ISSUE RESOLUTION STATUS REPORT

KEY TECHNICAL ISSUE: CONTAINER LIFE AND SOURCE TERM

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

Revision 2

September 1999

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Change history of "Issue Resolution Status Report, Key Technical Issue: Container Life and Source Term"

Revision #	Section/ Paragraph	Date	Modification
Rev. 0	All	March 1998	None. Initial issue.
Rev. 1	All	November 1998	Wholly revised
Rev. 2	Acknowledgments	September 1999	Revised to include names of authors, reviewers, and QA related information
Rev. 2	1.0/1, 3–6	September 1999	Revised to update NRC strategy for prelicensing, new NRC regulation, and DOE VA information
Rev. 2	2.1/1–3	September 1999	Revised to include new regulation and DOE information on VA design
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Rev. 2	3.0/1	September 1999	Minor modifications related to introduction of integrated subissues and changes in Figure 2
Rev. 2	3.1/1	September 1999	Minor modifications to include information on new DOE design for EBS
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Revision #	Section/ Paragraph	Date	Modification
Rev. 2	3.2.2/1	September 1999	Minor modifications due to new DOE design
Rev. 2	3.2.5/all	September 1999	Revised to clarify issues related to performance
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Rev. 2	3.3.1/2–5	September 1999	Revised to include updated information on TSPA-VA
Rev. 2	3.3.3/1–3	September 1999	Revised to include updated information on TPA 3.2
Rev. 2	3.4/1–4	September 1999	Major changes to include updated results of sensitivity analysis. Figures 3, 4, and 5 are incorporated.
Rev. 2	4.0/1	September 1999	Addition of ASTM Standard Practice for long-term behavior of EBS components
Rev. 2	4.1/Title	September 1999	Redefinition of subissue
Rev. 2	4.1.2/1	September 1999	Minor modification related to ENFE KTI
Rev. 2	4.2/Title	September 1999	Redefinition of subissue to include initial defects
Rev. 2	4.2.1/5	September 1999	Addition of a new acceptance criterion

Revision #	Section/ Paragraph	Date	Modification
Rev. 2	4.2.2/1–3, 5, 6	September 1999	Revised to include modifications on DOE design and effect of initial defects
Rev. 2	4.3/Title	September 1999	Redefinition of subissue
Rev. 2	4.3.1/2–3	September 1999	Addition of two new acceptance criteria
Rev. 2	4.3.2/1–2	September 1999	Minor modifications related to environment inside WPs
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Rev. 2	4.4.2/1, 2	September 1999	Minor modification related to environment inside WPs and scope of resolution
Rev. 2	4.5/Title, 1	September 1999	Redefinition of subissue according to performance based regulation
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Rev2	4.5.2/1-7	September 1999	Revised to provide technical bases for new acceptance criteria
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Revision #	Section/ Paragraph	Date	Modification
Rev. 2	4.6.1/5, 6	September 1999	Addition of two new acceptance criteria to consider effects of new DOE design
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Revision #	Section/ Paragraph	Date	Modification
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Rev. 2	5.6.1/Title, 1, 2	September 1999	New section to reflect DOE design changes
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Rev. 2	5.6.3/Title, 1 2	September 1999	New section to reflect DOE design changes
Rev. 2	6.0	September 1999	New references added according to listed changes

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

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

ANALYSES AND CODES: The Total system Performance Assessment (TPA) code Version 3.2 was developed under CNWRA Technical Operating Procedure (TOP)–018.

1.0 INTRODUCTION

The U.S. Nuclear Regulatory Commission (NRC) strategic planning assumptions for the prelicensing period of the high-level radioactive waste (HLW) repository proposed at Yucca Mountain, Nevada, call for the early identification and resolution of issues at the staff level. A principal mean for achieving this goal is through informal, prelicensing consultation with the U.S. Department of Energy (DOE) regarding site characterization, experimentation, and design activities that are conducted consistent with the NRC geologic repository regulations and the proposed rule (U.S. Nuclear Regulatory Commission, 1999a). All these consultations required by law, occur in an open manner that permits observation by the State of Nevada, Tribal Nations, affected units of local government, and other interested members of the public. Obtaining input and striving for consensus from the technical community and interested parties help the issue resolution process. This process attempts to reduce the number of subissues and to better define those that may be in dispute during the NRC licensing review.

Consistent with NRC regulations and a 1992 agreement with the DOE, staff-level issue resolution can be achieved during the prelicensing consultation period. However, resolution at the staff level would not prevent the issue from being raised and considered during licensing proceedings. Staff level issue resolution during prelicensing is achieved when the staff has no further questions or comments (i.e., open items), as to how the DOE program is addressing an issue. There may be some cases in which resolution at the staff level is limited to documenting a common understanding regarding differences in the NRC and the DOE technical positions. Additional pertinent information could raise new questions or comments regarding a previously resolved issue.

The NRC HLW program was realigned during fiscal year (FY) 1996-1997 in response to: (i) a reduction in Congressional budget appropriations for NRC in FY 1996; (ii) the reorganization of DOE's geologic repository program at Yucca Mountain, Nevada; and (iii) a 1995 report issued by the National Academy of Sciences to advise the U.S. Environmental Protection Agency regarding the technical bases for new geologic disposal standards for Yucca Mountain. As a result of these developments, the NRC HLW program was realigned to focus prelicensing work on those topics most critical to the post-closure performance of the proposed geologic repository. These critical topics are identified as Key Technical Issues (KTIs). This approach is summarized in Chapter 1 of the FY 1996 Annual Progress Report (U.S. Nuclear Regulatory Commission, 1997).

The Division of Waste Management (DWM) current approach is to focus most activities on issue resolution at the staff level. DWM activities have been reprioritized to improve the integration of the technical work necessary to achieve staff-level resolution. Regulatory attention is focused on those technical uncertainties that have the greatest effect on the assessment of repository safety. This focus is accomplished by identifying KTIs, integrating their activities into a risk-informed approach, and evaluating their significance for post-closure repository performance. Early feedback among all parties is essential in determining what is known, what is not known, and if additional information is likely to make a significant difference in understanding future repository safety.

An important step in the DWM approach to issue resolution is to provide DOE with feedback regarding issue resolution. Issue Resolution Status Reports (IRSRs) are the primary mechanism used to provide DOE with feedback on KTI subissues. IRSRs focus on: (i) acceptance criteria for issue resolution; and (ii) the status of resolution, including areas of agreement or comments or questions. Feedback is also contained in the Annual Progress Report (e.g., U.S. Nuclear Regulatory Commission, 1997), which summarizes the significant technical work toward resolution of all KTIs during the preceding fiscal year. Finally, open meetings and technical exchanges with DOE provide additional opportunities to discuss issue resolution, identify areas of agreement and disagreement, and develop plans to resolve such disagreements. In addition, the staff is using the IRSRs to develop the Yucca Mountain Review Plan (YMRP) for the potential repository license application (LA). The YMRP, Revision 0, to be completed in FY 2000, will contain the acceptance criteria and review methods for all KTIs, presented in the context of Integrated Subissues (ISIs). The next revision of the IRSRs, to be completed at the end of FY 2000, will update information on progress in subissue resolution for each KTI.

Furthermore, the IRSRs were the basis for the review of information in the DOE Viability Assessment (VA) (U.S. Department of Energy, 1998b,c). Comments on the VA are intended to facilitate the DOE's efforts to focus its program and develop a high-quality LA. The preliminary design concept, the total system performance assessment (TSPA), the LA Plan, and supporting documents (U.S. Department of Energy, 1998b,c) were reviewed. Through these reviews, a set of technical comments regarding the supporting data and models within the TSPA were identified. Detailed comments on the VA are provided in this revision of the IRSR. Section 5 of the IRSR documents the application of review methods and acceptance criteria to the VA and updates the status of resolution.

Each IRSR contains six sections. This Introduction is Section 1.0. Section 2.0 defines the KTI, all the related subissues, and the scope of the particular subissue that is the subject of the IRSR. Section 3.0 discusses the importance of the subissue to repository performance including: (i) qualitative descriptions; (ii) relationship to total system performance; (iii) results of available sensitivity analyses; and (iv) relationship to the DOE Repository Safety Strategy (see U.S. Department of Energy, 1998a), that is, its approach to the VA. Section 4.0 provides the review methods and acceptance criteria, which indicate the technical basis for resolution of the subissue that will be used in subsequent reviews of DOE submittals. These acceptance criteria are guidance for the staff and, indirectly, for DOE as well. Technical basis for the acceptance criteria are also explained in detail to further document the rationale for the staff decisions. Section 5.0 concludes the IRSR with the status of resolution of the subissues. Section 6.0 presents a list of pertinent references. The appendix summarizes those items resolved at the staff level and those items remaining open. These open items will be tracked and resolution will be documented in future revisions of the IRSRs.

2.0 KEY TECHNICAL ISSUES AND SUBISSUES

2.1 PRIMARY ISSUE

The primary issue of the KTI on Container Life and Source Term (CLST) is adequacy of the engineered barrier subsystem (EBS) design, to provide reasonable assurance that containers will be adequately long-lived, and radionuclide releases from the EBS will be sufficiently controlled, such that container design and packaging of spent nuclear fuel (SNF) and HLW glass will make a significant contribution to the overall repository performance. The site-specific regulation for the proposed YM repository, to be issued as 10 CFR Part 63 (U.S. Nuclear Regulatory Commission, 1999a), currently in the public commenting period, is a performance-based regulation. This IRSR is focused on the containers and waste forms as the primary engineered barriers, but it also considers other engineered subsystem enhancements (i.e., ceramic coatings, drip shields, backfill) incorporated as options in the EBS design. For the purpose of this IRSR, the physical boundary of the EBS is defined by the walls of the waste package (WP) emplacement drifts.

The reference DOE design for the EBS in the VA (U.S. Department of Energy, 1998b) consisted of double-wall WPs placed on steel supports in concrete-lined drifts. The WP consisted of a 10-cm-thick outer overpack made of a corrosion-allowance material and a 2-cm-thick inner overpack made of a corrosion-resistant Ni-base alloy, in addition to the waste form and other materials for criticality control, heat transfer, and structural support of fuel assemblies. The reference materials for the VA design of the WP were A516 Grade 55 steel (a wrought C-Mn steel) for the outer overpack and Alloy 22 for the inner overpack. Additional metallic components, such as the pour canister for defense HLW glass and the canister for DOE or Navy SNF, both made of Type 316L stainless steel, will be present in some WP designs. The WP and EBS designs have been significantly revised after the completion of the VA. The reference WP design recommended for the proposed SR (Howard, 1999), called the Enhanced Design Alternative II (EDA-II), consists of an outer overpack made of 2-cm-thick Alloy 22 surrounding an inner overpack made of type 316NG (Nuclear Grade) stainless steel that is 5 cm thick. The main purpose of the inner overpack is to provide structural mechanical strength to the WP. Additionally, a mailbox-shaped drip shield made of 2-cm-thick titanium Grade-7 extends over the length of the emplacement drifts to enclose the top and sides of the WP. The drift will have steel sets and lagging (or in some cases, rock bolts and mesh) for ground support, and the WPs will be resting on a steel invert with granular ballast. The design of the EBS in the EDA-II includes the use of backfill to cover the drip shield, even though a design without backfill is still being considered (Blink, 1999).

There are several design concepts for SNF and HLW glass containers in the VA (U.S. Department of Energy, 1998b). These concepts include uncanistered and canistered designs. There are eight different designs for the uncanistered fuels in the VA, but these designs all have the same container materials and wall thicknesses. The length, diameter, and interior of the eight designs vary for the different designs. The uncanistered SNF disposal container will be fabricated in two sizes. The large size will hold either 21 pressurized-water reactor (PWR) or 44 boiling-water reactor (BWR) assemblies. The small size will hold 12 PWR or 24 BWR assemblies. The design to hold commercial spent fuel and the Navy SNF consists of a single canister inside a disposal container. The co-disposal container for DOE-owned SNF and HLW glass canisters will hold five HLW glass canisters with a DOE-owned SNF disposal canister inserted in the center of the container and surrounded by the HLW glass canisters. Although the overpack materials have changed in the SR design, the uncanistered and canistered container design concepts for the various waste forms are not expected to change substantially. However, there are several design concepts not yet defined and the elimination of small PWR/BWR WP designs has been recommended (Blink, 1999).

2.2 SUBISSUES

Figure 1 identifies six subissues deemed important to the resolution of this KTI.

(1) The effects of corrosion processes on the lifetime of the containers

(2) The effects of phase instability of materials and initial defects on the mechanical failure and lifetime of the containers

(3) The rate at which radionuclides in SNF are released from the EBS through the oxidation and dissolution of spent fuel

(4) The rate at which radionuclides in HLW glass are leached and released from the EBS

(5) The effect of in-package criticality on WP and EBS performance

(6) The effects of alternate EBS design features on container lifetime and radionuclide release from the EBS

Each of these six subissues may, in turn, be addressed in terms of its principal components. Subissue 1 considers failure of outer and inner overpacks as a result of various corrosion processes affecting both WP materials, such as dry-air oxidation, humid-air and uniform aqueous corrosion, localized corrosion, microbially influenced corrosion (MIC), stress corrosion cracking (SCC), and hydrogen embrittlement. Subissue 2 examines long-term degradation of mechanical properties of container materials as a result of prolonged exposures of the WPs (thousands of years) to elevated temperatures. Mechanical failure due to phase instability of WP materials is highly dependent on material chemical composition and processing history. Examples of material instability that can degrade mechanical properties include segregation of metalloid elements such as phosphorus and sulfur, precipitation of carbides or intermetallic phases, and long-range ordering. Fabrication defects that may lead to early failure of container materials are also the subject of this subissue, as well as the effects of disruptive events, such as seismicity, faulting, and igneous activity. Mechanical failure of containers caused by disruptive events has been partially considered in repository design and thermal-mechanical effects (RDTME), structural deformation and seismicity (SDS), and igneous activity (IA) IRSRs. Subissue 3 considers degradation of SNF and subsequent radionuclide release from SNF in both dry air and aqueous environments following cladding failure. These processes involve radionuclide release by aqueous dissolution of the SNF matrix, limited by solubility or enhanced by colloid formation, and also includes dry oxidation of SNF and gaseous transport. In a similar manner, Subissue 4 deals with radionuclide release from the HLW glass form following aqueous dissolution of the borosilicate matrix. The degradation of WPs may give rise to criticality problems, which may occur either within the WP or outside the WP after transport and redeposition of fissile radionuclides in the repository environment.

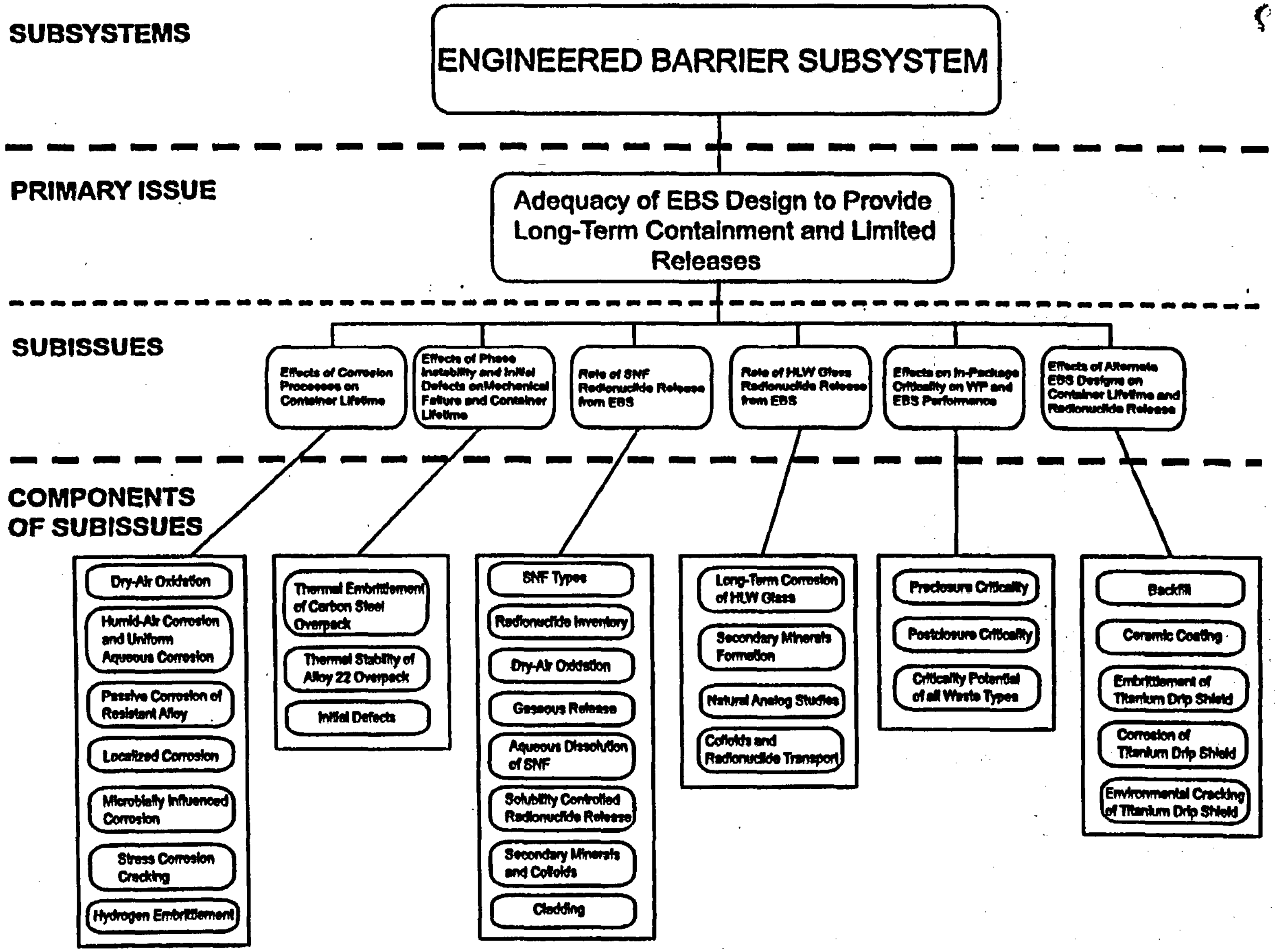


Figure 1. Flow diagram for container life and source term subissues

Criticality inside the WPs is assessed as Subissue 5 in this IRSR. Criticality outside the WP is evaluated as a relevant subissue in both the Evolution of the Near-Field Environment (ENFE) and Radionuclide Transport IRSRs. Subissue 6 examines alternate design features of the EBS, such as drip shields, backfill, and ceramic coatings, that DOE is considering for extending container life and decreasing radionuclide release from the EBS. Any other design feature adopted by DOE for improvement of the engineered barriers performance will be considered in this subissue.

This version of the IRSR addresses all the subissues listed above and describes the extent to which DOE has provided adequate technical bases for resolution of each subissue. Furthermore, in this and in subsequent versions of the CLST IRSR the staff will evaluate whether DOE's technical bases reflect important physical phenomena and processes, consistent assumptions and definitions, consideration of alternative models, bounding approaches, adequate abstraction of process models, appropriate expert judgments, and quality assurance documentation.

3.0 IMPORTANCE TO REPOSITORY PERFORMANCE

The primary goals of the DOE RSS (U.S. Department of Energy, 1998a) are the near-complete containment of radionuclides within the containers for several thousand years and acceptably low annual doses to the average member of a receptor group living near the site. The staff has developed a strategy (U.S. Nuclear Regulatory Commission, 1998a) for assessing the performance of a proposed HLW repository at YM. As currently visualized by the staff, the elements of this strategy that are necessary to demonstrate repository performance are defined as ISIs. Figure 2 illustrates the ISIs for this KTI within the solid line block pertaining to the EBS. The acceptance criteria, on which staff review of key elements in the DOE VA and LA will be based, are currently under development. These acceptance criteria are indicated in Section 4.0 of this IRSR. Subject to review and updating, these acceptance criteria and their technical bases will be incorporated in the YMRP. As noted in Section 2.0 of this report, the subissues related to container lifetime and radionuclide release rates from the EBS are considered important factors in the repository performance. For DOE to adequately demonstrate and quantify the consequences that container failure and radionuclide release have on repository performance, its LA must include the effects of the near-field environment on corrosion of containers; the mechanical disruption of containers; the degradation effects of both quantity and chemistry of the water contacting the various waste forms, the processes that affect solubility, formation of colloids and radionuclide release rates, and the effects of potential criticality processes and events.

3.1 RELATIONSHIP OF SUBISSUES TO THE U.S. DEPARTMENT OF ENERGY REPOSITORY SAFETY STRATEGY

The performance of the engineered barriers after emplacement is extremely important in the DOE RSS for the YM site (U.S. Department of Energy, 1998a). In this strategy, DOE has formulated several hypotheses that, if correct, would demonstrate that radioactive waste can be isolated at the proposed YM site for long periods of time. These hypotheses state that: (i) heat produced by emplaced waste will reduce the relative humidity (RH) at the WP surface; (ii) corrosion rates will be low at low RHs; (iii) double-walled WPs will significantly increase containment times because of protection of the inner barrier by the outer barrier; (iv) engineered subsystem enhancements can extend even further the containment period of the inner barrier; (v) containment time will be sufficiently long to prevent dry air oxidation of SNF during the thermal period (at the elevated temperatures prevailing in the initial period following waste emplacement); (vi) the amount of water that may contact the waste can be limited; (vii) release rate of soluble radionuclides will be controlled by slow dissolution of the waste form; and (viii) release rate of actinides will be controlled by solubility limits rather than by colloidal stability. Hypotheses (i), (ii), and (iii) may be removed from further detailed consideration because the EDA-II proposed to the DOE relies on a relatively low thermal loading and a single corrosion resistant alloy overpack. The staff needs to evaluate the CLST KTI subissues to determine the merits of each of these DOE hypotheses and may then perform its evaluation using, as appropriate, methodologies independent of the ones used by DOE.

3.2 IMPORTANCE OF SUBISSUES TO TOTAL REPOSITORY SYSTEM PERFORMANCE

Staff has implemented a strategy for assessing the performance of the proposed HLW repository at YM. The framework for this strategy is discussed in the Total System Performance Assessment and Integration IRSR (U.S. Nuclear Regulatory Commission, 1998a). The four ISIs related to the performance of the engineered barriers and influenced by the subissues of the CLST KTI are highlighted in Figure 2. Figure 1 identifies the six subissues of the CLST KTI considered most important by NRC staff in determining the adequacy of container design, EBS features, and waste form performance for long-term containment and limited radionuclide release. The container is the primary design element that provides radionuclide containment. After loss of containment, radionuclide release rates from the EBS are limited by waste form characteristics and transport processes throughout the container and the EBS. The combination of long-lived containers and low degradation rate waste forms can make a significant contribution to the performance of the repository system. The importance of the CLST subissues to repository performance is discussed in detail in the next sections.

3.2.1 Corrosion Effects—Importance to Performance

Under anticipated repository conditions, corrosion is expected to be the dominant failure mode limiting container life (Cragnolino and Sridhar, 1991, 1992; Sridhar, et al., 1995). Container life is defined as "the time lapsed until through-wall penetration of both outer and inner metallic barriers by corrosion or by mechanical failure." Loss of containment allows the release of radionuclides to the environment surrounding the WPs. Mechanical failures associated with material instability and container failures resulting from disruptive events are both considered in Subissue 2.

In recent performance assessment (PA) studies (Wilson, et al., 1994; TRW Environmental Safety Systems, Inc., 1995; U.S. Nuclear Regulatory Commission, 1995; Kessler and McGuire, 1996; TRW Environmental Safety Systems, Inc., 1997b, 1998a,b; U.S. Department of Energy, 1998c), container life is determined mainly by the failure time as a result of the penetration of the outer and inner overpacks, from corrosion. Leaving aside those cases related to disruptive events and/or mechanical failure it is assumed in these studies that radionuclide release cannot take place until the container is breached by through-wall penetration as a result of corrosion.

