and radionuclide sorption on engineered barrier system materials. These models should consider not only J-13 water, but also evolved water and gas phases that are the output of coupled THC calculations.

DOE should use the results from their laboratory experiments, field heater test program, and their natural analog program to test their abstracted models for consistency with observations. We recommend 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.

5.4.4.5 Integration for Subissue 4

The following four acceptance criteria address integration. DOE's approach to evaluate and abstract coupled THC effects on radionuclide transport in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 DOE considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, physical phenomena, and couplings, and used consistent and appropriate assumptions throughout.

The approach that DOE used for the VA did not include a formal screening process for features, events, and processes (FEP). As a result many important design features, physical phenomena, and couplings were not evaluated in a performance assessment framework. Many uncertainties associated with radionuclide transport in the near field are comparable to those identified in Section 5.4.2 concerning the WP chemical environment. In general, DOE recognizes the significance of coupled THC processes for prediction of the evolution of this environment. Efforts to model these phenomena represent an advance over prior TSPA efforts. Nevertheless, these models retain notable uncertainties and limitations such as neglect of concrete, cladding, basket, container, and waste form alteration products on water chemistry; and neglect of coupled gas and aqueous phase chemical evolution.

Although other engineered materials are likely to interact with radionuclides, DOE considered sorption only on the invert material which was assumed to be represented by intact concrete (U.S. Department of Energy, 1998b). Interactions with corrosion products of the metallic WP are assumed represented by equilibrium with goethite. However, goethite is not identified as a corrosion product in the models for WP corrosion. Other phases could predominate, and the metals are not necessarily predominantly iron. Corrosion of metals such as nickel, molybdenum, and aluminum will likely result in minerals sufficiently different from goethite that the water chemistry contacting the waste form will be influenced strongly. Other possibly important materials include iron oxidation products from the WP, secondary alteration products from the waste form, and hydrothermally altered cement. In addition, extensive use of the C-22 alloy may make nickel and molybdenum corrosion products significant.

In the TSPA-VA, in-drift colloid transport of plutonium-239 and plutonium-242 is modeled using calculated near-field geochemical conditions. An empirical relationship between colloid stability and ionic strength is stochastically incorporated into the RIP code (CRWMS M&O, 1998d; Section 4.4.3.3). The in-drift colloid model is relatively independent of the other parts of the near-field geochemical model (CRWMS M&O, 1998d). Colloid concentration is linked through the calculated ionic strength, but otherwise there is no strict coupling of THC processes.

Criterion 2 Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on transport of radionuclides in the near field.

DOE did not include some important design features, physical phenomena, and couplings that cause temporal and spatial variations in conditions. For instance, spatial variation in the thermal reflux is expected and DOE did not evaluate the potential variation. Thus the TSPA-VA models do not account for known temporal and spatial variations in conditions affecting coupled THC effects on the transport of radionuclides. For the TSPA-VA, stepped temporal changes in the chemical composition of water entering and traversing the emplacement drift were modeled. This is a significant advance.

Criterion 3 If potentially important couplings were neglected, DOE provided a technical basis for doing so.

DOE has neglected the effect of alkaline plume migration on sorption in the unsaturated zone. However, DOE recognizes that migration of alkaline fluids into the unsaturated zone may alter the siliceous host rock, both along fracture pathways and in the matrix, which could produce changes in the amount of fracture-matrix interaction and the sorption of radionuclides within the tuff rock units (CRWMS M&O, 1998d, Section 4.2.1.3.2). Although the DOE TSPA-VA process models did not include explicit representation of these potential changes to minerals along radionuclide migration pathways, the potential effect of alkaline plume migration was evaluated in sensitivity analyses by setting the unsaturated zone sorption coefficients for actinides (uranium, neptunium, plutonium, and protactinium) to zero (U.S. Department of Energy, 1998b, Section 5.6). The results indicate that total dose rates at early times are not affected by changes in sorption coefficients, primarily due to the dominance of early dose by unretarded technetium. Thus, DOE's neglect of the effect of alkaline plume migration on sorption in the unsaturated zone is acceptable.

Interactions with corrosion products of the metallic waste package are assumed represented by equilibrium with goethite. However, goethite is not identified as a corrosion product in the models for WP corrosion. Other phases could predominate, and the metals are not necessarily predominantly iron. Corrosion of metals such as nickel, molybdenum, and aluminum will likely result in minerals sufficiently different from goethite that the water chemistry contacting the waste form will be influenced strongly.

