



UNITED STATES  
NUCLEAR REGULATORY COMMISSION  
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August 28, 1998

Dr. Stephan J. Brocoum  
Assistant Manager for Licensing  
U.S. Department of Energy  
Office of Civilian Radioactive Waste Management  
Yucca Mountain Site Characterization Office  
P.O. Box 30307  
North Las Vegas, Nevada 89036-0307

SUBJECT: ISSUE RESOLUTION STATUS REPORT (KEY TECHNICAL ISSUE.  
EVOLUTION OF THE NEAR-FIELD ENVIRONMENT, REVISION 1)

Dear Dr. Brocoum:

As you know, the staff of the U.S. Nuclear Regulatory Commission (NRC) has developed a program for early resolution of technical issues at the staff level. The previous version of this Issue Resolution Status Report (IRSR) on the Key Technical Issue (KTI) of Evolution of the Near-Field Environment (ENFE) focused on defining those coupled thermal-hydrologic-chemical (THC) processes that will occur as the near-field environment evolves and affects a repository at Yucca Mountain (letter dated November 7, 1997, from N. K. Stablein to S. Brocoum). This revision assesses the potential importance of microbial processes on repository performance; evaluates the relevance of the large self potentials measured in the thermal tests on waste package performance; updates the discussion of the subissue on the chemical environment for the waste package to reflect the latest design of the waste package; and introduces a new subissue on criticality in the near-field environment. The KTI addressed by this IRSR is similar to, but broader than, the U.S. Department of Energy's (DOE's) treatment of the near-field geochemical environment in the Total System Performance Assessment-Viability Assessment (TSPA-VA). The recognition by both NRC and DOE of this issue's (evolution of the near-field environment) potential to affect repository performance indicates the importance of resolving it.

Consistent with NRC regulations on preclicensing consultations and a 1992 agreement with DOE, staff-level issue resolution can be achieved during the preclicensing consultation period; however, such resolution at the staff level would not preclude the issue being raised and considered during the licensing proceedings. Issue resolution at the staff level during preclicensing 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 the resolution at the staff level may be limited to documenting a common understanding regarding differences in NRC and DOE points of view. Further, pertinent additional information could raise new questions or comments regarding a previously resolved issue.

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Sections 4 and 5 of the enclosed IRSR summarize an independent pre-licensing review and analysis of coupled THC processes resulting from the evolution of the near-field environment and their impact on repository performance. While work is still ongoing to develop the technical basis for incorporation of the effects of coupled THC processes in a performance assessment, this IRSR provides the acceptance criteria that the staff will be using to evaluate this issue. DOE has indicated that self potentials and microbial activity may affect the performance of the repository. Based on its analysis to date, the staff concludes (1) that self potentials are unimportant to waste package performance, and (2) that the effects of microbial processes on seepage and flow need not be considered in performance assessments of the repository. In addition, preliminary analysis suggests that the effects of microbial processes on the chemical environment of the waste package, chemical environment for radioactive release, and radionuclide transport through the engineered and natural barriers may not need to be considered in performance assessments, provided that the mass-balance engineering-style calculations proposed in DOE's TSPA-VA *Methods and Assumptions* report are completed and results show that there is limited potential for microbial growth. Finally, the staff's comment on DOE's Thermohydrology Testing and Modeling Program, concerning whether their testing and modeling strategy included means for bounding the effects of thermal-hydrologic-chemical coupled processes, is resolved.

We would like to note that we have had very successful interactions with DOE project personnel on the thermal testing program and process-level modeling of coupled geochemical processes. However, there has been limited opportunity for exchange of information between DOE and NRC on the evaluation of the effects of coupled THC processes and the evolution of the near-field environment within a performance assessment framework. Previous DOE TSPAs have evaluated a limited range of effects on repository performance resulting from geochemical processes. DOE has indicated that an ambitious program will be undertaken to evaluate the effects of coupled THC processes on the near-field geochemical environment in the TSPA-VA. This information, along with the supporting data and analysis, needs to be evaluated in order to determine areas of potential agreement and disagreement. In the meantime, this IRSR will give DOE an understanding of NRC's approach to evaluating the effects of coupled THC processes on repository performance prior to the planned September Appendix 7 meeting on the potential effects of cementitious materials on repository performance. This IRSR should help facilitate the exchange of ideas between DOE and NRC, as well as provide DOE with an understanding of the criteria that NRC will be using to evaluate the information presented on this subject in DOE's VA. The planned Appendix 7 and review of the VA will allow NRC to more completely evaluate the status of resolution of the ENFE KI.

The enclosure should be viewed as a status report that provides the staff's most current views on issues related to the evolution of the near-field environment affecting the repository at Yucca Mountain. NRC plans to update this report in FY99 to reflect progress on all of the subissues. We welcome a dialogue on the potential effects of the evolution of the near-field environment on the repository with DOE, the U.S. Nuclear Waste Technical Review Board, State of Nevada,

S. Brocoum

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and other interested parties. If you have any questions about this letter, please contact Bret Leslie of my staff at (301) 415-8063, or via internet mail service (bwl@nrc.gov).

Sincerely,

Original Signed By

N. King Stablein, Acting Chief  
Engineering and Geosciences Branch  
Division of Waste Management  
Office of Nuclear Material Safety  
and Safeguards

Enclosure: As stated

cc: See attached list

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and other interested parties. If you have any questions about this letter, please contact Bret Leslie of my staff at (301) 415-8063, or via internet mail service (bwl@nrc.gov)

Sincerely,

A handwritten signature in black ink that reads "King Stablein". The signature is written in a cursive style with a large initial "K".

N King Stablein, Acting Chief  
Engineering and Geosciences Branch  
Division of Waste Management  
Office of Nuclear Material Safety  
and Safeguards

Enclosure As stated

cc See attached list

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**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 1**

**August 1998**

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

<u>Revision #</u>	<u>Section/ Paragraph</u>	<u>Date</u>	<u>Modification</u>
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 3.3.5 in Rev0

<u>Revision #</u>	<u>Section/ Paragraph</u>	<u>Date</u>	<u>Modification</u>
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	August 1998	Minor modification to reflect revised subissue definition

<u>Revision #</u>	<u>Section/ Paragraph</u>	<u>Date</u>	<u>Modification</u>
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
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

<u>Revision #</u>	<u>Section/ Paragraph</u>	<u>Date</u>	<u>Modification</u>
Rev1	4.4/1	August 1998	Minor modification to reflect revised subissue definition
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 open items' status and their description to reflect 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

<u>Revision #</u>	<u>Section/ Paragraph</u>	<u>Date</u>	<u>Modification</u>
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
Rev1	6.0/all	August 1998	Provide References for Rev1

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## ACKNOWLEDGMENTS

This report was prepared jointly by the U.S. Nuclear Regulatory Commission (NRC) and the Center for Nuclear Waste Regulatory Analyses (CNWRA) staffs. Primary authors of the report are Bret Leslie (NRC), William Murphy (CNWRA), and Roberto Pabalan (CNWRA). The authors thank John Bradbury (NRC), Richard Codell (NRC), Gustavo Cragnolino (CNWRA), Jennifer Davis (NRC), James Firth (NRC), Peter Lichtner (CNWRA), English Percy (CNWRA), David Pickett (CNWRA), Narasi Sridhar (CNWRA), and David Turner (CNWRA) for their assistance in technical discussions and efforts at integration between the other Key Technical Issue (KTI) teams. The review efforts of David Turner (CNWRA), Wes Patrick (CNWRA), David Brooks (NRC), and N. King Stablein (NRC) are appreciated by the authors. Preparation of this report would not have been possible without the assistance of Arturo Ramos (CNWRA) and Carrie Crawford (NRC).

## 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 CNWRA Quality Assurance Manual. Sources for other data should be consulted for determining the level of quality for those data.

**ANALYSES AND CODES:** The TPA code Version 3.1 was developed under TOP-018 procedures.

## **1.0 INTRODUCTION**

One of the primary objectives of the U.S. Nuclear Regulatory Commission (NRC) refocused precicensing program is to direct all its activities towards resolving the 10 key technical issues (KTIs) it considers to be most important to performance of the proposed high-level nuclear waste (HLW) repository at Yucca Mountain (YM). This approach is summarized in Chapter 1 of the fiscal year (FY) 1996 Annual Progress Report, NUREG/CR-6513 (Sagar, 1996). Other chapters address each of the 10 KTIs by describing the scope of the issue and subissues, path to resolution, and progress achieved during FY 1996.

Consistent with 10 CFR Part 60 requirements and a 1992 agreement with the U.S. Department of Energy (DOE), staff-level issue resolution can be achieved during the precicensing consultation period; however, such resolution at the staff level would not preclude the issue being raised and considered during the licensing proceedings. Issue resolution at the staff level during precicensing 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 NRC and DOE points of view. Furthermore, pertinent, additional information could raise new questions or comments regarding a previously resolved issue.

An important step in the staff approach to issue resolution is to provide DOE with feedback regarding issue resolution before the viability assessment. Issue Resolution Status Reports (IRSRs) are the primary mechanism that the staff will use to provide feedback to DOE regarding progress toward resolving the subissues comprising the KTIs. This report is the first revision of the IRSR on Evolution of the Near-Field Environment (ENFE). Revision 1 of this IRSR completely supersedes Revision 0, which becomes obsolete. IRSRs include: (i) acceptance criteria and review methods for use in issue resolution and regulatory review; (ii) technical bases for the acceptance criteria and review methods; and (iii) the status of resolution including where the staff currently has no comments or questions as well as where it does. Additional information is also contained in the staff Annual Progress Report that summarizes the significant technical work toward resolution of all KTIs during the preceding fiscal year. Finally, open meetings and technical exchanges with DOE provide opportunities to discuss issue resolution, identify areas of agreement and disagreement, and develop plans to resolve such disagreements.

In addition to providing feedback, the IRSRs will serve as guidance for the staff's review of information in the DOE viability assessment (VA). The staff also plans to use the IRSRs in the future to develop the Yucca Mountain Review Plan for the repository license application (LA), and subsequently to review the VA and LA.