3.2.2 Materials Stability and Mechanical Failure—Importance to Performance

Because the VA design had carbon steel as the outer overpack, the effect of WP temperature on material stability of carbon steel was evaluated. Staff evaluations (Sridhar, et al., 1994; Cragnolino, et al., 1996) indicated that carbon and low-alloy steels, such as A516 Grade 55 and A387 Grade 22 steels, may experience a substantial decrease in toughness as a consequence of long-term thermal aging at repository temperatures. This phenomenon, which is similar to temper embrittlement, may contribute to premature mechanical failure of outer overpacks. Thermal stability of corrosion-resistant Ni-base Alloy 22, used as inner overpack materials in the VA design and proposed as the outer overpack material in EDA-II, can also be compromised by prolonged exposures to elevated temperatures. In this case, generation of

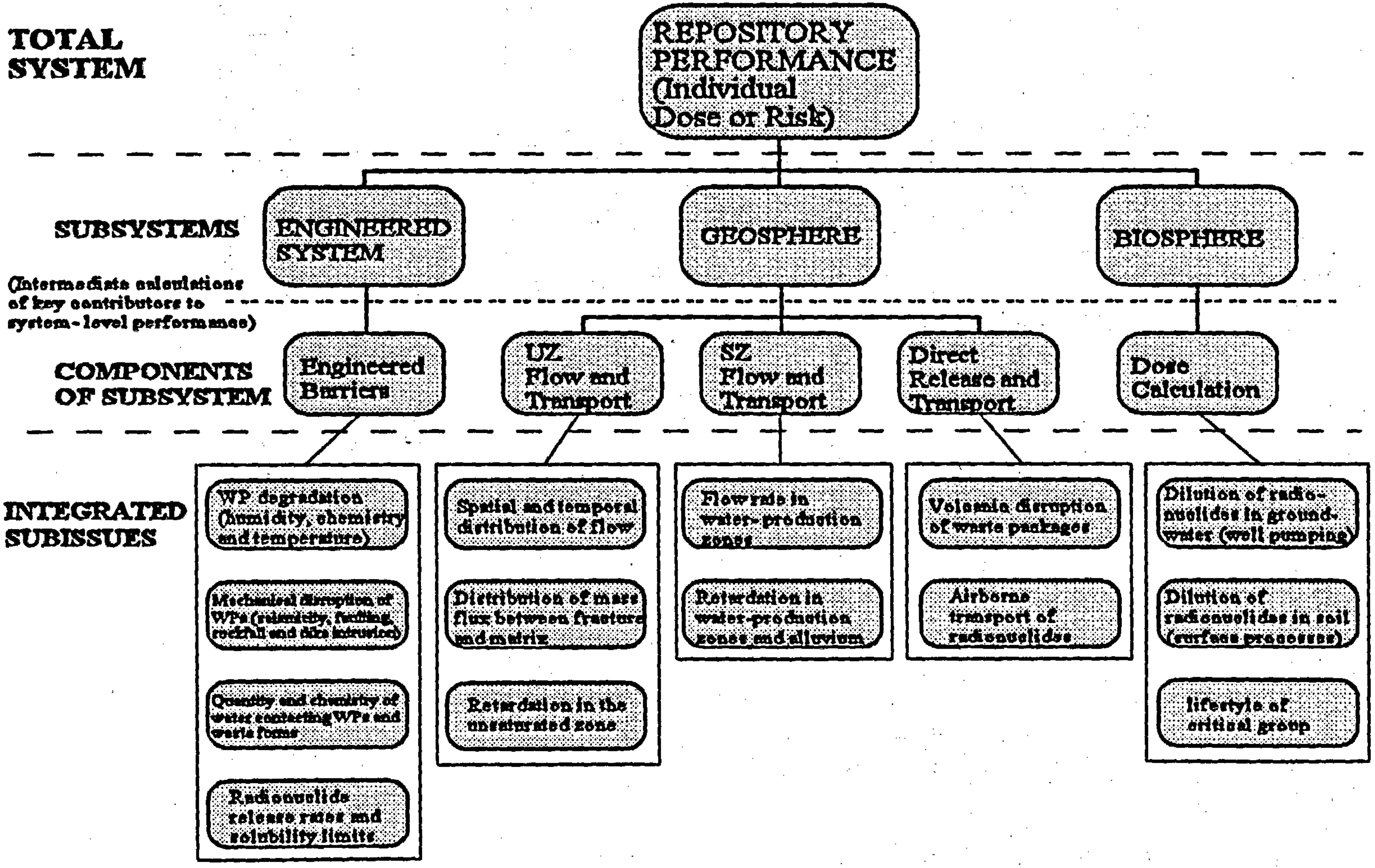


Figure 2. Flow diagram for total system performance assessment. The subissues of container life and source term provide input to the highlighted key elements.

ordered structures or formation of brittle intermetallic phases may affect mechanical properties or facilitate degradation processes, such as hydrogen embrittlement. Degradation of mechanical properties leading to mechanical failure from residual and/or applied stresses can adversely affect container performance and, ultimately, performance of the repository system. The consequences of disruptive events, such as seismic activity, volcanic activity, and faulting, in terms of their effects on mechanical failure of WPs, which is part of this subissue, will be considered in detail in subsequent revisions of this IRSR. Aspects of these disruptive events that may affect the mechanical integrity of containers are currently discussed in IRSRs on "Repository Design and Thermal-Mechanical Effects" (RDTME), "Structural Deformation and Seismicity" (SDS), and "Igneous Activities" (IA).

3.2.3 Spent Nuclear Fuel Degradation—Importance to Performance

Following container failure, SNF will be exposed to the potentially degrading effects of the environment in contact with the WP. Possible degradation processes for commercial SNF include dry-air oxidation and aqueous dissolution of the irradiated UO_2 matrix, followed by the formation of colloids and secondary minerals, that can result in the mobilization of radionuclides and subsequent release from the EBS (Manaktala, 1993; Manaktala, et al., 1995; Ahn, 1996a,b). For DOE-owned SNF, potential degradation processes include preferential dissolution of U-containing intermetallic phases. Resistance of the SNF to environmental degradation could contribute substantially to controlling radionuclide release from the EBS and could enhance overall performance of the repository.

3.2.4 High-Level Waste Glass Degradation Effects—Importance to Performance

After container failure, the glass waste form will be exposed to the potentially degrading effects of the environment in contact with the WP. A possible degradation process is dissolution of the borosilicate glass matrix, accompanied by the formation of colloids and secondary minerals, that can result in the mobilization of radionuclides followed by release from the EBS (Manaktala, 1992; Manaktala, et al., 1995). In this regard, the staff recognizes that glass wastes will constitute only about 3 percent of the total radionuclide inventory in the repository. However, if the glass waste form performs poorly in the repository environment, it could conceivably make a significant contribution to the overall radionuclide source term from the EBS. Accordingly, the resistance of the glass waste form to environmental degradation could contribute substantially to controlling radionuclide release from the EBS, enhancing the overall performance of the repository.

3.2.5 Criticality within the Waste Packages—Importance to Performance

Criticality within the WP during pre-closure and post-closure could have a significant effect on repository performance. The most likely effect of a criticality during the pre-closure period would be an increase in dose to workers. Depending on the dynamics of the criticality condition, off-site releases could be possible. The likely effects of in-package criticality with regard to repository post-closure performance are increases in the radionuclide inventory, WP heat output, and WP degradation.

For in-package criticality, steady-state criticality events would lead to increased radionuclide inventories. However, repeated fast reactivity insertion over a long period of time could also be important from a radionuclide production standpoint. Depending on the power level and duration of critical conditions, significant amounts of radionuclides, especially fission products including Tc-99, Np-237, and I-129, will be produced. The impact on the repository performance would be an increase in radionuclide inventory available for release from the WP, and a potential subsequent increase in dose to the critical group.

The second aspect of in-package criticality, with regard to the repository performance, is heat production, from the additional fission reactions taking place during criticality conditions, in addition to the expected decay heat. The additional heat produced could impact repository performance indirectly through its effect on the near field environment, and could increase WP corrosion rate.

The third aspect of in-package criticality is its impact on WP integrity, which includes the waste form. This factor is directly linked to the repository performance. Configurations initiated by a sudden mechanical disturbance such as an earthquake or rockfall, can result in a step reactivity insertion that could result in a mechanical failure of the already corroded WP and/or ruptures of the SNF cladding. The temperature and kinetic energy generated by the step reactivity insertion could also increase the exposed surface area and degradation rate of the SNF matrix. Other parts of the EBS could also be affected, including stability of the surrounding rock which could then cause further damage to the WP.

3.2.6 Alternate Engineered Barrier Subsystem Design Features—Importance to Performance

The DOE discussed several major design alternatives in the VA (U.S. Department of Energy, 1998b). These alternatives include ceramic coating on containers, dual corrosion-resistant materials, drip shield, backfill over and under the drip shield, WP filler, steel-lined drifts, lower thermal loading, and ventilation. Subsequently, EDA-II proposed for the SR design consists of a dual overpack with Alloy 22 as the outer overpack and type 316NG (Nuclear Grade) as the inner overpack (mainly to provide structural mechanical strength) (Howard, 1999). These design changes may have a significant effect on performance. For example, absence of a carbon steel outer overpack may obviate concerns related to thermal aging-induced embrittlement and stresses arising from corrosion products. However, the thinner overpacks proposed for the SR design may give rise to greater radiolysis of the near-field environment and less mechanical strength to sustain the effects of seismic events, including rock fall. The DOE is considering alternate design features for improving the performance of the EBS. These features include the use of drip shields made of Titanium Grade 7 to avoid direct contact of water dripping on the WP surfaces, and backfill to modify the environment surrounding the WPs (Howard, 1999).

3.3 CONSIDERATION OF CONTAINER LIFE AND RADIONUCLIDE RELEASE IN PREVIOUS PERFORMANCE ASSESSMENTS

The evaluation of container life and radionuclide release from the EBS has been performed in recent PA studies for the proposed YM repository in which the current WP conceptual design was considered. These studies include DOE's TSPA-95 (TRW Environmental Safety Systems, Inc., 1995); the Electric Power Research Institute (EPRI) Yucca Mountain Total System Performance Assessment, Phase 3 (Kessler and McGuire, 1996); the DOE TSPA-VA (U.S. Department of Energy, 1998c) and the NRC/Center for Nuclear Waste Regulatory Analyses (CNWRA) Total System Performance Assessment (TPA).

3.3.1 U.S. Department of Energy Total System Performance Assessment

DOE evaluated container life in TSPA-95 (TRW Environmental Safety Systems, Inc., 1995) using Version 1.0 of the stochastic Waste Package Degradation (WAPDEG) code (Atkins and Lee, 1996). WAPDEG is a probabilistic code designed to run stochastic simulations in which random values are sampled to represent parameters in the corrosion models for determining the WP failure time. This PA focused on WP failure by corrosion, and did not address WP degradation/failure resulting from mechanical stresses or juvenile failures.

The TSPA-VA (U.S. Department of Energy, 1998c), as well as the supporting Technical Basis Document (TRW Environmental Safety Systems, Inc., 1998a), included mechanical failure and juvenile failures, in addition to significant enhancements in the WAPDEG code for the consideration of corrosion processes.

The WP environment in WAPDEG was assumed to be humid air at elevated temperatures for an extended period followed by an aqueous environment, a hypothesis consistent with the DOE RSS (U.S. Department of Energy, 1998a). Dry-air oxidation of container materials was considered to be negligible and hence not included in the calculations. Humid-air corrosion was distinguished from aqueous corrosion by using two threshold values of RH, one for each corrosion process. For the carbon steel outer overpack, active general corrosion was assumed to occur in humid air and was modeled using a parametric equation exhibiting a dependence of the corrosion rate on time, RH, and absolute temperature. The parameters were obtained by multiple linear regression analysis of atmospheric corrosion data from tropical, urban, rural, and industrial locations. Aqueous corrosion of the outer overpack was evaluated through a similar approach, using literature data acquired from polluted river water and tropical lake water. Pitting corrosion of both overpacks was modeled based on input from an expert elicitation process. It was assumed that pitting corrosion of the Alloy 22 inner overpack did not occur below 80°C, the critical pitting temperature being distributed between 80 and 100°C. The general corrosion rate of Alloy 22 was obtained from expert elicitation based on consideration of three environmental conditions: (i) a moderately oxidizing (340 mV vs. Standard Hydrogen Electrode) environment with a pH of 3–10, (ii) a moderately oxidizing environment with a pH of 2.5, and (iii) a highly oxidizing environment (640 mV vs. Standard Hydrogen Electrode) environment at a pH of 2.5. These environments were assumed to form under localized corrosion sites even though the corrosion itself was considered to be uniform. The effect of galvanic coupling was ignored, but cladding failure was evaluated as part of this PA.

Radionuclide release calculations were conducted as part of the Repository Integration Program, the computer code used for the PA of the repository (TRW Environmental Safety Systems, Inc., 1998b). WP failure times, along with matrix alteration/dissolution rates determined by using a parametric equation that depends on environmental factors, were used in the code to compute the rate at which radionuclides are released, taking into consideration their solubility as a constraint. The matrix alteration/dissolution rates were determined by using a parametric equation from tests in Ca- and Si-free, sodium carbonate solutions for spent fuel and in simulated groundwater for HLW glass. The rates depend on environmental factors such as carbonate concentration, pH, temperature, and oxygen fugacity. The alteration/dissolution by the groundwater was assumed to take place under dripping conditions. In addition, cladding protection was considered for spent fuel, decreasing the exposed surface area of the bare spent-fuel matrix. The mass transfer of dissolved radionuclides out of and away from the WPs was computed as a sequence of processes. By using certain simplifications, release and transport of colloids were also considered.

The sensitivity analyses included in TSPA-VA (U.S. Department of Energy, 1998c) indicated that uncertainty and variability in the Alloy 22 general corrosion rate were important for 10,000 and 100,000 years. Additional factors important to total system performance were the fraction of wetted surface area, high pH water, microbiologically influenced corrosion, and juvenile failures.

3.3.2 Electric Power Research Institute Performance Assessment

An alternative assessment of WP performance was conducted by EPRI in their Yucca Mountain Total System Performance Assessment, Phase 3, using the Integrated Multiple Assumptions and Release Calculations code, as reported by Kessler and McGuire (1996). This is a deterministic code in which an event tree approach is used and the container life is assumed to be governed by a series of Weibull distributions that are dependent on the heat transfer mechanism (conduction only, conduction and convection, and heat pipe) and the temperature history. Corrosion processes considered in this statistical approach are general corrosion; localized corrosion (pitting and crevice); SCC; degradation from a metastable microstructure; embrittlement caused by hydride formation; and MIC. Galvanic protection is not considered. The Weibull distributions employ a feature that allows for the possibility that a small fraction of the containers may have failed at emplacement, or shortly thereafter, because of manufacturing flaws, construction errors, or emplacement mishandling. The parameters for the distribution in the case of aqueous corrosion processes were obtained through correlations derived from underground corrosion tests in soils.

The source term is modeled as a set of compartments in which it is assumed that all waste form surfaces are wetted immediately after the container fails. Advection and diffusion between the following compartments can be modeled: waste form; corrosion products found in the corroded section of the container; gravel backfill below and, sometimes, above the container; concrete invert (concrete matrix and fracture); and the rock matrix and fractures immediately surrounding the drift. The flux entering the container is assumed to be 5 percent of the water percolation rate times the horizontal cross-sectional area of the container. An opening at the bottom of the container is assumed to be equal in size to the opening at the top, so a flow-through model is used to mobilize waste inside the container. Radionuclides are assumed to be released congruently with the degradation of cladding and dissolution/alteration of the SNF matrix, but are constrained by their solubilities.

Results of the container failure rate model used in this PA indicate container lifetimes in the range of 10^4 to 10^5 years, including the effects of temperature, humidity and microbiologically influenced corrosion on WP degradation. Mechanical failures were not considered.

3.3.3 Nuclear Regulatory Commission/Center for Nuclear Waste Regulatory Analyses Total System Performance Assessment

The engineered barrier subsystem failure (EBSFAIL) module in TPA Version 3.1 (U.S. NRC, 1999b) and in Version 3.2 is used to calculate the failure times of the WPs from various corrosion processes. Below a critical value of RH only dry-air oxidation takes place. Above this value, humid-air corrosion occurs and at a higher critical value, aqueous corrosion begins. The aqueous environments considered in EBSFAIL are those derived, adopting several simplifications, from coupled thermal-hydrological-chemical calculations. The aqueous corrosion processes for both the outer and inner overpack are governed by the corrosion potential and the critical potential required to initiate localized corrosion. This approach uses well-established electrochemical kinetics equations for calculating the corrosion potential, which depends on environmental variables, such as temperature, oxygen partial pressure, and pH, as well as experimentally measured values of the critical potentials. The repassivation potential (E_{rp}), which depends on temperature and chloride concentration, is the critical potential used to define the occurrence of localized corrosion. Once the corrosion potential exceeds the E_{rp}, the initiation time for pitting corrosion is assumed to be negligible, but pit growth rates are calculated by using experimentally determined expressions and parameters. Failure of the WP is defined as penetration of both overpacks by a single pit or by general dissolution. The beneficial effect of galvanic coupling on the inner overpack is assessed through an equation that computes the couple corrosion potential using experimental values from the literature and an efficiency coefficient as an input parameter. A simplified mechanical failure model is included in EBSFAIL to consider the propensity of the outer steel overpack to fracture as a result of thermal embrittlement. Different container materials can be assessed by changing values of several input parameters. The E_{rp}, rates of uniform and localized corrosion, and threshold Cl^- concentration for localized corrosion can be listed, among others, as the corrosion-related parameters for the overpack materials. Yield strength and fracture toughness are the mechanical properties included as input parameters.

The engineered barrier subsystem release (EBSREL) module in TPA Version 3.2 calculates the time-dependent release of radionuclides after EBSFAIL determines the failure time of the containers. These release computations are based on the congruent dissolution of the SNF, limited by solubility. Four alternative models are considered for the dissolution of SNF. The first model consists of an empirical, parametric equation for the dependence of the dissolution rate on environmental factors obtained in flow-through tests conducted in pure carbonate solutions. These environmental factors are temperature, pH, carbonate concentration, and oxygen partial pressure. The dissolution rate in the second model is derived from data obtained in tests conducted in groundwater with the presence of Ca and Si ions acting as inhibiting species. This model provides a slower dissolution rate and represents the flow and chemical conditions expected in the case of the WP "bathtub" model (i.e., water accumulates inside the WP until it reaches a certain level, determined by the location of an exit opening). The third model allows for the use of a specified dissolution rate (e.g., derived from natural analogue studies or from drip tests). The fourth model is based on calculating the total U concentration considering five aqueous UO_2^{2+} species in equilibrium with a secondary uranyl phase, schoepite, assuming that all the radionuclides contained in the SNF matrix are incorporated in this solid phase and released congruently. Release from a perforated container can be evaluated through either a WP bathtub or flow-through models, which are considered as alternative conceptual models for the source term. Advective and diffusive transport of radionuclides away from the EBS is computed based on radionuclide mass balance in the water contacting the WP. Both EBSFAIL and EBSREL are deterministic modules incorporated in the TPA code, which provides for sampling parameter distributions in a probabilistic framework.

TPA 3.2 is the most recent update to the NRC TPA code and is currently being run. Results of sensitivity analyses are described below.

3.4 SENSITIVITY ANALYSES

Sensitivity analyses were conducted using Version 3.2 of the NRC TPA code. Specifically, the sensitivity of repository performance to the effects of container failure and radionuclide release was analyzed. (In this case, repository performance is defined as individual dose to the average member of the receptor group.) These effects, as well as the importance of parameter values used in the various models, can be determined by systematically performing sensitivity analyses. Both process-level models and the abstracted models in the TPA code can be used to ascertain the effects of container failure and radionuclide release on the performance of the repository system. Process-level models are detailed models based on fundamental principles and empirical correlations and expressions that govern container failure and radionuclide release for the range of expected conditions at the repository. Abstracted models within the NRC TPA code are designed to represent the physical processes by extracting only higher-order effects identified in process-level models. The CLST process-level models have been described in the Engineered Barrier System Performance Assessment Code (EBSPAC) Version 1.1, "Technical Description and User's Manual" (Mohanty, et al., 1997).

Details of process-level model sensitivity analyses will be provided in future revisions of this IRSR to assess the effects of container failure and radionuclide release on repository performance. Corresponding abstracted model sensitivity analyses are to be presented in the Total System Performance Assessment and Integration KTI IRSR on model abstraction. These studies are currently underway. Preliminary results of sensitivity analyses applied to the VA design of the WP and obtained with the TPA Version 3.2 code are summarized in the following paragraphs.

There are two parameters, related to the conditions prevailing in the near-field environment, that have a significant effect on container failure as a result of corrosion and, correspondingly, to radionuclide release from the EBS and to dose to a receptor group. These parameters are oxygen partial pressure and chloride concentration in the water contacting the WPs. Their main effects are to reduce the lifetime of the carbon steel overpacks after they exceed the threshold values required to promote localized corrosion, instead of uniform corrosion, at the relatively high pHs expected in the presence of a concrete liner in the emplacement drifts. Although not directly evaluated, RH has an important influence on container life because containers become wet at an earlier time when RH is higher, promoting the occurrence of aqueous uniform or localized corrosion.

(i) Although carbon steel is not considered in the SR design, sensitivity analyses were conducted to determine effects of various parameters associated with carbon steel

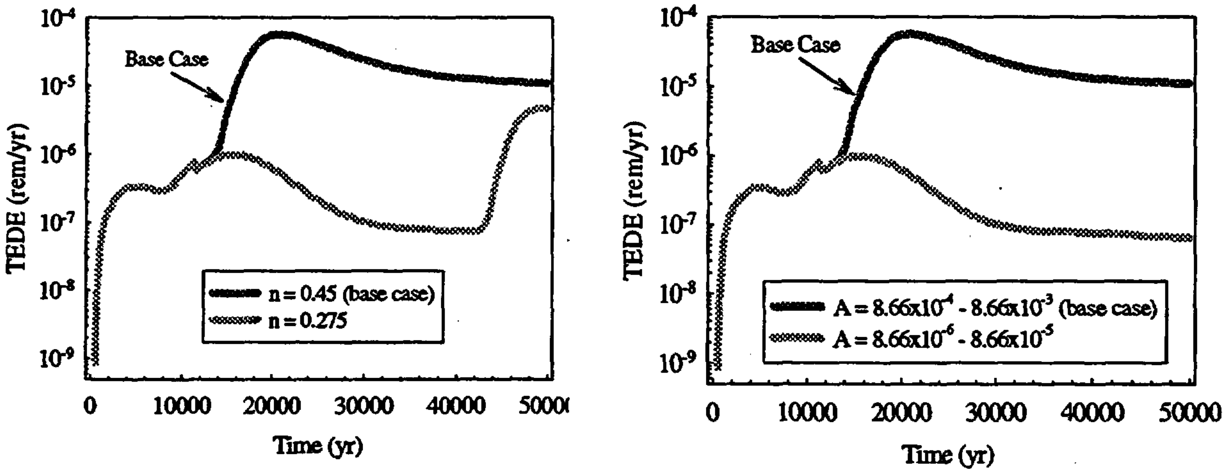


Figure 3. Effect of pit growth rate parameters for carbon steel in the VA design on dose. Sensitivity analyses were performed using TPA 3.2

corrosion in the VA design. These parameters are important for their effects on container life and, therefore, on dose. They include the critical value of RH selected for the occurrence of aqueous corrosion on the outer carbon steel overpack, the pre-exponential coefficient, and the exponent in the expression of the pit growth rate for the outer carbon steel overpack. The effect on dose of changing the pre-exponential coefficient and the exponent in the pit growth rate expression for carbon steel is illustrated in Figure 3.

(ii) Another parameter of significance is the passive corrosion rate of the inner overpack material, if localized corrosion does not take place. For Alloy 22, the inner overpack material selected by DOE in the VA design, the container failure time is significantly delayed, as demonstrated in the system-level sensitivity analysis. In this case, the effect of the passive corrosion rate value is even more important for its influence on dose, as illustrated in Figure 4 by comparing the base case with the modified case. The results shown in Figure 4 were obtained for the VA design. The observation that the reverse VA design exhibits a median failure time similar to that of the VA design in the modified case reveals that the resistance of Alloy 22 to localized corrosion is the major contributing factor to performance.

(iii) Other parameters that affect radionuclide release from failed containers and, as a consequence, dose, are SNF surface area as related to irradiated UO_2 particle or grain size, solubility of Am, and cladding as a barrier to release. Particles are fragments of irradiated UO_2 pellets, and a large number of grains in a particle can be exposed to groundwater during SNF dissolution. As expected, radionuclide releases assuming a fuel grain model are larger than those for a fuel particle model. Also, the presence of

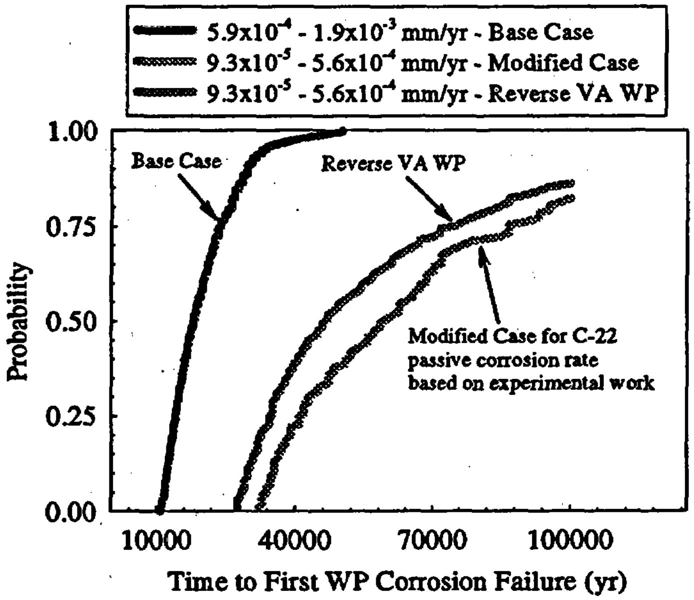


Figure 4. Effect of assumed passive current density for Alloy 22 in the VA design on predicted WP life time. Sensitivity analysis using TPA 3.2

cladding significantly decreases radionuclide release by decreasing the exposed area of the bare SNF matrix.

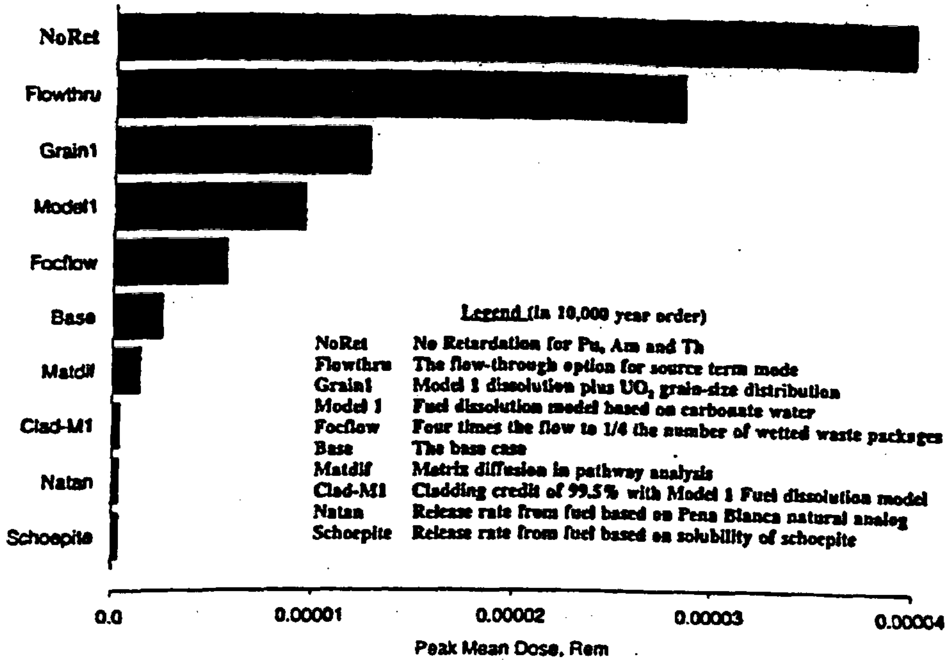
(iv) The contribution of HLW glass to dose, calculated based on a steady-state leaching model of HLW borosilicate glass, was found to be insignificant.

(v) The contribution of colloids (mainly Pu) to dose was also evaluated. The contribution of colloidal transport of Pu appears to be insignificant by considering the amount of Pu released from SNF in contact with J-13 well water. However, if Pu release increases as expected in the presence of altered groundwater, its contribution to dose can be significant.

Figure 5 shows the sensitivity of the peak mean doses at 10,000 and 50,000 years to the different dissolution models for SNF (see Section 5.3.4.1) and to various release-related parameters described above, ranked in order of importance to dose after 10,000 years. Among others, the parameters include no retardation of Pu, Am, and Th (as a simulation of potential colloid contribution), flow-through conditions, dissolution of SNF grains in pure carbonate solution, dissolution of SNF particles in pure carbonate solution, dissolution of SNF particles in groundwater with Si and Ca (base case), and cladding protection.