There must be a technical basis for omitting potentially important couplings that may significantly affect radionuclide transport. Although DOE considered many pertinent important couplings in their near field geochemical environment model, these abstractions were not used to evaluate the effects of coupled THC processes on transport of radionuclides. DOE should provide a technical basis for omitting these processes or include these effects in the performance analyses via abstracted models or bounding assumptions.

The FEP analysis should include the processes described above and those listed in Section 5.4.4. Presentations at the IDGE Workshop indicate that DOE will complete a FEP analysis for the TSPA that could be used for DOE's Site Recommendation and a potential License Application. The FEP analysis should meet the acceptance criteria in the TSPAI IRSR (U.S. Nuclear Regulatory Commission, 1998c) for scenario analysis. This FEP analysis will an important action for DOE to take to facilitate resolution on this subissue.

Criterion 4 The bases used for modeling assumptions and approximations were documented and justified, where simplifications for modeling coupled THC effects on radionuclide transport in the near field were used for performance assessment analyses instead of detailed process models.

DOE should continue to document the bases for simplifications used in modeling coupled THC effects on radionuclide transport.

5.4.4.6 Quality Assurance and Expert Elicitation Concerns for Subissue 4

The following three acceptance criteria address quality assurance and expert elicitation. DOE's approach to evaluate and abstract coupled THC effects on radionuclide transport in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Data and models were collected, developed, and documented under acceptable quality assurance (QA) procedures.

The assessment of DOE's QA program relative to this criterion is presented in Section 5.4.1.6 and is not repeated here.

Criterion 2 Deficiency reports concerning data quality on issues related to coupled THC effects on radionuclide transport were closed.

As of May 18, 1999, three QA deficiency reports concerning issues related to coupled THC effects on radionuclide transport remain open. These are the same reports mentioned in Section 5.4.2.6. Different aspects of the Tracers, Fluids and Material (TFM) Control program are covered by these deficiency reports. The TFM program is one of the main bases for abstracting the effects of microbial processes on the transport of radionuclides. Thus, DOE should pursue closing these deficiency reports.

Criterion 3 Expert elicitations, if used, were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

Concerns arising from the expert elicitations that DOE has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c) have already been documented (U.S. Nuclear Regulatory Commission, 1998h). These concerns were not directly associated with the radionuclide transport through the near field subissue. However, the expected use of the elicited expert judgements needs to be understood prior to convening the expert panel. The results from the near-field/altered-zone expert elicitation were not used to develop the initial and boundary conditions for the radionuclide transport abstraction. A positive aspect of the near-field/altered-zone expert elicitation was that experts were elicited for recommendations on what actions should be taken to reduce uncertainties (CRWMS M&O, 1998c). Based on presentations at the THCP Workshop and the IDGE Workshop, it is not clear that these recommendations are being considered.

5.4.5 Subissue 5: Effects of Coupled Thermal-Hydrologic-Chemical Processes on Potential Nuclear Criticality in the Near Field

The acceptance criteria of this new subissue have been presented and our preliminary conclusion is that sensitivity and uncertainty analyses should be completed to determine whether criticality will impact repository performance. 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. Provided that the sensitivity studies indicated that these potential consequences of criticality in the near field ill not adversely impact repository performance, then criticality need not be considered within a isk-informed assessment of repository performance.

Nuclear criticality is not considered as a Key Element of Subsystem Abstraction by the NRC (U.S. Nuclear Regulatory Commission, 1998c). 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 under the scenario analysis subissue of the Total System Performance Assessment and Integration Issue Resolution Status Report (IRSR; U.S. Nuclear Regulatory Commission, 1998c). Revision 1 of the Evolution of the Near-Field Environment (ENFE) IRSR (U.S. Nuclear Regulatory Commission, 1998g) was completed prior to development of scenario analysis acceptance criteria. The approach taken in Revision 1 of the ENFE IRSR (U.S. Nuclear Regulatory Commission, 1998g) was to focus on the evaluation of the consequences of criticality on repository performance.