Each IRSR contains six sections, including an Introduction in Section 1. Section 2 defines the KTI, the related subissues, and the scope of the particular subissue or subissues that are addressed in the IRSR. Section 3 discusses the importance of the subissues to repository performance, including: (i) qualitative descriptions; (ii) reference to a total system performance (TSP) flowdown diagram (U.S. Nuclear Regulatory Commission, 1998a); (iii) results of available sensitivity analyses, and (iv) relationship to the DOE Repository Safety Strategy (RSS), which

supersedes the DOE Waste Containment and Isolation Strategy (WCIS; U S Department of Energy, 1996a). Section 4 provides the staff review methods and acceptance criteria, which indicate the basis for resolution of the subissues and that will be used by the staff in subsequent reviews of DOE submittals. These acceptance criteria are guidance for the staff and indirect for DOE as well. The staff technical bases for the acceptance criteria are also included to further document the rationale for their decisions. Section 5 concludes the revision 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 the staff, and resolution will be documented in future IRSRs. Finally, Section 6 includes a list of pertinent references

## **2.0 KEY TECHNICAL ISSUE AND SUBISSUES**

Both NRC and DOE are evaluating the potential Yucca Mountain repository using a performance assessment (PA) approach. The NRC site-specific regulations for the proposed YM repository to be issued as 10 CFR Part 63, which are currently in preparation, will be performance-based (U.S. Nuclear Regulatory Commission, 1998b). The near-field environment at the proposed repository at YM 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, whereas other processes may have effects only close to or within the engineered barrier system (EBS). 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 and Repository Design and Thermal Mechanical Effects IRSRs, respectively.

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 PAs. 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, participation in meetings with DOE to discuss issues related to the KTI, observation of Quality Assurance (QA) audits of DOE, independent technical investigations, and performing sensitivity studies related to the effects of coupled TH processes on total system performance.

Four system attributes have been identified by DOE in their RSS (U.S. Department of Energy, 1998) 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 RSS (U.S. Department of Energy, 1998) 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, 1998).

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

- Coupled THC effects on seepage and flow
- WP chemical environment
- Chemical environment for radionuclide release
- Effects of THC processes on radionuclide transport (RT) 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 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 PA (TRW Environmental Safety Systems, Inc., 1996a), and continue to be studied as part of the 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 PA. The performance assessment should not provide overoptimistic estimates of performance. The adequacy of DOE's treatment of coupled processes in the PA 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 addresses all the subissues and provides acceptance criteria that 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. The staff 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 created 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 PA 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 RSS (U.S. Department of Energy, 1998), and this relationship 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. Consideration of the effects of coupled processes in the near-field environment evaluated in previous PAs is addressed 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 (SCP) (U.S. Department of Energy, 1988). DOE updated that strategy in the RSS as a result of additional site characterization data, advances in the engineered system design, and a changing regulatory framework (U.S. Department of Energy, 1998). The RSS 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, 1998). The updated strategy continues to rely on engineered and natural barriers to contain and isolate the waste from the public. For 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 RSS (U.S. Department of Energy, 1998).

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 TSP, and they 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 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. Although the RSS (U.S. Department of Energy, 1998) notes that environmental characteristics of the WP affect its corrosion rate, 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 (SF); (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 soluble radionuclides is controlled by solubility rather than colloid stability. Finally, for the fourth system attribute, concentration reduction during RT through engineered and natural barriers, two testable hypotheses are also addressed in this IRSR. The DOE hypotheses affected by the ENFE are that physical properties of engineered and natural barriers reduce concentrations during transport, and that chemical properties of engineered and natural barriers reduce concentrations during transport. Although unspecified among DOE hypotheses, criticality is noted in the RSS 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 RT away from the drifts. DOE will need to demonstrate adequately and to quantify the consequences of coupled processes, resulting from evolution of the near-field chemical environment, that affect repository performance as determined by its TSPA. 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.

NRC staff has developed a strategy for assessing the performance of the potential HLW repository at YM (U.S. Nuclear Regulatory Commission, 1998a). The TSPA Methodology IRSF 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 KESAs. Therefore, the approach that the staff will use to independently evaluate the DOE TSPA will focus on 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 (SZ). 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, 1998a). 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), Radionuclide Transport (RT), Thermal Effects on Flow (TEF)). 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

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

The effects of coupled THC processes on seepage into the drift and flow in the unsaturated zone (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 EBS. 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 by the loss of host rock volume associated with the dehydration process, creating new fractures and widening existing fractures, thereby, leading to possible increases in fracture flow.

The second 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. Because small changes in porosity can produce orders of magnitude changes in permeability (Lichtner and Walton, 1994), 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 performance of the repository is likely to be spatially limited to near the drifts of the repository. The reference 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, 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<sub>2</sub>. The carbonation of the concrete will affect mechanical properties (Parrott, 1994) of the ground control system. In addition, the concrete drift liners will have mechanical stability that 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). 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, 1998a), the effects of coupled THC processes on seepage and flow will need to be considered.

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

The effect of coupled THC processes on 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 EBS, 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 PA framework, which is primarily addressed in the CLST IRSR, 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<sup>-</sup>, 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 spatial and temporal distribution of WPs that could be 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. The effects of coupled processes on the KESA necessary to describe radionuclide release are presented below

Prior to water contacting the SF, degradation of the cladding must occur. If credit for cladding were to be taken in PA, the effects of coupled THC processes on the chemical environment of cladding would need to be considered. Both the chloride concentration and the redox potential at the cladding surface control whether localized corrosion of fuel cladding by pitting will occur (Cragolino and Galvele, 1978; Maguire, 1984). Under the environmental and potential conditions leading to pitting, stress corrosion cracking (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 as a result of the greater stability of the ZrF<sub>6</sub><sup>2-</sup> complexes compared to that of the passive ZrO<sub>2</sub> film.

Corrosion of the SF (predominantly UO<sub>2</sub>) pellets by contact with the groundwater, as modified by chemical and physical interactions in the near field, is the most important process affecting the long-term performance of this waste form. Several environmental factors are known to affect the dissolution rate of UO<sub>2</sub> in aqueous environments. The nature and concentrations of

the anionic species present in the groundwater are extremely important in determining the rate of corrosion of SF. Dissolved  $\text{CO}_3^{2-}$ ,  $\text{F}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ , and  $\text{Cl}^-$  can accelerate the corrosion rate (Blesa, Morando, and Regazzoni, 1994; Grambow, 1989). A major near-field environment factor affecting  $\text{UO}_2$  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 EBS.

The effect of pH on the rate of dissolution of SF 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 SF will be influenced by hydrolysis of highly charged cations such as  $\text{Cr}^{3+}$  from the degradation of the WP, air mass fraction in the WP, reaction with metallic components of the EBS, and radiolysis.

In addition, the modification of the pH of the leachate attributed to the formation of  $\text{HNO}_3$  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, SF will eventually oxidize, forming a large quantity of uranyl ( $\text{UO}_2^{2+}$ )-bearing solids. Other species, such as  $\text{SiO}_2(\text{aq})$ ,  $\text{H}_3\text{SiO}_4$ ,  $\text{H}_2\text{SiO}_4^{2-}$ , and product 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 SF. 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 SF 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 including: (i) physical disruption of structural components (e.g., cladding or degraded containers), due to the large volume increase accompanying oxidation and hydration of  $\text{UO}_2$ ; (ii) plugging porosity and reducing permeability because of volume expansion; (iii) incorporation by coprecipitation of Np, Pu, and other radioactive waste species, that will exist as trace components in the altered system relative to U, iron, and possibly other components from the engineered and geologic system, such as Ni, Al, and  $\text{SiO}_2$ ; (iv) limiting ingress of water and oxidants to unaltered wastes; and (v) controlling by solubility or dissolution rate, 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 SF with the near-field environment, rather than unaltered SF, 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,  $\text{F}^-$ , and  $\text{Fe}^{2+}$ . Ultimate glass waste form alteration products are likely to be clay or zeolite minerals, 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 EBS, such as Fe and Ca. Clay minerals generally have low solubilities

Some quantity of radioactive waste species are likely to be incorporated through solid solution in mineral alteration products of glass waste forms. Contribution of the 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 SF.

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 EBS 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 RT 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 RT subissue are: (i) fracture versus matrix flow; (ii) retardation in fractures; and (iii) retardation in the SZ. 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.

A major concern for PA in the near field is the transport of radionuclides through the EBS and the geologic setting as gaseous species, as species in colloidal form, or dissolved in aqueous solution. Each RT process is influenced by several geochemical parameters; thus, an assessment of the relative importance of each process 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 EBS materials and natural system minerals. Coupled THC processes will cause changes in system chemistry parameters, such as Eh, pH, and component concentrations that will influence the solubilities of radionuclide-bearing minerals.

Another retardation mechanism, which is strongly controlled by geochemical parameters, such as solution pH, is sorption. For example, sorption of actinide species, such as  $\text{UO}_2^{2+}$ ,  $\text{NpO}_2^+$ , and  $\text{Am}^{3+}$ , on oxides and oxyhydroxides through surface complexation mechanisms is characterized by a sharp sorption "edge," where sorption increases sharply with increasing pH from essentially zero over a relatively narrow, low pH range (Sagar, 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  $\text{Cs}^+$ ,  $\text{Sr}^{2+}$ , and  $\text{UO}_2^{2+}$ , can also occur through an ion exchange mechanism, which depends on the nature and concentration of competing cations present in solution (Sagar, 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 ( $\text{TcO}_4^-$ ) and does not sorb strongly, whereas, under reducing conditions,  $\text{Tc}^{4+}$  is predominant and sorbs more strongly (Lieser and Bauscher, 1988). Elevated temperatures expected for the near-field environment may also affect sorption, but there are few data for evaluating the magnitude of the effect. In the near field, boiling in response to thermal loading would tend to partition  $^{14}\text{CO}_2$  into the gas phase, enhancing gas transport. Local fluctuations of reducing and oxidizing conditions in the near field due to an unstable hydrologic regime could also induce secondary chemical effects, such as the formation of colloids (Buddemeier and Hunt, 1988; McCarthy and Zachara, 1989). Thus, the redox conditions, which also affect sorption behavior, will be influenced by coupled THC processes.

The potential importance of colloids in the transport of radionuclides is also of concern in PA in the near field. 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 RT 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 RT.