More detailed sensitivity analyses will be performed using improved models for radionuclide release, and the results will be updated in future revisions of this IRSR.

Peak Mean Dose for 10,000 Years, Rem



Peak Mean Dose for 50,000 Years, Rem

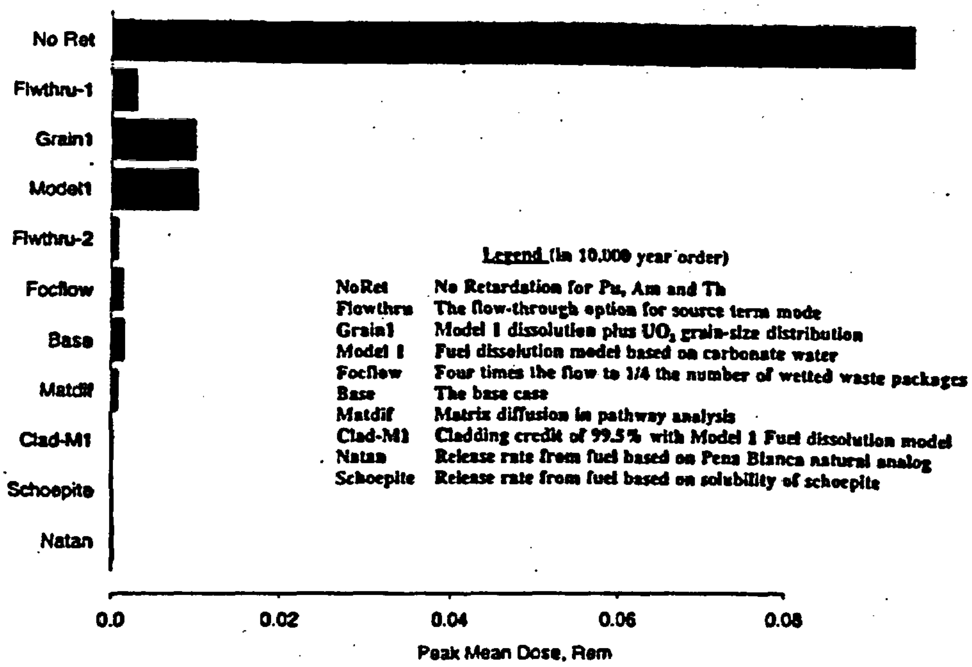


Figure 5. Effect of several spent fuel dissolution models and various input parameters on the peak mean dose

4.0 ACCEPTANCE CRITERIA AND REVIEW METHODS

The current Commission policy on repository performance is that DOE must be able to demonstrate that both engineered and natural barriers make a contribution to overall repository system performance. In this regard, the CLST primary issue (Section 2.0) relates to the contribution of the EBS to performance. Specifically, the adequacy of the EBS design will depend, in part, on DOE's demonstration that the containers will be sufficiently long-lived and that radionuclide releases will be sufficiently controlled such that the EBS contributes to overall repository system performance. DOE should address the six subissues described in Section 2.0, all of which relate directly to processes and events that affect container lifetime and radionuclide release. Resolution of these subissues will also involve the requirements set forth in the site-specific rule currently in preparation. The staff has developed acceptance criteria that, if satisfied, would resolve the CLST primary issue, the six subissues and, ultimately, questions related to the adequacy of the EBS design. The acceptance criteria are of two types—general and specific. The general or broader-level acceptance criteria are applicable to all the CLST subissues and are identified below. The general acceptance criteria are consistent with the methodology described in the ASTM Standard Practice C-1174 for prediction of the long-term behavior of EBS components in a geologic repository (American Society for Testing and Materials, 1997). These general criteria are supplemented by additional specific acceptance criteria developed for each of the six subissues, as provided in Sections 4.1 through 4.6.

<u>Acceptance Criteria Applicable to All Six Subissues</u>

(1) The collection and documentation of data, as well as development and documentation of analyses, methods, models, and codes, were accomplished under approved quality assurance and control procedures and standards.

(2) Expert elicitations, when used, were conducted and documented in accordance with the guidance provided in NUREG–1563 (Kotra, et al., 1996) or other acceptable approaches.

(3) Sufficient data (field, laboratory, and natural analog) are available to adequately define relevant parameters for the models used to evaluate performance aspects of the subissues.

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

(5) Parameter values, assumed ranges, test data, probability distributions, and bounding assumptions used in the models are technically defensible and can reasonably account for known uncertainties.

(6) Mathematical model limitations and uncertainties in modeling were defined and documented.

(7) Primary and alternative modeling approaches consistent with available data and current scientific understanding were investigated and their results and limitations considered in evaluating the subissue.

(8) Model outputs were validated through comparisons with outputs of detailed process models, empirical observations, or both.

(9) The structure and organization of process and abstracted models were found to adequately incorporate important design features, physical phenomena, and coupled processes.

4.1 SUBISSUE 1: THE EFFECTS OF CORROSION PROCESSES ON THE LIFETIME OF THE CONTAINERS

This subissue relates to the adequacy of DOE's consideration of the effects of corrosion processes on the lifetime of the containers. Resolution of this subissue will be accomplished through the application of the generic acceptance criteria specified in Section 4.0 and the particular acceptance criteria identified in Section 4.1.1.

4.1.1 Acceptance Criteria for Subissue 1

(1) DOE has identified and considered likely modes of corrosion for container materials, including dry-air oxidation, humid-air corrosion, and aqueous corrosion processes, such as general corrosion, localized corrosion, MIC, SCC, and hydrogen embrittlement, as well as the effect of galvanic coupling.

(2) DOE has identified the broad range of environmental conditions within the WP emplacement drifts that may promote the corrosion processes listed previously, taking into account the possibility of irregular wet and dry cycles that may enhance the rate of container degradation.

(3) DOE has demonstrated that the numerical corrosion models used are adequate representations, taking into consideration associated uncertainties, of the expected long-term behaviors and are not likely to underestimate the actual degradation of the containers as a result of corrosion in the repository environment.

(4) DOE has considered the compatibility of container materials, the range of material conditions, and the variability in container fabrication processes, including welding, in assessing the performance expected in the container's intended waste isolation function.

(5) DOE has justified the use of data collected in corrosion tests not specifically designed or performed for the YM repository program for the environmental conditions expected to prevail at the YM site.

(6) DOE has conducted a consistent, sufficient, and suitable corrosion testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further

testing to reduce any significant area(s) of uncertainty as part of the performance confirmation program.

(7) DOE has established a defensible program of corrosion monitoring and testing of the engineered subsystems components during the performance confirmation period to assure they are functioning as intended and anticipated.

4.1.2 Technical Bases for Acceptance Criteria for Subissue 1

Repository regulatory requirements recognize that the engineered barriers provided to isolate radioactive wastes for long periods of time will eventually degrade. It is anticipated that the primary cause for engineered barrier degradation under normal conditions (i.e., in the absence of disruptive events, such as seismicity, faulting, or volcanism) will be caused by one or more corrosion processes. Both DOE and the staff evaluated the most likely forms of materials degradation for the candidate container materials of interest (Farmer, et al., 1988; Gdowski, 1991; Cragnolino and Sridhar, 1991, 1992; Geesey, 1993; Sridhar, et al., 1994; U.S. Nuclear Regulatory Commission, 1997; Cragnolino, et al., 1999). These degradation modes include dry-air oxidation of container materials during the initial hot, dry period following emplacement of the WPs in the drifts. After this initial period, the drift wall and container surface temperatures will decrease and the container materials will be subjected to humid-air corrosion and various modes of aqueous corrosion, i.e., general corrosion, localized corrosion, MIC, SCC, and hydrogen embrittlement. It is hypothesized that each form of corrosion can initiate at a time when the RH exceeds a critical value. Under these conditions, it is expected that humid-air corrosion will occur in the presence of a thin surface film of condensed fluid in contact with water vapor above the surface. As the temperature continues to decrease, aqueous corrosion will occur as a result of the formation of a thicker film of condensed fluid that behaves as bulk water. In addition, groundwater enriched in salts through evaporation and rock-water interactions may drip on the WPs through fractures in the rock. The influx of water is determined by various thermo-hydrological processes, such as heat-pipe effects, gravity-driven refluxing, and percolation of meteoric water. A detailed discussion of these coupled thermo-hydrological-chemical processes is presented in the ENFE KTI IRSR. The importance of various modes of materials degradation and the corresponding effects on barrier performance are dependent on WP design and construction, materials selection, and the environment interacting with those materials. The chemical composition of the environment contacting the WP components depends on the evolution of the near-field environment. Information about this evolution is, therefore, a requirement for evaluating WP corrosion. The specific consideration of corrosion modes associated with weldments is necessary because it is well known that in many engineered structures and components, welded joints are more prone to corrosion failure than the base metal.

Resolution of Subissue 1 will necessitate identification of the most important modes of container degradation resulting from corrosion processes; numerical estimates of the effects of corrosion on container lifetime; assessment of the performance of the WPs, including material and fabrication methods for the containers; adequacy and validity of the available corrosion database; and confirmation of the intended performance, through monitoring and testing during the preclosure period.

4.2 SUBISSUE 2: THE EFFECTS OF PHASE INSTABILITY AND INITIAL DEFECTS ON THE MECHANICAL FAILURE AND LIFETIME OF THE CONTAINERS

This subissue relates to the adequacy of DOE's consideration of container materials stability and mechanical failure. Container failures resulting from degradation of mechanical properties because of material instability, combined with the action of residual and/or applied stresses, can lead to release of radionuclides from the EBS. Resolution of this subissue will be through the application of the generic acceptance criteria specified in Section 4.0 and the particular acceptance criteria identified in Section 4.2.1.

Disruptive events, such as seismic activity, volcanism, and faulting may promote premature failure of the containers through different processes. For example, as a consequence of seismic events, mechanical stresses may promote either brittle or ductile fracture of the container, depending on the degree of embrittlement of the container material and the magnitude and location (weldments, base metal) of the applied stresses. The component of this subissue related to the coupling of disruptive events and container material properties will be covered in future revisions of this IRSR.

4.2.1 Acceptance Criteria for Subissue 2

(1) DOE has identified and considered the relevant mechanical failure processes that may affect the performance of the proposed container materials.

(2) DOE has identified and considered the effect of material stability on mechanical failure processes for the various container materials as a result of prolonged exposure to the expected range of temperatures and stresses, including the effects of chemical composition, microstructure, thermal treatments, and fabrication processes.

(3) DOE has demonstrated that the numerical models used for container materials stability and mechanical failures are effective representations, taking into consideration associated uncertainties, of the expected materials behavior and are not likely to underestimate the actual rate of failure in the repository environment.

(4) DOE has considered the compatibility of container materials and the variability in container manufacturing processes, including welding, in its WP failure analyses and in the evaluation of radionuclide release.

(5) DOE has identified the most appropriate methods for nondestructive examination of fabricated containers to detect and evaluate fabrication defects in general and, particularly, in seam and closure welds.

(6) DOE has justified the use of material test results not specifically designed or performed for the YM repository program for environmental conditions (i.e., temperature, stress, and time) expected to prevail at the proposed YM repository.

(7) DOE has conducted a consistent, sufficient, and suitable materials testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further

testing to reduce any significant area(s) of uncertainty as part of the performance confirmation program.

(8) DOE has established a defensible program of monitoring and mechanical testing of the engineered subsystems components, during the performance confirmation period, to assure they are functioning as intended and anticipated, in the presence of thermal and stress perturbations.

4.2.2 Technical Bases for Acceptance Criteria for Subissue 2

Repository regulatory requirements recognize that the engineered barriers provided to isolate radioactive wastes for long periods, in addition to being degraded by corrosion processes, may be affected by material instability (i.e., degraded mechanical properties) owing to prolonged exposure to relatively high temperatures. The staff evaluated the most likely processes affecting material stability for both outer and inner overpack materials (Sridhar, et al., 1994; Cragnolino, et al., 1996; 1999).

Thermal embrittlement of carbon and low-alloy steels occurs when impurities originally present in the steel, mainly P, segregate to grain boundaries during thermal exposure. The segregation of P may result in reduction of fracture toughness due to a change in the low-temperature fracture mode from transgranular cleavage to intergranular fracture, promoting container failure that can be initiated at flaws under the effect of an impact. Preliminary calculations (Cragnolino, et al., 1998) suggest that significant grain boundary P segregation and, hence, the potential for a substantial degradation in toughness of steels, may occur only as a consequence of long-term thermal aging at repository temperatures anticipated for the high heat-loading concept (i.e., temperatures greater than 200 °C for several thousand years). At lower temperatures, it appears that thermal embrittlement should not be a matter of concern, even though it should be noted that low-alloy steels are more susceptible to this phenomenon than plain carbon steels. Although A-516 carbon steel was the material of choice for the overpack in the VA, it is no longer considered for the SR design. It is discussed here, however, for completeness and to document the results of confirmatory studies of the earlier design.

Alloy 22, the material currently selected for the outer overpack, experiences an ordering transformation when heated in the temperature range of 250 to 550 °C (Sridhar, et al., 1994; Cragnolino, et al., 1999). The result is an increase in the work-hardening rate and, as a consequence, an enhanced susceptibility to SCC and hydrogen embrittlement. Another possible cause of thermal instability in Alloy 22 arises from the precipitation of brittle intermetallic phases. The existence of long-range ordering of Alloy 22 and the absence of μ phase for aging times of 30,000 and 40,000 hours (3.4 and 4.6 years) at 425 °C has been reported recently (Rebak and Koon, 1998). However, the effect of cold work before thermal treatment on ordering transformation or intermetallic precipitation has not been studied. For Alloy 22, as for carbon steels, these effects are more likely to be a concern at high heat-loading.

The necessary stresses for mechanical failure to occur as a consequence of processes that cause material instability may arise from residual stresses generated as a result of welding operations; from stresses associated with the buildup of corrosion products in the gap between the outer and the inner overpacks; and applied stresses from the effect of disruptive events, such as seismic activity, volcanism, faulting, or a combination of these events.

The detection of fabrication defects associated, among other causes, with welding operations for both seam and closure welds requires the application of appropriate nondestructive examination methods. This fact or is important in reducing the incidence of initial failures in the performance of the WPs, a critical factor in calculated doses (Sridhar, 1999).

Resolution of Subissue 2 will necessitate identification of the most important modes of mechanical failure, an evaluation of the potential for materials instability, the effect of disruptive events on the probability of failures, numerical estimates of the effect of these processes on container lifetime, assessment of the performance of the containers regarding materials selection and fabrication methods, adequacy and validity of the available database of mechanical properties, including toughness, identification of appropriate nondestructive examination methods, and confirmation of the intended performance through monitoring and testing during the preclosure period.

4.3 SUBISSUE 3: THE RATE AT WHICH RADIONUCLIDES IN SPENT NUCLEAR FUEL ARE RELEASED FROM THE ENGINEERED BARRIER SUBSYSTEM THROUGH THE OXIDATION AND DISSOLUTION OF SPENT FUEL

This subissue relates to the adequacy of DOE consideration of the effect of the rate of degradation of SNF on the subsequent release of radionuclides and the rate of release from the EBS. Resolution of this subissue will be through application of the generic acceptance criteria specified in Section 4.0 and the particular acceptance criteria identified in Section 4.3.1.

4.3.1 Acceptance Criteria for Subissue 3

(1) DOE has considered all categories of SNF planned for disposal at the proposed YM repository.

(2) DOE has adequately justified the selection of radionuclides tracked in the release models from SNF and their related release parameters.

(3) DOE has identified the range of environmental conditions to be expected inside breached WPs.

(4) DOE has identified and considered likely processes for SNF degradation and the release of radionuclides from the EBS, as follows: dissolution of the irradiated UO_2 matrix, with the consequent formation of secondary minerals and colloids; prompt release of radionuclides; degradation in the dry air environment; degradation and failure of fuel cladding; preferential dissolution of intermetallics in DOE SNF; and release of radionuclides from the WP emplacement drifts.

(5) DOE has demonstrated that the numerical models used for SNF degradation and radionuclide release from the EBS are adequate representations, including

consideration of uncertainties, of the expected SNF performance and are not likely to overestimate the actual performance in the repository environment.

(6) DOE has considered the compatibility of SNF and the internal components of the WP, such as the basket materials, in the evaluation of radionuclide releases. Specifically, the SNF should not compromise the performance of the WP.

(7) DOE has justified the use of SNF test results not specifically collected for the YM site for the environmental conditions expected to prevail after breaching of the containers at the YM site.

(8) DOE has conducted a consistent, sufficient, and suitable SNF corrosion and radionuclide release testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further testing to reduce any significant area(s) of uncertainty as part of the performance confirmation program.

(9) DOE has established an adequate program of monitoring radionuclide release from the WP during the performance confirmation period, to assure that assumptions and calculations of SNF dissolution and radionuclide release from the WP are appropriately substantiated.

4.3.2 Technical Bases for Acceptance Criteria for Subissue 3

It is expected that after the failure of the metallic container, there will be a gradual release of radionuclides from the EBS over long periods of time. It is anticipated that a primary controlling factor for this slow release is the low rate of dissolution of SNF. Both DOE and the staff evaluated the most likely forms of SNF degradation (Manaktala, 1993; TRW Environmental Safety Systems, Inc., 1997a; U.S. Nuclear Regulatory Commission, 1997). These degradation modes include dry-air oxidation of the fuel cladding and matrix inside any breached container (i.e., by juvenile failure) during the initial period following repository closure, when the environment contacting the WP is considered to be hot and dry air. After failure of containers by any of the various corrosion or mechanical processes described in Sections 4.1 and 4.2, the fuel cladding will be subjected to modes of failure somewhat similar to those anticipated for the failure of containers. As the cladding is breached, the fuel matrix consisting of irradiated UO_2 will be dissolved either by groundwater partially filling the failed containers, or by groundwater dripping on the SNF surfaces. As an alternative process, fuel dissolution may occur by the action of humid air condensed as a thin film of water on the SNF. The chemical composition of the water inside breached WPs needs to be known or at least bounded to evaluate the dissolution of SNF. Initially, radionuclides in the gap between cladding and the SNF matrix and in the fuel grain boundaries will be released promptly. Following this prompt release, radionuclides will be released as the SNF matrix dissolves. Whereas high-solubility radionuclides, such as ^{99}Tc or ^{129}I will be released congruently with the dissolution of the matrix, U and low-solubility radionuclides, such as ^{239}Pu or ^{241}Am, will be reprecipitated on the surface of the irradiated UO_2 as secondary minerals. Consequently, the release of low-solubility radionuclides from the EBS will be determined thermodynamically by solubility limits or kinetically by steady-state concentration values controlled by the formation of secondary minerals. The low-solubility radionuclides may also form colloids that become a vehicle for the mobilization of radionuclides in larger concentrations than those of soluble species limited by solubility. Once radionuclides are mobilized, they can be transported away from the EBS either by diffusion or by advection, depending on the various modes of groundwater contact with the SNF, such as dripping, partial or full immersion, or vapor condensation.

Resolution of Subissue 3 will require consideration of all the categories of SNF to be disposed, the identification of the most important modes of degradation owing to various forms of water contact and water chemistry inside WPs for the different types of SNF; numerical estimates of the rate of SNF dissolution and the rate of radionuclide release from the EBS; assessment of the effects of the compatibility of SNF and internal components of the WP on radionuclide release; adequacy and validity of the available SNF database, including data on interactions with groundwater; and confirmation of the intended performance, through monitoring and testing during the preclosure period.

4.4 SUBISSUE 4: THE RATE AT WHICH RADIONUCLIDES IN HIGH-LEVEL WASTE GLASS ARE RELEASED FROM THE ENGINEERED BARRIER SUBSYSTEM

This subissue relates to the adequacy of DOE's consideration of the effects of degradation of HLW glass, taking into account the rate of degradation and its effect on the rate of radionuclide releases from the EBS. Resolution of this subissue will be accomplished through the application of the generic acceptance criteria specified in Section 4.0 and the particular acceptance criteria identified in Section 4.4.1.

4.4.1 Acceptance Criteria for Subissue 4

(1) DOE has taken into account all types of HLW glass planned for YM disposal.

(2) DOE has adequately justified the selection of radionuclides tracked in the release models from HLW glass and their related release parameters.

(3) DOE has identified the range of environmental conditions to be expected inside breached WPs containing HLW glass and eventually certain types of SNF as in the codisposal WPs.

(4) DOE has identified and considered likely processes for the degradation of HLW glass and the release of radionuclides from the EBS, i.e., dissolution of the primary phase; formation of secondary minerals and colloids; microbial action; and radionuclide releases and transport from the WP emplacement drifts.

(5) DOE has demonstrated that the numerical models used for determining the rate of dissolution of HLW glass and the rate of radionuclide release from the EBS are adequate representations, taking into consideration the associated uncertainties, of the expected HLW glass performance, and are not likely to underestimate the actual rate of degradation of the HLW glass and the subsequent rate of release in the repository environment.

(6) DOE has assessed the compatibility of HLW glass with internal components of the WP in the evaluation of radionuclide release, taking into consideration co-disposal with DOE-owned SNF in the same WP. Specifically, HLW glass should not compromise the performance of the WP.

(7) DOE has justified the use of test results for HLW glass not specifically collected for the YM site for environmental conditions expected to prevail after breaching of the containers at the YM site.

(8) DOE has conducted a consistent, sufficient, and suitable HLW glass and radionuclide release corrosion testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further testing to reduce any significant area(s) of uncertainty as part of the performance confirmation program.

(9) DOE has established an adequate program of monitoring radionuclide release from the WP during the performance confirmation period to assure that assumptions and calculations regarding HLW glass dissolution and radionuclide release are appropriately substantiated.

4.4.2 Technical Bases for Acceptance Criteria for Subissue 4

After the failure of the metallic container, it is expected that the EBS would provide gradual release of radionuclides over long periods. It is anticipated that limited mobilization of radionuclides arising from the low rate of dissolution of HLW glass will contribute to the slow release of radionuclides from the EBS. Both DOE and staff evaluated the most likely modes of HLW glass degradation (TRW Environmental Safety Systems, Inc., 1997a; Manaktala, 1992). These degradation modes include hydration and leaching of primary HLW glass phases and formation of secondary minerals and colloids. High-solubility radionuclides, such as ^{99}Tc, will be released congruently with the silica dissolution under slow water-replenishing conditions. The silica will dissolve at long-term dissolution rates under repository conditions. On the other hand, low-solubility radionuclides, such as ^{239}Pu or ^{241}Am will be reprecipitated with secondary minerals on the surface of the primary HLW glass phase. Consequently, the release of low-solubility radionuclides will be controlled by solubility limits or steady-state values that are constrained by the solubility of secondary minerals. These low-solubility radionuclides may also form mobile colloids that can carry larger amounts of radionuclides than those transported in the form of dissolved species. Once radionuclides are mobilized, they will be transported either by diffusion or by advection, depending on the water contact modes with the WP (i.e., dripping, partial or full immersion, or condensation of water vapor) under conditions that can be altered by backfill emplacement. The chemical composition of the water inside breached WPs should be known or properly bounded.

Resolution of Subissue 4 will require consideration of all types of HLW glass planned for YM disposal, identification of the most important modes of degradation of HLW glass, numerical estimates of the rate of HLW glass dissolution and the rate of radionuclide release from the EBS taking into consideration the water chemistry, assessment of the effects of the compatibility of HLW glass and internal components of the WP on radionuclide release, and confirmation of the intended performance through monitoring and testing during the preclosure period.

4.5 SUBISSUE 5: THE EFFECTS OF IN-PACKAGE CRITICALITY ON WASTE PACKAGE AND ENGINEERED BARRIER SUBSYSTEM PERFORMANCE

This subissue addresses whether DOE has sufficiently analyzed the effects of potential in-package nuclear criticality on repository performance during the repository operations period, and over the postclosure time frame of interest. If the WP is designed to prevent any nuclear criticality over the preclosure and postclosure periods, the adequacy of the design of the WP and other components of the EBS must be demonstrated. If allowing in-package criticality is allowed during post-closure, with a probability greater than or equal to the event probability defined in the draft 10 CFR Part 63, the consequences of the in-package criticality, and the contribution to total repository risk, must be determined. Resolution of the in-package criticality subissue will be through the application of the generic acceptance criteria specified in Section 4.0 and the particular acceptance criteria identified in Section 4.5.1 for in-package criticality.

4.5.1 Acceptance Criteria for Subissue 5

(1) DOE has used sound technical bases for selecting the design criteria for components to mitigate any potential effects of in-package criticality on the repository performance. These design criteria may include development of subcritical limit, probability and consequence of criticality, and any other design criteria considered to be necessary by DOE.

(2) DOE has identified all the features, events, and processes that may increase the reactivity of the system inside the WP. The acceptance criteria provided for the Scenario Analysis subissue in the Total System Performance Assessment and Integration (TSPAI) IRSR must also be considered.

(3) DOE has identified the configuration classes and configurations that have potential for nuclear criticality. If models are used to develop the configurations, approach and accuracy in modeling verification and validation will be evaluated.

(4) DOE has developed a technically defensible, transparent, and traceable method in assigning probability values to each of the scenario classes, scenarios, configuration classes, and configurations.

(5) DOE has developed appropriate computer models, input parameters, and determined quantitative values for calculating the effective neutron multiplication factor (k_{eff}), including appropriate biases and uncertainties in the model.

(6) DOE has developed appropriate computer models, evaluated input parameters, and determined quantitative values for calculating the radionuclide inventory, heat, kinetic energy, and other parameters that would change as a result of k_{eff} exceeding the subcritical limit developed under Criterion (1).

(7) DOE has determined the risk contribution from the in-package criticality to the total repository system performance appropriately.

4.5.2 Technical Bases for Acceptance Criteria for Subissue 5

The majority of the SNF, currently planned to be placed in the WP, is commercial PWR, BWR, and some Mixed Oxide (MOX) fuel. Navy SNF and DOE-owned SNF, which includes spent fuels from research reactors, are also planned to be placed in the repository. Vitrified HLW glass logs with, and without, excess weapons fissile materials, are other types of DOE waste destined for the repository.

To minimize the potential for, and consequences of, criticality, DOE should develop a set of design criteria to support a demonstration that the design of the WP and EBS is adequate. Given the different types of waste to be placed in the WPs, the DOE may choose different design criteria for different waste types. For example, if DOE chooses to take credit for the burn-up of commercial spent fuel, the subcritical limit established for the commercial spent fuel must adequately incorporate the biases and uncertainties associated with the neutronics code with regard to isotopic and k_{eff} prediction. The approach in incorporating these biases and uncertainties must have a sound and defensible basis. On the other hand, if DOE chooses not to take any credit for burn-up of DOE SNF, because of the difficulty in determining the burn-up history, the subcritical limit established as part of the design criteria may include only biases and uncertainties associated with the neutronics code in predicting k_{eff} for the fresh fuel. Other design criteria established by DOE must be technically defensible to assure that in-package nuclear criticality events will not have an unacceptable effect on the repository system.

