

August 13, 2001

The Honorable Richard A. Meserve  
Chairman  
U. S. Nuclear Regulatory Commission  
Washington, DC 20555-0001

Dear Chairman Meserve:

SUBJECT: REVIEW OF CHEMISTRY ISSUES AND RELATED NRC STAFF CAPABILITY  
FOR THE PROPOSED HIGH-LEVEL WASTE REPOSITORY AT YUCCA  
MOUNTAIN

The ACNW conducted a review of chemistry issues associated with the performance of the engineered barrier system (EBS) and the release and transport of radionuclides for the proposed Yucca Mountain high-level waste (HLW) repository. As part of this review, an ACNW Working Group on Chemistry Issues for the HLW repository at Yucca Mountain met on February 21 and 22, 2001, at NRC headquarters in Rockville, Maryland. The chemistry review is part of the ACNW's "vertical slice" approach to evaluating the NRC's process for the identification and resolution of issues related to the proposed Yucca Mountain HLW repository. This letter presents observations and recommendations based on the ACNW's review of the Working Group's report, which is attached.

The Working Group focused on waste package and drip shield corrosion, near-field (i.e., inside the drifts) and in-package chemistry, radionuclide mobilization and transport, and coupled thermal-hydrologic-chemical (THC) processes. The Working Group addressed the adequacy of process-level modeling to address specific chemical issues, the abstractions of process models into the Total System Performance Assessment for the Site Recommendation (TSPA-SR), and the technical bases of the TSPA-SR. The Working Group also commented on the degree to which "conservative" assumptions about chemistry might compromise the credibility of the analyses of the near-field and natural environments, and on how well the NRC is prepared to address contingencies, such as changes in DOE's repository design (e.g., from a "hot" to a "cold" repository) or changes in the DOE safety-case strategy.

### **Observations and Recommendations**

#### **Observation 1**

The NRC staff has developed a process for reviewing issues in connection with the proposed Yucca Mountain repository. The process is based on a set of Key Technical Issues (KTIs) and related subissues that are documented in the Issue Resolution Status Reports (IRSs). This iterative process for reviewing issues in technical exchanges and establishing specific agreements with DOE appears to be appropriate. The staff is addressing the important chemistry issues on

engineered barrier lifetimes and radionuclide release and transport through the KTIs. The issue resolution process is neither designed nor intended to serve as a basis for unearthing new, potentially important chemistry issues that DOE has not considered. In general, the NRC and Center for Nuclear Waste Regulatory Analyses (CNWRA) staff seem to be well positioned to deal with the impacts of evolutionary repository design changes. Major design changes featuring new components, materials, and approaches could, however, have a significant impact on the staff's ability to complete a thorough review in a timely fashion.

### **Recommendation 1**

The staff should continue exploring the chemical issues associated with major repository design changes such as a "hot" versus a "cold" repository or the use of backfill.

### **Observation 2**

The KTIs dealing with chemistry-related issues are generally comprehensive. There are numerous specific subissues and questions that are intended to address chemistry issues in detail. These are addressed by DOE with uneven quality and thoroughness. DOE tends to use very "conservative" assumptions in some cases but not in others, and does not integrate differing approaches in a consistent way. The inconsistent use of "conservatism" throughout the TSPA-SR models makes it difficult to identify issues important to risk and to ascertain if particular errors or problems are significant to overall performance. In addition, the complexity of the TSPA-SR model and code make it difficult to evaluate.

The NRC staff (including the CNWRA) is employing its own analytical tools (e.g., the Total-System Performance Assessment [TPA] model) and expertise on repository chemistry to reach conclusions on DOE's ability to meet regulatory requirements for licensing.

### **Recommendation 2**

- The NRC staff should continue to be supported and encouraged in their efforts to conduct independent analyses using the NRC's TPA code, process models, and internally developed confirmatory data and information in reviewing the proposed Yucca Mountain repository.
- The staff needs to continue evaluating DOE's TSPA computer code and supporting process models in as much detail as practicable to fully understand how the models are implemented in the codes and the important results.
- The staff needs to have sufficient independent computer capability to evaluate probabilistic codes for TSPA and process models and to defend its conclusions about DOE's ability to meet regulatory limits.

### **Observation 3**

The long-term extrapolation of short-term experimental corrosion data to material performance continues to be a concern to the ACNW. For example, engineered barriers are modeled as primarily corroding in humid air, but the data used is derived from experiments and tests in fully saturated conditions. This assumption may be appropriate, but there are uncertainties about the longevity of the corrosion-resistant oxide layers on the metals. This may lead to a lack of confidence in the validity of the results obtained. An understanding of waste package corrosion

mechanisms relevant to the expected repository environment is central to projecting repository performance over long times. Planned upgrades in the NRC's TPA code and ongoing experimental work will be useful for understanding the consequences of these issues.

### **Recommendation 3**

We reiterate previous ACNW recommendations that staff continue developing a better understanding of corrosion mechanisms for Alloy 22 and other engineered barrier materials to judge the validity of extrapolating short-term experimental results on material performance to very long time frames.

### **Observation 4**

We continue to be concerned about the treatment of coupled chemical processes due to the difficulty in incorporating them into the modeling, the complexity of the processes, and the potential for negative impacts on performance. Although considerable effort has been expended both by NRC and DOE on coupled processes, the inherent complexity of phenomena and the paucity of important data have limited the benefit of what has been done. In addition, continuing uncertainty about final repository design introduces uncertainty in the identification of important contributors to coupled processes such as temperature effects and the impacts of material interactions on chemistry, barrier degradation, and possible radionuclide releases and transport. It is likely that such processes can have important influences on repository performance.