### **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 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-SiO<sub>2</sub>-H<sub>2</sub>O and Pu-tuff-H<sub>2</sub>O, but concluded that the probability of forming such conditions is extremely low. Choi and Pigford (1997) concluded from their technical analysis of the potential for autocatalytic criticality that, based on simplified geometries, there is a theoretical potential

for appreciable energy release from autocatalytic (or self-enhancing) criticality, but that additional analysis is required to determine the extent and consequences of such an event. Potential effects on repository performance of criticality in the near field include an increase in the fission product inventory, a decrease in the fissile radionuclide inventory, and an increase in thermal output.

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

Some limited consideration of the effects of coupled processes on the performance of the proposed YM repository have been included in past PA studies. 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 CNWRA staffs using the TPA code (Manteufel, et al., 1997). In addition, 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 TSPA-93 described as part of its source-term model 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, SZ 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 considered in TSPA-93.

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  $\text{RaSO}_4$  and cassiterite ( $\text{SnO}_2$ ) for a range of possible water chemistries and temperatures were near  $10^{-6}$  M. 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  $\text{RaSO}_4$  or  $\text{SnO}_2$  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 UZ. 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 Jarzempa, 1997).

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

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

The DOE Methods and Assumptions (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,  $p\text{CO}_2$ ,  $\text{CO}_3^{2-}$ , Cl<sup>-</sup>, F<sup>-</sup>, Si,  $\text{PO}_4^{3-}$ , and  $\text{SO}_4^{2-}$ . Section 6.3 of TRW Environmental Safety Systems, Inc., (1997b) provides a fairly comprehensive description of coupled 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 variation: 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/CNWRA 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 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, as 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 RT 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 inert materials). The aqueous and colloidal RT through the EBS components and radionuclide sorption along the EBS transport pathways as a function of pH and temperature will be abstracted. The output from the EBS transport model will be a release of radionuclides from the EBS 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 to assess the performance of the individual components that contribute to the performance of the repository system (Kessler and McGuire, 1996). This

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 (performed by setting distribution coefficients,  $K_d$ s, for all radionuclides to zero for distances of 10 and 100 m into the UZ, and for the entire UZ), and another that 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 evaluated at a fluid pH of 11. The results of these simulations were compared to the TSPA-95 base case results for a single realization, expected-value calculated dose at the accessible environment at  $10^4$ ,  $10^5$ , and  $10^6$  yr, and 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.

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, but 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<sup>4</sup> and 10<sup>6</sup> 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<sup>6</sup> 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<sub>2</sub> gas evolution and transport, and calcite precipitation in a transient thermal field. The model was used to explore possible mechanisms for <sup>14</sup>C retardation and gas phase release. An initial pulse of <sup>14</sup>CO<sub>2</sub> was modeled to exit the mountain after less than 2,000 years of transport. Subsequently, <sup>14</sup>C releases to the atmosphere were small and some <sup>14</sup>C remained trapped in precipitated calcite. However, these results were not incorporated in analyses of CCDFs.

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

In sensitivity analyses conducted by NRC and the CNWRA, 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 efforts. The results of the preliminary efforts are summarized in the following sections.

### **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 (CSH) phases along fractures and reduce fracture porosity and hydraulic conductivity. In addition, mineral precipitation could seal fracture surfaces and enhance RT 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  $K_d$ s of radionuclides, particularly actinides, in cementitious environments are high (Bradbury and Sarott, 1995; Campbell and Krupka, 1997). For example, a comparison of  $K_d$ s of different elements specified in the TPA code input file with  $K_d$ s of those elements in cement environments taken from Bradbury and Sarott (1995) indicate that, except for Tc, Ra, Cs, and Se, radionuclide  $K_d$ s 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 its value 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**

Results to be determined in FY99 to more thoroughly analyze potential sensitivity.

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 UZ is less than a specified amount, then the travel time for RT in the UZ is approximated to be zero. In that case, performance is independent of the flow and transport properties of the UZ below the emplacement horizon. The parameters used in the calculations resulted in short travel time in the UZ. Thus, the sensitivity of repository performance to the parameters of interest was not, in fact, thoroughly tested in the calculation. Revisions to the TPA code are planned that will include providing for 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.

#### **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 RT. If the flow is predominantly through the matrix, it is likely that iron oxyhydroxides will be effectively filtered from suspension and will act as an additional barrier to transport. 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 RT were studied by using radionuclide distribution coefficients,  $K_d$ 's, applicable to the cases. As an

approximation of the spatial extent of iron oxyhydroxide transport, and to be consistent with the physical situation considered in the cement sensitivity analyses, the zone of transport was assumed to extend vertically downward from the emplacement horizon to the bottom of the Topopah Spring Tuff. Because of large uncertainties in estimating the amount of colloid-size iron oxyhydroxide generated by 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 ( $0.9 < R_d < 1.0$ ). 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 (Percy, 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  $10^n$  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**

Results to be determined in FY99 to more thoroughly analyze potential sensitivity.

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 the parameters of interest was not, in fact, thoroughly tested in the calculations. Revisions to the TPA code are planned that will include providing for 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.

### **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 ( $E_r$ ), 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,  $E_r$  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<sub>2</sub> 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 provide quantitative concentrations of dissolved aqueous and gaseous species in the matrix pore space, in tabular form, to the TPA 3.1 code (Lichtner, 1997).

To address uncertainties in the MULTIFLO calculations, 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 results of MULTIFLO are multiplied by a chloride multiplication factor. 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  $2.1 \times 10^{-5}$  atm, reflecting carefully deaerated 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 EBS 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 EBS release was found to be highly sensitive to chloride multiplication factor within a very narrow range of variation, exhibiting a step function response near value of 10.0, where cumulative releases increase by approximately one order of magnitude. A similar step function is also apparent for oxygen partial pressure, with higher releases occurring for partial pressures greater than about  $3.0 \times 10^{-2}$  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 EBS release 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.

### **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 SF) have been calculated (Murphy and Percy, 1992; Murphy, Percy, and Pickett, 1997). The rate is a maximum because a maximum limit was used for the infiltration that removed 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 SF 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 SF, the oxidation rate was assumed to limit their release rate.

Murphy, Percy, and Pickett, (1997) calculated a maximum bounding oxidation rate (i.e., the maximum average oxidation rate) for uraninite at Peña Blanca based on the approach of Murphy and Percy (1992) and a revised minimum time for oxidation based on radiometric ages of late forming uranophane (an oxidized 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 Percy, 1992)

$$R_o = (U_o/t) + FC \quad (1)$$

where  $R_o$  stands for the oxidation rate (e.g., grams year<sup>-1</sup>),  $U_o$  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<sup>-1</sup>), 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<sup>-1</sup> 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 SF oxidizes at a comparable rate as that at Peña Blanca, the total rate is given by  $172 \times 140 \text{ g yr}^{-1} \text{ (kg/1,000 g)} = 24 \text{ kg yr}^{-1}$ . 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  $\beta$  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 Percy (1992) adopted a U solubility of  $10^{-7}$  mole liter<sup>-1</sup>. 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<sup>-1</sup>) 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<sup>-1</sup> corresponds to UserLeachRate[kg/yr/m<sup>2</sup>] = 5 × 10<sup>-6</sup> kg yr<sup>-1</sup> m<sup>-2</sup>. The release rate for each individual radionuclide is generally given by the U oxidation rate times its fractional inventory of the radionuclide in SF.

#### **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.

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

This IRSR and its acceptance criteria focus on providing a path to resolution and will be revised to become more specific as PA 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, 1998b) are finalized. A systematic approach to resolving subissues is necessary because, in the past, there has been little concerted effort to evaluate, within a PA 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 PA 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 PA.

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, 1998a; 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, 1998a).

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 UZ 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 PA 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, 1998a). 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 *overoptimistic, 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.

This group of acceptance criteria addresses data and model justification.

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

- 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 EBS, 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.

This group of acceptance criteria addresses data uncertainty and verification.

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

- 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.

This group of acceptance criteria addresses model uncertainty.

- 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 CSH phases along fracture surfaces as a result of migration of a hyperalkaline fluid that could affect hydraulic properties.
- Given the current design of the repository, it will be acceptable to ignore the potential effects of microbial processes on seepage and flow
- Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered
- 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 EBS, 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.**

**This group of acceptance criteria addresses model verification of abstractions.**

- **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.**
- **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.**
- **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 EBS, 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 nondefensible predictions

This group of acceptance criteria addresses integration.

- 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.
- Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on seepage and flow.
- 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.
- 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/data abstractions, and track the progress made in resolving deficiencies/non-conformities 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.

These programmatic acceptance criteria address quality assurance (QA) issues

- Data and models were collected, developed, and documented under acceptable quality assurance (QA) procedures.
- Deficiency reports concerning data quality on issues related to coupled THC effects on seepage and flow were closed

**Review Method:**

- If DOE uses this approach, NRC will attend, as observers, the elicitation workshops and review the documentation to ensure that the expert elicitations

followed the guidance in NUREG-1563 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

This programmatic acceptance criterion addresses expert elicitations.

- 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  $\text{SiO}_2$ ; (Byers, 1985)] These rocks are variably vitric, devitrified to an assemblage of silica minerals and alkali feldspar, or 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 SZ groundwater is a dilute, oxidizing, sodium bicarbonate solution rich in dissolved silica. The 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, 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 units at YM, primary glass has been altered to analcime  $\pm$  kaolinite rather than clinoptilolite. This mineralogic change, which is generally associated with the disappearance of cristobalite with depth, is consistent with a decrease in the activity of aqueous silica (Kerrisk, 1983). Fracture and cavity mineralogy is characterized by calcite, smectite, silica minerals, zeolites, and manganese oxides (Carlos, 1989; Carlos, Bish, and Chipera, 1991; Paces, et al., 1996).