DOE must identify all the features, events, and processes that have the potential to increase the reactivity of the in-package system. The features, events, and processes considered for the TSPA must also be evaluated in terms of their effects on producing conditions favorable for in-package criticality. For example, events such as igneous activity (extrusive and intrusive), seismic shaking (high-frequency, low-magnitude, and rare large magnitude events), tectonic evolution (slip on existing faults and formation of new faults), climatic change (change to pluvial conditions) mentioned in TSPAI IRSR must be evaluated in terms of their effect on WP geometry and content with respect to in-package criticality.

Uncertain parameters associated with package configurations resulting from events and processes must be identified. Selection of the distribution and range of values for uncertain parameters associated with a configuration must be technically defensible. For example, the value chosen for the amount of iron oxide, produced from corrosion of the WP internal components remaining inside the WP must be technically defensible. If models are used to predict the iron oxide inventory in the WP, the acceptability of the model through validation and verification must be established.

The probabilities for those configurations that result in critical conditions must be determined for screening purposes. In developing the approach for the probability calculations, DOE needs to consider the range of parameters that could affect the probability distribution. If the parameter values are based on expert elicitation and the values span a wide range, the resulting probabilities can be skewed. Any approach selected in developing the probability values for critical configurations must be based on technically defensible ranges of parameter values.

In determining the consequences of critical (i.e., exceeding the subcritical limit established under design criteria) and supercritical configurations, DOE must consider different processes that could cause an increase in k_{eff}, leading to varying rates of reactivity insertion that could cause a criticality event. Criticality must be analyzed under steady-state, transient, and autocatalytic conditions if it is credible that these conditions exist within the WP. The rate of reactivity insertion must be determined and consequence calculations performed under slow and step insertion conditions if these conditions are credible. The consequences for critical and supercritical configurations must be analyzed with respect to radionuclide production, heat generation, and WP degradation, which includes that of the waste form. The consequences of additional heat generation and large energy release over very small time frames must be analyzed in terms of their potential effects on the WP, EBS, and repository environment.

To determine the risk that may result from in-package criticality, DOE must consider the direct and indirect consequences, and the probabilities associated with them. Increase in radionuclide inventory, production of additional heat, and increase in WP degradation are the direct consequences of configurations that may result from criticality events. Indirect effects include those affecting other components of the EBS and the near-field environment that could result in additional release of radionuclides. For example, a step reactivity insertion could affect the rock stability, especially when there is no backfill. Further instability of the rock formation would result in higher probability of rockfall on the WP. Therefore, the calculated risk must be based on the primary and the secondary consequences of the critical configurations.

4.6 SUBISSUE 6: THE EFFECTS OF ALTERNATE ENGINEERED BARRIER SUBSYSTEM DESIGN FEATURES ON CONTAINER LIFETIME AND RADIONUCLIDE RELEASE FROM THE ENGINEERED BARRIER SUBSYSTEM

This subissue is designed to address the effects of alternate EBS design features, such as backfill, drip shields, and ceramic coatings, on container lifetime and radionuclide release from the EBS. These design features were presented as options for the improvement of the EBS performance in the VA (U.S. Department of Energy, 1998b). Consideration of this subissue in the CLST IRSR, Revision 2, is limited because the design proposed SR design is not completed (TRW Environmental Safety Systems, 1999), NRC is focusing its review on the alternative design features listed in the VA, and the most recent information regarding EDA-II (Howard, 1999). Therefore, some discussion is warranted because some of these items will be included in the final LA design.

4.6.1 Acceptance Criteria for Subissue 6

(1) DOE has identified and considered the effects of backfill, and the timing of its emplacement, on the thermal loading of the repository, WP lifetime (including container corrosion and mechanical failure), and the release of radionuclides from the EBS.

(2) DOE has identified and considered the effects of ceramic coating on WP lifetime, including negative consequences as a result of breakdown of the ceramic coating (cracking, spalling, or delamination) in response to the action of environment,

manufacturing defects, mechanical impacts and stresses arising from a multiplicity of sources, and the potential for enhanced localized corrosion of the containers that might occur at cracks or perforations in the ceramic coating layers.

(3) DOE has evaluated the compatibility of ceramic coating materials with outer overpack materials and the combined effect of ceramic coating with backfill on container lifetime.

(4) DOE has identified and considered the effects of drip shields (with backfill) on WP lifetime, including extension of the humid-air corrosion regime, environmental effects, breakdown of drip shields and resulting mechanical impacts on WP, the potential for crevice corrosion at the junction between the WP and the drip shield, and the potential for condensate formation and dripping on the underside of the shield.

(5) DOE has evaluated the effect of design changes in container wall thickness that may increase γ-radiolysis of the water contacting WPs and, therefore, enhance the possible occurrence of localized corrosion processes.

(6) DOE has identified the chemical composition of the water in the environment surrounding the WPs and its evolution with time.

(7) DOE has justified the use of test results for drip shields, ceramic coatings, and backfill materials not specifically collected for the YM site for the environmental conditions expected to prevail at the proposed YM repository.

(8) DOE has conducted a consistent, sufficient, and suitable corrosion testing program at the time of the LA submittal. In addition, DOE has identified specific plans for further testing to reduce any significant area(s) of uncertainty as part of the performance confirmation program.

4.6.2 Technical Bases for Acceptance Criteria for Subissue 6

If ceramic coatings are placed on the WP, it is believed that the initiation of humid-air corrosion can be delayed, extending the lifetime of the containers. Ceramics have the advantage of being thermodynamically stable; they are the endpoint of oxidation. Natural analogs of ceramics are millions of years old, and some of the oldest human artifacts are ceramics, such as Egyptian terra cotta vases dated to 5000 B.C. (Bose, 1947). Thus, it is expected that a ceramic component on the WP could provide defense-in-depth by protecting the outer overpack material from corrosion from moisture or water drips. For a ceramic to perform this function, it must: i) be seamless and cover the entire outer overpack without any cracks; ii) have sufficient thickness to minimize diffusion of oxygen and water; iii) have minimum interconnectivity of pores and sufficient density to avoid penetration of oxygen and water; and iv) be chemically durable under repository conditions. The ceramic coating must be sufficiently robust to withstand mechanical loads (e.g., falling off a degraded pedestal, seismic loads, and loads caused by the mass of the collapsed drift on the WP, at a later time). It is assumed that ceramic coatings will not be used without backfill because impact loading from rock fall in the absence of backfill may crack the ceramic coating. The elimination of carbon steel as a material of choice for the disposal containers implies that ceramic coatings are currently not being considered as an alternative feature for improving the performance of the WPs.

Nevertheless, for the sake of completeness, a brief discussion on ceramic coatings is included in this IRSR, Revision 2.

Use of a drip shield design option is intended to minimize the possibility of water dripping on containers. Corrosion of the containers can be enhanced by the presence of flowing liquid water that may also remove corrosion products facilitating localized penetration. In addition, liquid water can mobilize and advectively transport most radionuclides. Where active flowing fractures in the repository couple with sharp drift wall edges, seeps (drips) into the drift can occur. It is the function of the drip shield to divert these drips from the WP surface. Although moisture condensation between the WP and the drip shield cannot be prevented, drip shield will reduce water contact arising from fracture flow. If the diverted seepage flux is drained without passing through the part of the invert contaminated by the radionuclides from failed WPs, diffusive transport will be controlled within the EBS, and dose rates will be greatly reduced. Thus, part of the design of an effective drip shield is to physically separate the water that it sheds from water that condenses under it and flows through breached WPs. For VA, the drip shield may be a cylinder or half cylinder surrounding or supported by the WP itself. It is assumed that backfill will be included whenever drip shields are considered because rock fall can degrade the performance of an otherwise long-lived ceramic or corrosion-resistant-metal drip shield.

The description of the two previous alternative design components includes scenarios in which the concrete liner and parts of the near-field rock fall into the drift. Covering the WP with backfill protects other components from damage from such mechanical loads. Therefore, DOE management has directed that backfill be included whenever drip shields or ceramic coatings are considered. Backfill must also be considered in relation to areal mass loading because the introduction of backfill promotes an increase in the temperature inside the emplacement drifts. Increased temperature in the drifts can delay the onset of humid-air corrosion and aqueous corrosion, but may also affect cladding lifetime and thermal stability of WP materials.

Resolution of Subissue 6 will require identification of the effects of selected design option(s) on WP lifetime and the release of radionuclides from the EBS.

In EDA-II, which has been recommended to DOE for the SR, a design consisting of a 2-cm thick Alloy 22 outer overpack surrounding a 5-cm-thick type 316NG stainless steel overpack that provide structural strength was adopted. The introduction of a thinner overpack as compared to the VA design raises concerns related to the effect of γ-radiolysis and the resulting oxidizing conditions. Additionally, the presence of steel beams on the floor, instead of concrete, despite the possible addition of a nonelectrically conductive ballast, may result in differences in the electrochemical conditions in the vicinity of the WPs and the drip shield (including the effects of corrosion products) that need to be considered.

4.7 REVIEW METHOD FOR ALL SUBISSUES

Issue resolution with DOE will be pursued through continuation of the ongoing prelicensing consultation and interaction. Staff will review the following items: DOE's site characterization progress reports, in relation to the further development of container design and materials selection; EBS design documents, such as the "Mined Geologic Disposal System Advanced Conceptual Design Report" (TRW Environmental Safety Systems, Inc., 1996a); the "Viability

Assessment of a Repository at Yucca Mountain" (U.S. Department of Energy, 1998b), the planned FY2000 SR; future repository iterative PAs and sensitivity studies; data from ongoing research and testing on container materials and waste forms; outcomes of peer reviews or expert elicitations on EBS components; and results of independent staff analyses, studies, and evaluations of the EBS. Staff will focus its review on the leading candidates for the container materials and likely modes of degradation, the overall design characteristics or features of the containers, the design basis for the containers, and the container fabrication process, as well as the numerical assessments of container performance.

For the waste forms (Subissues 3 and 4), the staff will review waste form degradation processes and comparative releases in both dry air and aqueous environments, including fuel dissolution, HLW glass leaching, formation of secondary minerals and colloids, cladding degradation, and mobilization of radionuclides in the EBS.

Numerical assessments will be performed using the most up-to-date version of the TPA code. The acceptance criteria will be used to evaluate DOE's demonstration that the containers will be sufficiently long-lived and radionuclide releases sufficiently controlled and that the EBS contributes to overall repository system performance.

When necessary, separate auxiliary analyses will be conducted to evaluate, in detail, process models and sensitivity of the performance to the variation of critical parameters that cannot be adequately assessed by using the TPA Code. If required, independent and confirmatory testing will be used to ascertain the reliability and applicability of the data provided by DOE.

Specific attention will be given to the DOE plans for monitoring and testing during the performance confirmation period as a result of concerns regarding the anticipated lack of sufficient data at the time of the LA.[1] This effort will focus on evaluating and assessing techniques and tools identified by the DOE for testing and monitoring the performance of the WPs and the EBS, including the quantity and chemistry of the water contacting the WPs, container corrosion, and gaseous and aqueous radionuclide release.

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[1] Letter (June 2, 1999) to Lake Barrett from Carl Paperiello concerning U.S. Nuclear Regulatory Commission Staff Review of the U.S. Department of Energy Viability Assessment for a High-Level Radioactive Waste Repository at Yucca Mountain, Nevada.

5.0 STATUS OF SUBISSUE RESOLUTION AT THE STAFF LEVEL

In prior years, staff raised detailed concerns and questions about DOE's site characterization and PA program in areas related to this KTI. These concerns and questions were documented in "NRC Staff Site Characterization Analysis of the U.S. Department of Energy's Site Characterization Plan, Yucca Mountain Site, Nevada" (U.S. Nuclear Regulatory Commission, 1989).

Recently, the staff recognized the need to refocus the prelicensing repository program on resolving issues most significant to repository performance. The status of the staff refocused efforts, including general descriptions of the primary issues and concerns in the 10 HLW program subject areas of interest (i.e., 10 KTIs), was documented in "NRC High-Level Radioactive Waste Program Annual Progress Report: Fiscal Year 1996" (U.S. Nuclear Regulatory Commission, 1997).

Additional comments and concerns related to the 10 KTIs were reported in four documents related to DOE's 1995 iterative PA: (i) NRC/CNWRA Audit Reviews of DOE's TSPA-95 (Austin, 1996a,b,c); and (ii) "NRC/CNWRA Detailed Review of Selected Aspects of Total System Performance Assessment—1995" (Baca and Jarzemba, 1997).

Continuing staff efforts to resolve the issues, concerns, and questions identified in these reports resulted in further refinement and clarification of the primary issue and subissues in the CLST subject area of interest, as described in Section 2.0 of this IRSR. Comments arising from the review of DOE VA were documented in the attachment to the letter to Barrett from Paperiello.[2]

In the following sections, a summary is provided on the status of resolution for each of the subissues described in Section 2.0, including the detailed open items resulting from the staff site characterization analysis.

5.1 STATUS OF RESOLUTION OF SUBISSUE 1 AND RELATED OPEN ITEMS

5.1.1 Dry-Air Oxidation of Outer Overpack

Carbon steel is not considered as an outer overpack material in any of the EDA and is not considered as either an inner or outer overpack material in EDA-II, the most favored WP design in the DOE SR plan. Additionally, a relatively low thermal loading strategy is envisioned by DOE for the proposed SR. Hence, dry oxidation of carbon steel is no longer a component of this subissue. However, the discussion on dry oxidation of carbon steel is summarized in this section to provide completeness in addressing the VA design and to document the results of confirmatory studies on this design.

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[2] Letter (June 2, 1999) to Lake Barrett from Carl Paperiello concerning U.S. Nuclear Regulatory Commission Staff Review of the U.S. Department of Energy Viability Assessment for a High-Level Radioactive Waste Repository at Yucca Mountain, Nevada.

After emplacement and for an extended period that may last several thousand years, the environment in contact with the WPs is expected to be hot, dry air. Under dry conditions corresponding to RHs lower than approximately 65 percent, the carbon steel may undergo dry oxidation. Currently, DOE postulates that dry-air oxidation of carbon steel would be negligible for the Mined Geological Disposal System planned at the YM repository site, with predicted metal penetrations of about 2 µm after 10,000 years at 200 °C (Stahl, 1993). Henshall (1996) predicted, assuming a parabolic growth law, a uniform oxidation penetration of 127 µm after exposure to temperatures decreasing from 280 to 210 °C, over a 5000-year period. Assuming periodic spalling of the oxide, a uniform penetration of 350 to 600 µm was estimated over 5000 years. For these reasons, dry-air oxidation is not considered in the DOE TSPA-VA.

Based on literature reviews, CNWRA and NRC (Larose and Rapp,1997; Ahn, 1996c) calculated that the penetration of C-Mn steels by dry oxidation will range from 4 µm in 1,000 years at 250 °C to 100 µm at 200 °C for 10,000 years. Based on data from studies of other iron-base alloys (containing in most cases 10 to 20 percent Cr and 14 to 34 percent Ni) conducted above 600 °C (Shida and Moroishi, 1992; Otsuka and Fujikawa, 1991; Newcomb and Stobbs, 1991; Tasovac, et al., 1989; Mayer and Smeltzer, 1973; Raman, et al., 1992), it was suggested that deeper grain boundary penetration may occur at lower temperatures (Ahn, 1996c).

Based on the previous arguments, it can be concluded that for C-Mn steels dry-air oxidation is not a significant failure process. Accordingly, the staff considers resolution has been achieved on the issue related to the potential significance of dry oxidation of carbon steel as a failure process. However, since Alloy 22 is considered to be a prime candidate material for the outer overpack in EDA-II, internal and particularly intergranular oxidation may need to be re-evaluated. As reviewed by Ahn (1996c), these oxidation processes have been observed in nickel-base alloys at high temperatures (>600 °C), but in short exposure times.

5.1.2 Humid-Air Corrosion of Outer Overpack

DOE has modeled humid-air corrosion of carbon steel by using atmospheric corrosion data to derive a parametric equation in which corrosion depth depends on time, RH, and temperature (TRW Environmental Safety Systems, Inc., 1995, 1996b, 1998a; U.S. Department of Energy, 1998c). The coefficients in the equation were obtained using data for carbon steel from atmospheric corrosion exposures extended to 16 years in different locations (excluding marine environments) considering only the time fraction in which RH was greater than 70 percent. There are differences, however, between atmospheric and humid-air corrosion related to the effects of both temperature and heat transfer, as well as the regularity and frequency of wet and dry cycles, as discussed by Cragnolino, et al. (1998).

Atmospheric corrosion of most metals occurs above a critical RH at which the metal surface is covered by a water film of sufficient thickness (several monolayers) to sustain electrochemical reactions. Iron and steel exhibit a primary critical RH of around 60 percent, similar to most metals (Fyfe, 1994). Corrosion proceeds at a slow rate above 60 percent RH, but at 75–80 percent RH, the corrosion rate sharply increases. This secondary critical RH is attributed to capillary condensation of water in the pores of the solid corrosion products. The water films that form on the metal surface usually contain a variety of contaminants, including trace amounts of Cl^- and other soluble species, such as CO_2 that increase the electrical conductivity and decrease the pH of the film, leading to an increase in the dissolution of the iron or steel (Leygraf, 1995). Alternate wet-dry cycles can result in the formation of iron oxides enriched in Fe^{3+} ions during the dry cycle which then undergo reduction to Fe^{2+} during the subsequent wet cycle (Misawa, et al., 1971; Stratmann, et al., 1990; Dunn, et al., 1999). Increase in corrosion rate due to wet-dry cycling results from (i) a decrease in diffusion distance of oxygen in the electrolyte layer due to evaporation of water and (ii) increase in the redox potential due to secondary Fe^{3+}/Fe^{2+} redox reactions. The corrosion rates, after numerous wetting and drying cycles, were reported as 0.2 mA/cm^2 (2.3 mm/yr) when drying and as high as 0.5 mA/cm^2 (5.8 mm/yr) during the reduction of Fe(III) to Fe(II) immediately after rewetting (Nishikata, et al., 1994; Tsuru, et al., 1995). There is no experimental evidence indicating these high corrosion rates can be sustained over long periods.

Nevertheless, the time interval during which humid-air corrosion takes place could be limited to a few hundred years, and limited uniform penetration of the carbon steel can be expected under such conditions. It appears that DOE is satisfactorily addressing humid-air corrosion through an extensive experimental program that combines thermogravimetric experiments with prolonged tests in the vapor phase in an attempt to simulate conditions that may prevail in the emplacement drifts (Gdowski, 1998). Because carbon steel is de-emphasized in the SR design or is considered only as an inner overpack material in one of the EDAs, humid air corrosion is no longer an important component of this subissue.

5.1.3 Uniform and Localized Corrosion of Carbon Steel Overpack in Aqueous Environments

The corrosion of carbon steel in aqueous environments is relevant to those designs that have the carbon steel as an outer (VA design) or inner (one of the EDA designs) overpack. Although carbon steel is not a candidate material in the EDA-II proposed as a basis for the SR design, the factors affecting its corrosion behavior are summarized briefly in this section to document the results of previous investigations.

The mode and rate of corrosion of carbon steel in chloride solutions simulating natural groundwaters are strongly dependent on pH and the chemical composition of the aqueous environment (Sridhar, et al., 1994; Brossia and Cragnolino, 1999). Uniform or general corrosion of carbon steel occurs below a pH of about 9.3, whereas localized corrosion (crevice corrosion) may occur above this pH if the potential is higher than the repassivation potential. The tendency for localized corrosion is also dependent on the concentration of anionic species and temperature. For example, localized corrosion occurred only below a chloride / total carbonate concentration ratio of 0.03 at a temperature of 65 °C (Brossia and Cragnolino, 1999), whereas at a temperature of 95°C, localized corrosion occurred over a wide range of chloride / total carbonate concentration ratios that extends up to 0.1. Uniform corrosion rates of steels in neutral and slightly alkaline environments are dependent on the rate of oxygen reduction in aerated solutions. The uniform corrosion rate increases linearly with temperature to 75 °C and then decreases from 75 to 100 °C as a result of the reduction in solubility of oxygen in aqueous solutions with increasing temperature. The corrosion rate at ambient temperature also rises with the concentration of NaCl to about 3–5 weight percent, but diminishes at higher concentrations as a result of the decrease in the dissolved oxygen concentration through the salting-out effect on gas solubility. The reduction in wall thickness of a disposal container can be predicted with relative accuracy from uniform corrosion rates in neutral and slightly alkaline environments even after long exposure times.

Marsh, et al. (1985) have developed potential-pH diagrams defining regions of uniform corrosion, passivity, and pitting corrosion of carbon steel for carbonate/bicarbonate solutions containing chloride at 50 °C. At low (HCO_3^- + CO_3^{2-}) concentrations, the corrosion of carbon steel is uniform. By increasing the (HCO_3^- + CO_3^{2-}) concentration, a region of passivity is observed and, as a consequence, pitting corrosion in the presence of chloride occurs above certain critical potentials. Pitting corrosion was observed in 0.001 M (HCO_3^- + CO_3^{2-}) with the addition of 10 ppm Cl^- at a pH of 9.2 above a potential of −600 mV_{SCE}. Increasing the Cl^- concentration shifted the regions of passivity and pitting to lower potential and higher pH values. Similar results were obtained at the CNWRA (Cragnolino, et al., 1998) measuring critical (initiation and repassivation) potentials for assessing the susceptibility to localized corrosion of ASTM A516 Grade 60 carbon steel. The repassivation potentials values are used in EBSPAC Version 1.1 (Mohanty, et al., 1997) and in the NRC/CNWRA TPA 3.2 code.

Pitting corrosion of carbon steels under natural corroding conditions has not been investigated in sufficiently prolonged tests. Marsh and Taylor (1988) measured pit depth distribution on carbon steel in 0.1 M $NaHCO_3$ solution (pH 8.4) with the addition of 1000 ppm Cl^- at 90 °C under potentiostatic conditions (−400, −200, and 0 mV_{SCE}) in tests lasting 10,000 hours (417 days). The expression given by Marsh and Taylor (1988) using an extreme value statistics approach to predict the maximum pit depth expected in the surface of a single container, exhibits a time dependence consistent with a growth process controlled by diffusional mass transport inside the pit. This expression is being used in EBSPAC Version 1.1 (Mohanty, et al., 1997) and in the NRC/CNWRA TPA 3.2 codes. According to this expression, the outer steel overpack can be penetrated in approximately 200–300 years. It is, however, uncertain if such propagation rates can be maintained. Even more important is to establish whether the behavior exhibited by pits about 5-mm deep can be extended to depths of about 100 mm, or whether other factors lead to arrest of pit propagation. The aspect ratio of the pit can change substantially by dissolution of the pit walls, causing the propagation to slow. As noted by Marsh and Taylor (1988), the ohmic drop within the pit could become so great that initiation of new pits on the surface will be more favorable than the continuous growth of deep pits.

MIC is currently acknowledged as a phenomenon that may affect the performance of WP materials (Geesey and Cragnolino, 1995). Microbial populations can survive exposures to temperatures on the order of 120 °C. They turn active only when the temperature declines and water and nutrients become available in sufficient quantities for bacterial growth. Classically, bacteria involved in MIC have been divided into three broad phenotypic groups: (i) acid-producing bacteria (APB); (ii) sulfate-reducing bacteria (SRB); and (iii) iron-oxidizing bacteria (IOB) (Little, et al., 1991). Bacteria representatives of each of these phenotypic groups have been identified as part of the natural flora at the proposed HLW repository site at YM (Pitonzo, et al., 1996; Horn, et al., 1998). Microbial biofilms are known to grow in environments where nutrients are present only at growth-limiting levels (Costerton, et al., 1995). Bacteria are able to grow because of the efficiency with which biofilms are capable of scavenging the available nutrients. However, as noted in the ENFE IRSR, before a determination of the effects of bacteria on corrosion of container materials, a nutrient and energy inventory calculation should be used by DOE to evaluate the potential for microbial activity.

General aspects of the MIC of carbon steels in the partially saturated environment of YM have been reviewed previously in some detail (Geesey, 1993). Groundwater in the vicinity of the proposed repository site at YM has 25 ppm sulfate and, therefore, microbial action by SRB could lead to sulfide concentration in the range necessary to induce a significant decrease in the E_{rp} for carbon steel, and together with the presence of chloride, could be an important factor in inducing localized corrosion. In contrast, it seems that the main effects of APB are the promotion and enhancement of uniform corrosion. It should be noted that the effect of axenic and mixed cultures of bacteria isolated from YM on the corrosion rate of AISI 1020 carbon steel has been reported (Pitonzo, et al., 1996). A mixed culture of IOB, APB, and SRB was the only one to show significant corrosion rates (1.3 mm yr^{-1}). This rate was three to four times that of the abiotic control. Additional DOE experimental work should be performed to reach more definite conclusions about the effect of microbial action on the integrity of carbon steel containers.

SCC of low- and medium-strength (< 690 MPa) ferritic steels, a class of steels to which A 516 steel belongs, occurs in a variety of environments, such as acidic and alkaline nitrate (NO_3^-); acidic phosphate ($H_2PO_4^-/HPO_4^{2-}$); bicarbonate/carbonate (HCO_3^-/CO_3^{2-}); and caustic (OH^-) solutions (Kowaka, 1990; Parkins, 1977; Ford, 1983). No cracking has been reported in chloride solutions for ferritic steels of strength lower than 690 MPa, but high-strength steels are extremely susceptible to environmentally assisted cracking in chloride solutions as a result of hydrogen embrittlement. Hydrogen embrittlement of carbon steels is not considered a plausible failure mode under repository conditions even for steel weldments (Sridhar, et al., 1994) unless high hydrogen concentrations are attained—a situation that does not seem possible in the oxidizing environment at YM.

Of all the species involved in the SCC of carbon and low-alloy steels, as summarized previously, only bicarbonate anions may be present in a tuff repository environment at high concentrations approaching those required to promote cracking within the pH range of 6.5–9. The bicarbonate concentration in the groundwater is approximately 2×10^{-3}M. The range of potentials for SCC of carbon steels in carbonate solutions diminishes with decreasing temperature, indicating that cracking becomes less severe at lower temperatures (Sutcliffe, et al., 1972). The nitrate concentration in the groundwater is approximately one order of magnitude lower and similar to that of chloride, but evaporation processes may lead to high concentrations. It appears, however, that high pHs ranging from 10 to 14 are also required to induce cracking in nitrate solutions.