### **Recommendation 4**

The staff should continue evaluations of coupled THC processes, especially with respect to the chemical environment on the waste packages and drip shields, to ensure that important phenomena that affect the degradation of barrier performance are not overlooked.

### **Observation 5**

The overall TSPA-SR treatment of in-package chemistry, which controls the release rates of radionuclides, does not address important complexities and the technical bases are not adequately described and justified. In addition, it is not necessarily a "conservative" approach for waste packages that have some liquid water present inside. It is not clear that process model abstractions capture the important uncertainties of in-package and near-field chemical conditions (e.g., pH and oxidation-reduction reactions).

### **Recommendation 5**

The staff needs to more fully address in-package chemistry issues as it develops an integrated chemistry model to be implemented in TPA. Current plans for upgrading the chemistry capabilities of the NRC's TPA code emphasize the chemistry of water on the exterior of the waste packages.

### **Observation 6**

The TSPA-SR employs conflicting physical and chemical processes and conditions to model the radionuclide source term. For example, the in-package chemistry model is based on the assumption that the waste package is full of liquid water. This is not physically realizable for the

vast majority of waste packages that are only coated with a thin film of moisture. The source term model, however, employs a “nonmechanistic release scenario” that does not account for varying amounts of moisture inside the waste packages. While this appears to be a very “conservative” approach, it is not clearly discussed and leads to confusion and uncertainty as to its importance to dose. This approach may be a significant contributor to relatively high dose values at long time frames in TSPA-SR, and when waste packages are neutralized in DOE’s analyses for its Repository Safety Strategy. This may lead to perceptions that early waste package failures would result in doses greatly exceeding regulatory limits and that only the waste package outer barrier stands between the public and the waste at Yucca Mountain. The NRC and CNWRA have identified a number of concerns with the implementation of the source term model in TSPA-SR, and DOE is likely to put forth a more realistic source term model in the next iteration of TSPA.

### **Recommendation 6**

The staff should continue to develop an appropriate source term model for NRC’s TPA code to be prepared to review a more realistic source term model from DOE and should continue identifying important parameters, issues, and concerns.

### **Observation 7**

Transport of radionuclides as both ionic and colloidal species in TSPA-SR is represented by unchanging parameters (e.g.,  $K_d$ s) that provide little predictive capability or insight into the controlling chemical mechanisms for retardation. DOE has agreed to address numerous staff concerns about colloid formation and movement and the development of appropriate information for radionuclide transport and attenuation.

### **Recommendation 7**

NRC and CNWRA staffs should continue efforts to gain better insights into radionuclide transport and attenuation processes in TSPA.

### **Concluding Remarks**

The NRC’s prelicensing process for resolving issues, as it pertains to chemical phenomena affecting performance of the proposed Yucca Mountain repository, is well documented in the IRSRs. The staff appears to be addressing the chemistry issues that are likely to be important and the issue resolution agreements with DOE appear appropriate. Currently, limited information exists on the chemical processes that may mobilize radionuclides and transport them through the EBS. In subsequent iterations of TSPA, DOE needs to conduct a thorough reevaluation and documentation of the information bases, modeling approaches, and calculations of in-package chemistry, and to develop more realistic approaches for radionuclide release and transport. We continue to be concerned about the treatment of coupled chemical processes because of the potential for negative impacts on performance in the compliance period. We believe that

these issues are solvable in a timely fashion but will require a careful evaluation of models and approaches.