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

Under elevated temperatures, the rates of alkali feldspar dissolution and growth of secondary phases, such as smectite, clinoptilolite, silica minerals, and calcite, would be accelerated. Thermodynamic analyses for smectites (Ransom and Helgeson, 1994) and clinoptilolites (Bowers and Burns, 1990) have quantified their decreasing stability with increasing temperature and decreasing aqueous silica content. In addition, the swelling capacity of 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 end 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 (Knauss, et al., 1984; Knauss, Beiriger, and Peifer, 1987; Knauss, 1987) show evidence for dissolution of primary minerals, precipitation of secondary phases, and variations in water chemistry. Secondary precipitation of clay minerals, zeolites, cristobalite, and calcite have been observed. Experiments in which CO<sub>2</sub> loss occurred showed more extensive secondary mineralization and particularly more calcite precipitation than in pressurized closed-system experiments. Water chemistry variations were generally small, and achieved approximately steady-state conditions in long-term experiments. The aqueous silica concentration was observed to increase substantially at elevated temperatures corresponding to the increased

solubility of silica minerals. The water chemistry in selected experiments was reasonably represented as a function of time with partial equilibrium and kinetic reaction path models of the water-rock interactions (Delany, 1985). Application of the results of these experiments to the near-field environment at 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<sub>2</sub> pressure have been shown to be particularly important in defining the solid-phase assemblage and the aqueous-solution composition. At present, these models are limited principally by the lack of thermodynamic and kinetic data. However, there have been several recent attempts to obtain such data by experimental and estimation techniques (e.g., Bowers and Burns, 1990; Johnson, et al., 1991; Ransom and Helgeson, 1994; Murphy, et al., 1996; Chipera and Bish, 1997) which, in some cases, appear to yield inconsistent results. Calculations of time-dependent processes are further hampered by the difficulty in realistically characterizing reactive surfaces in geologic environments.

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

Gas flow from the near field, driven by vaporization of water, is predicted to be away from the near field in all directions (Pruess, Wang, and Tsang, 1990; Tsang and Pruess, 1987), and would tend to purge air containing O<sub>2</sub> from the near-field environment. Because the vapor pressure of water at temperatures above 95 °C exceeds the hydrostatic pressure of less than 0.1 MPa at YM, the gas phase in the near field would tend to be dominated by H<sub>2</sub>O. Given the temperature-dependent solubility of different minerals, it is possible that solutions (both liquid and gas phase) moving by thermally driven convection will redistribute chemical components such as pH, chloride, oxygen, CO<sub>2</sub>, silica, and calcium.

Silica redistribution in the 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  $1.3 \times 10^{-14} \text{ m}^2$  to about  $10^{-17} \text{ m}^2$  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). Because small changes in porosity can effect orders of magnitude changes in permeability (Lichtner and Walton, 1994), 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 attempting 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. Because 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  $\text{CO}_2$  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. The staff 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 this FY.

Interaction of cement with the tuffaceous host rock and ambient groundwater could have an important effect on seepage. The chemistry of pore fluids in contact with hydrated cement phases is characterized by persistent alkaline pH (>10). Hyperalkaline cement pore water is thermodynamically incompatible with silica, a major component of the proposed YM repository host rock unit. Thus, migration of the high-pH cement pore water into the host rock is likely to result in strong alteration of the tuff. The Maqarin natural 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<sub>2</sub>-high Ca cement pore fluid mixes with the ambient groundwater containing high CO<sub>2</sub> concentrations (Lichtner and Eikenberg, 1995; Steefel and Lichtner, 1994)

The results of Lichtner, et al., (1998) indicate that calcification of the cement could occur as would be expected under ambient conditions. In the case of counter-diffusive transport, calcification is more pronounced in the case of a partially saturated system compared to a fully saturated one. Both the large block test and drift scale test thermal tests could provide important insights that could constrain the potential effects on seepage resulting from cementitious materials interacting with the tuff. 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). Processes that subsurface bacteria may mediate and that may impact a potential repository's performance include canister corrosion; leaching of the glass waste form; supply of organic by-products as potential complexants; direct uptake of radionuclides and either immobilization or mobilization depending on whether the microorganism is attached to an immobile surface; alteration of pore-water and gas chemistry; and production of 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, 1995). Extensive research and modeling programs are ongoing in other countries (Christofi and Philp, 1997), particularly in Switzerland (Brown and Sherrif, 1998), Canada (Stroes-Gascoyne, 1996), United Kingdom (West, 1995), and Sweden (Pedersen, 1996; Pedersen and Karlsson, 1995). While more work has been focused on low-level and intermediate-level waste repositories; as the quantity of organic carbon is greater than that expected for 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). It was concluded that 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 RT at YM were summarized and evaluated (Hersman, 1996). One conclusion from this study was that the elevated subsurface concentrations of CO<sub>2</sub>, relative to the atmospheric concentration, is the result of biogenic activity. Finally, an initial attempt to model the potential effects on repository performance of microbial processes in the near field 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<sub>2</sub>) 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<sup>-1</sup> 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<sup>-1</sup>, 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<sub>2</sub> 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 SZ, the US site is located in the oxidizing 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 Karland, 1985).

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 stimulation of microbial activity. Addition of organic carbon to the crushed tuff samples stimulated growth more than just added water. Thus, there is a potential for increased microbial activity at 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  $\text{CO}_2$  equilibrium with pore fluids in the UZ. Volatilization of  $\text{CO}_2$  as water evaporates is expected to increase gas phase  $\text{CO}_2$  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). Although ambient gas chemistry appears to be controlled by near surface microbial processes (Thorstenson, et al., 1998), the  $\text{CO}_2$  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.

Having already stated that based on the groundwater studies, organic C is expected to be limiting for microbial growth and activity; 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 (CRW 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 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 OF THE WASTE PACKAGE CHEMICAL ENVIRONMENT**

DOE identified long WP lifetime as a key factor in its RSS (U.S. Department of Energy, 1998). WP lifetime is embodied in a PA 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, depends strongly on the near-field chemical environment. As discussed in Section 4.1, the ENFE will help to determine the quantity and chemistry of water contacting WPs. Contributions from the CLST KTI, RDTME KTI, USFIC, 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 overoptimistic, 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.

This group of acceptance criteria addresses data and model justification.

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

- 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.
- Should microbial activity be sufficient to evaluate 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).
- 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.
- 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; 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.

This group of acceptance criteria addresses data uncertainty and verification.

- 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

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

**This group of acceptance criteria addresses model uncertainty.**

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

- **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.**

**This group of acceptance criteria addresses model verification of abstractions.**

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

This group of acceptance criteria addresses integration

- 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.
- Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on WP chemical environment.
- 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.
- Where simplifications for modeling coupled THC effects on WP chemical environment 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/data abstractions, and track the progress made in resolving deficiencies/nonconformities 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, 19'8a,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.

These programmatic acceptance criteria address QA issues.

- Data and models were collected, developed, and documented under acceptable QA procedures.
- Deficiency reports concerning data quality on issues related to coupled THC effects on WP chemical environment were closed

### Review Method:

- If DOE uses this approach, NRC will attend, as observers, the elicitation workshops and review the documentation to insure that the expert elicitations followed the guidance in NUREG-1563 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.

This programmatic criterion addresses expert elicitations.

- 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 PAs 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**

It has been shown that 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. 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 single heater tests 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 discussed 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. 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. 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), and would tend to purge air containing  $O_2$  and  $CO_2$  from the near-field environment. Because the vapor pressure of water at temperatures above 95 °C exceeds the hydrostatic pressure of less than 0.1 MPa at YM, 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: (i) electrochemical reduction of Fe is irreversible (far from equilibrium) and is diffusion-limited in the aqueous phase, (ii) 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  $O_2$ ; and (iii) the

initial formation of  $\alpha$ -FeOOH can lead to a secondary reduction reaction to  $\text{Fe}_3\text{O}_4$ , which is - argued to be the case for alternating wet and dry environments (Nishikata, et al., 1994). During the dry period,  $\text{Fe}^{2+}$  oxides or oxyhydroxides are oxidized by air to  $\alpha$ -FeOOH, and the cyclic process proceeds due to the electronic conductivity of the inner layer of  $\text{Fe}_3\text{O}_4$ .

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

As discussed in the CLST IRSR, 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,  $\text{NO}_3^-$ , and  $\text{HCO}_3^-/\text{CO}_3^{2-}$ . 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  $\gamma\text{-Fe}_2\text{O}_3$ , on carbon steel. This passive film slows down uniform corrosion rates by several orders of magnitude, but makes the metal more prone to the localized breakdown of passivity leading to the occurrence of pitting or crevice corrosion. Should the pH of the water contacting containers be neutral (pH = 7) or acidic, then active uniform corrosion will be important to WP lifetime calculations. SCC can occur in a  $\text{HCO}_3^-/\text{CO}_3^{2-}$  environment at a pH of about 8, when the corrosion potential (related to the Eh of the environment) reaches a critical value.

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  $\text{Fe}^{2+}$  ions; (ii) reduction of oxygen that diffuses from the outside; (iii) reduction of water or  $\text{H}^+$  inside the crevice created by the crack or localized corrosion front; (iv) the hydrolysis of  $\text{Fe}^{2+}$ ; and (v) the electromigration of anionic species, such as Cl and  $\text{SO}_4^{2-}$ , 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  $\text{Fe}^{2+}$  and  $\text{H}_2\text{O}$  or  $\text{H}^+$  reduction, the pH inside the crevice can be mildly acidic or alkaline (Sridhar, Lichtner, and Dunn, 1996). If  $\text{Fe}^{2+}$  is oxidized to  $\text{Fe}^{3+}$  by  $\text{O}_2$  dissolved in water, lower pH conditions could be generated locally due to  $\text{Fe}^{3+}$  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 ( $\beta$ -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  $\text{Cr}^{3+}$ . 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**

Staff 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  $\text{HCO}_3^-/\text{CO}_3^{2-}$  concentrations low. A study by Atkinson, Eventt, 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 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 MIC of the WP has been one of the primary concerns (Pedersen and Karlsson, 1995; Stroes-Gascoyne, 1996; Christofi and Philp, 1997). One of the few experiments 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), it was 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), and this topic is addressed in the CLST IRSR.

Horn and Meike (1995) concluded that microbial activity could potentially compromise the integrity of WPs at YM. Subsequent to that study, research has focussed 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 address 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 and 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 due to the increased nutrient loading associated with the backfill (Stroes-Gascoyne and Gascoyne, 1998; Wilder, 1996). In addition if spills of organic fluids are not minimized, then the potential for microbial activity would increase as the microbial activity in the tuff system has been demonstrated to be water and organic carbon limited (Kieft, et al., 1997). 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, the potential for microbial growth, and the potential for MIC (CRWMS M&O, 1997b). A complete 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), and an evaluation of energy-producing reactions 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) and, thus, not all the materials can 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). It is important to note that for the three countries (Switzerland, Canada, and United Kingdom) that have applied this extremely conservative (West, 1995; McKinley, et al., 1997) approach to their HLW repositories, they have 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).