Residual stresses resulting from fabrication processes, in particular welding, are the predominant mechanical factors to be considered for assessing the possible effect of SCC on the performance of WPs. Although no studies on the beneficial influence of stress relief have been reported in the open literature for the case of SCC in carbonate solutions, it is reasonable to assume that postweld heat treatments are beneficial for preventing SCC in these solutions, as is the case in nitrate and caustic solutions (Parkins, 1985). Although temperatures as low as 400 °C have an effect through partial stress relief, optimum resistance to SCC can be only restored by heating around 650 °C—the temperature at which full stress relief can be attained (Parkins, 1985). Whereas full stress relief treatment of a large container or overpack should not be difficult since it is customarily applied to large pressure vessels, heat treatment after closure would require an additional consideration in the case of WPs because high heat inputs may raise the cladding temperature well above 350 °C. DOE may need to clearly demonstrate that the absence of substantial residual stresses and the composition, pH, and redox potential of the environment, in contact with the WPs, preclude the possibility of SCC of the carbon steel overpack.

In its TSPA-VA, DOE considered the critical pH above which localized corrosion would occur to be 10, based on an expert panel assessment. DOE also cited long-term experiments performed at LLNL in which localized corrosion was not observed at about a pH of 9.7 (U.S. Department of Energy, 1998c). The growth rate for localized corrosion was also derived from an expert panel elicitation. The effect of microbial organisms on corrosion was considered in terms of a multiplication factor, ranging from 1 to 5, which increased the uniform corrosion rate. In general, DOE is satisfactorily addressing uniform and localized corrosion of steel outer overpacks through experimental and modeling work. The possible elimination of carbon steel in the proposed WP design for the SR may make these components of subissue 1 moot. However, if carbon steel becomes a candidate material, a focused effort by the DOE is recommended in determining environmental conditions leading to the occurrence of localized corrosion, the confirmation of pit growth expressions through prolonged tests, the combined effects of uniform and localized corrosion on through-wall penetration, the evaluation of the possible occurrence of MIC and SCC, and the appropriate consideration of the effects of welds and welding procedures on aqueous corrosion.

5.1.4 Localized and Uniform Passive Corrosion of the Corrosion-Resistant Alloy Overpack

Alloy 22, a Ni-Cr-Mo alloy, was selected by DOE in the VA design as the candidate material for the inner overpack of the double-shelled WP. In the proposed design for the SR, however, the outer overpack will be made of Alloy 22, surrounding an inner overpack of type 316NG stainless steel, which will provide structural strength. The performance of Alloy 22 has become one of the most important factors in determining the sensitivity of the repository system to the performance of a variety of engineered and natural barrier components. Hence, this component of the subissue is discussed in greater detail. DOE has not taken credit for the performance of type 316NG stainless steel in the proposed SR design. Hence, issues associated with the corrosion behavior of type 316NG stainless steel are not discussed.

The corrosion behavior of the Ni-Cr-Mo alloys was reviewed in detail by the CNWRA on the basis of literature data (Cragnolino and Sridhar, 1991; Sridhar, et al., 1994) and the available information is currently being updated (Cragnolino, et al., 1999). A similar review was conducted by DOE (Gdowski, 1991). Initially, Alloy 825 was the primary candidate material, replaced after several years by the more corrosion resistant Alloy 625, and at the end of 1997, by the even more corrosion-resistant Alloy 22. In this context, corrosion resistance implies increasing resistance to localized corrosion (pitting and crevice) in chloride solutions promoted by the combined effect of Cr as the main alloying element (about 22 percent in weight) and the increasing Mo (and W for Alloy 22) content. To quantitatively assess the beneficial effect of these alloying elements and in particular to develop a methodology for PA, the CNWRA used

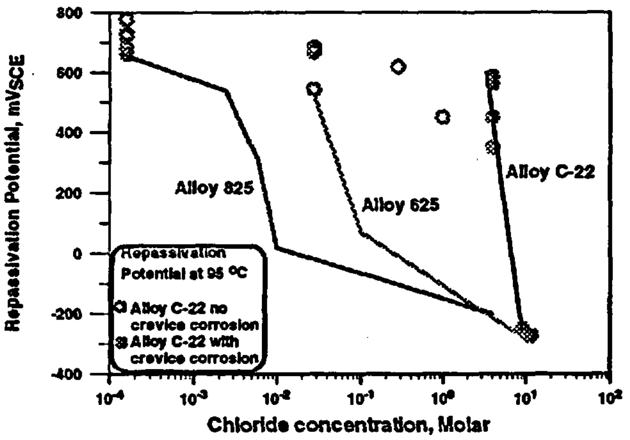

Figure 6. Effect of chloride concentration on repassivation potential of three different CRAs

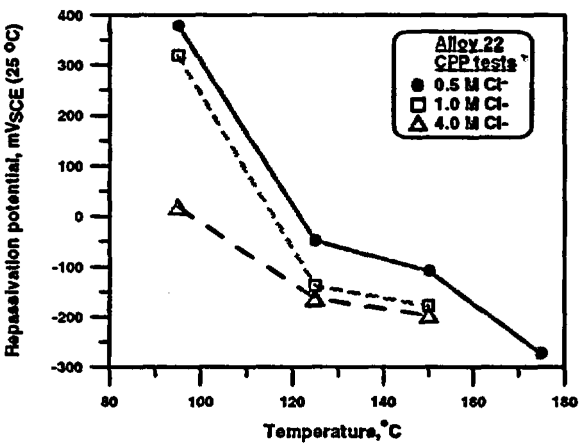

Figure 7. Effect of temperature on localized corrosion of Alloy 22

the E_{rp} as a criterion for the occurrence of localized corrosion. This potential is measured in chloride-containing solutions simulating concentrated groundwater assumed to be present in the environment surrounding the WPs. Extensive experimental work was conducted on Alloy 825 to develop and confirm the validity of this methodology (Sridhar, et al., 1995; Dunn, et al., 1996). A similar approach is currently being developed for Alloys 625 and 22 (Gruss, et al., 1998). The short-term data on localized corrosion of these alloys is illustrated in Figure 6. As shown in this figure, E_{rp} for Alloy 22 is significantly higher than that of Alloys 825 and 625 at chloride concentrations less than about 4 M, corresponding to saturated NaCl solution. At higher chloride concentrations, E_{rp} decreases significantly. As shown in Figure 7, at any chloride concentration, E_{rp} is a function of temperature. An abrupt decrease in E_{rp} is observed above 95°C. It would be useful to conduct prolonged tests to confirm the results obtained in short-term tests.

The approach adopted by the CNWRA consists of measuring the E_{rp} to compare the values obtained and associated uncertainties with the corrosion potential expected over a wide range of environmental conditions. This approach was used in EBSPAC (Mohanty, et al., 1997) and in the TPA Version 3.2 code, to determine the electrochemical conditions leading to the initiation of localized corrosion. Calculations conducted with Alloys 825 and 625 demonstrated the initiation of localized corrosion and the failure of the WP because of the pit penetration of both the outer and inner overpack in a few thousand years after repository closure. In the case of Alloy 22, however, computations reveal the absence of localized corrosion for more than 10,000 years. As a consequence, a precise determination of the rate of uniform corrosion under passive conditions becomes important to assess container life. Measured values of passive current densities, and the values of corrosion rates calculated using Faraday laws are shown in Table 1 in which the range of values assumed in TPA 3.2 are also listed. The values of passive current density listed are the result of a single determination using as received, base metal specimens (not welded). Uncertainties associated with the measurements of extremely low corrosion rates by weight loss preclude a reliable and precise prediction of container life. This calculation becomes important for radionuclide release and the subsequent estimation of dose to the average member of a critical group. A reasonable

Table 1. Measured passive current density and calculated corrosion rates of Alloy 22 under various conditions

Starting Condition of Alloy 22	$[Cl^-]$ (M)	pH	Temp (° C)	Potential (mV_{sce})	Anodic Current Density (A/cm^2)	Corrosion Rate (mm/yr)	Lifetime of 20-mm thick WP Barrier (yr)
As received	0.028	8	20	200	2×10^{-9}	2×10^{-5}	1,007,455
As received	0.028	8	95	200	3×10^{-8}	3×10^{-4}	67,163
As received	0.028	0.7	95	200	7×10^{-8}	7×10^{-4}	28,784
As received	4	8	95	200	3×10^{-8}	3×10^{-4}	67,163
As received	4	8	95	200	4×10^{-8}	4×10^{-4}	50,372
Low dissolution rate assumed in TPA 3.2					6×10^{-8}	6×10^{-4}	33,581
High dissolution rate assumed in TPA 3.2					2×10^{-7}	2×10^{-3}	10,074

approach to resolve these uncertainties, could be the use of fundamental models describing the growth of anodic oxide films on metals, to establish at least an upper bound value for the rate of dissolution under passive conditions. The fundamental models for passive film growth can be compared to measured passive current density values.

The technical bases for modeling the general and localized corrosion of Alloy 22 is provided in TSPA VA (U.S. Department of Energy, 1998c) and supporting documents (TRW Environmental Safety Systems, 1998a). A generalized expression for corrosion rate was obtained from expert elicitation as well as consideration of experimental data developed under a wide range of conditions (U.S. Department of Energy, 1998c). For developing the distribution of corrosion rates from expert elicitation, three types of environments were delineated in TSPA VA. It was assumed that 84 percent of the WP area will be contacted by a moderately oxidizing environment at a pH range of 3–10. Moderately oxidizing environment was defined as 340 mV vs. Standard Hydrogen Electrode (SHE), although it was not specified if this Eh was that of the environment or the corrosion potential of Alloy 22. Thirteen percent of the WP area was assumed to be contacted by the same moderately oxidizing environment at a pH of 2.5. Three percent of the surface area was assumed to be dripped on by a highly oxidizing environment (640 mV vs. SHE) at a pH of 2.5. These two last environments were rationalized in terms of conditions prevailing in local areas such as crevices. However, as noted in the ENFE IRSR, Revision 2, the set of environments selected by expert elicitation is not based on WP modeling conducted as part of the VA (TRW Environmental Safety System, Inc., 1998d). Based on these environmental assumptions, the expert panel provided a cumulative distribution of the corrosion rate ranging from 2×10^{-8} mm/y to 10^{-2} mm/y. This range of general corrosion rate was justified in terms of experimental data being developed by LLNL as well as data published by alloy manufacturers. Localized corrosion of Alloy 22 was assumed not to occur below a critical temperature of 80°C, with a uniform probability distribution ranging from 0 to 1 from 80 to 100°C. The localized corrosion rate was assumed to be dependent on temperature and time. The Arrhenius law was used to express the dependence on temperature and the expression for the localized corrosion rate was obtained using a database that includes corrosion rates measured gravimetrically under conditions leading to uniform and eventually to localized corrosion in $FeCl_3$ solutions.

Many concerns remain with respect to the DOE approach to modeling the corrosion of Alloy 22. These are summarized as follows.

(1) The assumptions regarding the three types of environments arising from localized corrosion are inconsistent with the assumption regarding the rate of general corrosion. While acidic environments have been shown to occur under localized corrosion conditions in which the anodic sites are spatially separated from the cathodic sites, these conditions do not occur under general corrosion conditions in which the anodic and cathodic sites are spatially contiguous. Because soluble corrosion products from anodic reactions generate acidic pH due to hydrolysis and cathodic reactions generate alkaline pH, their contiguous presence does not result in overall acidification of the environment. Additionally, while oxidizing conditions drive localized corrosion, the environment inside the localized corrosion regions is generally quite reducing (Sridhar and Dunn, 1994). This environment again results from spatial separation of anodic and cathodic sites during localized corrosion. Radiolysis may contribute to oxidizing conditions, but the technical basis for the assumption of 640 mV vs. SHE is not clear.

(2) The technical basis for the wide range of general corrosion rates assumed by DOE in TSPA-VA is not clear. The experimental data cited to justify the use of such a wide range of corrosion rates include environments that promote localized corrosion ($FeCl_3$), active corrosion (H_2SO_4, HCl), and passive corrosion (NaCl). Because the model assumes that at temperatures below 80–100°C, no localized corrosion occurs, the general corrosion rate should be consistent with passive behavior. As shown in Table 1, the passive corrosion rates in well-constrained experiments do not exhibit the wide range of values adopted in the TSPA-VA. The wide range of values results in container failures distributed over an extremely long time interval and, as a consequence, radionuclide release may be spread in time leading to non-conservative estimates of the peak dose.

(3) The critical temperature for localized corrosion is assumed in the TSPA-VA to be independent of chloride concentration or corrosion potential. As shown in Figure 6, the critical temperature is dependent on both chloride concentration and potential.

(4) The database used to estimate localized corrosion rates provides an extremely wide distribution of rates because it comprises conditions leading to both uniform and localized corrosion. No specific measurement of corrosion rates in actively dissolving pits or crevices was considered.

The effect of welding on the localized corrosion resistance of Alloy 22 should be evaluated because the weld metal exhibits a critical temperature for pitting corrosion significantly lower than that of the base metal (Sridhar, 1990). The lower localized corrosion resistance of the weld metal has been related by Sridhar (1990) to the segregation of alloying elements, such as

Mo and W in the interdendritic regions formed during the solidification of the fusion zone (Cieslak, et al., 1986). Because of its low carbon content, Alloy 22 is not susceptible to intergranular corrosion as a result of sensitization induced by carbide or intermetallic phase precipitation (Sridhar, 1990). Inappropriate welding procedures, however, can result in increased susceptibility to intergranular corrosion. In addition, the effect of cold work followed by long-term thermal exposure on the localized corrosion of Alloy 22 should be evaluated, as noted, for the case of mechanical properties in Section 5.2.2.

A concern arising from the DOE testing program is the possibility of SCC of Alloy 22 in an acidified (pH 2.5) NaCl solution deaerated with nitrogen at 90 °C (Roy, et al., 1998a). DOE tests used precracked double cantilever beam (DCB) specimens. The observation of SCC contradicts findings reported in the literature as reviewed by Sridhar, et al. (1994), in which tests using two-point bend specimens did not reveal SCC below a critical temperature of about 200 °C in 20.4 percent $MgCl_2$ solution. Recent experimental results from CNWRA using DCB specimens of Alloy 22 in various chloride solutions including $MgCl_2$ did not indicate any evidence of SCC (Dunn, 1999). The procedures used in these tests were similar to those used by Roy, et al. (1998a). It should be noted, however, that $MgCl_2$, which is a strong SCC promoter, is not expected to be present in the repository environment.

In CLST IRSR, Revision 1, the need to evaluate better the effects of galvanic coupling between carbon steel and Alloy 22 was pointed out. However, in view of the design changes proposed in the EDA, galvanic effects between steel and Alloy 22 are not considered important. If the titanium drip shield is eliminated from consideration, accidental contact between Alloy 22 and steel sets on the drift wall may occur, and this aspect should be reconsidered.

There is limited information available on MIC of Ni base alloys, with high Cr and Mo content, such as Alloy 22. It appears that these alloys are not susceptible to MIC in the expected repository conditions (Geesey, 1993). However, DOE should provide updated information excluding the possibility of MIC of Alloy 22.

DOE is satisfactorily addressing many aspects of the localized and uniform passive corrosion of Alloy 22 as the candidate material for the inner overpack covering a reasonable range of environmental conditions through experimental and modeling activities (Stahl, 1998). However, several unresolved aspects of this component of Subissue 1 relate to the effects of welds on localized corrosion, the appropriate values for general corrosion rate, the susceptibility of the material to SCC, and confirmation of the lack of susceptibility to MIC. Undue reliance has been placed on the use of expert elicitation even though experimental data can be obtained. DOE is developing an extensive corrosion testing program. Results from the DOE experimental program should be used in TSPA to support the validity of model assumptions and abstractions. Regardless of the approach used, DOE should provide adequate data confirming the resistance of Alloy 22 to localized corrosion and the validity of long-term extrapolation of uniform, passive corrosion rates.

5.2 STATUS OF RESOLUTION OF SUBISSUE 2 AND RELATED OPEN ITEMS

5.2.1 Thermal Embrittlement of Carbon and Low-Alloy Steel Overpack

As mentioned previously, the VA design included carbon steel outer overpack and a relatively high thermal loading strategy. Prior to the focus on carbon steel in the VA, DOE considered other steels including 2.25%Cr-1%Mo steel and cast steel as alternative outer overpack materials. Depending on the thermal loading strategy, container materials can be exposed to temperatures well above 100 °C for several thousand years. In addition, backfilling the drifts after a certain period of emplacement can induce a relatively sharp increase in the temperature of the WP surface above 100 °C, followed by a gradual temperature decrease with time. Prolonged exposures of outer overpacks at these temperatures may significantly affect the thermal stability and specific mechanical properties of carbon and low-alloy steels. In the proposed SR design, carbon steel is no longer considered as the outer overpack material. However, thermal embrittlement of steel is summarized in this section for completeness and to document the results of work related to previous design alternatives.

Thermal embrittlement is related to the phenomenon of temper embrittlement that affects tempered low-alloy steels as a result of isothermal heating or slow cooling within the temperature range 325–575 °C (Briant and Banerji, 1983). Temper embrittlement is a major concern to the integrity of engineering components that operate within that critical temperature range. It is also a concern for heavy section components that are slowly cooled through the critical temperature range after heat treatment or welding operations. Examples of such components are pressure vessels and turbine rotors. This phenomenon leads to a shift in the ductile to brittle transition temperature (DBTT) to higher temperatures, increasing the propensity of the steels toward brittle fracture at low impact energies. The potential for embrittlement of carbon and low-alloy steels as a result of extended exposures (hundreds to thousands of years) at temperatures lower than 300 °C, as expected under repository conditions, has never been experimentally investigated (Cragnolino, et al., 1996). Lack of information is undoubtedly related to the extended times that are required to induce this phenomenon—if it occurs—at these relatively low temperatures.

Temper embrittlement occurs when impurities originally present in the steel, such as Sb, Sn, P, Si, and As, segregate along prior austenite grain boundaries as observed experimentally during exposure to temperatures ranging from 300 to 600 °C for a few years. Of these elements, P is the most common embrittling element found in commercial low-alloy steels. The segregation of P promotes the fracture of notched specimens on impact and leads to a change in the low-temperature fracture mode from transgranular cleavage to intergranular fracture.

Preliminary calculations performed at the CNWRA (Cragnolino, et al., 1998) suggest that significant grain boundary P segregation, and, hence, the potential for a substantial degradation in toughness, may occur only as a consequence of long-term thermal aging at repository temperatures anticipated for the high heat-loading concept (i.e., temperatures greater than 200 °C for several thousand years). At lower temperatures, it appears that thermal embrittlement should not be a matter of concern. It should be noted, however, that low-alloy steels are more susceptible to this phenomenon than plain carbon steels. Calculations performed by DOE (Farmer, 1998), also using the approach adopted by the CNWRA, suggest that at 200 °C and below, the amount of P segregated to grain boundaries in approximately 2000 years is too low to induce embrittlement. On the basis of these arguments, it can be considered that thermal embrittlement of the carbon steel is not a significant failure mode unless a high areal mass loading (with the eventual use of backfilling) is adopted, or additional data at relatively low temperatures (250–300 °C) for pressure vessel steels lead to a reconsideration of the evaluation provided previously, or both. This conclusion agrees with DOE's current assessment of the effect of thermal embrittlement on mechanical failure of carbon and low alloy steels.

5.2.2 Thermal Stability of Alloy 22 Overpack

The thermal stability of Ni-Cr-Mo alloys under repository conditions has been reviewed by Sridhar, et al. (1994) and by Gdowski (1991). It is well established that alloys such as Alloy 22 are prone to the formation of M_6C-type carbides and intermetallic phases, such as μ, σ, P, and Laves (Raghavan, et al., 1982).

All the Ni-Cr-Mo alloys of the class consisting of C-4, C-276, and Alloy 22 experience an ordering transformation when heated in the temperature range of 250–550 °C (Tawancy, 1981; Raghavan, et al., 1982; Hodge and Ahluwalia, 1993). Ordering in this temperature regime involves an initial short-range ordering followed by a long-range ordering. The long-range ordering follows the same type of crystallographic modification found in pure Ni_2Cr (Klein, et al., 1970) and involves formation of an orthorhombic crystal structure from the (420) or the (220) planes of the parent γ-phase. Six orientations of this orthorhombic structure are possible with respect to the parent γ-phase. Because of this definite orientation requirement, a twinning type deformation mode is preferred rather than dislocation slip. The result is an increase in the work hardening rate and as a consequence an increased susceptibility to SCC and hydrogen embrittlement. The effect of aging at lower temperatures for long periods on the mechanical properties was reported by Hodge and Ahluwalia (1993). A significant embrittlement as measured by the CVN impact energy was found for specimens aged at 538 °C for 20,000 hours (2.3 years) and even at 425 °C for 45,000 hours. Rebak and Koon (1998) confirmed the existence of the long-range ordering of Alloy 22 and the absence of the μ phase for aging times of 30,000 and 40,000 hours (3.4 and 4.6 years) at 425 °C. Hodge and Ahluwalia (1993) concluded, however, that long-range ordering in Alloy C-4 and related alloys is not likely under the thermal conditions expected in the repository (container temperatures lower than 250 °C) in a 1000-year period.

As noted by Sridhar, et al. (1994), cold work before aging can increase markedly the kinetics of long-range ordering, as observed for Alloy 276 (Sridhar, et al., 1980). Local areas of cold-work may occur in the containers through rock impingement, mechanical damage during handling, or on areas where surface defects, such as weld spatter have been mechanically removed by grinding operations. Cold work in the form of residual stresses can also result from fabrication during the cylinder rolling operations. Exposure to temperatures ranging from 200 to 500 °C may result in grain boundary segregation of metalloid elements, such as P (Berkowitz and Kane, 1980). Effects on mechanical properties and susceptibility to embrittlement of Ni-Cr-Mo alloys, however, have not been studied in sufficient detail. Issues related to weldments of Ni-Cr-Mo alloys have been briefly reviewed by Sridhar, et al. (1994). Segregation of Cr and Mo has been reported, but the effect on mechanical properties is unknown. Gdowski (1991) also concluded that long-term aging of Alloy 22 should be studied at temperatures of 250 °C or less, to determine the formation of intermetallic phases and the occurrence of long-range ordering, as well as possible effects on mechanical properties and corrosion resistance. Although the studies of Hodge and Ahluwalia (1993) and Rebak and Koon (1998) suggested that repository temperatures are too low to promote degradation of mechanical properties, the effect of cold work before long-term aging has not been addressed yet.

The thermal stability of Alloy 22 is presently being investigated by the DOE in collaboration with Haynes International. A decreased impact strength concurrent with increased yield strength and decreased ductility for Alloy 22 as a result of μ-phase formation was recently reported by Edgecumbe-Summers, et al. (1999). At 700 and 750°C, sharp decreases in the impact strength were observed after aging for 2,000 and 16,000 hours. Unfortunately, reduction in impact strength for shorter aging times, as might be expected during welding and annealing operations, were not reported. The need to examine the effect of low-temperature aging on the phase stability of Alloy 22 has been identified (McCright, 1998). Rebak and Koon (1998) emphasized the need for long-term Alloy 22 aging tests so that extrapolations of phase stability can be performed within the temperature range of 100 to 250 °C over a 10,000-year period. The conceptual design of a 300-year aging experiment has also been discussed (McCright, 1998) where samples would be aged at temperatures of 427 and 300°C. By extending the performance confirmation period of the repository, the 300-year aging test results could be used to determine if the containers need to be removed from the emplacement drifts so that the waste may be repackaged in the event of significant deterioration of mechanical properties.

Mechanical failure of containers caused by disruptive effects has been partially considered in other IRSRs (i.e., RDTME, SDS, IA). In future revisions of this IRSR more details on mechanical failure of overpack materials will be discussed and an assessment of DOE progress in resolving this subissue will be presented. The effect of phase instability on corrosion and SCC resistance is considered in Subissue 1.

5.2.3 Initial Defects

Initial or premature failures of containers are those that occur essentially instantaneously (in comparison to the expected period of performance) due to one or more initial defects. These defects are considered to be undetected during fabrication, handling operations, and the preclosure period. Many of these defects are produced during fabrication, such as lack or fusion of weld penetration, surface contamination (e.g., poor degreasing), laps and iron contamination, improper filler metal in welds, iron dilution (e.g., in bimetallic welds), voids, and inclusion/primary carbides. Other premature failures may be related to improper heat treatments (e.g., temperature evolution in large components leading to sensitization or temper embrittlement) or material mix-up. Improper handling operations may lead to dents or scrapes that may induce localized cold work or become initiation sites for pits or cracks.

The consideration of initial failures in performance assessment codes (both TSPA-VA and TPA, Version 3.2) has a very limited technical basis (Sridhar, 1999). DOE needs to provide a better link between initial defects and failure rates by considering detectability of small defects and performance of defective containers. The appropriate evaluation of the attainable resolution of nondestructive examination methods requires a focused effort by the DOE. It is worthwhile to note that the dominant influence of defects on premature failures is a consequence of the long lifetimes estimated for the Alloy 22 containers, as shown in Table 1.

5.3 STATUS OF RESOLUTION OF SUBISSUE 3 AND RELATED OPEN ITEMS

5.3.1 Spent Nuclear Fuel Types

It is anticipated that 70,000 metric tons heavy metal (MTHM) of waste, consisting of 63,000 MTHM commercial SNF, 2,600 MTHM DOE SNF, and 4,400 MTHM (equivalent) of vitrified HLW will be emplaced in the repository (TRW Environmental Safety Systems, Inc., 1997b). The commercial fuels are nominally UO_2, clad with Zircaloy. The DOE-owned SNF, which falls into 34 groups (Duguid, et al., 1997), has been, for the purposes of PA, grouped into 14 categories. Most of the DOE inventory is metallic fuel in the form of U or U alloyed with Al, C, or Si—the most predominant being the N-reactor fuel from Hanford, which is metallic U. The cladding is either Al or Zr. These fuels have higher enrichment than the commercial fuels and are classified as high enriched (U ranging from 20 to 95 percent) and low enriched (U less than 20 percent). The commercial fuels, in contrast, have enrichments ranging from 2.5 to 4 percent. However, the radionuclide inventory of DOE-owned SNF is small compared to that of commercial SNF. This revision of the CLST IRSR focuses on the commercial light water reactor fuels, although most of the subissues are equally valid for other fuels.