Sincerely,

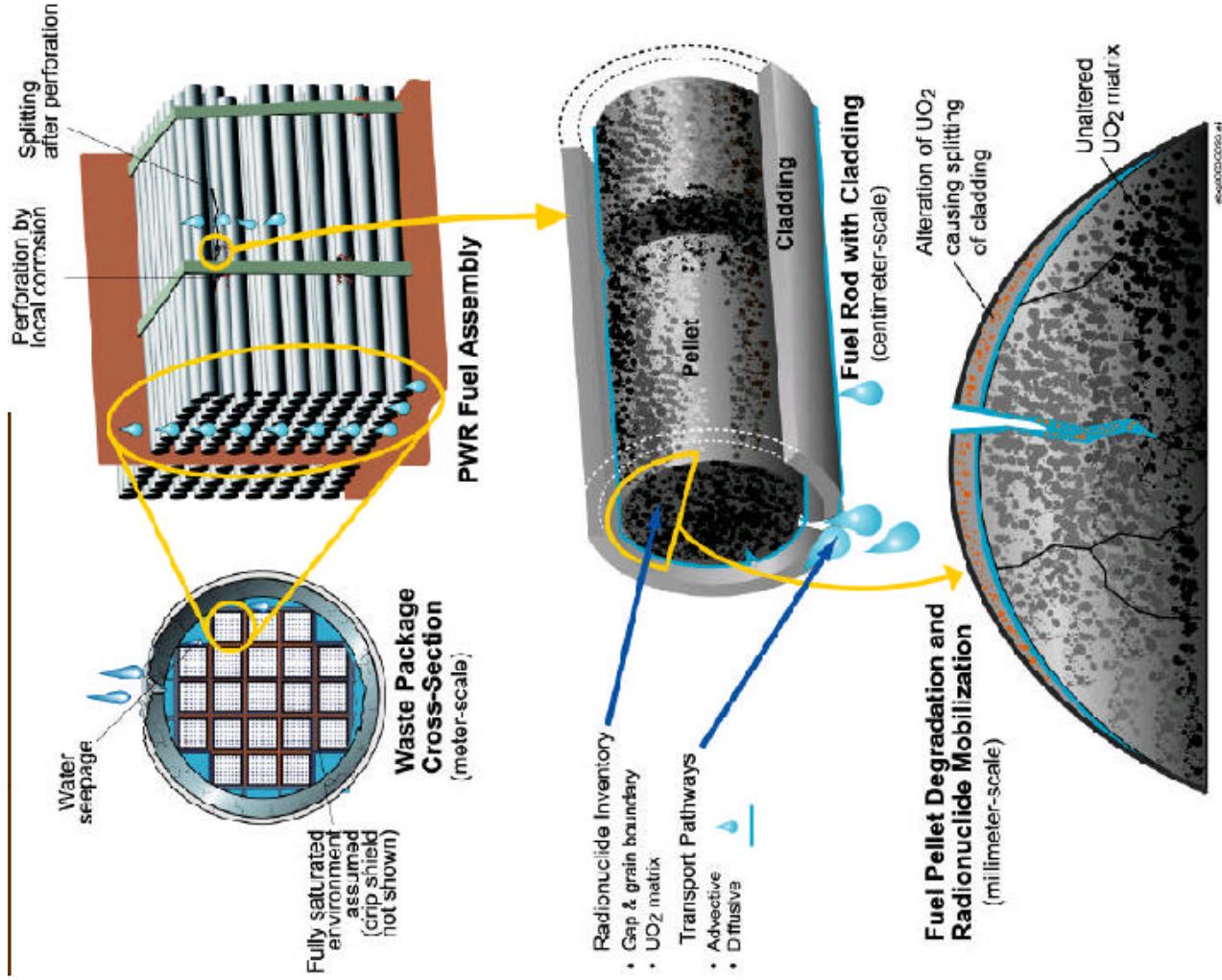
/RA/

George M. Hornberger  
Chairman

Attachment:

Report of the ACNW Working Group on Chemistry Issues and Related NRC Staff Capability for the Proposed High-Level Waste Repository at Yucca Mountain by Andrew C. Campbell, James H. Clarke, Paul G. Shewmon, Martin J. Steindler, and Raymond G. Wymer, dated August 2001

Figure 1. Waste form degradation model in TSPA-SR.



**Report of the ACNW Working Group on Chemistry Issues  
and Related NRC Staff Capability for the Proposed High-Level Waste  
Repository at Yucca Mountain**

**Andrew C. Campbell, James H. Clarke, Paul G. Shewmon,  
Martin J. Steindler, and Raymond G. Wymer**

**August 2001**

## **INTRODUCTION**

The Department of Energy's (DOE's) post-closure safety case for the proposed Yucca Mountain High-Level Waste (HLW) repository has shifted from an original focus on the geology of the site to emphasis on the engineered barrier system (EBS). This is reflected in the nominal scenario of the DOE's Total System Performance Assessment for the Site Recommendation (TSPA-SR)<sup>1</sup>. The scenario depends on a system of corrosion-resistant nickel-chromium-molybdenum alloy waste containers and titanium alloy drip shields to delay any potential releases from the EBS beyond the 10,000-year compliance time.<sup>i</sup> The performance of these barriers is based on extrapolating measured or estimated chemical properties and processes, derived from short-term laboratory testing, to very distant times. Chemical mechanisms and processes also play a dominant role in the performance of other engineered and natural barriers (e.g., cladding degradation, waste form dissolution, and mobilization and transport of radionuclides). It is important for the Nuclear Regulatory Commission (NRC) staff to understand the technical bases and performance impacts of chemical processes affecting the EBS and natural system to determine with reasonable expectation that the repository will meet the regulatory requirements. Given the time scales involved, the NRC is faced with the task of making judgments about phenomena and processes that cannot be tested directly and that are outside the framework of scientific endeavors where the outcome of an experiment and the test of a model or of a hypothesis can be directly observed. Thus, it is essential to assess the technical context for the TSPA modeling and the NRC's review approach, which are based on relevant information, data, and analyses. The lines of evidence that support the modeling results and regulatory conclusions need to be clear and traceable to valid scientific data.

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<sup>i</sup> The 10,000 year compliance time is specified in both NRC's proposed high level waste (HLW) regulation, 10 CFR Part 63, and the Environmental Protection Agency's final HLW standard, 40 CFR Part 197.

## **BACKGROUND**

The NRC is charged in the site recommendation process for the proposed Yucca Mountain HLW repository with providing comments on the sufficiency of the subsurface and waste form data for a license application. An important concern to the Commission is ensuring that the staff's review of approaches, methods, expertise, and capability are adequate for the sufficiency comments and for review of a license application. An Advisory Committee on Nuclear Waste (ACNW) Working Group evaluated the chemical issues associated with the engineered barriers and natural system and the adequacy of the related NRC issue resolution activities.<sup>ii</sup> The Working Group, chaired by Raymond G. Wymer, met on February 21 and 22, 2001, at NRC headquarters in Rockville, MD. The Working Group's activities are part of the ACNW's effort to evaluate the NRC's pre-licensing process for identification and resolution of issues related to licensing the proposed Yucca Mountain HLW repository. This report presents the findings and conclusions resulting from the Working Group meeting and from independent studies of NRC and DOE documents by the working group members.