Only after the potential for microbial activity has been modeled, and if the results indicate that microbial activity was likely for YM, would the potential for MIC need to be assessed as was done 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

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

DOE has identified slow rate 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, 1998). Radionuclide release rates and solubility limits constitute one of the NRC KESAs for a TSPA. Radionuclide release from waste forms and from the EBS 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 KESAs (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.

##### 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 overoptimistic, 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.

This group of acceptance criteria addresses data and model justification.

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

- 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.
- 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.
- 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 microbially enhanced dissolution of the HLW glass form (Staudigel, et al., 1995)
- 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.
- If the testing program for coupled THC processes on the chemical environment for radionuclide release from the EBS 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; EBS 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.

This group of acceptance criteria addresses data uncertainty and verification.

- 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.
- 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.
- 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
- 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.
- 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 from the EBS.

#### Review Method:

- 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.

This group of acceptance criteria addresses model uncertainty.

- 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.

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

This group of acceptance criteria addresses model verification of abstractions.

- 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
- 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.
- 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 SF, 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 nondefensible predictions.**

**This group of acceptance criteria addresses integration.**

- **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.**
- **Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release.**
- **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.**
- **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/data abstractions and track the progress made in resolving deficiencies/nonconformities 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.**

**These programmatic acceptance criteria address QA issues.**

- **Data and models were collected, developed, and documented under acceptable QA procedures.**
- **Deficiency reports concerning data quality on issues related to coupled THC effects on the chemical environment for radionuclide release were closed.**

**Review Method:**

- **If DOE uses this approach, NRC will attend, as observers, the elicitation workshops and review the documentation to insure that the expert elicitations followed the guidance in NUREG-1563 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.**

**This programmatic criterion addresses expert elicitations.**

- **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 EBS. As mentioned previously in Section 3, limited analyses in past PAs 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 as EQ3 (Wolery, 1992) or others, that the solubility range chosen for its reference case PA 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<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and HCO<sub>3</sub><sup>-</sup>, and F<sup>-</sup>, which are governed by near-field THC processes, affect degradation modes of 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<sub>2-x</sub>) 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 (Santanon, Raghavan, and Chin, 1992). Above a certain critical potential, Zircaloy is susceptible to pitting corrosion in chloride-containing environments (Cragolino and Galvele, 1978). Such a potential can be attained naturally under slightly oxidizing conditions (i.e., in the presence of Fe<sup>3+</sup>). 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 as a result of the greater stability of the ZrF<sub>6</sub><sup>2-</sup> complexes compared to that of the passive ZrO<sub>2</sub> film.

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

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, whereas at pH values between 4 and 8, the rate 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<sub>2</sub>O system in the presence of certain anions (Paquette and Lemire, 1981) over which specific domains for the dominant degradation modes can be superimposed. Temperature increases the rate of dissolution of UO<sub>2</sub>, 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<sub>3</sub><sup>2-</sup>, that form stable soluble complexes with U<sup>6+</sup> cations, substantially increase the rate of oxidative dissolution (Blesa, Morando, and Regazzoni, 1994). At low CO<sub>3</sub><sup>2-</sup> concentrations (0.001 M), the rate of dissolution is proportional to the total concentration because the rate-determining step is the surface complexation of CO<sub>3</sub><sup>2-</sup> (Blesa, Morando, and Regazzoni, 1994). At intermediate concentrations (0.5 M), the rate depends on the square root of the total concentration because solution transport of CO<sub>3</sub><sup>2-</sup> to the surface is rate controlling, or dissolution of an initially-formed UO<sub>2</sub>CO<sub>3</sub> film controls the overall rate (Grambow, 1989). At a higher CO<sub>3</sub><sup>2-</sup> concentration (1.0 M at 100 °C), the corrosion rate reaches a constant value, but at even higher concentrations, the 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<sub>4</sub><sup>3-</sup> < SO<sub>4</sub><sup>2-</sup> < F < CO<sub>3</sub><sup>2-</sup>, although in the case of PO<sub>4</sub><sup>3-</sup> and SO<sub>4</sub><sup>2-</sup>, a maximum in the corrosion rate is observed at intermediate concentrations (about 1.5 × 10<sup>-2</sup> M) (Blesa, Morando and Regazzoni, 1994).

Other species, such as SiO<sub>2</sub>(aq), H<sub>3</sub>SiO<sub>4</sub><sup>-</sup>, and H<sub>2</sub>SiO<sub>4</sub><sup>2-</sup>, can react with U(VI) to precipitate complex uranyl silicates, which 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, accelerating the precipitation of secondary phases and, eventually, the preferential release of 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) with the exception of Np. Wilson (1990) used semi-static experiments (i.e., involving periodic removal of a leachant aliquot and replacement with fresh solution) and observed that actinide (U, Pu, Am, Cm, and Np) concentrations reached constant values rapidly, suggesting that steady-state conditions between SF dissolution and secondary-phase formation are established. Formation of U<sup>6+</sup> secondary phases, such as uranophane, was confirmed. Actinide concentrations, with the exception of Np, measured at 85 °C were lower than at 25 °C, suggesting 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, and 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, leading 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 rates (Bates, et al., 1995), but 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 UZ 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 including: (i) physical disruption of structural components (e.g., cladding or degraded containers), due to the large volume increase accompanying oxidation and hydration of  $UO_2$ ; (ii) plugging porosity and reducing permeability because of volume expansion; (iii) incorporation by coprecipitation of Np (e.g., Buck, et al., 1998), Pu and other radioactive waste species that will exist as trace components in the altered system relative to U, Fe, and possibly other components from the engineered and geologic system, such as nickel, aluminum, and silica; (iv) limiting ingress of water and oxidants to unaltered wastes; and (v) controlling by solubility or dissolution rate the source term for radionuclide (not just U) releases from the EBS. With regard to long-term performance of the proposed repository, secondary alteration products resulting from interactions of SF with the near-field environment, rather than unaltered SF, will control releases of many radionuclides from the EBS.

Experimental (Holland and Brush, 1980) and theoretical (Murphy, 1997) studies indicate that the solubilities of likely 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<sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, H<sub>3</sub>SiO<sub>4</sub><sup>-</sup>, F<sup>-</sup>, and Fe<sup>2+</sup>. 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 because silicon, boron, and aluminum, which are the principal network formers of borosilicate glasses, do not undergo changes in oxidation state within the range of expected Eh values under repository conditions. The effect of pH is far more important. The rate of dissolution is strongly accelerated at alkaline pH due to matrix dissolution. At pH lower than 4, the rate is accelerated by diffusion-controlled hydrogen ion exchange for alkali ions. Whereas many anions have a minor effect on the solubility and rate of dissolution of borosilicate glasses, fluoride accelerates the dissolution substantially through the formation of SiF<sub>6</sub><sup>2-</sup> complexes. The value of RH is important in the durability of glasses in humid air. Glasses are also susceptible to environmentally assisted cracking in aqueous environments (McCauley, 1995), but 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, although other components of the glass (e.g., B) will also affect its stability. Alkalinity produced by interactions of water with cementitious materials and lowered 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, analogous to alteration products of the natural volcanic glasses existing at YM. However, they are likely to incorporate augmented quantities of components of the EBS, 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**

Staff 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 than 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).

Corrosion products from metallic components, mostly in the form of metal cations, can affect corrosion rates 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  $Fe^{3+}$ , whereas, the pH can decrease by the hydrolysis of highly-charged cations, such as  $Cr^{3+}$  and  $Fe^{2+}$ , among others. In addition, the presence of low molecular weight organic compounds, including carboxylic acids produced by degradation of fuel, lubricants, or other organic materials, either by chemical or biochemical mediated processes, may accelerate the rate of corrosion of SF due to the formation of complexing species, especially those able to form chelates.

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, and is discussed in the Advanced Conceptual Design report (TRW Environmental Safety Systems, Inc., 1996b). 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. 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 EBS.

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**

Aside from autoradiolytic effects on pre-release waste characteristics, consideration of radiolysis is most important in its potentially complex effects on aqueous oxidation-reduction conditions. Although radiation may affect pre-failure chemistry outside the WP depending on

package shielding, to an extent that corrosion rates are affected (Reed and Van Konynenburg, 1993), this section is concerned with post-failure effects on radionuclide mobilization because of the massive multipurpose container design.

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

Radiolytic oxidizing species, such as  $\text{OH}^\bullet$ , where  $^\bullet$  denotes a free radical,  $\text{H}_2\text{O}_2$ ,  $\text{HO}_2^\bullet$ , and  $\text{O}^2$  (Spinks and Woods, 1976), could oxidize reduced species (e.g.,  $\text{Fe}^0$  in the WP to  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ,  $\text{N}_2(\text{aq})$  to  $\text{NO}_2$  or  $\text{NO}_3$ , and  $\text{U}^{4+}$  to  $\text{U}^{6+}$ ). Molecular hydrogen ( $\text{H}_2$ ) produced as the result of the combination of two  $\text{H}^\bullet$ , in contrast, is relatively non-reactive, and 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, and Bates, 1991; Wronkiewicz, et al., 1993; Sunder, et al., 1992; Sunder and Christensen, 1993). As pointed out by Van Konynenburg (1986), such processes are enhanced by unsaturated conditions expected in the proposed 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 and these topics are addressed in the CLST IRSR. 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  $0.2 \text{ g L}^{-1}$  of  $^{239}\text{Pu}$  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 in the repository 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. DOE has stated that they 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).

#### **4.4 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, 1998). Distribution of mass flux between fracture and matrix and retardation of radionuclides in fractures in the UZ and in the SZ constitute three of the NRC KESA for TSPA. Each of these KESA (see Appendix A) will be affected by coupled THC processes and are necessary to appropriately describe RT 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 RT 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 RT 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 overoptimistic, 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.

This group of acceptance criteria addresses data and model justification

- Available data relevant to both temporal and spatial variations in conditions affecting coupled THC effects on RT in the near field were considered
- 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 RT in the near field.

- 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 RT in the near field.
- 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 RT through engineered and natural barriers.
- Should microbial activity be sufficient to potentially cause adverse microbial effects on RT 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.
- 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.
- If the testing program for the effects of coupled THC processes on RT 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 UZ, 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; EBS 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 effects 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

This group of acceptance criteria addresses data uncertainty and verification.