The scenario analysis acceptance criteria allow scenarios to be screened out of a performance assessment based on probability, consequence, or regulatory arguments (U.S. Nuclear Regulatory Commission, 1998c). 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 technical acceptance criteria for abstraction listed in Section 4.5. DOE's efforts to assess the effects of criticality on repository performance are addressed in the Container Life and Source Term (CLST), ENFE, and Radionuclide Transport (RT) IRSRs. The CLST IRSR (U.S. Nuclear Regulatory Commission, 1998d) lists the acceptance criteria that will be used to evaluate criticality within the waste package. 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 ithin and surrounding the emplacement drifts that is affected by coupled thermal-hydrologic-hemical processes. The RT IRSR (U.S. Nuclear Regulatory Commission, 1998e) only addresses criticality that could occur under isothermal conditions or at greater distances from the repository.

Since publication of Revision 1 of the ENFE IRSR, DOE has made a major step toward resolution of the nuclear criticality subissue. The Disposal Criticality Analysis Methodology Topical Report (U.S. Department of Energy, 1998e) was published. This report describes the approach DOE will use to evaluate the potential for and consequences of repository criticality as an input to the TSPA-LA. The DOE 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 those configurations exceeding the probability criterion. The 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 being external to the WP but inside the drift wall (U.S. Department of Energy, 1998e).

NRC is reviewing the overall approach presented in the methodology topical report (U.S. Department of Energy, 1998e). The review will be documented in a safety evaluation report (U.S. Nuclear Regulatory Commission, 1999c). Therefore, a review of the near-field portions of ie topical report is not presented here. The DOE 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 th to calculate its consequences only if the probability exceeds a defined threshold. The consequence analyses are limited to considering effects on radionuclide inventory and repository thermal profile (U.S. Department of Energy, 1998e).

DOE's approach in dealing with criticality in the near field differs from that proposed in Revision 1 of the ENFE IRSR (Section 4.5.1). NRC stated that, prior to including near-field criticality in PA, "...sensitivity and uncertainty analyses should be completed [by DOE] to determine whether criticality will impact repository performance." However, DOE's approach satisfies the NRC's scenario analysis approach (U.S. Nuclear Regulatory Commission, 1998c) and is acceptable. We review DOE's treatment of near field criticality in the TSPA-VA below. Based on DOE's work in the Viability Assessment, the four step process DOE will use to evaluate the potential for repository criticality and the consequences DOE will investigate (U.S. Department of Energy, 1998e), we have no further questions, at this time, concerning potential nuclear criticality in the near field. This subissue is resolved at the staff level. We will continue to review DOE's Disposal Criticality Analysis Methodology Topical Report and will evaluate DOE's analysis supporting their screening of disposal criticality in the near field as part of the safety evaluation report process.

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) data and model justification; (ii) data uncertainty and verification; (iii) model uncertainty; (iv) model verification; (v) integration; (vi) quality assurance; and (vii) expert elicitation (U.S. Nuclear Regulatory Commission, 1998c)

5.4.5.1 Scenario Screening for Subissue 5

The following acceptance criterion address scenario screening. DOE's approach to evaluate whether coupled THC effects on potential criticality needs to be abstracted in a TSPA for the proposed repository at YM will be acceptable provided that the acceptance criterion is met.

Criterion 1 Sensitivity and uncertainty analyses (including consideration of alternative conceptual models) were completed to determine whether criticality will impact repository performance, and whether additional new data are needed to better define ranges of input parameters.

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), the 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 concludes, based on a more detailed discussion (CRWMS M&O, 1998a, Section 10.6.2.2 and 10.6.4), that the external criticality mechanisms are exceedingly unlikely. The near-field scenario involves transport of fissile material (U and Pu) out the bottom of a breached waste package. The scenario then envisions 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

0.6.2.2.1). DOE uses high solubilities for U (6,000 ppm) and Pu (78 ppm), estimated for high-H 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 U and Pu 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 is that resulting 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 3 percent after 20,000 yrs. (CRWMS M&O, 1998e). Due to the remote probability of the event, and the negligible effect on total radionuclide inventory, DOE concludes that external criticality has insignificant effect on repository performance (U.S. Department of Energy, 1998d, page O-3).

DOE did not consider the effects on the near field from heat output from criticality. DOE 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). Pending the results of the staff review of DOE's Disposal Criticality Analysis Methodology Topical Report, we have no questions concerning potential nuclear criticality in the near field at this time.

.4.5.2 Data and Model Justification for Subissue 5

The following three acceptance criteria address data and model justification. DOE's approach to evaluate and abstract coupled THC effects on potential nuclear criticality in the near field in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Available 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 were considered.