## **CHEMISTRY REVIEW ISSUES**

We conducted an audit review of the following chemistry topics: waste package and drip shield corrosion, near-field chemical environment, in-package chemistry, radionuclide release, and radionuclide transport. Our review builds upon previous ACNW reviews of the near-field environment.<sup>2,3</sup> Many of the chemical processes, particularly those in the EBS, need to be

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<sup>ii</sup> Members of the Working Group were Dr. Andrew C. Campbell (ACNW Senior Staff Scientist), Dr. James H. Clarke (ACNW Consultant), Dr. Paul G. Shewmon (ACNW Consultant), Dr. Martin J. Steindler (ACNW Consultant), and Dr. Raymond G. Wymer (ACNW Member). Representatives from the NRC staff and the Center for Nuclear Waste Regulatory Analyses (CNWRA) attended and participated in the meeting, and a telephone link was open to the CNWRA. The meeting was announced in the *Federal Register* and open to the public. Representatives of DOE and the State of Nevada were also present.

considered in the context of coupled thermal-hydrologic-chemical processes that affect the performance of specific subsystems. We have sought to understand with a reasonable level of detail the underlying science, assumptions, models, and abstractions as provided by DOE, and the corresponding issues and review comments by the NRC staff for this limited set of topics. During the course of our review we considered issues in the context of relevant Key Technical Issues (KTIs), subissues in the NRC's HLW program<sup>4</sup>, and relevant system attributes and principal factors of the DOE Yucca Mountain Repository Safety Strategy (RSS).<sup>5</sup> The Working Group addressed the process-level modeling for specific issues, the abstractions of process models into TSPA-SR, and the technical bases for TSPA-SR. In this context, the Working Group evaluated the adequacy of the NRC review capabilities and methods to deal with the DOE safety case for the specific chemistry issues and discussed regulatory issues, such as the multiple-barriers requirement. The Working Group also commented on the degree to which "conservative" assumptions challenge the validity and the utility of the analyses in the near field and in the natural environment. A final point addressed was the flexibility of the NRC to deal with contingencies, such as changes in the repository design.

## **DISCUSSION**

### Corrosion of the Waste Packages and Drip Shields

The Alloy 22 waste package outer barrier and titanium drip shields are key engineered elements of the repository system. ACNW has previously reviewed the corrosion of Alloy 22.<sup>6</sup> The predicted long-term performance of the drip shield and waste package barrier system is based upon the slow general corrosion rates and resistance to localized corrosion and stress corrosion cracking (SCC) of the materials within the expected chemical and physical environment of the

repository. The Working Group looked at the technical bases for modeling the corrosion of these features, including general and localized corrosion of the Alloy 22 waste package, SCC of Alloy 22, and hydride cracking of the titanium alloy drip shield.

### **Corrosion of Alloy 22 Waste Package**

The calculated life of the waste package is determined in most scenarios by the general corrosion rates of Alloy 22 measured in experiments.<sup>7</sup> The general corrosion rate appears to be extremely slow under a wide range of temperature and chemical conditions.<sup>8</sup> General corrosion rates are derived from measurements in aqueous solutions (liquid water), whereas the majority of waste packages will be covered only with an adsorbed layer of water in equilibrium with humid air. The corrosion process is assumed to proceed at the rate of dissolution measured in the aqueous solutions. For example, in experiments at the Center for Nuclear Waste Regulatory Analyses (CNWRA), the materials are subjected to elevated temperatures, aggressive chemical constituents (e.g., high concentrations of chloride ions and low pH), applied voltages to facilitate corrosion, and stirring to avoid the buildup of corrosion products. Alloy 22 develops a protective Cr-rich oxide layer and the titanium alloy develops a protective oxide film that result in these materials corroding very slowly in experiments, making it difficult to discern variations in corrosion rates for different conditions. The use of aqueous corrosion data appears to conservatively bound waste package degradation. For example, humid air corrosion rates for 316L stainless steel are about 10 times slower than aqueous corrosion rates.<sup>9</sup> Measurement of general corrosion rates for Alloy 22 under humid air conditions has not been done by NRC, but DOE is conducting limited experiments.

Alloy 22 is much more resistant to crevice and pitting corrosion in chloride solutions than are stainless steels. The time frame for initiating such corrosion should be longer than that calculated for failure by general corrosion. The resistance to SCC around and in the Alloy 22 welds is less clear. Some tests indicate somewhat faster corrosion here than away from the welds, and some show little difference. The matter is still under study and NRC is concerned about the effects of welding and post-weld treatments on localized corrosion and SCC. There is evidence that there can be SCC failure in the weld zone after a surface layer of the Alloy 22 (with compressive stress) has been removed by general corrosion. Under extreme laboratory conditions, which are not representative of expected repository conditions, Alloy 22 can undergo SCC failures.<sup>iii</sup> DOE will be evaluating the SCC impacts of trace elements (potential catalysts) on Alloy 22 in accord with issue resolution agreements with the NRC.