- Reasonable or conservative ranges of parameters or functional relations were used to determine effects of coupled THC processes on RT in the near field. Parameter values, assumed ranges, probability distributions, and bounding assumptions are technically defensible and reasonably account for uncertainties.
- Uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on RT in the near field were considered.
- 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 RT in the near field.
- The initial conditions, boundary conditions, and computational domain used in sensitivity analyses involving coupled THC effects on RT in the near field were consistent with available data.
- 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 RT 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 UZ, 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 bound 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.

This group of acceptance criteria addresses model uncertainty

- 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 UZ and 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 RT in the near field; and (vii) changes in hydraulic and sorptive properties of the natural system resulting from interactions between cementitious materials and groundwater.

- Alternative modeling approaches consistent with available data and current scientific understanding were investigated, and their results and limitations were appropriately considered.
- DOE provided a reasonable description of the mathematical models included in its analyses of coupled THC effects on RT 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 UZ, 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

This group of acceptance criteria addresses model verification of abstractions

- The mathematical models for coupled THC effects on RT 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.
- DOE appropriately adopted accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on RT in the near field
- 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 abstract: the distribution of mass flux between fracture and matrix, retardation in fracture in the UZ, 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 to take credit for certain design features that have been demonstrated in NRC's or DOE's, or PA 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 NRC TPA code to selectively probe DOE's approach for these three abstractions for potential inconsistency in the analysis and nondefensible predictions

This group of acceptance criteria addresses integration.

- 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.
- Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on RT in the near field
- 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 could include activities, such as independent modeling, laboratory or field data, or sensitivity studies.
- Where simplifications for modeling coupled THC effects on RT in the near field 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/data abstractions, and track the progress made in resolving

deficiencies/nonconformities 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.

These programmatic acceptance criteria address QA issues.

- Data and models were collected, developed, and documented under acceptable QA procedures.
- Deficiency reports concerning data quality on issues related to coupled THC effects on RT in the near field were closed.

#### Review Method:

- If DOE uses this approach, NRC will attend, as observers, the elicitation workshops and review the documentation to ensure that the expert elicitations followed the guidance in NUREG-1563 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.

This programmatic criterion addresses expert elicitations

- 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 RT. As mentioned previously in Section 3, the dearth of analysis in past PAs on the effects of coupled geochemical processes on the potential RT 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 RT 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 UZ. 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  $K_d$ s associated with cementitious repositories. If substantial negative changes in the performance of the repository resulted from the use of these alternative  $K_d$ s, 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 for PA in the near field is the transport of radionuclides through the EBS and the geologic setting as gaseous species, as species in colloidal form, or dissolved in aqueous solution. Each RT process is influenced by several geochemical parameters; thus, an assessment of the relative importance of each process 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 or coprecipitation as an impurity in other minerals. 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 RT in the UZ, and in the SZ beneath the repository. In addition to its effect on RT, changes in porosity and permeability of the uppermost portion of the SZ could affect the extent of vertical mixing of fluids leaving the UZ. 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 and altered under conditions analogous to the proposed repository near field. These effects are reported to seal the matrix from the fractures, which could lead to enhanced fracture flow and minimal matrix diffusion.

## Sorption

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

Oxides and oxyhydroxides of metals, such as Fe, Mn, and Si, are common fracture lining minerals in the YM system (Carlos, et al., 1993) and may also be created by oxidation of materials introduced during the construction and operation of the repository (e.g., steel containers and rock bolts). The oxidation state in the near field may affect sorption behavior. For example, under oxidizing conditions, technetium is principally present as pertechnetate ( $TcO_4^-$ ) and does not sorb strongly, whereas, under reducing conditions,  $Tc^{4+}$  is predominant 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). For cations, such as  $UO_2^{2+}$ ,  $NpO_2^+$ , and  $Am^{3+}$ , oxyhydroxides exhibit a sharp sorption edge where, 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; 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 suggesting the potential importance of these minerals as a sorbent phase. In addition, the potential for forming a sorptive oxide coating on less sorptive particles, such as quartz or feldspar, suggests an additional role for these minerals in radionuclide sorption (Robert and Terce, 1989)

Other sorptive phases, such as clays, occur at YM as secondary replacement products. In addition, clays may also develop in the near field as alteration products of vitrified waste and SF. Because of interlayer exchange sites and the large surface area resulting from their layered structure, clays 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, and 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 in a manner like 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 RT in the near field at YM. Zeolites exhibit a fixed charge developed by substitution of  $Al^{3+}$  for  $Si^{4+}$  in the zeolite structure that 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), particularly for the alkaline and alkaline earth elements such as the short-lived

radioisotopes of Cs<sup>+</sup> and Sr<sup>2+</sup>, but there also appears to be a component of pH-dependent surface charge 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<sup>2+</sup> or Mg<sup>2+</sup>) and the carbonate anion (CO<sub>3</sub><sup>2-</sup>). For this reason, sorption on carbonates is a complex function of pH, solution chemistry, and p(CO<sub>2</sub>). 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 (Bish, 1993; Murphy, et al., 1996), depending on temperature and fluid chemistry. In addition, removal of radionuclides from solution either due to the precipitation of stoichiometric radioelement compounds or coprecipitation as an impurity in other minerals, is also affected by the temperature and chemistry of the solution (Murphy and Prikryl, 1996). Walton, Ross, and Juhnke (1985) demonstrated this reaction experimentally by machining circular flow channels into granite blocks and constructing 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 (<sup>125</sup>Sb, <sup>60</sup>Co, and <sup>54</sup>Mn) were concentrated at the hot side of the experiment, probably by sorption on Fe oxyhydroxides. <sup>144</sup>Ce and <sup>99</sup>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), and the effects of elevated temperature are poorly understood. Machesky, Palmer, and Wesolowski (1994) indicate that the zero-point-of-charge (pH<sub>ZPC</sub>) of rutile decreases with increasing temperature. This change suggests that negative charge development is enhanced for oxyhydroxides with increasing temperature, and 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 empirical nature of the limited data make it difficult to extrapolate over ranges in physical and chemical conditions likely in the near field. The effects of temperature are likely to be greater for mineral precipitation and dissolution, but additional experimental information is necessary to constrain temperature effects on RT through the near field.

## Diffusion

A retardation mechanism that is potentially important in the near field at YM is 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 (Percy, Prikryl, and Leslie, 1995). Instead, U transport in fractured tuff appears to

be dominated by fracture flow and precipitation of a suite of secondary uranyl minerals that proceeds with time through hydrated uranyl oxides to uranyl silicates and coprecipitation 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 RT 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 that, after an early initial release of  $^{14}C$  to the gas phase,  $CO_2$  will dissolve into the aqueous phase, and calcite precipitation will serve 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), where it is 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 EBS, 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, and 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, including pH, Eh, ionic strength, and  $p(CO_2)$ . 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 and 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 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  $\text{HCO}_3^-$  and  $\text{SO}_4^{2-}$ , can also affect the formation of oxides by consuming metal ions in the precipitation of carbonate and sulfate solids. If changes in solution chemistry result in desorption of radioelements, they are free to sorb onto the immobile medium. In this case, colloid transport becomes less of an issue for PA.

In addition to pH controls on the sorptive capacity of colloidal particles, the stability of the colloidal suspension of charged particles varies as a function of ionic strength, solution chemistry, and pH. At higher ionic strengths that may occur in the near field the double layer will collapse, and 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, resulting in increased bonding of positively charged crystallite edges to negatively charged planar sites. Positively charged oxides will also bond to negatively charged clay surfaces and organic macromolecules (Ryan and Gschwend, 1990). Under these conditions, dispersion is low, flocculation and agglomeration occurs, and the suspension is destabilized. As the pH increases towards the  $\text{pH}_{\text{ZPC}}$ , the positive surface charge of the oxides decreases and bonding to clays diminishes. At high pH, edge sites and oxyhydroxides exhibit a negative surface charge and actively repel the negatively charged clays, 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, and a mat or cake 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, either 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. Although London-van der Waals forces generally predominate at these ranges, energy provided from Born repulsive forces, or thermal and hydrodynamic energy, can overcome the attraction energy well and lead to particle erosion and re-entrainment. An additional possibility is that a decrease in the solution ionic strength may extend the electrostatic double layer, leading to particle release (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.

Although the size of colloids makes them vulnerable to several different filtration mechanisms, 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, while minimum velocity occurs at the fracture wall. Because of their size, colloids can never "experience" the minimum water velocity and, as such, 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, further enhancing the 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**

Staff analysis of the effects of engineered and man-introduced materials on RT 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, and can bind relatively highly charged cations, such as heavy metals and transuranic radionuclides, reducing sorption onto minerals. For example, studies of Kohler, Wieland, and Leckle (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, indicating that the concentration of organic matter in the near field will control effects on sorption. Also, organic molecules may become bound as gels and coatings to the surface of inorganic particles, such as clays and oxides, changing the sorptive behavior to reflect the organic coating (Robert and Terce, 1989).

RT 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 EBS 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. On the other hand, mineral alteration, due to alkaline solutions and precipitation of secondary phases, could reduce the sorptive and retardation ability of the geologic barrier and could also affect its hydraulic properties (porosity and permeability). For example, calculations by Lichtner and Eikenberg (1995), using a geochemical transport model (MPATH), indicated that interaction between a hyperalkaline plume released from a cement-based low-level radioactive waste repository and a marl host rock resulted in a rapid decrease in porosity of the host rock several meters from the repository due to precipitation of secondary phases, whereas porosity increased at the interface of the marl host rock and the cement due to mineral dissolution.

A potentially adverse scenario is that of flow of a hyperalkaline fluid along fractures in the tuffaceous host rock of the proposed repository. Dissolution of the tuff could lead to widening of the fractures and enhancement of groundwater flow and RT. Alternatively, precipitation of calcite and CSH phases along the fracture and matrix interface could seal the fractures from the matrix, producing isolated channels through which RT 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 transport through the repository. Preliminary calculations by

Lichtner, et al., (1997) suggest that strong alteration of the YM tuff host rock and of cement contact with the tuff could result from interaction of cement and tuff pore waters and the respective minerals.