The DOE treatment of external criticality in the TSPA-VA is preliminary and incomplete, but as discussed in Section 5.4.5, the methodology 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), the DOE lists 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 Pu out of the waste package. Should DOE abstract near field criticality in a future performance assessment, then they 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.

Criterion 2 DOE's evaluation of coupled THC processes properly considered 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.

Should DOE abstract near field criticality in a future performance assessment, then they should 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.

Criterion 3 Sufficient data were collected on 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 may affect nuclear criticality in the near-field environment.

Should DOE abstract near field criticality in a future performance assessment, then they 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.

5.4.5.3 Data Uncertainty and Verification for Subissue 5

The following four acceptance criteria address data uncertainty and verification. DOE's approach to evaluate and abstract coupled THC effects on potential nuclear criticality in the near field in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on potential nuclear criticality in the near-field environment. Parameter values, assumed ranges, probability distributions, and bounding assumptions were technically defensible and reasonably accounted for uncertainties.

All DOE criticality analyses to date have been preliminary with respect to planned analyses for the license application. Thus, a systematic data evaluation has not been performed. Sensitivity studies in an analysis of external Pu 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, then they 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.

Criterion 2 Uncertainties in data due to both temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality were considered.

Should DOE abstract near field criticality in a future performance assessment, then they should consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality.

Criterion 3 DOE's evaluation of coupled THC processes properly considered 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.

Should DOE abstract near field criticality in a future performance assessment, then they 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.

Criterion 4 The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on potential nuclear criticality in the near-field environment were consistent with available data.

Should DOE abstract near field criticality in a future performance assessment, then they 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.

5.4.5.4 Model Uncertainty for Subissue 5

The following two acceptance criteria address model uncertainty. DOE's approach to evaluate and abstract coupled THC effects on potential nuclear criticality in the near field in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.

Should DOE abstract near field criticality in a future performance assessment, then they should investigate alternative modeling approaches that are consistent with available data and current scientific understanding.

Criterion 2 DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on potential nuclear criticality. The description included a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model.

Should DOE abstract near field criticality in a future performance assessment, then they 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 the TSPA to model performance impact (U.S. Department of Energy, 1998e). These models should be adequately described.

5.4.5.5 Model Verification for Subissue 5

The following three acceptance criteria address model verification. DOE's approach to evaluate and abstract coupled THC effects on potential nuclear criticality in the near field in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 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.

Should DOE abstract near field criticality in a future performance assessment, then the mathematical models they use 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.

Criterion 2 DOE appropriately adopted accepted, and well-documented, procedures to construct and test the numerical models used to simulate coupled THC effects on potential nuclear criticality.

Should DOE abstract near field criticality in a future performance assessment, then they 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 (CL)—the value of k_{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).

Criterion 3 Abstracted models for coupled THC effects on potential nuclear criticality were based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted model results were verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results were compared with different mathematical models to judge robustness of results.

Should DOE abstract near field criticality in a future performance assessment, then they should verify abstracted model results through comparison to outputs of detailed process models and empirical observations.

5.4.5.6 Integration for Subissue 5

The following five acceptance criteria address integration. DOE's approach to evaluate and abstract coupled THC effects on potential nuclear criticality in the near field in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 DOE considered all the relevant features, events, and processes. The abstracted models adequately incorporated important design features, including criticality safety features; physical phenomena and couplings, including neutron absorbers; and used consistent and appropriate assumptions throughout.

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 DOE should consider all the relevant features, events, and processes. For instance, DOE should evaluate the impact of heat generated from a criticality on repository performance. DOE has indicated that they will do this as part of their disposal criticality analysis (U.S. Department of Energy, 1998e).

Criterion 2 Important mass transfer and mass transport processes and mechanisms considered for formation of both a critical mass and configuration were plausible for the YM near-field environment.

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, that are plausible for the YM near-field environment, should be considered.

Criterion 3 Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality.

Should DOE abstract near field criticality in a future performance assessment, then their models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality.

Criterion 4 If potentially important couplings and criticality in the near field are neglected, DOE provided a technical basis for doing so.

Should DOE abstract near field criticality in a future performance assessment, and if potentially important couplings in the near field are neglected, DOE should provide a technical basis for doing so.