According to DOE's sensitivity analyses for TSPA-SR, the SCC lid stress profiles and general corrosion of lid welds in waste packages are more important than any other variable until well beyond 100,000 years. The area of SCC cracks is extremely small relative to the area of general corrosion patches that are assumed to develop on the waste packages (by about 3-4 orders of magnitude). No water is allowed to flow through the SCC cracks and only very small releases of radionuclides occur via diffusion. Thus, SCC only affects the low dose region (well below 15 mrem) of the overall dose distribution and is a negligible contributor to overall risk. The

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<sup>iii</sup> Experimental data show very slow general corrosion under a wide range of temperature and chemical conditions. Consultants to the State of Nevada conducted experiments that showed Alloy 22 can be induced to undergo SCC for specific conditions of high acidity, temperature, and pressure, and in the presence of lead and mercury ions at about 0.5 wt. percent. The ACNW concluded that the range of experimental conditions used by consultants was not relevant to environmental conditions anticipated for the proposed Yucca Mountain high-level waste repository; however, there is a need to conduct confirmatory work to ensure the robustness of the Alloy 22 waste package (Ref. 6)

usefulness of these sensitivity measures to inform decision makers about the important contributors to risk from the repository is not apparent and needs to be clearly explained by DOE.

### **Corrosion of Titanium Alloy Drip Shield**

Hydride cracking of the titanium drip shield is excluded by DOE based on estimates that the hydrogen pickup would not be sufficient “. . . to attain critical hydrogen concentration in Ti drip shields before failure by general corrosion.”<sup>10,11</sup> The NRC staff did not accept this exclusion and requested that DOE provide the technical bases for it. The staff was concerned about the effect of fluoride-induced corrosion on hydrogen absorption. The Working Group discussed the critical hydrogen concentration and felt that the geometry of the shield and the mine drift would not allow enough mechanical work to be put into the titanium shield to break it (i.e., it is still robust enough to avoid fracture). An additional safety margin, for which no credit is taken, is provided by the 0.1% palladium that will be alloyed with the titanium. This will reduce the rate of hydrogen pickup by the titanium in the drip shield alloy to an even lower level than assumed in the “conservative” estimate.

### **Comments on the Adequacy of the NRC Staff Approach and Capability to Address**

#### **Resolution of Corrosion Chemistry Issues**

In reviewing the NRC and CNWRA evaluations and analyses for container and drip shield corrosion, the Working Group has concluded that no important corrosion areas or questions have been neglected. The Staff has considered a number of subissues in the Issue Resolution Status Reports (IRSRs) for the Container Life and Source Term (CLST) and Evolution of the Near-Field

Environment<sup>12</sup> (ENFE) KTIs related to drip shield and waste package corrosion. Within the issue resolution process, the NRC staff has raised a wide range of specific questions to satisfy itself that DOE's approaches, assumptions, and methods are appropriate and supported. These questions are the basis of the interactions with DOE in a series of technical exchanges focusing on each KTI and its subissues. These include questions about the following: general and local corrosion rates and SCC rates that are measured in laboratory experiments, the waste package chemical environment, the influence of trace metals on SCC, thermal treatments to relieve stress, passive oxide film stability, and the use of measured corrosion rates for projecting long-term performance. DOE has made agreements to address specific concerns and to provide more documentation to the NRC.<sup>13</sup> At this time all of the subissues related to waste package and drip shield performance in both CLST and ENFE are closed pending additional information, tests, and documentation from DOE. However, issues that relate to the use of this information in the TSPA-SR model (e.g., the abstraction of information into TSPA-SR) are currently open and will be addressed in the technical exchanges for Total System Performance Assessment and Integration (TSPAI) KTI.<sup>14</sup>

In general, we find the NRC's IRSRs to be well documented in terms of identifying important issues that are the focus of the DOE/NRC technical exchanges and agreements. In particular, we note that the CLST IRSR, Revision 3, provides a comprehensive treatment of the issues and carefully documents the agreements and the context of those agreements with respect to the issue areas most important to performance. The staff also appears to be well positioned in terms of approach and expertise to deal with evolutionary changes in design and their impacts on waste package corrosion.

## **Near-Field Chemical Environment**

The near-field (in-drift) chemical environment is an important element of the TSPA-SR analysis because it determines the chemistry of the water contacting the drip shield and waste package and influences the degradation rates.<sup>15</sup> The in-drift chemistry will be controlled by coupled processes involving thermal-hydrological-chemical (THC) interactions between the water entering the drifts and the emplaced materials.<sup>iv</sup> During the 10,000-year compliance time chemical reactions will be affected significantly by radioactive decay heat, especially in the first few thousand years. Beyond 10,000 years, the decay heat slowly decreases so that at 100,000 years the system is near ambient thermal conditions. At post-10,000-year time frames coupled hydrological-chemical (HC) processes are likely to dominate in-drift chemistry. The THC and HC interactions affecting waste package performance are followed by NRC in the ENFE KTI methods through subissue 2 -- the effects of coupled THC processes on the waste package chemical environment.

In the current DOE modeling approach, corrosion of the drip shield and waste package barriers is assumed to start when the relative humidity exceeds a critical value (between about 60 and 80%), at which point very thin layers of water will coat the surfaces.<sup>16</sup> Salts will precipitate on the drip shield where dripping water occurs and the evaporation rate exceeds the seepage rate.<sup>17</sup> Particulate material (rock dust and aerosol particles) will also coat the drip shield and the waste package. As cooling proceeds, brines will form by redissolution of the salts and reactions with particles.<sup>18</sup> As the temperature drops and the amount of sorbed water increases, these brines will

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<sup>iv</sup> The principal materials placed in the drifts are iron (steel drift supports, invert framework, and waste package inner barrier and internals) and uranium [spent nuclear fuel (SNF)], which constitute about 46% and 20%, respectively, of the material mass (excluding the drift rock and invert-fill material or backfill if it is used). The metals constituting the drip shield and waste package outer barrier make up another 32% of in-drift materials.

become increasingly diluted. The composition and temperature at which brines may form on the barrier surfaces may have an impact on local corrosion or SCC if sufficient stresses are present (e.g., from rock falls). In DOE's approach, coupled THC processes are treated separately and then recombined in the TSPA-SR analysis. The NRC staff has expressed concerns about this approach and DOE has agreed to provide further information.