One of the strongest effects on RT from engineered materials is likely to be on the potential colloidal transport. Oxides and oxyhydroxides of metals, such as Fe, Mn, and Si, found in fractures at YM, 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 sorbent minerals may form pseudocolloids for RT within the near field. Recent work suggests that colloids that could be formed within the WP or from the SF would most likely be agglomerate 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 hydrologic up gradient (i.e., in the WP) would be subject to alkaline pore fluids associated with the invert.

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

Experiments on SF leaching without imposed irradiation (Finn, et al., 1994a,b) are representative of potential autoradiolytic effects from SF alpha radiation. These experiments imply that nascent hydrogen,  $H^{\bullet}$ , plays a role in reducing carbonate in solution to formate an oxalate. During RT, 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 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), but 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 UC 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.

Although both oxidizing and reducing radiolytic effects on waste forms can be hypothesized, 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 RT. 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 PA 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 RT have been examined in foreign nuclear waste repository programs (Christofi and Philp, 1997; Brown and Sherrif, 1998; Lessart, et al., 1997) and in crushed volcanic tuff systems (Brown, Bowman, and Kieft, 1994). Potential impacts on RT 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, they 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) and has been demonstrated to be negligible for the Swedish program. The ability of microorganisms to be transported as colloids in an UZ 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 RT 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 RT in its TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b).

It will not be necessary to address microbial effects on RT 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 in the repository 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. 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 will be considered as part of the CLST KTI and criticality issues in the far field of the proposed repository will be considered as part of the RT KTI. 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 effects 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, have not yet been developed (U.S. Nuclear Regulatory Commission, 1998a). As a consequence, all the acceptance criteria necessary to evaluate credible scenarios that can affect the performance of the proposed repository over the compliance period, such as criticality in the near field, are not included in Revision 1 of the ENFE IRSR.

##### Review Method:

- 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 overoptimistic, 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.

This acceptance criterion addresses the need to evaluate whether criticality needs to be abstracted and included in a TSPA.

- 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

**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.

This group of acceptance criteria addresses data and model justification.

- 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.
- 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.
- 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.

This group of acceptance criteria addresses data uncertainty and verification.

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

This group of acceptance criteria addresses model uncertainty.

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

This group of acceptance criteria addresses model verification of abstractions.

- 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.
- 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.
- 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 nondefensible predictions

**This group of acceptance criteria addresses integration**

- **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.**
- **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.**
- **Models reasonably accounted for known temporal and spatial variations in conditions affecting coupled THC effects on potential nuclear criticality.**
- **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.**
- **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 model/data abstractions, and track the progress made in resolving deficiencies/nonconformities 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.**

**These programmatic acceptance criteria address QA issues**

- **Data and models were collected, developed, and documented under acceptable QA procedures.**
- **Deficiency reports concerning data quality on issues related to coupled THC effects on the potential for nuclear criticality were closed.**

### Review Method:

- If DOE uses this approach, NRC will attend, as observers, the elicitation workshops and review the documentation to ensure that the expert elicitations followed the guidance in NUREG-1563 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.

This programmatic criterion addresses expert elicitations.

- 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 (HEU) from naval and research reactors, and small amounts of plutonium and enriched U from commercial and DOE-owned SF.

##### **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 Cragolino, 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  $k_{eff}$ , which 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  $k_{\infty}$ . The value of  $k_{eff}$  is always less than or equal to the value of  $k_{\infty}$ . Typically, an upper bound for subcritical conditions is to have a calculated  $k_{eff}$  value of less than 0.95, with a 95-percent confidence level.

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

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  $^{239}\text{Pu}$  and  $^{235}\text{U}$ ; (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  $^{239}\text{Pu}$  concentration of 7.2 g/L is generally reported as the minimum critical concentration of Pu (Knief, 1992).

### **Neutron Absorbers**

Neutron absorbers reduce the reactivity of a fissile mixture by reducing the thermal neutron flux. These materials generally absorb neutrons and release gamma or alpha particles, which do not contribute to further fission events. There is a unique minimum absorber to fissile material mass ratio for all absorbers, above which the system will remain subcritical, independent of any other influences. One of the most effective absorber materials, boron, is likely to be engineered into waste forms for disposal. Exclusion of neutron absorbers, such as B, in model calculations leads to conservative estimates of the subcritical margin of safety.

If a mechanism for concentrating Pu exists that can lead to Pu concentrations above the 7.2 g/L required for a  $k_{\infty}$  of 1 for a pure water  $^{239}\text{Pu}$  system (Knief, 1992), the required absorber concentration to guarantee subcriticality becomes quite large. For example, for a mixture composed solely of iron,  $^{239}\text{Pu}$ , and water for a situation that has concentrated  $^{239}\text{Pu}$  to a level of 10.0 g/L, an iron concentration of 1,600 g/L is necessary to ensure subcriticality. The required amount of iron would increase for larger Pu concentrations.

## 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 infinite conditions, any geometry of finite size will be 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 by returning neutrons that have leaked out of the system to the fissile material where they are able to contribute to further fissions. Reflectors will reduce losses from geometry effects, so for conservative calculations that assume infinite dimensions, reflectors have no effect. Although it is unknown whether reflectors will be present around the fissile material, calculations that are done that take credit for neutron leakage losses from the system must also 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, and 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  $^{239}\text{Pu}$ ,  $^{235}\text{U}$  and other fissile materials are mobilized from waste forms in the YM repository environment, and the fissile material 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, as the system reaches criticality, water would boil off and disperse Pu into a greater volume of rubblized rock, eventually creating a geometry that is autocatalytic (or self-enhancing).

In general, Bowman and Venneri (1995) considered a spherical geometry of homogeneous mixtures of  $^{239}\text{Pu}$  with water and  $\text{SiO}_2$  as a proxy for rock. For the idealized spherical Pu-H<sub>2</sub>O-SiO<sub>2</sub> 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, and depending on the

mole fraction of silica, Bowman and Venneri (1995) proposed that a spherical mass of as little as 2 kg of  $^{239}\text{Pu}$  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  $^{235}\text{U}$  would require an even larger critical mass than  $^{239}\text{Pu}$ . 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, and several critiques of the hypothesis proposed by Bowman and Venneri (1995) have been prepared (Murphy, Jarzempa, and Lichtner, 1995; Parks, Williamson, and Hyder, 1995; Van Konynenburg, 1995). Most of these critiques focus on several key aspects of the Bowman-Venneri hypothesis that limit its applicability in the YM environment, including:

- The lack of specificity on plausible RT 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 UZ 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  $^{239}\text{Pu}$  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  $^{239}\text{Pu}$  by means of dissolution and redeposition (Murphy, Jarzempa, and Lichtner, 1995).
- The poor analogy between  $\text{SiO}_2$  and the host rock at YM. Other constituents in the waste (e.g.,  $^{238}\text{U}$ ), waste canister (e.g., Fe), rock (e.g., K, Al, and Na), and in the groundwater (Cl) would serve as neutron absorbers, requiring 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  $^{239}\text{Pu}$ ) 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 YN system would be necessary to evaluate the masses of  $^{239}\text{Pu}$  or  $^{235}\text{U}$  required to achieve criticality.

#### **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 in sufficient quantity, in an adequate configuration with respect to neutron reflection and escape, with 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, or precipitation from a solution may 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 YN 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.

## **5.0 STATUS OF ISSUE RESOLUTION AT THE STAFF LEVEL**

NRC staff has reviewed and commented on the DOE site characterization and PA programs in areas related to ENFE. The site characterization concerns were documented in the Staff Site Characterization Analysis (SCA) of DOE's SCP, YM Site, Nevada (U.S. Nuclear Regulatory Commission, 1989). Additionally, a letter from NRC to DOE with results of the staff review of DOE's thermohydrology testing and modeling program (U.S. Nuclear Regulatory Commission, 1997) described concerns on DOE's coupled THC modeling efforts. Staff's concerns on near field topics within DOE's PA program have been documented in the NRC audit review of DOE's 1995 TSPA (TSPA-95; U.S. Nuclear Regulatory Commission, 1996) and in NRC's comments on the DOE's TSPA (U.S. Nuclear Regulatory Commission, 1998c). The site characterization, thermohydrology, and performance assessment comments, and their status of resolution, are described in the following sections. Following their presentation a summary is provided on the status of resolution for each of the subissues described in Section 2.0.

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

The NRC review of the DOE SCP (U.S. Nuclear Regulatory Commission, 1989) resulted in eight comments and one question related to the ENFE KTI. Since the time of the SCA, DOE has adopted a revised program plan (U.S. Department of Energy, 1996b) and the RSS (U.S. Department of Energy, 1998). The RSS developed a number of hypotheses concerning safety attributes of the proposed HLW repository (U.S. Department of Energy, 1998). DOE's refocused program, a result of Congressional direction (U.S. Department of Energy, 1996b, page 11), incorporates the predecessor to the RSS (WCIS; U.S. Department of Energy, 1996a) and focuses on the remaining technical questions that have been demonstrated, through TSPAs, to be important to waste containment and isolation. As a result of the refocused program, many of the study plans have changed in scope, being deferred, or canceled (U.S. Department of Energy, 1997a; Appendices A and G). In addition, NRC has refocused its pre-licensing program to address those issues most significant to repository performance (Sagar, 1996), as the NRC site-specific regulations for the proposed repository will be performance based (U.S. Nuclear Regulatory Commission, 1998b). Some of the 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 CNWRA staff has become available to close open items. As a result, two of these comments were closed in Revision 0 of the ENFE IRSR. NRC's disposition of the remaining comments and question is provided below.

**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 EBS) and Subissue 4 (effects of coupled THC processes on RT 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. Staff 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 WP degradation products on sorption, and interaction among and between radionuclides on sorption. Based on the low solubilities of radionuclides, the staff considers interaction between and among radionuclides insignificant in sorption processes. NRC technical concerns are embodied in DOE's RSS 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).

While DOE is addressing resolution of this comment in a site characterization framework through submittal of a study plan, DOE is also evaluating the potential impact to performance of WP degradation products within its TSPA-VA, as part of the EBS transport module (TRW Environmental Safety Systems, Inc., 1997b). The staff still considers the effects of degradation products on repository performance important. To resolve this comment, the staff will apply the acceptance criteria in this IRSR to DOE's approach to evaluate the effects of WP degradation products on RT in the TSPA-VA.

**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. Staff 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 RSS 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).