Criterion 5 The bases used for modeling assumptions and approximations were documented and justified, where simplifications for modeling coupled THC effects on potential nuclear criticality were used for PA analyses instead of detailed process models.

Should DOE abstract near field criticality in a future performance assessment, then the bases used for simplifying modeling assumptions and approximations should be documented and justified.

5.4.5.7 Quality Assurance and Expert Elicitation Concerns for Subissue 5

The following three acceptance criteria address quality assurance and expert elicitation. DOE's approach to evaluate and abstract coupled THC effects on potential nuclear criticality in the near field in a TSPA for the proposed repository at YM will be acceptable provided that each of the acceptance criteria are met.

Criterion 1 Data and models were collected, developed, and documented under acceptable quality assurance (QA) procedures.

The assessment of DOE's QA program relative to this criterion is presented in Section 5.4.1.6 and is not repeated here.

Criterion 2 Deficiency reports concerning data quality on issues related to the effects of coupled THC processes on potential near field criticality were closed.

As of May 18, 1999, there are no QA deficiency reports concerning issues related to coupled THC effects on potential nuclear criticality in the near field.

Criterion 3 Expert elicitations, if used, were conducted and documented in accordance with the guidance in NUREG-1563 (Kotra, et al., 1996) or other acceptable approaches.

Concerns arising from the expert elicitations that DOE has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c) have already been documented (U.S. Nuclear Regulatory Commission, 1998h). These concerns were not directly associated with the criticality subissue. However, the expected use of the elicited expert judgements needs to be understood prior to convening the expert panel. The results from the near-field/altered-zone expert elicitation were not used to develop the scenarios for criticality abstraction. A positive aspect of the near-field/altered-zone expert elicitation was that experts were elicited for recommendations on what actions should be taken to reduce uncertainties (CRWMS M&O, 1998c).

5.5 SUMMARY OF ISSUE RESOLUTION AT THE STAFF LEVEL

As noted in Section 5.1 of this revision, two items relating to the ENFE KTI, resulting from our review of the DOE SCP, have been previously resolved (Comments 29 and 92). Comments 81 and 84 have been judged to be outside the scope of the ENFE KTI. Comment 90 is considered closed. The remaining ENFE-related items from the review of the DOE SCP (Comments 25, 79, and 89; Question 30) are considered to be open.

The potential impact of microbial processes affecting seepage and flow need not be considered in performance assessments of the proposed repository. Likewise, the large self potentials measured in Yucca Mountain thermal tests, although speculated to be important to waste package performance, have been demonstrated to be unimportant to waste package performance and need not be considered in performance assessments of the proposed repository.

The acceptance criteria for each subissue in the ENFE IRSR have been completed and the Revision 1 IRSR will be used to review the DOE TSPA-VA. Revision 2 of the IRSR will document the review of the TSPA-VA and its supporting documentation to determine whether DOE's evaluation of the potential impacts of coupled thermal-hydrologic-chemical processes on repository performance is acceptable and will serve as the basis for the LA review.

The ENFE KTI remains unresolved from the perspectives of the staff and the DOE (Table 3). However, general consensus prevails concerning the nature of technical problems to be addressed and appropriate approaches to be undertaken in resolution of technical issues. Coupled THC processes are recognized by DOE to have potentially important effects on repository performance with regard to each of the subissues identified in the ENFE KTI.

Preliminary models, which are supported by a variety of experimental and field data, show potentially large effects from coupled THC processes on the flow field. No effects of coupled THC processes on seepage, flow, and radionuclide transport were abstracted for the TSPA-VA performance calculations. This constitutes an important unresolved technical issue for both the seepage and flow, and the radionuclide transport subissues. Advances in modeling coupled THC effects on the waste package chemical environment have illustrated that large deviations from ambient water chemistry and broad temporal variations in water chemistry are to be anticipated. Omission of modeled near-field chemistry in establishing environmental conditions for waste package corrosion processes in the TSPA-VA appears to be an important limitation in the abstraction process. Integration of coupled THC effects on gas and water chemistry and incorporation of the variety of engineered materials in evaluations of the chemical environment for radionuclide release is required for resolution of the radionuclide release subissue. Based on DOE's work in the Viability Assessment, the four step process DOE will use to evaluate the potential for repository criticality and the consequences DOE will investigate (U.S. Department of Energy, 1998e), we have no further questions concerning potential nuclear criticality in the near field. The criticality subissue is resolved at the staff level.