To evaluate the status of resolution of this subissue, several components are important, including: (1) radionuclide inventory and distribution in the fuel; (2) dry oxidation of the SNF and its effects on subsequent performance in aqueous environment; (3) dissolution in aqueous environment; (4) solubility of radionuclides; (5) secondary mineral formation and co-precipitation; (6) formation of colloids; (7) cladding performance; and 8) conceptual models for release. Each of these components will be addressed in this section according to current knowledge and information needs for subissue resolution. Unlike the container materials project for which NRC generated independent experimental data, DOE generated most of the data regarding radionuclide release from SNF. The issue resolution in this context is achieved through examination of the adequacy of DOE data for determining the radionuclide release from SNF and consistency of the information with evidence generated by studies in other countries.

5.3.2 Radionuclide Inventory and Distribution in Spent Nuclear Fuel

The radionuclide inventory becomes distributed among the fuel matrix, boundaries between UO_2 particles (usually called grain boundaries), the fuel-cladding gap, the cladding itself, and corrosion products, such as crud deposited on the outer surface of the cladding during reactor operation, or secondary minerals formed after exposure of the fuel to the repository environment. The fractional distribution of the actinides in the fuel matrix is more than 99 percent, whereas up to 6 percent of the volatile fission products, such as ^{135}Cs and ^{129}I tend to migrate to the fuel cladding gap (Johnson and Tait, 1997). According to Johnson and Tait (1997) approximately 1 percent of ^{99}Tc accumulates at the gap and grain boundaries. Although part of the inventory of ^{99}Tc is apparently associated with ε-phase particles (Gray and Thomas, 1994), these particles are not very soluble. This circumstance explains the conclusion of Gray and Wilson (1995) that the inventory at those locations is less than 0.2 percent since the ε-phase particles may not dissolve in the SNF dissolution studies. Although considerable uncertainties may exist regarding the grain boundary inventory of ^{99}Tc, the current 2 percent approximation used in the TSPA-VA (TRW Environmental Safety

Systems, 1998b) may be conservative. More recent measurements of ^{129}I and ^{135}Cs (Gray, 1999) show wider variations in the gap and grain boundary inventories. The combined gap and grain boundary inventories of ^{129}I approximately equal the fission-gas release fraction. For ^{135}Cs the combined gap and grain boundary inventories are approximately one third of the fission-gas release fraction. Because the fission-gas release fractions vary as much as 20 percent, the gap and grain boundary inventories can be more than 20 percent for ^{129}I and more than 5 percent for ^{135}Cs. It is important that the DOE determines accurately the prompt release of ^{99}Tc because of its potential effect on early peak doses.

Radionuclides important to system-level performance depend somewhat on the assumptions regarding the flow pathways, transport parameters, and dilution (Jarzemba and Pickett, 1995). In the TSPA performed by NRC, the radionuclides contributing the greatest to the dose at 10,000 and 100,000 years were ^{237}Np, ^{234}U, ^{99}Tc, ^{129}I, ^{36}Cl, and ^{79}Se from a total of 20 radionuclides considered. In the DOE TSPA-VA (TRW Environmental Safety Systems, Inc., 1998c), the six predominant radionuclides contributing to 99 percent of the dose after 1,000,000 years among the nine radionuclides considered were ^{99}Tc, ^{129}I, ^{237}Np, ^{234}U, ^{239}Pu, and ^{242}Pu. Resolution of the differences in the importance of specific radionuclides will depend on the assumptions regarding radionuclide distribution between gap, grain boundaries, and matrix, releases including solubility and coprecipitation of certain radionuclides in secondary U minerals, and transport, including stability of colloids and sorption.

In the case of DOE fuels, distribution of radionuclides is dependent on the microstructure of the fuel material and there is, at present, considerable uncertainty regarding distribution of radionuclides among the various metallurgical phases. The distribution is important since these fuels dissolve preferentially along the boundaries between the second-phase particles and matrix, releasing particles of the second phase that could contain most of the radionuclides of importance to performance (Westinghouse Savannah River Company, 1997). A preliminary DOE PA indicates that ^{237}Np, ^{129}I, and ^{99}Tc are the most important radionuclides contributing to dose (Duguid, et al., 1997).

NRC acknowledges the adequacy of DOE selection of radionuclides of importance to performance. However, since detailed PA models developed by DOE and NRC differ, DOE should provide a rationale for deciding which radionuclides are important for their contribution to dose in TSPA calculations.

5.3.3 Dry-Air Oxidation of Spent Nuclear Fuel

The dry oxidation of SNF may be important if there is a premature container failure exposing the fuel to an oxygen-rich environment at temperatures higher than 100 °C with RH below approximately 30 percent. This process may occur if containers exhibit juvenile failures or fail as a result of seismicity-induced fracture. The effect of dry air oxidation can be manifested as gaseous release of certain radionuclides (e.g., ^{14}C); alteration of the oxidation state of the SNF such that subsequent aqueous dissolution rate is enhanced; and spallation of SNF grains, caused by volume expansion, to increase the surface area. Since ^{14}C is a weak beta emitter, its contribution to dose may be small. Two other radionuclides that may be released are ^{36}Cl and ^{129}I (Ahn, 1994).

The static air oxidation of SNF at temperatures ranging from 175 to 195 °C has been studied as a function of burn-up, fission-gas release, and grain size (Einziger, et al., 1992). It was found that air oxidation of SNF is quite different from oxidation of unirradiated UO_2. Unirradiated UO_2 oxidizes by the formation of a surface layer of U_3O_7 that proceeds inward and is followed by the formation of U_3O_8 at the surface. SNF was shown to undergo grain boundary oxidation, which then progresses into the grain matrix. The oxide stoichiometry can be as high as $UO_{2.4}$ (U_4O_{9+x}), which causes a 3-percent reduction in volume compared to UO_2 (Thomas, et al., 1989). This reduction in volume can create small areas of grain boundary cracking. Additionally, because the oxide is hyperstoichiometric with respect to oxygen, it would be expected to be more conductive than a stoichiometric oxide and, hence, dissolve more readily (Blesa, et al., 1994). Extrapolation of the kinetics of $UO_{2.4}$ from 175 °C to lower temperatures indicates that this oxide may not form for periods extending to several thousand years (Einziger, et al., 1992; Ahn, 1996b).

The effect of $UO_{2.4}$ formation on subsequent aqueous dissolution was examined by Gray, et al. (1993), who found that prior oxidation to $UO_{2.4}$ did not increase the intrinsic dissolution rate significantly in oxygenated, flowing carbonate solutions. The apparent dissolution rate (i.e., dissolution rate calculated by assuming the apparent particle area), however, was higher by a factor of 10 for the preoxidized SNF, possibly because of grain boundary opening (Gray, 1997). No difference in intrinsic dissolution rate was observed between oxidized and unoxidized SNF particles on the basis of U release. Lack of sensitivity of the dissolution rate to prior oxidation may be consistent with the finding that the surface layer of UO_{2+x} attains an oxidation state dependent only on the corrosion potential of the fuel which, in turn, is determined by the redox condition of the environment (Shoesmith and Sunder, 1992). For example, in aerated solutions, Shoesmith and Sunder (1992) found that the surface oxidation state corresponded to $UO_{2.5}$.

Above 250 °C, $UO_{2.4}$ (with an average burn-up of 28 to 48 GWd/MTU) oxidizes rather rapidly to U_3O_8, as observed by Eizinger, et al. (1992) in laboratory tests and the oxidation is accompanied by a volume expansion of about 36 percent. It appears there is a threshold temperature of about 250 °C below which U_3O_8 is not formed in 10,000 years (Ahn, 1996b). This threshold temperature increases at higher burn ups (Hanson, 1998) and may decrease at lower burn-ups (Eizinger and Cook, 1985). The volume expansion is expected to increase the surface area by two orders of magnitude. Preliminary experiments by Gray, et al. (1993) on unirradiated fuel that was oxidized to produce U_3O_8 showed that the effect of increased surface area on aqueous dissolution rate due to the volume expansion was, at most, about a factor of 4. However, more recent results using a different fuel exhibited a 10-times increase in the intrinsic dissolution rate, resulting in a fractional release rate (rate for unit specimen weight) equal to 150 times that of unoxidized particles (Gray and Wilson, 1995). Oxidation of fuel to U_3O_8 can lead to disintegration of fuel into individual grains because of volume expansion. Nevertheless, recent tests of intact whole rod (fuel plus cladding) in inert and air atmospheres at 325–570 °C did not result in cladding rupture (Einziger, 1997), presumably because the cladding acted as an oxygen getter preventing pellet oxidation. Splitting of cladding was noted at low burn-up ends of rods with defective cladding (Einziger, 1997).

Based on the limited data published by DOE, prior dry air oxidation to temperatures up to 250 °C would not be expected to significantly increase (at most, a factor of 10 in certain SNF) the subsequent aqueous dissolution rate. Such an expectation is consistent with the dissolution model proposed by Shoesmith and Sunder (1992), who argue that the surface oxidation state of SNF is conditioned by the redox nature of the aqueous environment contacting it, regardless of the prior oxidation state. Gaseous release of ^{14}C, ^{36}Cl, or ^{129}I is likely to occur during dry oxidation of SNF (Van Konynenburg, 1994; Ahn, 1994; Ahn 1996b). These releases may be important for juvenile failure of containers and cladding.

NRC considers that DOE has satisfactorily addressed dry air oxidation as a likely process for SNF degradation and radionuclide release through a complete experimental program and has developed adequate process and abstracted models.

5.3.4 Radionuclide Release

The rate of release of U and other species from breached WPs is controlled by a series of processes, such as transport of oxidants and flux of water, oxidative dissolution of SNF, uranyl mineral precipitation, uranyl mineral dissolution or transformation, and transport of radionuclides, and is affected by the condition of the fuel cladding. Thermodynamic analysis of some of the secondary uranyl minerals indicates that their solubility may decrease with an increase in temperature, thus exhibiting retrograde solubility (Murphy, 1997). However, increasing temperature may increase the dissolution and transport rates. This effect of temperature may mean that at high temperatures (for example in prematurely failed containers), radionuclide release will occur under conditions of fast dissolution rate and low solubility of secondary minerals. If some radionuclides co-precipitate with secondary uranyl minerals, their release may be governed by solubility of secondary minerals. At low temperatures, the dissolution and transport rates will be low, but the solubility may be high leading to dissolution or transport-limited release of radionuclides. Lastly, fuel cladding may act as a partial barrier to radionuclide release.

5.3.4.1 Dissolution Rate Controlled Release of Radionuclides in Aqueous Environments

Under oxidizing conditions, UO_2 dissolves as uranyl species, which then react with other species in solution to form uranyl oxyhydroxides or silicates, depending on the chemistry of the solution (Wronkiewicz, et al., 1992; Finn, et al., 1998). The release of highly soluble radionuclides, such as ^{99}Tc and ^{129}I, occurs at a relatively constant fractional rate (Finn, et al., 1998) and, therefore, it has been assumed that the dissolution rate of the fuel matrix controls the release rate of highly soluble radionuclides (Wilson and Gray, 1990a). The dissolution behavior of SNF and the release of radionuclides depend on the mode of contact of water with the fuel and the chemistry of the water. Four different techniques have been used to determine the fuel dissolution rate: (i) flow-through tests (Gray and Wilson, 1995), where a column of the SNF particles is exposed to a solution whose flow rate is adjusted so the concentration of uranyl species is maintained below solubility limits; (ii) semistatic or, batch-type immersion tests, where fuel pellets or particles are exposed to replenished solutions for specific periods of time and the solution then analyzed (Wilson, 1990a,b); (iii) electrochemical tests, where rotating disc specimens are subjected to a potential scan and the electrochemical polarization behavior analyzed to yield a corrosion rate (Shoesmith, et al., 1989); and (iv) drip tests, where J-13 water drips on fuel particles and the resultant effluent is collected periodically to measure radioactive release (Bates, et al., 1995; Finn, et al., 1998). Of these techniques, the electrochemical technique measures the dissolution rate of the fuel matrix directly, whereas the other techniques estimate the matrix dissolution rate from the leachate analysis.

Using the flow-through system, UO_2 dissolution rates ranging from $(0.6 \pm 0.4) \times 10^{-14}$ to $(2.1 \pm 1.2) \times 10^{-14}$ moles/($cm^2 \cdot s$) were measured in three laboratories [Whiteshell Laboratories, Pacific Northwest National Laboratory (PNNL), and Lawrence Livermore National Laboratory (LLNL)], over a test period of 300 days in air-saturated 0.1 M NaCl plus 0.01 M $NaHCO_3$ solution (pH 9.0) at 25 °C (Gray, et al., 1994). By using electrochemical techniques, Shoesmith, et al. (1989) estimated steady-state corrosion rates of UO_2 at 25 °C. In air-saturated solutions ($[O_2] = 2.5 \times 10^{-4}$ M), rates ranging from 0.4×10^{-15} to 3.9×10^{-15} moles/($cm^2 \cdot s$) were obtained in 0.1 M $NaClO_4$ at 25 °C. With the addition of 0.01 M and 0.5 M $NaHCO_3$, rates increased to 2.1×10^{-13} moles/($cm^2 \cdot s$) and to 5.4×10^{-13} moles/($cm^2 \cdot s$). On the other hand, in N_2-deaerated ($[O_2] < 3 \times 10^{-8}$ M), plain 0.1 M $NaClO_4$ solution, the corrosion rate was estimated to range from 4×10^{-21} to 4×10^{-19} moles/($cm^2 \cdot s$). The corrosion rates in the flow-through tests are about an order of magnitude lower than those estimated by Shoesmith, et al. (1989) under almost equivalent environmental conditions. This difference in corrosion rates was attributed to variations in the value of E_{corr}, which seems to be extremely sensitive to the reactivity of the surface, particularly in carbonate-containing solutions (Gray, et al., 1994). It appears that flow-through tests gave practically the same corrosion rate with and without the presence of NaCl in solution (Gray, et al., 1994). These observations suggest that, under the conditions of these tests, the dissolution rate is kinetically controlled by charge transfer at the UO_2/solution interface rather than by mass transport processes in solution or by the solubility of U compounds.

The flow-through tests in sodium carbonate solution with or without the presence of NaCl in solution, are accelerated and conservative tests for the YM repository. The actual groundwater chemistry in the YM repository is expected to contain cations such as Ca and Si species, which may inhibit the dissolution rate of the primary phase (Wilson and Gray, 1990b). However, whether Ca and Si remain in solution or interact with WP degradation products has not yet been demonstrated for SNF. Also, the flow-through tests are accelerated tests used to determine the intrinsic dissolution rate of the primary phase without involving secondary minerals. In addition, conditions in drip tests may be also severe, presumably giving rise to conservative estimations of dissolution rate compared with bathtub/immersion conditions. Some experimental results suggest that dissolution rates obtained under immersion conditions in J-13 well water and other groundwaters containing Ca and Si can be 10 to 100 times lower than those obtained under flow-through conditions in sodium carbonate solutions as discussed by Ahn (1999).

The dissolution kinetics of the primary phase are dependent on the effective reactive surface area of SNF. The effective reactive surface area is determined by the volume of groundwater entering the failed container; the mode of contact of the water (i.e., immersion or drip conditions), the breached area of the cladding; the penetration of water into the gap between cladding and fuel; and the exposed grain boundary and subgrain boundary areas resulting from prior dry oxidation or corrosion. Additionally, it has been argued that if the RH is sufficiently high, all the SNF surfaces may be covered by a film of water, increasing the surface area. Therefore, DOE needs to provide an adequate basis for the assumed surface area.

Several environmental factors are known to affect the dissolution rate of UO_2 in aqueous environments.

(1) The pH of the aqueous environment has an effect on the rate of dissolution of SNF. Under oxidizing conditions, only a slight dependence of corrosion on pH has been observed at pH values lower than 4.0, whereas at pH values between 4.0 and 8.0, the rate decreases linearly with pH (Grambow, 1989). At higher pH values, the rate of dissolution seems to be unaffected by pH changes. At the proposed YM repository, slightly alkaline pHs are expected under nominal conditions. However, container corrosion (e.g., crevice corrosion) and cement dissolution will modify the pH that may range from acidic to alkaline depending on location and conditions inside WPs.

(2) Temperature increases the rate of dissolution of UO_2 although a functional dependence is not well-established over a wide range of temperatures extending from ambient temperatures to the boiling point of water.

(3) The nature and concentrations of the anionic species present in the groundwater are extremely important in determining the rate of corrosion of SNF. Anions, such as CO_3^{2-} that form stable soluble complexes with UO_2^{2+} cations, substantially increase the rate of oxidative dissolution (Needes, et al., 1975; Blesa, et al., 1994). At low CO_3^{2-} concentrations (0.001 M), the rate of corrosion is proportional to the total concentration because the rate-determining step is the surface complexation of CO_3^{2-} (Blesa, et al., 1994). DOE's flow-through tests were conducted in the regime of low carbonate concentrations that is expected at the YM repository (Gray, et al., 1992; Steward and Weed, 1994; Gray and Wilson, 1995). At intermediate concentrations (0.5 M), the corrosion rate depends on the square root of the total concentration because the solution transport of CO_3^{2-} to the surface is rate-controlling, or dissolution of an initially formed UO_2CO_3 film controls the overall rate (Grambow, 1989). At a high CO_3^{2-} concentration (1.0 M at 100 °C), the corrosion rate reaches a constant value, but at even higher concentrations, the rate decreases, probably because of the formation of surface films that limit the overall rate (Needes, et al., 1975). The rate of corrosion increases in the sequence $Cl^- < SO_4^{2-} < F^- < CO_3^{2-}$, although, in the case of SO_4^{2-}, a maximum in the rate is observed at intermediate concentrations (about 1.5×10^{-2} M) (Blesa, et al., 1994). Other species, such as Ca^{2+}, $SiO_2(aq)$, $H_3SiO_4^-$, and $H_2SiO_4^{2-}$, which are able to induce the precipitation of complex uranyl silicates, tend to reduce the corrosion rates (Gray, 1992; Mohanty, et al., 1997).

(4) Corrosion products from metallic components, mostly in the form of metal cations, affect corrosion rates directly through precipitation reactions forming secondary minerals that may slow down dissolution. However, corrosion rates can be increased by indirect action of corrosion products that may change the redox potential, the pH of the environment, or both. 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+}, among others (Sridhar, et al., 1995). These two effects may lead to higher rates of SNF corrosion.

(5) Both α- and γ-radiolysis of water may accelerate the corrosion rate of SNF via an increase in the E_{corr} because of the generation of H_2O_2, O_2, and short-lived oxidizing radicals (i.e., HO, HO_2). Except in the case of early failures, the γ-radiation fields may decay to almost negligible levels by the time (thousands of years) the containers fail and fuel contacts groundwater (Shoesmith and Sunder, 1992). Therefore, α-radiation appears to be the only significant source of radiolytic oxidants and may play a role in occluded regions inside failed containers, in which reducing conditions can prevail as a result of oxygen depletion caused by oxidation of steel WP components.

(6) The presence of low molecular weight organic compounds, including carboxylic acids produced by degradation of vehicle fuels, lubricants, or other organic materials, either by chemical or biochemical mediated processes, may accelerate the rate of corrosion of SNF through the formation of complexing or chelating species. The same action can be exercised by humic substances, such as humic acid and fulvic acid that act as polyelectrolytic weak acids. On the other hand, detergents or similar compounds can act as inhibiting species by blocking active sites on the SNF surface. It has been claimed that the temperatures prevailing around WPs will promote the oxidation of organic substances, such as lubricants or vehicle fuels to CO_2, removing them from the media as potential nutrients for microbial activity (Meike, 1996; Wilder, 1996). Additional uncertainties arise from the effect of biochemically mediated processes in the modification of the near-field environment once conditions are established for temperature and availability of water for microbial growth.

A key hypothesis of TSPA-VA (TRW Environmental Safety Systems, Inc., 1997b) is that the SNF intrinsic dissolution rate will determine the aqueous release of highly soluble radionuclides and the rate of SNF alteration to form secondary minerals and colloids. The product of the intrinsic dissolution rate and the wetted surface area provides the source term for radionuclide transport through the unsaturated zone. Whereas the intrinsic dissolution rates derived from flow-through tests are used by DOE as input to the release of radionuclides in TSPA-VA (TRW Environmental Safety Systems, Inc., 1998b), the drip test results from Argonne National Laboratory (ANL) are used to estimate the retention of certain highly soluble radionuclides (e.g., Cs) in the secondary minerals. The rate of secondary mineral formation will be assumed to be proportional to the intrinsic dissolution rate of the SNF.

Flow-through dissolution tests have been used to determine the intrinsic dissolution rate of UO_2 and irradiated SNF matrix mostly in sodium carbonate solutions (free of Ca- and Si-species) because secondary mineral precipitation is avoided as a result of the fast medium flow. The intrinsic dissolution rate determined in tests using a relatively small range of environmental variables is a function of temperature, pH, and total carbonate concentration (Gray, et al., 1992; Steward and Weed, 1994; Gray and Wilson, 1995). Unsaturated vapor or drip tests may result in intrinsic dissolution rates different from the semi-static and flow-through tests. Dissolution rates in semi-static tests seem to be lower than in flow-through tests as a result of the precipitation of secondary minerals, particularly in the presence of Ca and Si species (Gray, 1992; Mohanty, et al., 1997). The presence of a thin secondary phase appears to block possible reactive surfaces whether they are external or in grain boundaries. Unlike drip tests, cation depletion is unlikely when abundant groundwater is present (Wilson, 1990a). The pH of the thin film of solution in the drip tests may be more acidic than that of the bulk solution because of radiolysis and lack of dilution by a large volume of solution (Finn, et al., 1994). Furthermore, accumulation of some of the fission products (such as ^{14}C) in the thin film may increase the dissolution rate of SNF. In addition, inhibiting effects by Si species or Ca cations may be diminished by the depletion of these species as the reaction proceeds with a limited influx of groundwater in the drip tests. It would be useful to compare the behaviors of unirradiated UO_2 and SNF in drip tests in assessing the effect of α-radiation under such conditions. Hence, a careful DOE examination of the consistency among assumed dissolution rates from flow-through tests and the measured radionuclide concentrations in the drip tests (which will be used to calculate the retention factors), or in the semi-static tests, is needed. For this comparison, a good marker for matrix dissolution (e.g., ^{99}Tc, ^{90}Sr, ^{129}I, or ^{135}Cs) could resolve this issue. The results of natural analog studies, which indicated a similar paragenetic sequence of minerals to the drip tests (Pearcy, et al., 1994), should also be considered in estimating the fuel dissolution rate. In DOE tests in sodium carbonate solutions, pH, carbonate concentration, oxygen fugacity, and temperature are varied to obtain a parametric equation for the rate of dissolution. In reality, the water chemistry near the waste form can be more complicated, depending on the mode of water contact. Under immersion conditions, the chemistry inside a failed WP can be highly concentrated in chloride and metal chloride complexes as a consequence of corrosion in crevices. On the other hand, dilution of these solutions with groundwater can also occur. In addition, the chemistry of bulk groundwater dripped onto WPs can vary, depending on EBS design options such as emplacement of drip shield, concrete, steel, and backfills. Current DOE assessments include these variations to a certain extent. However, the details, particularly in relation to immersion conditions, have not been studied.

Overall, DOE is adequately addressing dissolution-rate-controlled release of radionuclides as a likely process through experiments and modeling. However, DOE needs to provide realistic intrinsic dissolution rates in the chemical environments expected to exist inside WP, a realistic estimation of surface area in irradiated fuel pellets and an evaluation of the consistency among various test results to be used in model calculations.

5.3.4.2 Solubility Controlled Release of Radionuclides

In DOE studies, the solubility of radionuclides, with the exception of Np species, is currently constrained by solubility values derived primarily from the expert elicitation supporting TSPA-93 and TSPA-95 (Wilson, et al., 1994; TRW Environmental Safety Systems, Inc., 1995). For example, the solubility of Pu species varies from a minimum of 1×10^{-8} M to a maximum of 1×10^{-6} M (TRW Environmental Systems, Inc., 1998b) The uncertainties in the solubility of several radionuclides have been addressed by NRC (Murphy, 1991). For example, the Pu solubility varied from 5.63×10^{-13} to 6.6×10^{-7}M depending on whether the mineral phase in equilibrium was assumed to be PuO_2 (at pH of 8.5), or $PuO_2(OH)_2$ (at pH of 6.9). Murphy (1991) recommended that a critical evaluation of near-field chemistry, including deviation from chemical equilibrium be conducted, and that uncertainties in the thermodynamic database be assessed. The range of solubility values for Np species used in DOE TSPA-1993 and TSPA-1995 was about 2 orders of magnitude higher than that used in TSPA VA. The higher value of solubility was attributed to the possible presence of metastable phases in the Np solubility experiments performed by Nitsche, et al. (1993, 1994). It was argued that the solubility measurements, which were performed using solutions supersaturated with respect to NpO_2, did not adequately represent the conditions in the SNF dissolution experiments or for fluid compositions similar to J-13 water anticipated in the WP. More recently, the distribution of solubility values for Np species was revised by DOE to lower values based on a review of experimental SNF dissolution and Np solubility measurements (Sassani and Siegmann, 1998). These solubility limits, used in TSPA-VA (U.S. Department of Energy, 1998c) need to be reevaluated by DOE as the water chemistry inside the WP becomes better known. Also, DOE needs to provide experimental confirmation of the solid Np compounds assumed to be in equilibrium with the dissolved Np species.

5.3.4.3 Effects of Secondary Minerals and Colloids on Radionuclide Release

Secondary minerals precipitate on or near the SNF surface through the homogeneous reaction of uranyl species with other species present in the environment contacting the WP. Secondary minerals may mitigate radionuclide release by sequestering or coprecipitating other radionuclides, thus reducing their concentration in the aqueous environment, and by partially blocking the SNF surface, thus reducing the access of water. This effect could be particularly important in solutions containing Ca and Si species. It has been suggested (Wilson and Gray, 1990b) that a protective film can be formed under such conditions on unirradiated UO_2 surfaces. Since the partial molar volume of the uranyl minerals is higher than that of uraninite, periodic spallation of the dissolution products may occur, exposing fresh SNF surface to further dissolution. Tests of SNF in dripping J-13 well water show that the predominant secondary mineral is Na-Boltwoodite ($Na[(UO_2)(SiO_3OH)](H_2O)$; the other minerals being uranophane $Ca[(UO_2)(SiO_3OH)_2](H_2O)_5$; dehydrated schoepite $(UO_2)(O)_{0.1}(OH)_{1.8}$; and Cs-Mo uraninite (Wronkiewicz, et al., 1992; Finn, et al., 1998). The appearance of these minerals on the SNF surface is consistent with the secondary mineral formation on uraninite found in the natural analog site at Peña Blanca. The drip tests indicate that Ru, Mo, Cs, Np, and Ba are incorporated in the alteration products and Pu appears to be concentrated at the fuel surface (Buck, et al., 1998; Bates, 1998b). In general, DOE is adequately addressing the effect of secondary minerals and colloids on radionuclide release through experiments and modeling. However, consistency in the assumptions of dissolution rates and retention factors for radionuclides must be examined further, as noted previously. The protective role of secondary minerals (Mohanty, et al., 1997) has not been considered in TSPA (TRW Environmental Safety Systems, Inc., 1997b). In addition, DOE needs to consider how corrosion products affect secondary mineral formation and retention of radionuclides.