Concern has been expressed about the use of J-13 well water as a proxy for the water entering the drift. Water dripping on the drip shield and waste package will contain dissolved chemicals derived from reactions with the rock as it passes through the mountain, as well as gases that are absorbed from air in the drift [including oxygen ( $O_2$ ) and carbon dioxide ( $CO_2$ )]. The presence of some of these chemicals, which are not necessarily represented by J-13 well water, could have deleterious effects on the drip shield or waste package. For example, interaction of percolating water with soluble minerals in the high-silica rhyolite may introduce certain trace elements such as fluoride ( $F^-$ ) and lead ( $Pb^{2+}$ ) ions to the water. These aqueous solutions will also react with particles deposited on the drip shield and waste package, further complicating the potential chemical mix reacting with the barriers. Chemicals in the water dripping on the drip shield and waste package may also include constituents derived from emplaced materials of construction such as steel corrosion products and leachate from cement grout in the drifts. Currently, the effects of emplaced materials on water chemistry are generally excluded from consideration because DOE believes that most water contacting the drip shields and waste packages will not have interacted significantly with drift components. In addition, design changes may significantly alter the types and amounts of chemicals in the water (e.g., from material used as backfill). The DOE testing program includes evaluation of evaporative effects on chemistry; CNWRA has done some confirmatory work. However, the influence of the chemistry of the incoming water to long

term barrier performance is not well constrained at this time. This is an area of concern to the staff in the ENFE subissue 2 and is the focus of a number of agreements with DOE.<sup>19</sup>

Comments on the Adequacy of NRC Staff Approach and Capability to Address Resolution of Near-Field Chemistry Environment Issues

The chemical environments on the drip shield and waste package are a concern to the NRC staff in subissue 2 of the ENFE KTI -- “the effects of coupled thermal-hydrologic-chemical processes on the waste package chemical environment.” To a limited extent, in-drift chemistry impacts are also addressed in the CLST KTI through subissue 1 -- “the effects of corrosion processes on the lifetime of the containers.” In the ENFE technical exchange NRC and DOE agreed that DOE will provide further documentation of the geochemical environment to help predict corrosion behavior of the drip shield and waste package. This includes DOE providing the technical basis for bounding the impacts of trace metals (e.g., lead) and fluoride and the impacts of engineered materials. Other agreements by DOE with the NRC include issues dealing with the following: the technical bases for certain features, events, and processes (FEPs), model and data uncertainties for the in-drift geochemical environment, local chemistry effects (including cement-water interactions, range of water composition, kinetic effects of chemical processes), and impacts of dust and salts on waste package degradation. DOE has also agreed to provide additional documentation and analyses to support modeling approaches.

In our review we found the staff positions to be comprehensive and thoroughly documented in the relevant IRSRs. At this juncture, we do not believe that the NRC and CNWRA have missed significant issues in this area. Questions about the integrated subissue on the chemistry and

quantity of water on the waste package as implemented in the TSPA-SR model are currently open and will be addressed in the technical exchange for the TSPA1 KTI.

### **In Package Chemistry**

Another area of concern to the Working Group is the in-package chemistry affecting radionuclide release.<sup>20</sup> The slow corrosion rates of the waste package and drip shields have lead DOE to conclude that the waste package system will meet the compliance requirements in 10,000 years. However, simplifying assumptions used for the source term appear to be extremely “conservative” and result in significant releases once the waste packages become degraded. The degree to which the source term assumptions bias overall performance toward a “conservative” result is not considered. Without the benefit of more realism about waste and material chemistry inside the waste packages, the waste package may be perceived as the only significant barrier protecting public health and safety.

The Working Group evaluated in-package chemical conditions and processes, focusing on water chemistry properties, such as pH, and chemical processes, like reduction and oxidation (redox) reactions, that affect the chemical speciation of radionuclides. The TSPA-SR model for the in-package chemistry is a simplified approach assuming a fully saturated environment at ambient temperatures (see Figure 1). This approach, which DOE maintains is “conservative,” is used because of uncertainties about the composition of water that could contact waste forms, and because of the complexities of predicting interactions between water penetrating the waste package and materials within it.<sup>21</sup> However, DOE has not evaluated the effects of reacting more limited amounts of water with a subset of materials within the waste package at elevated temperatures. Because of the complexities and difficulties in calculating meaningful pH values for

chemical processes, we conducted an evaluation of approaches and assumptions that affect the pH of water inside the waste package and the resulting impact on waste form degradation and radionuclide solubility.<sup>v</sup>

### **In-package pH Effects**

In DOE's TSPA-SR model, the calculated in-package pH<sup>vi</sup> plays a fundamental role in determining the release of radionuclides from the EBS. The pH is used in calculating radionuclide solubility (e.g., of Np), spent fuel degradation rates, which congruently releases non-solubility-limited radionuclides (e.g., of Tc and I), HLW glass dissolution rates, and the stability of colloidal species (e.g., of Pu). The calculated pH thus serves as a "master variable" controlling the release of the most significant radionuclides in TSPA-SR.