Preliminary investigations of process models for radionuclide sorption on engineered barrier system materials and colloid transport represent an advance in the TSPA-VA. Efforts are being undertaken by DOE to address effects of coupled THC processes in the near field through studies of coupled models, laboratory testing, field testing, and natural analog studies. DOE has made considerable progress in addressing the effects of coupled THC processes on performance in the TSPA-VA relative to prior performance assessments. Present limitations in these analyses are recognized and generally well documented by DOE, particularly in the TSPA-VA Technical Basis Documents (CRWMS M&O, 1998d). Resolution of the subissues with the ENFE KTI is hindered by the changing and preliminary design of the engineered barrier system and thermal loading regime.

DOE's consideration of Enhance Design Alternative II (EDA II), a lower heat load repository design, could substantially reduce the impact of coupled THC processes on repository performance. The EDA II design is substantially different than the TSPA-VA design and many of the effects of coupled THC processes on repository performance expected at a higher heat load may no longer be applicable. NRC will review DOE's Feature, Events, and Processes (FEP) screening analysis, and its database, to determine whether an adequate technical basis has been provided for those features associated with coupled THC processes that DOE has screened out from the performance assessment. The DOE's FEP analysis and the staff's review of the analysis will provide the best near-term opportunity to further resolution of subissues within the ENFE KTI.

Table 3. Summary of issue resolution for ENFE KTI subissues

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

.0 REFERENCES

Altman, W.D., J.P. Donnelly, 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, 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_2-Al_2O_3-MgO-SO_3-H_2O 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. Jarzemba, *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.

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.

Bertetti, F.P., R.T. Pabalan, and M.G. Almendarez, *Studies of neptunium^{V} sorption on quartz, clinoptilolite, montmorillonite, and a-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.

Bowers, T.S., and R.G. Burns, *Activity diagrams for clinoptilolite: Susceptibility of this zeolite to further digenetic 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, 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.

Buchwald, V.F., and R.S. Clarke, *Corrosion of Fe-Ni alloys by Cl-containing akaganeite (ß-FeOOH): The Antarctic meteorite case*, American Mineralogist, Vol. 74, pp. 656–667, 1989.

Buck, E.C., et al., *Retention of neptunium in uranyl alteration phases formed during spent fuel corrosion*, Scientific Basis for Nuclear Waste Management XXI, Materials Research Society, Pittsburgh, Pennsylvania, Symposium Proceedings 506, pp. 87–94, 1998.

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.

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-95-851, Los Alamos National Laboratory, Los Alamos, New Mexico, 1995.

Carlos, B., *Fracture-Coating Minerals in the Topopah Spring Member and Upper Tuff of Calic 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.

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*, B00000000-01717-5700-00008, Revision 0, Volume 1 of 3, 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, Rev 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,* BABEAF000-01717-2200-00011 Revision 2, 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,* B00000000–01717–4301, Revision 01, 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,* BBA000000–01717–5705–00018, Revision 01, North Las Vegas, Nevada, MOL.19980318.0412, 1998e.

Codell, R.B., and W.M. Murphy, *Geochemical model for ^{14}C 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.

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.

Cragnolino, 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.

Cragnolino, G.A., et al., *Hanford Tank Waste Remediation System Familiarization Report,* CNWRA 97-001, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1997.

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.

Davis, M.A., et al., Sustaining native microbial growth with endogenous nutrients at Yucca Mountain, Proceedings of the Eighth International Conference on High Level Radioactive Waste Management, American Nuclear Society, La Grange Park, Illinois, pp. 662–665, 1999.

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_2-O_2-bearing fluid inclusions in three U deposits*, Geochimica et Cosmochimica Acta, Vol. 52, pp. 1,155–1,167, 1988.

Ebert, W.L., and J.K. Bates, *A comparison of glass reaction at high and low SA/V:PCT vs 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.

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., *Matrix Properties of Hydrogeologic Units at Yucca Mountain, Nevada*, Water-Resources Investigation Report 96-XX, Draft, U.S. Geological Survey, Denver, Colorado, 1996.

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.

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.

Horn, J.M., et al., *MIC evaluation and testing for the Yucca Mountain repository*, Paper 152, CORROSION98, NACE International, Houston, Texas, 1998.