Colloids may enhance or mitigate radionuclide release from the EBS, depending on composition, stability, and sorptive properties (Manaktala, et al., 1995; Ahn, 1996a). The formation and stability of colloidal material are examined briefly in this IRSR, and transport of colloidal material is addressed in the "Radionuclide Transport" IRSR (U.S. Nuclear Regulatory Commission, 1998b). Sensitivity studies for colloid effects were performed using Version 3.2 of the NRC TPA code. A bounding case for Pu and Am colloid transport showed minimal effect on dose in the 10,000 year compliance period. This is largely due to the extended waste package lifetimes. As long as waste package lifetime exceeds the compliance period of 10,000 years, there will be no significant contribution to dose from the transport of colloid material at Yucca Mountain. Therefore, further examination of colloidal effects on transport and release are not considered necessary. A summary of colloidal effects is summarized in this section for completeness.

Colloid formation during SNF dissolution was first identified in semistatic tests (Wilson, 1990a,b). Low solubility radionuclides, such as ^{239}Pu, ^{240}Pu, ^{241}Am, and ^{244}Cm were found as colloids when the medium, resulting from immersion tests in J-13 well water at 85 °C, was filtered through 0.4 µm and 1.8 µm pore filters. In drip tests using J-13 well water at 90 °C, ^{239}Pu and ^{241}Am were retained after filtering the medium (Bates, et al., 1995).

Four different processes of colloid formation have been reported in the dissolution of SNF (Manaktala, et al., 1995; Ahn, 1996a). The first process is nucleation and condensation from supersaturated solutions leading to continuous colloid formation under near-static replenishing conditions. The second process is dispersion arising from the mechanical detachment or spallation of secondary minerals formed on the surface of the altered SNF. Colloids will form continuously by this process, independent of radionuclide concentration in groundwater. The third process is sorption of dissolved radionuclides onto preexisting colloidal material, such as corrosion products or colloids in the groundwater. Groundwater in the vicinity of YM is known to have about 0.3 to 1.4 µg/mL of colloids ranging in size from 0.03 to 1.0 µm and composed mainly of silicate- or Fe-based minerals (Kingston and Whitbeck, 1991). The sources of corrosion products include containers, canisters, and basket materials. More than 10^5 MT of iron oxide can potentially be generated by the corrosion of the steel outer containers (Ahn and Leslie, 1998). A significant fraction of these corrosion products is expected to be present as colloidal matter. The fourth process is individual grain detachment from SNF by preferential dissolution of grain boundaries. This process decreases with time as the altered layer develops and covers the irradiated UO_2 surface (Wronkiewicz, et al., 1992).

Bates, et al. (1995) detected colloids of ^{239}Pu, ^{241}Am, and ^{244}Cm in drip tests conducted with SNF in J-13 well water at 90 °C and estimated their concentrations. Similar observations have been reported for immersion tests (Wilson, 1990a,b). In the TSPA-VA (U.S. Department of Energy, 1998b), estimations provided through expert elicitation were used that can be considered as bounding values. Although DOE is currently addressing adequately the processes of colloid formation in its general aspects, it has been recommended that DOE determine colloid contribution to actinide release resulting from these processes in PA calculations.

Once colloids are formed, they may flocculate and settle depending on groundwater properties, such as pH and ionic strength (Manaktala, et al., 1995). Currently, DOE is using empirical correlations to evaluate the stability of colloidal suspensions in terms of ionic strength and pH (Triay, 1998). From current assumptions and calculations, there is not expected to be a significant contribution to dose from the transport of colloid material at Yucca Mountain. Therefore, further examinations of colloidal effects on transport and release are not considered necessary.

5.3.5 Effect of Cladding on Release of Radionuclides

SNF cladding can act as a barrier to the release of radionuclides. Although cladding was not previously considered in PA codes, DOE (U.S. Department of Energy, 1998c) included the consideration of cladding as an additional metallic barrier in the TSPA-VA. Several potential degradation mechanisms of Zircaloy cladding, such as localized corrosion, creep, delayed hydride cracking, hydrogen embrittlement, mechanical failure owing to rock fall, SCC, and fuel and cladding oxidation can impair this beneficial action.

Pitting corrosion of Zr and Zr alloys (Zircaloy -2 and -4) occurs in acidic and neutral chloride solutions above a critical potential that is dependent on chloride concentration but independent of pH (Cragnolino and Galvele, 1978; Maguire, 1984). This potential, equivalent to the E_{rp} in the reported data, is easily reached in the presence of reducible species, such as Fe^{3+} ions, even at low concentrations (Maguire, 1984). Therefore, localized corrosion of fuel cladding may occur, depending on chloride concentration and temperature, in an oxidizing environment (as is presumably present around the SNF) if the corrosion potential is higher than the E_{rp}, which is about 0.34 V_{SHE} in 1 M NaCl solution at room temperature (Cragnolino and Galvele, 1978). If the corrosion potential is lower, the corrosion rate of Zircaloy would be extremely low as a result of the protective characteristics of the ZrO_2 passive film.

In addition, Zircaloy is susceptible to SCC in the presence of tensile stresses lower than the yield strength under the same environmental and electrochemical conditions that promote pitting (Cox, 1973; Cragnolino and Galvele, 1973; Mankowski, et al., 1984). Sufficiently high hoop stresses (60-70 percent of the yield strength) may be present in localized regions of the cladding as a result of fuel pellet expansion during irradiation. DOE has not considered the possibility of localized corrosion and SCC of Zircaloy in the presence of oxidizing chloride solutions. DOE needs to evaluate these processes in solutions simulating conditions inside the WPs and their effects on the integrity of fuel cladding, if cladding is considered as a metallic barrier to radionuclide release in the TSPA.

Both PNNL (Levy, et al., 1987) and LLNL (Schwartz and Witte, 1987; Thomas and Schwartz, 1996) developed mechanism-based creep models for predicting the creep rupture life of cladding under repository conditions. The PNNL model was developed under a DOE contract, whereas the LLNL model was supported by an NRC contract. The PNNL/DOE model is based on the deformation and fracture mechanism maps and treats several different creep deformation and failure modes (Chin, et al., 1986). For the low longitudinal stress and low temperature (<350 °C) anticipated for the cladding in the repository, the potential dominant creep failure mechanism is diffusion-controlled cavity growth (DCCG). McCoy and Doering (1994) reviewed previous models and proposed several refinements. The LLNL model, which treats DCCG, has been recently upgraded (Thomas and Schwartz, 1996). Both models have the same basic form traceable to the original model of Raj and Ashby (1975) and should predict similar results, providing similar values are used for the model constants. As reviewed by Ahn (1998), there is general consensus that cladding failure by creep rupture is unlikely under repository conditions, assuming no backfill. Creep rupture data from Germany (Peehs and Fleisch, 1986) are consistent with this assessment. There is, however, a lack of direct comparison of DCCG model prediction against experimental data and a lack of experimental evidence for DCCG in Zircaloy cladding materials (Pescatore, et al., 1989). Disagreement exists on the pertinent values of some constants in the DCCG models and on the validity of extrapolating to lower stresses and temperatures the results of calculations performed using creep models. Failure prediction is very sensitive to the effect of temperature. DOE should complete an evaluation of these discrepancies and assess the potential for creep failure, as a likely process for fuel cladding degradation, for alternate EBS designs.

Delayed hydride cracking is a time-dependent crack propagation process, under sustained-load conditions, that results from diffusion of hydrogen to the crack tip, and is followed by the formation and fracture of hydrides in the near-tip region (Dutton, et al., 1977). Most of the relevant experimental data on delayed hydride cracking in the literature are for Zr-2.5 weight percent Nb (Northwood and Kosasih, 1983); zirconium, Zircaloy-2, and Zircaloy-4 appear to be less susceptible or even unsusceptible to delayed hydride cracking. Both DOE (Cunningham, et al., 1987) and NRC (Ahn, 1998) reviewed the Canadian data and performed pertinent modeling. They concluded that delayed hydride cracking would not be important in the repository because the operating stress intensity, which is about 0.5–2 MPa·$m^{1/2}$ (Siegmann, 1997a), is less than the crack growth threshold, K_{IH}, for delayed hydride cracking. The value of K_{IH} decreases with hydrogen concentration in solid solution (Shi and Puls, 1994), and values ranging from 5 MPa·$m^{1/2}$ (Shi and Puls, 1994) to 12 MPa·$m^{1/2}$ (Cunningham, et al., 1987) have been reported. Based on the available evidence, it seems that delayed hydride cracking is not important under repository conditions, but this assessment can be strengthened by DOE if results regarding the probability of failure are quantified on the basis of the crack-size distribution in the cladding.

Zircaloy cladding generally contains some circumferential hydrides that have little or no effect on ductility or fracture toughness (Northwood and Kosasih, 1983). The amount of hydrogen pick-up in the repository is expected to be small. As a result, hydride embrittlement has not been studied closely. The occurrence of hydride embrittlement depends on the presence or absence of hydride reorientation from the circumferential planes to the radial planes. A few years after emplacement, the maximum cladding temperature for the design basis WP is expected to be about 330 °C (Siegmann, 1997b), which is above the solvus temperature of 290–300 °C (Northwood and Kosasih, 1983). On the other hand, the average temperature of the cladding is expected to be less than 237 °C (Siegmann, 1997b). Note that use of backfill would cause the temperature to increase. If the temperature of the cladding reaches more than 300 °C the circumferential hydrides are expected to redissolve in the Zr matrix. As the temperature drops below the solvus temperature, radial hydrides can reprecipitate at slow cooling rates if the cladding stress exceeds a critical value. The value of the critical stress required for hydride reorientation to occur is not well defined, but values ranging from 35 to 138 MPa have been quoted (Einziger and Kohli, 1984). The low critical stress value of 35 MPa is for Zr-2.5Nb (Hardie and Shanahan, 1975), whereas the value for Zircaloy is about 84–95 MPa (Marshall, 1967; Bai, et al., 1994; Chan, 1996). The cladding stress in the repository is anticipated to be 60–100 MPa (Siegmann, 1997b; Ahn, 1998). Under these stress levels, reorientation of hydride may be feasible at the slow cooling rates anticipated in the repository. The morphology of the radial hydrides formed under such conditions is conducive to embrittlement (Chan, 1996) and can lead to decreases in ductility and fracture toughness. Experimental data (Simpson and Cann, 1979; Kreyns, et al., 1996) indicate fracture toughness of Zircaloy cladding is reduced from about 44 MPa·$m^{1/2}$ without hydrogen to 7.5 MPa·$m^{1/2}$ at 4,000 wt. ppm hydrogen (Kreyns, et al., 1996); and to 1 MPa·$m^{1/2}$, at higher hydrogen contents (Simpson and Cann, 1979). Thus, hydrogen embrittlement may or may not be an important failure mechanism for fuel cladding in the repository environment, depending on the cladding temperature, which is a critical factor in determining the possibility of hydride reorientation.

Fuel and cladding oxidation are potential failure mechanisms if the disposal container fails when temperatures of the fuel assemblies are relatively high (>250 °C). Cladding oxidation would be the predominant failure mechanism if the fuel rods remain intact when the container fails, whereas fuel oxidation would be the dominant failure mode for perforated or failed fuel rods. The possible failure of fuel rods by cladding and fuel oxidation was examined by Einziger (1994); McCoy (1996); and Ahn (1996b). Cladding oxidation can be dismissed as a failure mechanism if the drifts are not backfilled because the rate of oxidation is not sufficiently fast at temperatures lower than 350 °C, as expected in the absence of backfilling. For perforated fuel rods, fuel oxidation is not expected to be a problem if the fuel temperature is lower than 250 °C because the fuel oxidation kinetics would be too sluggish. Above 250 °C dry air oxidation of irradiated UO_2 to U_3O_8, and the consequent volume increase, may split the cladding. Below this temperature, secondary mineral formation may occur in perforated fuels exposed to aqueous environments, leading to volume expansion and the potential for cladding failure. It appears that DOE is satisfactorily addressing this problem since experiments are currently being conducted at ANL to evaluate such possibilities under various environmental conditions, including tests in the vapor phase.

The possibility of mechanical failure of cladding through rock fall in the repository was considered by DOE assuming the following sequence of events, as described by McCoy (1998): (i) emplacement drifts collapse and become filled with rubble after repository closure; (ii) rocks fall and lie on top of the waste containers; (iii) waste containers corrode, become mechanically weak, and fail; and (iv) rocks fall from the container top and impact on the fuel assemblies inside. Mechanical failure of cladding is treated by considering a span of cladding from one spacer grid to the next as a simple elastic-plastic beam with clamped ends. Cladding is taken as a thin-walled tube subjected to an external load resulting from the rock fall at the middle of the span. The elastic-plastic behavior of the fuel rod is described by a piecewise linear stress-strain curve. The McCoy model takes into account the block size, block geometry, drop height, stack arrangement of the fuel assemblies, fracture geometry of the fuel rods and assemblies, and the load-displacement response of the fuel rod and assemblies. The choice of a failure criterion based on the uniform elongation of the fuel rod rather than using a fracture mechanics approach, which can be applied to partially degraded cladding with a distribution of flaws, is a limitation in the DOE analysis. In addition, to assess mechanical failure of cladding properly, the model should be extended by DOE to consider the effect of rock fall from drifts and seismic effects.

NRC agrees with the approach adopted by DOE to evaluate mechanical failure of fuel cladding with the understanding that most of the models used for the various processes are adequate. Nevertheless, some models require additional refinements to reduce uncertainties in the evaluation, particularly in the case of failure due to rock fall. However, the main concern that needs to be resolved refers to the temperature estimates for fuel cladding. Due to its important effect on various failure processes (e.g., creep, hydride reorientation and embrittlement), DOE should complete an evaluation of the range of cladding temperatures expected under repository conditions and the associated uncertainties to determine the probability of cladding failure and its consequence in terms of radionuclide release. Finally, DOE also needs to evaluate the extent of damage introduced during reactor operation and deterioration during transportation and storage that may affect the behavior under disposal conditions. Only limited data are available in this area.

5.3.6 Radionuclide Mobilization in Waste Packages

When radionuclides are released from the cladding, partially failed containers may be another barrier to radionuclide release (TRW Environmental Safety Systems, Inc., 1997b). The effectiveness of the container materials in reducing radionuclide release depends on the size and distribution of corrosion pits, presence of through-wall cracks, and the effect of corrosion products in the pits. The size of pits depends on the material and near-field environment. In the case of carbon steels, the pits are likely to be wide and filled with voluminous corrosion products (mainly various hydrated ferric oxides, such as akaganeite, lepidocrocite, magnetite, and ferrous chloride). In the case of stainless steels and Ni-base alloys, the pits will likely be narrow and deep with corrosion products not as voluminous as those of carbon steels. The distribution of pits depends on a number of geometric factors (presence of crevices, manufacturing defects, such as laps and grind marks) and metallurgical defects (sulfide and oxide inclusions, second-phase precipitates). In addition to through-wall pits developing from the outside, pitting may occur from the inside of the inner overpack after water flows in, through accumulation of fission products, such as ^{129}I, and ionic species already present in the groundwater. Extreme-value statistical treatments have been attempted in different applications, based on relatively short-term tests or field experience, to calculate the areal fraction of pits on carbon steel and aluminum (Aziz, 1956; McNeil, 1987). These approaches have not yet been applied to estimate the area of penetration of containers under repository conditions. On the other hand, if uniform corrosion takes place with scattered groundwater drips on the container, larger penetrations in the form of corrosion patches may develop (Lee, 1998). Corrosion products inside perforations or holes could provide diffusion barriers as in cladding. Although models of diffusion and convection are available to analyze restricted radionuclide release through perforations or holes (TRW Environmental Safety Systems Inc., 1995; U.S. Nuclear Regulatory Commission, 1995; Zwahlen, et al., 1990), experimental data are scarce to support the models. Wilson (1990a,b) compared the reduction of radionuclide release in J-13 water from SNF with cladding defects in the form of pinholes or slit flaws. DOE should provide adequate experimental data as a basis for the application of models of radionuclide transport through perforations both in containers and fuel cladding.

For colloidal transport, if the suspension of colloids is stable, release would take place by the transport of colloids through perforations in containers. However, colloidal transport could be limited because the penetrations may be small pits or hairline cracks filled with corrosion products (Johnson, 1998; Lee, 1998), as discussed above. DOE has not yet performed calculations to estimate the amount of colloids that could be transported through perforations. Bounding values could be acceptable if an appropriate scenario description is developed.

Depending on locations of the penetrations, radionuclide release may be modeled by an immersion (bathtub) or flow-through condition (TRW Environmental Safety Systems Inc., 1995; U.S. Nuclear Regulatory Commission, 1995). In the bathtub model, the critical parameters are the locations of the entry and exit ports for the water that determine the volume of water accumulating inside the container. There is great uncertainty regarding the likely geometry of these parameters because DOE has not conducted any relevant scale-down test to evaluate the location, morphology, and characteristics of corrosion penetrations. DOE should investigate, by conducting tests that simulate drift emplacement conditions, the probable location and distribution of penetrations in the containers because radionuclide release will be influenced by the amount of dripping water and the establishment of bathtub or flow-through conditions.

5.4 STATUS OF RESOLUTION OF SUBISSUE 4 AND RELATED OPEN ITEMS

5.4.1 Importance of High-Level Waste Glass for the Source Term

Projected radionuclide inventory for the HLW borosilicate glass is about 3 percent of the total radionuclide inventory to be disposed at the YM repository (U.S. Nuclear Regulatory Commission, 1995). Therefore, based solely on this percentage of the inventory argument, HLW glass could be neglected in the source-term analysis. Contribution of the HLW glass to the source term could be significant, however, if the rate at which the radionuclides can be released and transported from the glass is higher than that from the SNF (e.g., radionuclides released in colloidal form). The contribution could also be significant if radionuclides contained in the hydrated layer (corrosion product layer adhering to the glass surface) are released in larger quantities as a pulse, a situation that may not occur in the release from SNF.

DOE should provide confirmation that, in the above-mentioned cases, radionuclides are not released at rates greater than SNF. Otherwise, DOE should consider the effects of such processes and, hence, radionuclide release from HLW glass in PA. Note that DOE could use alternatives, such as glass ceramics, to solidify liquid HLW. Because details of these alternatives have not yet been reported, they will be discussed in future revisions of this IRSR, if proposed.

To evaluate the status of resolution of this subissue, several components are important, including HLW glass dissolution processes, formation of secondary minerals, natural analog studies, effects of colloids and microbes, and radionuclide transport in the EBS. Each of these components will be addressed in future revisions of the CLST IRSR. Unlike the metallic container materials, for which NRC has generated independent experimental data, DOE has generated most of the data regarding radionuclide release from HLW glass. Issue resolution in this context will be achieved through examination of the adequacy of DOE HLW glass data.

5.4.2 High-Level Waste Glass Corrosion Process

The HLW glass corrosion process involves: (i) transport and contact of reactant (i.e., groundwater or water vapor) to the HLW glass surface; (ii) chemical reaction between the reactants and glass surface; and (iii) transport of reaction products away from the reaction zone.

The HLW glass corrosion rate is controlled by the combination of these three processes and depends on factors, such as chemical composition of the glass and surrounding fluids; solubilities of the reaction products; exposed surface area; temperature; pH; and RH. The long-term corrosion behavior can be divided into three distinct stages as reviewed by Ellison, et al. (1994). In stage I, referred to as "the short-term stage," the chemical potential gradient between the glass components and local environment is the steepest. The glass components are released into the local environment at a comparatively high rate. The soluble components, such as boron and alkalis, are released at a higher rate compared with components, such as silica and aluminum oxide. This results in the formation of a layer on the glass surface that is depleted of soluble components, compared with the bulk glass. This layer is often called "the altered surface layer." In stage II, "the intermediate stage," the corrosion rate decreases as the concentration of reaction products, particularly silica, increases in solution. In addition, the reaction products in the altered surface layer reach saturation concentration of their crystalline phases and result in the formation of secondary phases, such as zeolites and clays. In stage III, "the long-term stage," glass corrosion rate is further affected because of the reprecipitation of secondary phases that exceed solubility limits at the altered zone. Physical processes, such as crystallization, cracking, or exfoliation of the altered surface layers, that occur in stage III, could influence the glass corrosion rate, as well as the release and transport of colloids and radionuclides. The change in dissolution rate also depends on the identity, distribution, and surface area of the secondary phases. In most cases the dissolution rate increases as a result of crystallization, exfoliation, and cracking of the altered surface layers. The transition from one stage to another is dependent on the glass composition and the local environment. A highly durable glass may take months to years to reach stage II, whereas a nondurable glass may reach stage II within hours or days.

It is evident, from this discussion, that the long-term stage cannot be characterized by a single reaction rate, as has been currently proposed by DOE. DOE has not taken into account different stages of the dissolution process in long-term glass dissolution models. In addition, models for matrix dissolution should cover a full range of the evolving environments that contact the WPs at the proposed YM repository because DOE has conducted tests using only demineralized or J-13 water.

5.4.3 Long-Term Glass Corrosion Studies

Several long-term HLW glass corrosion studies have been conducted in the last 20 years and research relevant to the YM repository is currently conducted at ANL on simulated HLW glasses, simulated glasses doped with plausible radionuclides, and fully radioactive glasses. Drip tests, designed to simulate slow flow through the breached canisters, have been used by Fortner and Bates (1996) and Fortner, et al. (1997) to study the long-term performance of actinide-doped West Valley Demonstration Project and Defense Waste Processing Facility (DWPF) HLW glasses, whereas the long-term product consistency test (PCT-B), designed to simulate fully immersed conditions, has been used by Ebert and Tam (1997) to study long-term performance of DWPF glasses. In addition, vapor hydration tests, designed to replicate a natural alteration process, are used by Luo, et al. (1997), to compare the dissolution behavior of DWPF glasses with that of naturally occurring basalt glasses. It is evident from the continuing studies at the ANL that the test conditions strongly influence glass corrosion rates and it is prudent to develop different tests to characterize various conditions that may exist in the repository. Although the previous studies may provide insight into corrosion mechanisms, a full range of the evolving environments in contact with the WPs at the proposed YM repository should be covered. In addition, the studies conducted by the ANL provide only limited data for a small set of simulated and actual HLW borosilicate glasses. DOE should ensure that the doped concentrations of radionuclides represent real concentrations in the HLW glass. Results of the ANL study should be confirmed by other laboratories to attain a reasonable level of confidence in the data to be used for PA calculations.

The dissolution rate of the HLW glass decreases as the groundwater environment in contact with the WPs becomes saturated with glass matrix components, such as silica. Even though the glass corrosion studies discussed previously confirmed that net dissolution rate decreases as the surrounding environment becomes rich in HLW glass matrix components, the drip test studies show a steep increase in radionuclide release rate for Pu and Am after 400 weeks. The steep increment in radionuclide release rate was attributed to the spalling of radionuclide containing colloids from the exposed HLW glass surface. The HLW glass corrosion models proposed by DOE do not account for such excursions in corrosion behavior. These excursions can have a significant effect on radionuclide release.

The dissolution kinetics of the primary phase, as commonly represented by the kinetics of B release, determine the release rate of high solubility radionuclides, such as Tc. The rate is dependent on the effective reactive surface area of HLW glass. The higher the exposed surface area, the higher the radionuclide release rate. DOE should provide the methodology used for estimating the effective reactive surface area for HLW glasses in the WP and incorporating the effect of surface area on the HLW glass dissolution rate.

The concentration of silica in the near-field environment may affect the degradation of SNF WPs. If there is abundant silica during SNF dissolution, uranosilicates may eventually form. If silica is depleted and groundwater in contact with WPs is limited, schoepites may form. Because the retention factors of radionuclides in uranosilicates and schoepites may be different, the silica release from HLW glass corrosion may affect the release behavior of radionuclides from SNF dissolution. DOE has not considered this effect in its PA. Clarification of the effects of HLW glass corrosion on radionuclide retention in secondary minerals during SNF dissolution is recommended.

5.4.4 Formation of Secondary Minerals during High-Level Waste Glass Corrosion

Long-term corrosion studies of HLW glasses indicate formation of secondary phases on the exposed surface of the HLW glasses. This process is dependent on the external environment. Long-term PCTs in J-13 water show formation of clay, Ca-phosphate, and (Th, U, Ca) titanate as secondary phases (Bates, 1998a), whereas the vapor hydration tests show accumulation of clay, zeolites, Ca-silicates, weeksite, and K-feldspar as secondary phases (Bates, 1998a). Formation of different phases under diverse test conditions is attributed to varying solution chemistries. These test conditions represent two environments that may exist at different times at the proposed YM repository. The formation of secondary phases may also be influenced by the corroding container materials. Secondary minerals play an important role in radionuclide release because they can incorporate low-solubility radionuclides, such as Pu and Am, and control their solubility limits. They may also act to block the reactive surface area of the primary phase.

It is important that the long-term radionuclide release rate in the corrosion models used by DOE include the influence of stage III behavior. The DOE models include the effect of temperature, pH, and dissolved silica; however, they do not consider the incorporation of radionuclides in the alteration phases that may result in periodic release spikes. DOE should provide experimental data on the formation of secondary phases under anticipated repository conditions, if the contribution of the HLW glass to the dose is significant. If the models are simply based on experimental dissolution data for stages I or II that exhibit significant retention of radionuclides in the secondary phases, evaluation of the long-term radionuclide release rates could be erroneous.

5.4.5 Natural Analog Studies of Glasses

Natural analog studies, coupled with experimental data and geochemical modeling, provide yet another method of gaining confidence in predicting long-term corrosion behavior of glasses. Natural analog studies are useful in evaluating the merits of extrapolating short-term experiments to longer time frames. There are several natural glasses, especially basalt, that have compositions comparable to the HLW glasses and that have been subjected to conditions similar to those expected in the proposed YM repository (Ewing, et al., 1998; McKenzie, 1990). The characterization of secondary phases formed on these natural glasses can provide insights into the long-term dissolution behavior of HLW glasses.