The pH is a complicated function of a number of competing reactions for the dissolution of materials in the waste package that either produce or consume hydrogen ions.<sup>vii</sup> The relationship

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<sup>v</sup> Note that DOE recently issued revised in-package chemistry and in-package chemistry abstraction model reports (references 21 and 22 respectively). The pH results in the revised reports are much more complicated than the previous calculations that were abstracted into TSPA-SR, Rev. 00. Our comments here focus on the more recent reports as a subsequent revision to TSPA-SR will likely show significant differences from the existing set of model results.

<sup>vi</sup> The pH of a solution is a measure of its acid content. The pH is defined as the negative log to the base 10 of the hydrogen ion (or proton) concentration (i.e.,  $pH = -\log_{10} H^+$ ). In pure water pH 7 is neutral, a pH less than 7 is acidic and a pH above 7 is basic. Because pH is a log scale, a one unit change in pH represents a 10 fold change in hydrogen ion concentration. Thus a solution with pH equal to 4 has a 1000 fold higher hydrogen ion concentration than a solution of pH 7. At pH values above 7 concentrations of the basic hydroxyl ion ( $OH^-$ ) become significant.

<sup>vii</sup> The pH is modeled as a function of the amount of water passing through the waste package and the reactions and reaction rates of the materials in the waste package (i.e., Fe and Al alloys, commercial spent nuclear fuel (CSNF), or HLW glass for co-disposal packages). The amounts of CNSF or HLW glass exposed to water were specified in the calculations. The dissolution reactions for Fe-containing materials (e.g., A-516 steel and 316 stainless steel) are acid-producing (i.e., the pH goes down) and the dissolution reactions for Al alloys, CSNF, and HLW glass are acid-consuming reactions (i.e., pH goes up). The balance achieved among these competing reactions determines the pH at any time as water reacts with the waste and package materials. (It is assumed that the waste packages contain 4,500 liters of water.) The change in pH values over time is a consequence of the progressive consumption of different materials in the waste package as water flows through the system. By about 300,000 years all the internal materials will have reacted. At this point the pH is determined by the chemistry of the incoming water, which is assumed to be about the same as J-13 well water.

among the material reaction rates and the overall pH is fundamentally nonlinear, and uncertainties, such as in those in the thermodynamic data bases, may strongly affect the calculated pH, and consequently the calculated chemical effects, in unanticipated ways. Moreover, coupled THC interactions between the water and the degrading materials at temperatures above ambient have been ignored. In addition, no comparisons with laboratory, field, or analog data are made to provide confidence that the approach used by DOE in fact bounds the pH values that could be expected to occur in the waste package. Because of the significant impacts on radionuclide release rates and transport properties, the coupled processes affecting pH need to be more carefully evaluated and the evidence bases supporting the approach need to be thoroughly described and documented.

Finally, abstraction of the pH into the TSPA-SR model relies on a complicated mix of response surfaces for shorter time frames and linear extrapolations for longer time frames.<sup>22</sup> The TSPA-SR abstraction for pH is focused on bounding the variability from the process model calculations. DOE assumes that the variability in the pH results obtained for a limited number of equilibrium chemistry model (EQ3/6) calculations represents the uncertainty of the in-package pH. While the response surface abstraction into TSPA-SR may bound this variability, it is not clear that the abstractions capture the important uncertainties in pH.

### **In-Package Reduction and Oxidation (Redox) Reactions**

DOE makes the assumption that an oxidizing environment exists within the waste packages at all times, and that this is a “conservative” assumption for times when most of the metals are still present and the ingress of water and gases (e.g., O<sub>2</sub> and CO<sub>2</sub>) would be limited by the slow development of general corrosion “patches” on the waste packages. In the current modeling

approach the chemical state of the water is set to be fully oxidizing by setting the partial pressure of dissolved oxygen in the water to a constant value that is representative of equilibrium with oxygen in air. Most of the important stable radionuclide solid phases under oxidizing conditions have significantly higher solubilities than the stable solid phases under reducing conditions.<sup>viii</sup> Aqueous corrosion of steel in the waste package could have a significant effect on radionuclide solubilities by producing a reducing environment when the ingress of oxygen is limited (e.g., from closure to well beyond the first 100,000 years of repository life). Redox gradients within the waste packages are acknowledged as possible by DOE. Taking credit for consumption of oxygen relative to the ingress of oxygen would require coupling chemical reaction kinetics with water and gas exchange through waste package breaches. At this time, no attempt has been made to evaluate or bound this scenario and so no conclusions can be made about the impact of this “conservatism” on radionuclide release rates.

The DOE assumption of an oxidizing environment, however, is not carried through the analysis in a consistent fashion. For example, it is assumed that the ionic species for I and Tc are in their most mobile forms -- iodide ( $I^-$ ) and pertechnetate ( $TcO_4^{2-}$ ). If oxidizing conditions prevail, however, the predominant iodine species may be iodate ( $IO_3^-$ ), which is capable of reacting with many materials in the EBS and natural environment. The assumption that iodide species prevails is inconsistent with the assumed chemical conditions.