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, Vol. 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.

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.

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. 3128–3133, 1997.

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.

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.

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 HLW 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, 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, 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.

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., I.R. Triay, C.R. Cotter, H.D. Kitten, and J. Bently, *Reversibility of Sorption of Plutonium-239 onto Colloids of Hematite, Goethite, Smectite, and Silica.* LA–UR–98–3057, Los Alamos, New Mexico, Los Alamos National Laboratory, 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^-, Br^-, and I^- 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.

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, Livermore, California, Lawrence Livermore National Laboratory, 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.

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 E.C. Pearcy, *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., 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 J56, In press, 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., 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. Pearcy, 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.

Noy, D.J. et al., *Computer modeling of microbial processes using MGSE - an overview*, Technical Report WE/96/26, British Geological Survey, Keyworth, United Kingdom, 1996.

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*, 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.

Organ, R.M., *The current status of the treatment of corroded metal artifacts*, B.F. Brown (ed.), Corrosion and Metal Artifacts, NBS Special Publication 479, U.S. Government Printing Office, Washington, D.C., 1977.

Pabalan, R.T., and D.R. Turner, *Sorption modeling for HLW 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^{VI} 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 subtererranean bacteria in deep crystalline bedrock and their importance for the disposal of nuclear waste*, Canadian Journal of Microbiology, Vol. 42, Num. 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.

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, U.S. Geological Survey (USGS), Washington, D.C., USGS Professional Paper 300, 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*, U.S. Geological Survey, Washington, D.C., USGS Professional Paper 454-G, pp. G1–57, 1964.

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 Act, Vol. 58, pp. 4,537–4,547, 1994.

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.

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, Center for Nuclear Waste Regulatory Analyses, San Antonio, Texas, 1996.

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.

Santanan, L., S. Raghavan, and B.A. Chin, *Zircaloy cladding rupture during repository storage*, Nuclear Technology, Vol. 97, pp. 316–322, 1992.

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., et al., *The corrosion of nuclear fuel (UO_2) in oxygenated solutions*, Corrosion Science, Vol. 29, pp. 1,115–1,128, 1989.

Spinks, J.W.T., and R.J. Woods, *Introduction to Radiation Chemistry*, Wiley-Interscience, New York, New York, 1976.

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(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.

Steefel, 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.

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., et al., *Oxidation of UO_2 fuel by the products of gamma radiolysis of water*, Journal of Nuclear Materials, Vol. 190, pp. 78–86, 1992.

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. 1507–1529, 1998.

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.

"RW Environmental Safety Systems, Inc., *Status/Summary Report for Fiscal Year 1996 ,ctivities 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, 1996a.

TRW Environmental Safety Systems, Inc., *Mined Geological Disposal System Advanced Conceptual Design Report. Vol. I–II*, B00000000-01717-5705-00027, Rev. 00, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1996b.

TRW Environmental Safety Systems, Inc., *Total System Performance Assessment—Viability Assessment (TSPA-VA) Plan*, B00000000–01717–2200–00179, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1996c.

TRW Environmental Safety Systems, Inc., *Reference Design Description for a Geologic Repository*, B00000000-01717-5707-00002, Rev. 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*, B00000000-01717-2200-00193, Rev. 01, TRW Environmental Safety Systems, Inc., Las Vegas, Nevada, 1997b.

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.

"urgoose, S., *The nature of surviving iron objects. Conservation of Iron*, National Maritime ,luseum Monograph, No. 53, National Maritime Museum, Greenwich, United Kingdom, pp. 1–7, 1982.

Turgoose, S., *Structure, composition, and deterioration of unearthed iron objects*, Current Problems in the Conservation of Metal Antiquities, Tokyo National Research Institute of Cultural Properties, Tokyo, Japan, pp. 35–53, 1989.

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, *[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, 1996a.

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., 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 'Jaste Management, U.S. Department of Energy, Washington, D.C., 1997a.

U.S. Department of Energy (DOE), *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 1, 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. 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 1)*, 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: 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, 1998e.

U.S. Nuclear Regulatory Commission, *Issue Resolution Status Report (Key Technical Issue: Total System Performance Assessment and Integration)*, enclosure to letter dated May 11, 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, *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, 1998g.

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, 1998h.