A recent study by Luo, et al. (1997) compared formation of secondary phases in the naturally occurring Hawaiian basaltic glasses with the results of vapor hydration tests that were conducted for 7 years on simulated basaltic and HLW borosilicate glasses. Luo, et al. (1997) concluded that secondary phases formed on both simulated natural glasses and HLW borosilicate glasses were similar to secondary phases observed in naturally occurring basaltic glasses, and vapor hydration tests could be used to simulate naturally occurring conditions.

DOE has not considered field data on naturally occurring glasses, combined with experimental data and models on dissolution of HLW glasses, to demonstrate that long-term dissolution behavior under repository conditions can be represented by extrapolation of results from short-term laboratory tests. Such data can be important to supplement and support the validity of the existing glass dissolution data generally obtained by short-term experiments.

5.4.6 Colloids, Microbes, and Radionuclide Transport in the Engineered Barrier Subsystem

Secondary phases formed on the HLW glass surface could be released as colloids and transported through the EBS (Manaktala, et al., 1995; Ahn, 1996a). These colloids could be hosts to radionuclides released from the glass. Sensitivity studies for colloid effects were performed using Version 3.2 of the NRC TPA code. A bounding case for Pu and Am colloid transport showed minimal effect on dose in the 10,000 year compliance period. Therefore, further examinations of colloidal effects on transport and release are not considered necessary. A summary of colloidal effects is summarized in this section for completeness.

Recent research by Fortner, et al. (1997) and Fortner and Bates (1996) have not only shown formation of colloids in the alteration phases, but also the transport of 100 percent of the actinides as colloids to the environment. Thus, colloids could lead to sudden increases in actinide concentrations in the environment present in the emplacement drifts. DOE has identified dominant colloid formation processes under anticipated repository conditions but has not modified the long-term dissolution models to account for such events. Currently, DOE has adopted empirical correlations of the colloidal suspension based on the ionic strength and pH

(Triay, 1998). If the suspension of colloids is stable, release would take place by the transport of colloids through perforations in containers. However, colloidal transport could be limited because the penetrations may be small pits or hairline cracks filled with corrosion products (Johnson, 1998; Ahn, 1994; Lee, 1998). DOE has not performed calculations to estimate the amount of colloids that can be transported through perforations.

Studies conducted to date have ignored the corrosion of HLW glasses by the action of microbes. The effect of microbes in the dissolution of natural glasses can be significant (Thorseth, et al., 1992). Microbes can also change the solubilities of radionuclides by the increased production of organic acids. DOE should study the effects of microbial activity on HLW glass corrosion if the viability and nature of the microbial activity at the proposed YM repository are demonstrated through the use of calculations based on a mass balance of nutrients and energy-generating chemical reactions, as indicated in the ENFE IRSR.

Transport of radionuclides released from HLW glass depends on several factors, such as size and shape of perforations (e.g., pits) in both the HLW glass pour canister and the disposal containers. The local environment around the container is based on location of the perforations. If perforation occurs as holes on the upper half of the container, immersion (bathtub) conditions will prevail; however, if the penetration occurs also at the bottom of the container, drip conditions would develop. Radionuclide release models for bathtub or drip scenarios have been developed (TRW Environmental Safety Systems Inc., 1995; U.S. Nuclear Regulatory Commission, 1995) but have not been validated using experimental radionuclide release data. DOE should validate these scenarios and models for the anticipated YM repository conditions. In addition, DOE needs to conduct a characterization of dissolved radionuclide species formed from HLW glass because these chemical characteristics will affect the transport of radionuclides through the EBS leading to enhanced release or retention of radionuclides.

5.5 STATUS OF RESOLUTION OF SUBISSUE 5 AND RELATED OPEN ITEMS

In January 1999, DOE submitted its "Disposal Criticality Analysis Methodology Topical Report, YMR/TR-004Q, Revision 0" (U.S. Department of Energy, 1998d) for the staff's review and approval. This report provides a description of DOE's methodology for demonstrating postclosure criticality control for the YM repository, and, correspondingly, protection of public health and safety. The methodology is risk-informed and, as such, focuses on probabilistic evaluations of the various processes and events that may result in configurations with a potential for criticality. The report also discusses the potential consequences of criticality and the related impacts to total repository system performance. A Request for Additional Information (RAI) has been sent to DOE (Letter from C.W. Reamer/NRC to S. Brocoum/DOE, August 18, 1999).

DOE plans to apply the methodology presented in the topical report "to the different waste forms: commercial SNF (including boiling water reactor, pressurized water reactor, and mixed oxide SNF); DOE SNF (including naval SNF); immobilized plutonium; and vitrified HLW glass." Specific application of the methodology to naval and DOE-owned SNF will be included in a series of addenda to the topical report, to be submitted for the NRC review and approval. The scope of the topical report includes a range of possible postclosure criticality locations (in-package, near-field, and far-field) over time. The following sections provide the status of the in-package criticality issues, which have been identified as the result of the staff's initial review of the DOE topical report. The issues are discussed with respect to the acceptance criteria provided in section 4.5.1.

5.5.1 Criticality Design Criteria

The design criteria proposed by the DOE for the in-package criticality scenario pertain mainly to commercial spent nuclear fuel. DOE has presented a set of design criteria with respect to 1) critical limit (CL); 2) probability; 3) criticality consequence; and 4) repository perfomance. The following paragraphs discuss only the design criteria with respect to issues that have been identified at this time.

Critical Limit is the value of k_{eff} at which the system is considered potentially to be critical. A system with the value of $k_{eff} = 1$ is critical. To identify the biases and uncertainties associated with modeling and computational tools which determine a critical limit value, critical experiments which have characteristics similar to that of the system of interest (i.e., EBS) are used to test and identify calculational tools and modeling biases and uncertainties. Since critical experiments which would involve actual spent fuel in a waste package may not be feasible, isotopic measurements from spent fuels combined with fresh fuel laboratory critical experiments or other critical experiments may be used to identify the criticality code biases and uncertainties. These biases and uncertainties basically indicate how much the criticality code would under predict the value of k_{eff} as functions of certain parameters.

Some of the major concerns with respect to DOE's approach in establishing the CL are:

> DOE is taking an integral validation approach which results in cancellation of errors for establishing both isotopic and criticality biases. DOE is performing separate isotopic and criticality validation by direct comparison of calculated to measured values establishes the true biases and uncertainties which are associated with the computational tools. This issue does not apply to the DOE-owned spent fuel if the fresh-fuel assumption is used as indicated by DOE.
>
> DOE is using the non-bounding parameter values for spent fuel exposure history and the uniform, instead of a more realistic, profile for axial burnup distribution. These assumptions result in higher CL or lower k_{eff} depending on how the biases and uncertainties are added. This issue does not apply to the DOE-owned spent fuel if the fresh-fuel assumption is used as indicated by DOE.
>
> DOE is redefining administrative margin (i.e., Δk_m) without accounting for the unanalyzed scenarios, operational errors, and the degree of subcriticality. The approach proposed by Navy include a 5% administrative margin which would eliminate this issue with respect to the Navy fuel.

DOE has proposed to use only the increase in radionuclide inventory in a single package as the criticality consequence criteria. This criteria will be used either for screening FEPS or design modifications. In addition to the radionuclide inventory, other criticality consequences, such as increase in heat and its effect on the waste package corrosion rate, and waste form alteration resulting from transient criticality, must be included in the total consequence analysis.

5.5.2 Criticality Scenarios

In its development of Master Scenarios List for in-package criticality, DOE has not explicitly considered some of the FEPS associated with the Yucca Mountain site. Inclusion of seismicity, faulting, and igneous activity in the in-package criticality scenario development is not apparent.

The seismic event is especially important in evaluating scenarios which involve step-insertion reactivity in a very short period. The consequences of this scenario become important in terms of mechanical disruption of the EBS and the waste form. Other seismic consequences include possible loss of neutron-absorbing material on which DOE may rely as part of the long term criticality control system.

Faulting is a feature which needs to be considered in combination with seismic event. Therefore, scenarios based on seismic events combined with movements on the faults in the vicinity of waste package need to be considered with respect to formation of critical configurations.

5.5.3 Criticality Configurations

In developing configurations with respect to in-package criticality, the range of parameter values for the waste form chemical, physical, and nuclear characteristics must be determined. Furthermore, the physical and chemical characteristics of the waste package internal components as a function of time must be determined.

DOE is proposing to rely on the presence of corrosion products which are formed from gradual corrosion of the various steel components within the waste package for criticality control. The issues identified by the staff at this point are on the assumptions with regard to the chemical composition and quantity of the corrosion product present within the waste package.

DOE has assumed that Fe_2O_3 is formed by the corrosion of iron. Another possible corrosion product is FeOOH. DOE assumes that FeOOH is equivalent to the water/Fe_2O_3 mixtures. Alternate forms of corrosion products such as FeOOH need to be considered explicitly.

Another issue is the assumption with regard to the quantity of the iron oxide remaining within the waste package, which would displace the water and function as neutron absorber. Given the uncertainty in the pH of the water and the thermal circulation within the package, the iron oxide can be gradually transported from the waste package. Another possibility is the flushing of the iron oxide in a flow through configuration. With inflow being greater than the outflow, the water will be present within the waste package with gradual removal of iron oxide.

5.5.4 Criticality Probability

DOE has proposed to use a Monte Carlo-based approach in order to determine if the probability of k_{eff} value for a waste package is higher than the CL for a given waste package and waste type.

In calculating the probability of internal criticality, the proposed approach is based on sampling the values for a number of parameters. These parameters include drip rates, barrier lifetimes, location of the penetration into the waste package, waste form parameters (e.g., burnup and enrichment), and degradation of waste package internal components and waste form.

Although the details of the method have not been reviewed, the overall approach has been examined. As the result of the review, some questions such as accounting for the correlation of the above sampled parameters have been raised.

5.5.5 Criticality Analysis

The tools proposed by the DOE for performing in-package steady state and transient criticality analyses for commercial SNF are SAS2H, MCNP4A, and RELAP5/MOD3.2 computer codes. The issues identified to this date are the validation and applicability of the tools for the in-package criticality analysis.

The issue with respect to integral validation of SAS2H and MCNP4A and establishing bounding parameter values for commercial spent fuel irradiation were discussed earlier with respect to CL under the design criteria.

The adequacy of SAS2H, which is a point depletion code, for calculating isotopic inventory in a commercial spent fuel assembly is the other concern. Radial and axial neutron spectra variations are the environments under which fuel assemblies are irradiated in reactor cores and SAS2H can not model these neutron spectra variations appropriately. Especially with respect to the BWR fuel characteristics (e.g., varying axial and radial enrichment and presence of integral burnable absorbers), the limitation of SAS2H becomes very apparent.

The use of RELAP5 for performing transient criticality analysis is another area of concern. RELAP5 is a thermal hydraulics code developed for reactor core-type of flow conditions, which is mainly flow parallel to the fuel assemblies. No major cross flow is allowed. The water flow in a transient condition within the waste package would be mainly perpendicular to the fuel assemblies. This is one of the issues with respect to criticality analysis which needs to be addressed by DOE.

5.5.6 Criticality Consequence

The in-package criticality consequence analysis presented in the topical report is with respect to steady state and transient conditions.

The consequence analysis method proposed by DOE considers only the radionuclide buildup from long-term steady state critical condition. Other consequences such as additional heat buildup and its effect on the waste package corrosion and waste form dissolution have not been considered. In addition, effects of the additional heat on the near-field environment need to be analyzed.

The consequence of transient criticality is mainly a function of the reactivity insertion rate. This is related to the scenarios such as seismic shaking which would create a rapid reactivity insertion rate. The scenario analyzed by DOE involves only reactivity insertion as a function of time needed for settling the iron oxide which has been estimated to be 36 seconds. As indicated earlier, the presence of iron oxide within the waste package is questionable. In addition, the reshuffling of the spent fuel assemblies from seismic shaking is a more constraining scenario, with respect to time, than the free fall of iron oxide particles. Therefore, consequences for transient criticality within the package with the reactivity insertion in one second or less needs to be also evaluated.

The consequences of other transient criticality conditions such as autocatalytic criticality which could result from over moderation within the waste package needs to be also considered.

5.5.7 Criticality Risk

The total risk associated with internal criticality is the combination of probability and consequences for all the possible scenarios and configurations in terms of the incremental dose to the member of the critical group at 20 km from the proposed Yucca Mountain site.

DOE's preliminary analysis for a single PWR waste package becoming critical at a steady state condition for 10,000 years indicates that the increase in the inventory of the 36 TSPA isotopes is 24%. The increase in the isotopes which are important to the repository performance, I-129, Tc-99, Np-237, and U-234, are between 4% and 11%. Tc-99 provides the highest and earliest incremental dose which is around 0.07 mrem/year. This appears to be an unrealistically high dose to result from only a 4% increase in the inventory of Tc-99 in a single waste package.

Furthermore, as indicated earlier, the above risk analysis has not taken into account the other consequences such as additional heat buildup and mechanical disruption from transient criticality. The risk from criticality must also include these consequences.

5.6 STATUS OF RESOLUTION OF SUBISSUE 6 AND RELATED OPEN ITEMS

The alternate design options of backfill, drip shields, and ceramic coatings have not yet been addressed in sufficient detail. DOE is conducting an experimental program for the evaluation of ceramic coatings (TRW Environmental Safety Systems, 1998d). DOE is evaluating the performance of titanium alloys through long-term corrosion tests. However, there is very little data pertinent to Ti grade 7, the alloy chosen as the candidate drip shield material. The Ti drip shield performance is important in determining when water contacts the containers and how radionuclide release occurs.

5.6.1 Thermal Embrittlement of Titanium Drip Shield

Similar to ferritic-pearlitic low-alloy steels (Cragnolino, et al., 1996), α-Ti alloys can also suffer from thermal embrittlement. Both temper embrittlement of steels and thermal embrittlement of α-Ti alloys occur as a result of a thermally activated redistribution of barely soluble impurities from grain interiors to grain boundaries (Nesterova, et al., 1980). Both phenomena are characterized by: (i) embrittlement of the material, (ii) nominally constant strength with widely variable ductility, and (iii) increased intergranular boundary fracture. Unlike temper embrittlement of iron in which only equilibrium segregation of impurities (i.e., P, As, Sn) occurs, impurity segregation in α-Ti results in the precipitation of finely dispersed particles at the grain boundaries (Nesterova, et al., 1980; Kryukov, et al., 1981; Ushkov, et al., 1984; Ushkov, et al., 1995).

Commercially pure (CP) Ti exhibits a minimum impact toughness after heat treatment in the temperature range of 350 to 600 °C for 500 hours. Embrittlement susceptibility starts at approximately 350 °C and, at higher temperatures, reaches a maximum (minimum in toughness) near 500 °C, decreasing at even higher temperatures. At temperatures greater than 700 °C, the embrittlement effect disappears, and the impact toughness approaches its original value. Similar results are shown for a Ti-2Al-1.5Zr alloy (Nesterova, et al., 1980). For Al-bearing α-Ti alloys, embrittlement occurs partially as a result of decomposition of the Al in solid solution with Ti and the precipitation of α_2 (Ti_3Al) throughout the interior of grains, which results in embrittlement of the grains themselves (Nesterova, et al., 1980; Ushkov, et al., 1984; Ushkov, et al., 1995). For CP Ti and α-Ti alloys that contain trace amounts of Fe and Ni as impurities[3], embrittlement results from the precipitation of sub-μm size face-centered-cubic (fcc) Ti intermetallics in the form of $Ti_2(Fe,Ni)$. The effect of longer thermal holds (> 500 hr) on embrittlement has received little attention, but has revealed significant segregation of Fe and Ni at grain boundaries at temperatures as low as 350 °C (Ushkov, et al., 1984). Thus, the reported minimum Fe concentration of 0.01 wt% for the observance of embrittlement may not be valid for longer thermal treatments. For near-α alloys, such as Ti Grade 12 (Ti-0.3Mo-0.8Ni), and alloys that contain a sufficient concentration of β stabilizing elements, thermal embrittlement is not generally observed. In these alloys, any Fe present as an impurity is adsorbed by the β phase that is present. As such, the effective concentration of Fe in the α phase of the alloy is reduced to a very low level (Nesterova, et al., 1980). The temperature experienced by the Ti drip shield is likely to be much lower than that of the containers. Nevertheless, considering the long exposure period, the susceptibility to mechanical failure by embrittlement should be examined further.

5.6.2 Uniform and Localized Corrosion of Titanium Drip Shield

Ti alloys have been long recognized for being highly resistant to corrosion as a result of their ability to form a protective oxide film when in contact with oxygen or water (Schutz, 1992). The presence of oxidizing agents, such as nitric and chromic acids, and the dissolved salts of iron, nickel, copper, and chromium can act as inhibitors and decrease corrosion. The oxidizing agents likely inhibit corrosion by accelerating growth of the oxide film because they increase the potential into the region of stability of TiO_2. The pH and chloride concentration have been found to have a relatively minor influence on the passive dissolution rate of some Ti alloys, although data are limited in this area. Decreasing the pH from near neutral to 1 in 0.6 M sodium chloride solutions raised the passive dissolution rate of Ti-15V-3Cr (a β-Ti alloy) by a factor of 2 (from 0.5 to 1.1 $\mu A/cm^2$). When tested in 5 M HCl, the passive current density increased only by an order of magnitude compared to that measured in 0.6 M NaCl (Kolman and Scully, 1994). In other work on pure Ti thin films, it was observed that the pH and chloride concentration had little to no influence on the passive dissolution rate (Kolman and Scully, 1996). Similar observations that pH and chloride concentration do not affect general corrosion

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[3] As impurities, typical concentrations for Fe and Ni in α-Ti alloys are ≤ 0.05 wt% and ≤ 0.005, respectively (Nesterova, et al., 1980; Ushkov, et al., 1984).

rates have been made during industrial experience using Ti alloys in geothermal and desalination plant applications (Conover, et al., 1980).

A critical review of the literature on the localized corrosion of Ti alloys indicates that while Ti alloys can be susceptible to crevice corrosion, insufficient experimental data are available for Ti-Pd alloys, such as Ti grade 7, under relevant conditions to adequately determine the conditions for various corrosion modes and their propagation rates. Furthermore, there is insufficient data that could be used to model long-term material performance using the TPA code. Crevice corrosion of CP Ti has been observed under conditions that could develop in the repository. At reasonably achievable potentials for the repository (−0.1 V_{SCE}), the minimum temperature at which crevice corrosion was observed is approximately 170 °C at a chloride concentration of 0.034 M, 80 °C at 0.2 M chloride, and 50 °C at 1 M chloride (Tsujikawa and Kojima, 1993). Based on these reported results, crevice corrosion of Ti is possible in the repository. The addition of Pd to Ti, though, does improve the crevice corrosion resistance of Ti. Given the lack of data for Ti-Pd alloys, however, further examination of the conditions conducive to crevice corrosion of Ti-Pd is warranted. Experiments are needed to determine if more aggressive conditions are required to nucleate and propagate crevice corrosion in these materials or if Pd additions just delay the onset of crevice corrosion through an increase in the incubation time.

5.6.3 Environmentally Assisted Cracking of Titanium Drip Shield

Environmentally assisted cracking (EAC) of Ti-Pd alloys has not been extensively examined as concluded in a recent review of alternative EBS designs (Cragnolino, et al., 1999). Many Ti alloys are susceptible to EAC due to hydrogen embrittlement associated with the precipitation of hydrides ahead of the crack tip. However, the Ti-Pd alloys may be highly resistant to EAC, especially those that have a low equivalent oxygen content (high O_{equiv} leads to higher strength and greater susceptibility). The addition of Pd to Ti has also been thought to enhance EAC resistance because hydrogen evolution as H_2 would preferentially take place at Pd-rich sites, thereby decreasing the available atomic hydrogen that could be absorbed into the Ti lattice. It is unclear if this mechanism is operable. In any event, the relatively few reported EAC failures of Ti-Pd alloys may be a result of very slow crack propagation rates, which become important in engineering structures intended to maintain functionality for extremely long periods of time (i.e., several thousands of years in the case of the drip shield and waste package). Thus, further DOE investigation of the EAC behavior of Ti-Pd alloys is needed, with particular emphasis on methodologies that would enable monitoring and measurement of slow crack propagation rates.

Also of importance are the possible interactions with other materials present in the repository and the effects of fabrication on degradation processes. Galvanic coupling of Ti to iron/carbon steels, stainless steels, and Ni-based alloys may lead to conditions that enhance the propagation rates of processes already occurring. Though some evidence shows that fabrication processes would not significantly affect material performance, the resolution limit of the methods used may mask propagation rates (in terms of SCC/HE crack propagation) that would be relevant on the time scale of repository performance.

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APPENDIX

Status of U.S. Nuclear Regulatory Commission Site Characterization Analysis Open Items on Waste Package and Release from Engineered Barrier System

Item ID	Source	Title	Status	KTI	Comment
Comment 5	SCA	Interpretation of substantially complete containment	Resolved 7/11/94	CLST	
Comment 6	SCA	Performance assessment: hypothesis testing table and alternative conceptual models		All	
Comment 25	SCA	Rationale on additional testing on waste and interactions between and among radionuclides on sorption		CLST ENFE RT	
Comment 28	SCA	Sorption on particulates and colloids	Resolved 7/31/91	RT ENFE CLST	
Comment 30	SCA	Solubility modeling	Resolved 7/31/91	ENFE RT CLST	
Comment 44	SCA	Overall goal is not consistent with substantially complete containment	Resolved 7/31/91	CLST	
Comment 47	SCA	Relationship of postclosure tectonics to the waste package and the engineering barrier system requirement		SDS CLST	
Comment 79	SCA	Adequacy of waste package corrosion tests for the repository	Resolved 2/12/98	ENFE CLST	
Comment 80	SCA	Performance goals consistent with interpretation and intent of substantially complete containment	Resolved 3/7/95	CLST	

Status of U.S. Nuclear Regulatory Commission Site Characterization Analysis Open Items on Waste Package and Release from Engineered Barrier System (cont'd)

Item ID	Source	Title	Status	KTI	Comment
Comment 81	SCA	Adequacy of program in stress corrosion cracking behavior of waste packages	2/12/98	CLST ENFE	
Comment 82	SCA	There is an inadequate discussion on how the waste package performance may be verified at the time of license application		CLST	
Comment 83	SCA	The term "uniform corrosion" is misleading	Resolved 7/31/91	CLST	
Comment 84	SCA	Issue resolution strategy and testing package for the waste package and engineering barrier system do not take into account the full range of likely natural conditions that might affect performance of the barrier	Resolved 2/12/98	CLST SDS ENFE	
Comment 85	SCA	Performance Assessment: Temporal changes in the state of stress due to corrosion of the container is not accounted for	Resolved 3/9/98	CLST	
Comment 86	SCA	Degradation modes of copper-based alloys do not appear to agree with scientific literature	Resolved 2/12/98	CLST	
Comment 87	SCA	Adequacy of effects of dissimilar metal contacts causing corrosion		CLST	
Comment 88	SCA	Assumption of reduced uncertainties because of the unsaturated zone		CLST	

Status of U.S. Nuclear Regulatory Commission Site Characterization Analysis Open Items on Waste Package and Release from Engineered Barrier System (cont'd)

Item ID	Source	Title	Status	KTI	Comment
Comment 89	SCA	Construction materials may change the local pH and affect the corrosion of the metal containers and the leach rates of radionuclides from the glass	Resolved 2/12/98	ENFE CLST	
Comment 90	SCA	Consideration of varying oxygen concentrations on the corrosion of metal containers	Resolved 2/12/98	CLST ENFE	
Comment 91	SCA	Waste Package/Performance Assessment: Consideration of alternative canisters for carbon-14 releases		CLST	
Comment 97	SCA	Adequacy of evidence to eliminate iodine as an important radionuclide	Resolved 7/31/91	RT CLST	
Comment 118	SCA	The monitoring and testing activities should include long-term in situ and long-term waste package activities		CLST TSPAI	
Question 30	SCA	Water quality as related to waste package design		ENFE CLST	
Question 31	SCA	Integrity of spent fuel cladding		CLST	
Question 32	SCA	Container "similarity" for borosilicate glass waste vs. spent fuel		CLST	
Question 34	SCA	Meaning of "undetected defective closures" in waste package fabrication and handling design goals		CLST	
Question 35	SCA	Acceptance criteria for helium leak results	Resolved 3/7/95	CLST	

Status of U.S. Nuclear Regulatory Commission Site Characterization Analysis Open Items on Waste Package and Release from Engineered Barrier System (cont'd)

Item ID	Source	Title	Status	KTI	Comment
Question 36	SCA	Explanation and justification for use of corrosive surface finishing chemicals on waste package prior to emplacement		CLST	
Question 37	SCA	Basis for 10 cm or more of free fall for canister and contents	Resolved 3/7/95	CLST	
Question 38	SCA	Basis for mm scratch criterion to avoid emplacement of damaged canisters		CLST	
Question 39	SCA	Meaning of "unusual process history" as a criterion to avoid emplacement of damaged canisters		CLST	
Question 40	SCA	Basis for using a factor of 2 for corrosion for rate for borehole liner in comparison to container material		CLST	
Question 43	SCA	Anticipated operational occurrences considered part of normal conditions on the preclosure design and analysis	Resolved 7/31/91	SDS CLST	
Question 44	SCA	Basis for assumed numbers of breached assemblies or canisters		RDTME CLST	
Question 45	SCA	Waste package: investigation of particulate source terms, retention factors, and plate-out of waste package during accident conditions		CLST	
Question 46	SCA	Basis for stricter containment of long half-life isotopes	Resolved 7/11/94	CLST	

Status of U.S. Nuclear Regulatory Commission Site Characterization Analysis Open Items on Waste Package and Release from Engineered Barrier System (cont'd)

Item ID	Source	Title	Status	KTI	Comment
Question 47	SCA	What is the origin of the stated definition of a container failure	Resolved 3/7/95	CLST	
Question 48	SCA	Selection of peer review panel on waste package	Resolved 7/31/91	CLST TSPAI	
Question 49	SCA	Effects of low temperature oxidation on containers	Resolved 3/9/98	CLST	
Question 50	SCA	Assumption that stress propagation results in corrosion	Resolved 7/31/91	CLST	
Question 51	SCA	Impacts of INEL and Hanford high-level wastes on the YM Program	Resolved 11/8/94	CLST	
Question 52	SCA	Leaching properties specification will require the producer to control leaching characteristics of the glass waste	Resolved 7/31/91	CLST	
Question 53	SCA	Specification of cooling rate of the glass waste	Resolved 3/7/95	CLST	
Question 54	SCA	Release rates of radionuclides from spent fuels in J-13 water	Resolved 7/31/91	CLST	
Question 58	SCA	Flexibility of the Exploratory Shaft Facility design to accommodate *in situ* testing of the waste package, if required	Resolved 9/97	RDTME CLST	