While DOE is addressing resolution of this comment in a site characterization framework through submittal of a study plan, 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). The DOE model is expected to include the interaction between inner corrosion resistant alloy and outer corrosion allowance material. In particular, crevice formation between the outer barrier, and the outer barrier corrosion product precipitates and the inner barrier could cause pH reduction in the crevice, due to hydrolysis of dissolved metal ions from corrosion of both barriers; and accumulation of corrosion products inside the crevice will be considered (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). With adoption of C-22 as the corrosion resistant alloy, the reduction of pH and generation of concentrated  $\text{FeCl}_3$  solutions that may occur in crevices (Sridhar, Lichtner, and Dunn, 1996) appears to remain the most plausible chemical environment under which this alloy can undergo localized corrosion (Brossia, Dunn, and Sridhar, 1998). Resolution of the chemistry in this environment will require input

from the CLST KTI. Thus, the staff still considers the test environment in WP corrosion tests important. To resolve this comment, the staff will apply the acceptance criteria in this IRSR, in combination with review by CLST staff, according to the acceptance criteria in the CLST IRSR, to DOE's approach to evaluate the effects of the chemical environment of the WP on performance in the TSPA-VA.

**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, the ENFE KTI considered that Comment 81 was related to Subissue 2 (chemical environment of WP), 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 CNWRA during this fiscal year, suggest that this comment does not require consideration of the chemistry of the WP 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.

**SCA Comment 84:** The issue resolution strategies and testing programs for design of the WF and the EBS 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. Staff 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, the ENFE KTI considered that Comment 84 was related to Subissue 2 (chemical environment of WP); Subissue 3 (chemical environment for radionuclide release from the EBS); and Subissue 4 (RT 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 CNWRA during this fiscal year, 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.

**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 WP); Subissue 3 (chemical environment for radionuclide release from the EBS), and Subissue 4 (RT 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. Staff considered Comment 89 open, based on DOE's response, which indicated that testing programs will investigate how water chemistry is changed by the WP and other repository materials, and 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)

While DOE is addressing resolution of this comment in a site characterization framework through submittal of a study plan, 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). The staff still considers the introduction of large quantities of cements potentially important to repository performance. To resolve this comment, the staff will apply the acceptance criteria in this IRSR to DOE's approach in the TSPA-VA to evaluate the effects of pH variation, resulting from the use of grouts, cements, and organic materials, on WP and glass form degradation.

**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 WP). 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 the response of DOE, which 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, based on the testing program

**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 WPs. 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 WP) and is related to concerns in Subissue 3 (chemical environment for radionuclide release from the EBS) and Subissue 4 (RT 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. Staff 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).

While DOE is addressing resolution of this comment in a site characterization framework (Walt et al., 1998), 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). The staff still considers the chemical environment (water quality) of the WP to be important to repository performance. To resolve this comment, the staff 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.

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

Staff commented on the DOE Thermohydrology Testing and Modeling Program (U.S. Nuclear Regulatory Commission, 1997). 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. The staff's 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, 1997) will be addressed in this IRSR and the other two comments are addressed in the TEF IRSR. 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 NRC staff participation in the thermal testing workshops and near-field/alterred 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). While progress has been made in process-level modeling and results have been obtained from the experiments and modeling efforts, 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). The staff considers the proposed sensitivity studies important in determining whether THC

processes significantly impact repository performance. The staff's 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, 1997) is considered resolved. The staff 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.

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

NRC staff has provided comments to DOE on its performance assessment activities (U.S. Nuclear Regulatory Commission, 1996; 1998c). The NRC audit review of TSPA-95 identified that although many conservatisms were considered, several potential WP failure modes were not considered, and, as a result, the calculations may be non-conservative. Staff concerns related to the DOE humid air corrosion model were raised because the model did not account for the effect of aggressive groundwater, the modifications of the environment by the presence of corrosion products, and the evolution of the chemical composition of the environment with time. Staff 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. Furthermore, staff will continue to evaluate how DOE accounts for the effect of aggressive groundwater, the hygroscopic nature of corrosion products, or other forms of capillary condensation in the pores of oxide scale formed on the container surface in its evaluation of WP and total system performance in TSPA-VA in light of anticipated design changes for the WP.

As a general comment, 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. Given the empirical relationships for solubilities of  $Np$ ,  $Pu$ , and  $Am$  as a function of pH and temperature from 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, 1998c). 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, although it will affect seepage while intact and may affect flow within and out of the drift after it has collapsed. The staff 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.

Another concern that the staff has is that uncertainty in near-field environmental conditions, as it impacts WP performance, does not appear to be fully captured by the modeling (U.S. Nuclear

Regulatory Commission, 1998c) Two concerns arising from the expert elicitations that C has conducted in support of its TSPA-VA (CRWMS M&O, 1997b, 1998c) are that the propagation of uncertainty in near-field environmental conditions is impeded by the use of values for the corrosion potential, and the expected use of the elicited expert judgements is not 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 because the WP elicitation was completed prior to the initiation of the near field elicitation. The staff's 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 focussed 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.

##### **5.4.1 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.

#### **5.4.2 Effects of Coupled Thermal-Hydrologic-Chemical Processes on Waste Package Chemical Environment**

The consequences of coupled 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 RT. 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, 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 processes 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.

#### **5.4.3 Effects of Coupled Thermal-Hydrologic-Chemical Processes on 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.

#### **5.4.4 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) TH effects on gas and water chemistry in the UZ and 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 RT in the near field, and (vii) changes in hydraulic and sorptive properties of the natural system resulting from interactions between cementitious materials and groundwater.

#### **5.4.5 Potential Nuclear Criticality in the Near Field**

The acceptance criteria of this new subissue have been presented and staff's 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 ;  
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  
will not adversely impact repository performance, then criticality need not be considered within  
a risk-informed assessment of repository performance

## **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 the  
staff 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 PAs of the proposed repository. Likewise, the large self potentials measured in Yucca  
Mountain thermal tests, although speculated to be important to WP performance, have been  
demonstrated to be unimportant to WP performance and need not be considered in PAs 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

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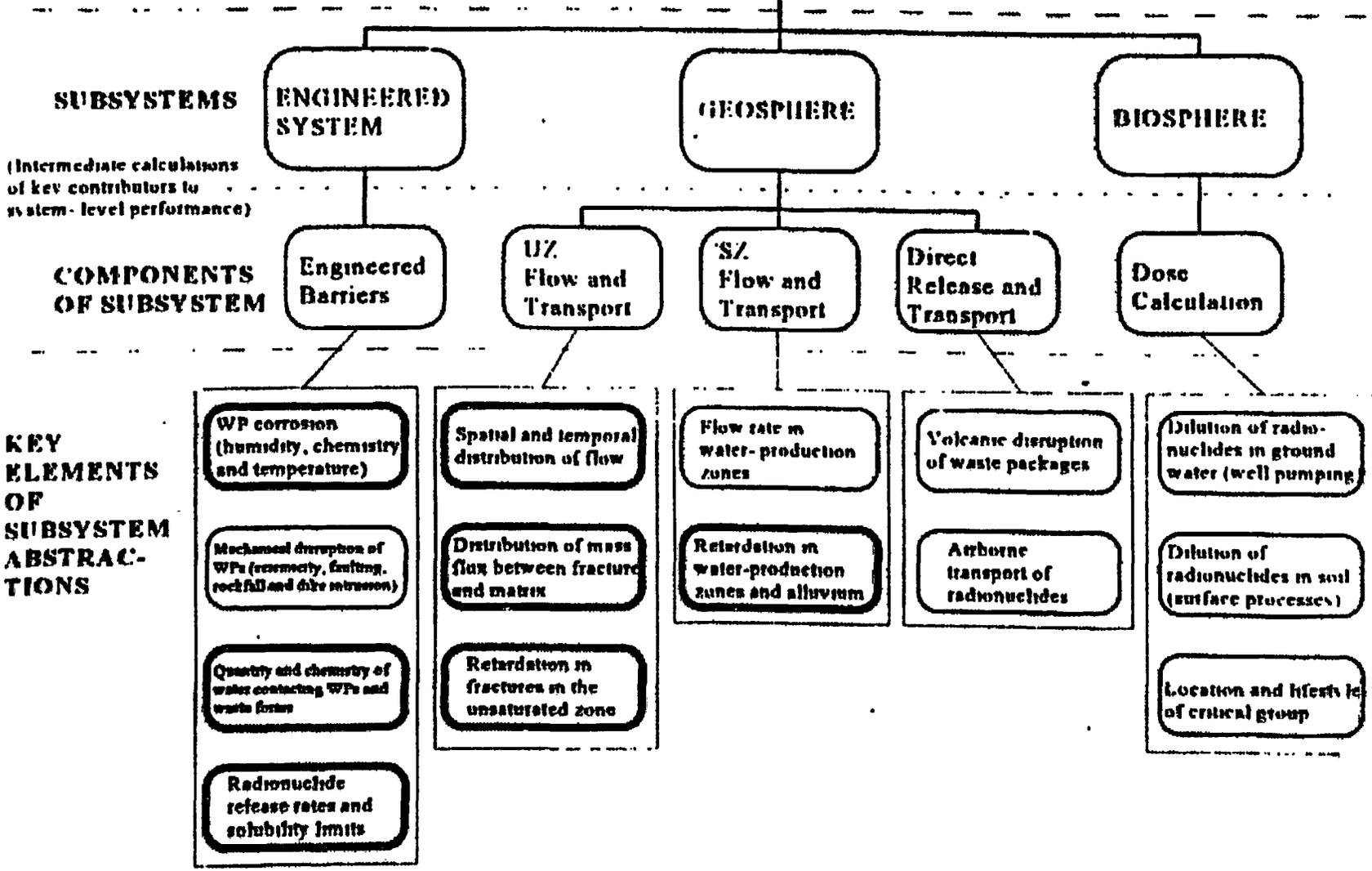
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## **APPENDIX A**

### **FIGURE ILLUSTRATING ELEMENTS OF THE NUCLEAR REGULATORY COMMISSION STAFF'S TOTAL SYSTEM PERFORMANCE ASSESSMENT**

**TOTAL SYSTEM**

**REPOSITORY PERFORMANCE**  
(Individual Dose or Risk)



**SUBSYSTEMS**  
(Intermediate calculations of key contributors to system-level performance)

**COMPONENTS OF SUBSYSTEM**

**KEY ELEMENTS OF SUBSYSTEM ABSTRACTIONS**