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, *Submittal of U.S. Department of Energy Topical Report on Disposal Criticality Analysis Methodology,* letter dated February 11, 1999, from C.W. Reamer, U.S. Nuclear Regulatory Commission, to S.J. Brocoum, U.S. Department of Energy, 1999c.

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, VA, 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*, Paper 162, CORROSION98, 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.

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 glas 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, 1993.

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 STAFF'S TOTAL SYSTEM PERFORMANCE ASSESSMENT

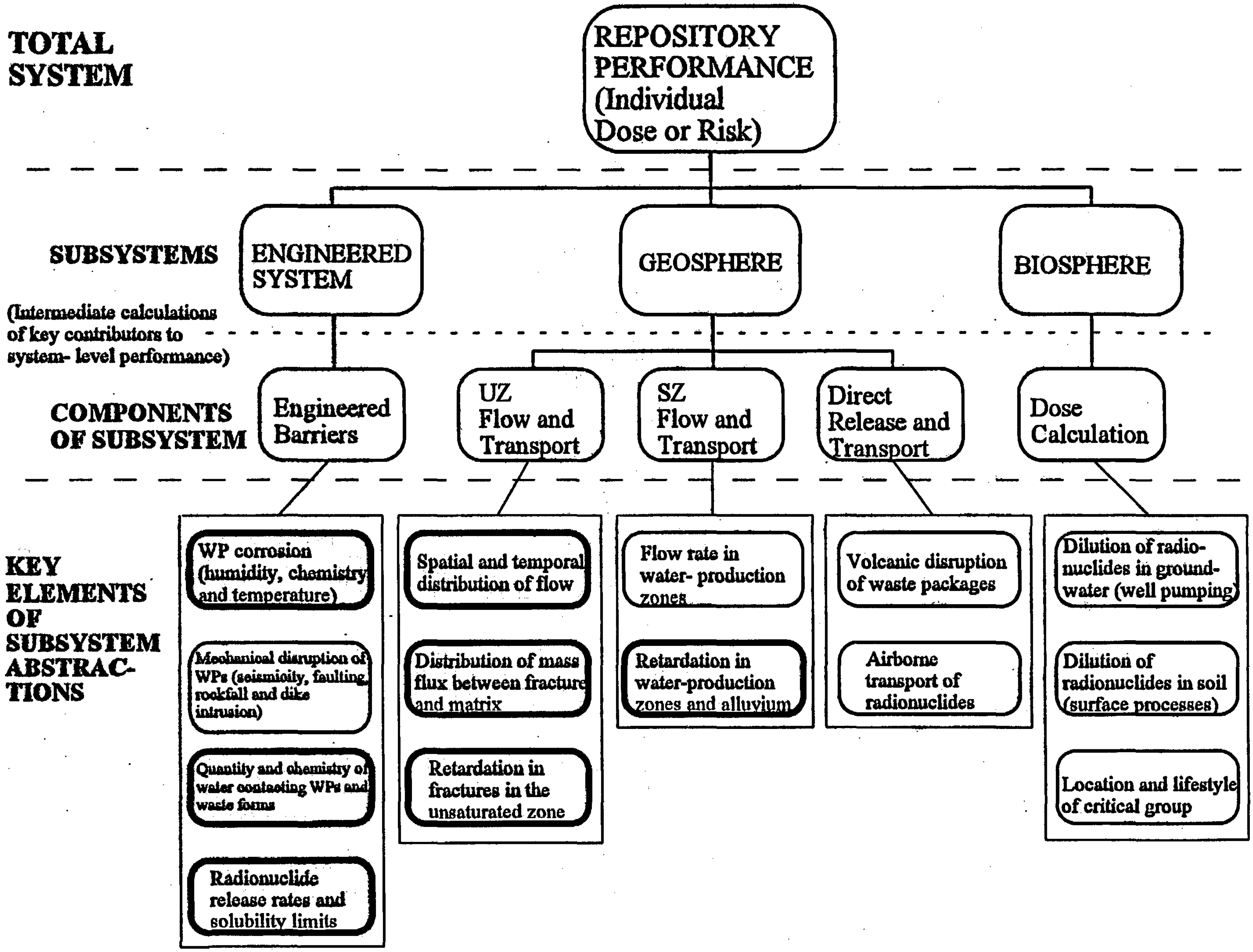

Figure A-1. Flowdown diag for total system performance assessment. This K Evolution of Near-Field Environment, provides input to the highlighted key elemen s.