It is not clear that the TSPA-SR models address chemical effects when some chemical species that are likely to be present in irradiated  $UO_2$  react with water at modest temperatures. Such

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<sup>viii</sup> For example, the stable form of Tc in a reducing environment is technetium oxide ( $TcO_2$ ) which is insoluble in water, whereas the stable form under oxidizing conditions is the pertechnetate ( $TcO_4^{2-}$ ) ion, which is highly soluble in water. In a reducing environment  $Np^{4+}$  will form, which will be less mobile than  $NpO_2^+$ , the stable form in an oxidizing environment.

speciation in the fuel could significantly affect the resulting reactions with modified groundwater. For example, if technetium oxide ( $TcO_2$ ) and/or sulfide ( $TcS_2$ ) are formed, the rate of release from dissolving fuel could be much slower than currently assumed. However, there seem to be no kinetic data to indicate how soon these would transform to the mobile  $TcO_4^-$ . Moreover, leaching experiments of spent fuel under oxidizing conditions provide some limits to the importance of this reaction. Furthermore, there is good evidence that part of the technetium in the spent fuel will be present as finely divided metal.

DOE continues to evaluate Np solubility limits, which are too uncertain for such an important isotope in TSPA-SR.<sup>23</sup> The selection of the most soluble form of crystalline neptunium oxide ( $Np_2O_5$ ) as the solubility limiting solid phase is "conservative."<sup>24</sup> Less soluble oxide phases of Np may be more appropriate for repository conditions. In addition, the formation of Np solid solutions and the incorporation of Np into secondary uranium oxide phases need to be further evaluated to establish more realistic limits for Np solubility.

### **Radionuclide Release Models**

The in-package chemistry model assumes all the void volume inside the waste package is filled with water (~4,500 liters) and that this water reacts with all available materials in the waste package (i.e., it behaves in a manner analogous to the way the same reactions would take place in a well-stirred bathtub). The approach taken in the model appears to be "conservative" because the radionuclide release model assumes much smaller amounts of water entering waste packages that intercept drips (between 15 and 0.15 liters per year). Moreover, most waste packages in the TSPA-SR model never have liquid water in them. DOE does claim that the approach is "conservative." However, DOE apparently has not quantitatively evaluated the impact of smaller

amounts of water with respect to the potential for producing more aggressive chemical conditions. Though less water dripping on the waste packages would imply less waste reacting with it, the possible tradeoffs in terms of enhanced radionuclide mobility (e.g., Np solubility, congruent release of Tc, or the stability of colloidal Pu) have not been addressed. On the other hand, mobilization and release of radionuclides from waste packages that are assumed never to have contained liquid water seem to be “conservative” to the point of absurdity. Given the complexity of the in-package chemistry and the importance in terms of radionuclide mobility, DOE needs to better document and support its approach.<sup>ix</sup>

To mobilize radionuclides from the EBS, the TSPA-SR model uses both diffusive releases (via moisture films in the EBS) and advective releases (via flowing water).<sup>25</sup> The TSPA-SR results show that diffusion dominates the overall radionuclide releases from the repository even in the absence of flowing water. The model of diffusive transport of radionuclides out of the waste packages and through the invert is decoupled from the amount of liquid water available in the waste package. The water chemistry model is developed for a fully saturated waste package and is used to provide concentrations for the diffusion model. A zero-concentration boundary condition is assumed outside the waste package and at the interface between the invert and the unsaturated zone rock. These assumptions imply significant water is present, whereas a diffusion only model implies extremely limited amounts of water are present. Thus, the TSPA-SR employs conflicting physical and chemical processes and conditions to model the source term release. While this appears to be a very “conservative” approach, it is not clearly discussed and leads to

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<sup>ix</sup>In general, using a “conservative” model or parameter value in a complex nonlinear model system does not necessarily produce “conservative” results. Such an approach requires that the outcome with a reasonable range of values be evaluated to bound the overall impacts. For example DOE uses a “conservative” (i.e., high) HLW glass dissolution rate that raises pH in the waste package as the glass dissolves. A more realistic lower dissolution rate produces more acidic conditions, which in turn would affect other aspects of the release model such as radionuclide solubilities, material degradation rates, colloid stability, and so forth. It is not clear that this type of coupled analysis has been done to assure that using “conservative” parameter values or model assumptions is truly “conservative” in terms of overall impacts on the TSPA-SR results.

confusion and uncertainty. What is more important, the use of this extreme "conservatism" in the model appears to be a significant contributor to relatively high dose values at long time frames and when waste packages are "neutralized"<sup>x</sup> in DOE's RSS analyses. This may lead to perceptions that early waste package failures would result in doses exceeding regulatory limits.

The problems of modeling these processes are surmountable and other nations' repository development programs have dealt with the complexities of modeling advection and diffusion into and out of breached waste packages. For example SKB -- the Swedish Nuclear Fuel and Waste Management Co. -- has conducted an extensive program of engineering development. This program includes experimental, modeling, and design development work focused on the waste package and internal processes that would attenuate radionuclide releases from their proposed repository system.<sup>26</sup>

**Comments on Adequacy of NRC Staff Approach and Capability to Address Resolution of In-Package Chemistry**

The chemical environment in the waste package is a concern to the NRC staff in subissue 3 of the ENFE KTI -- "the effects of coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release." The in-package chemistry impacts are also addressed in the CLST KTI through subissue 3 -- "the rate at which radionuclides in SNF are released from the EBS through the oxidation and dissolution of SNF" -- and subissue 4 --"the rate at which radionuclides in HLW glass are leached and released from the EBS." The staff has developed a significant number of specific questions for each of these subissue areas. At this time these

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<sup>x</sup> Neutralization of a barrier means that its effect is removed from the model.

issues are closed pending the additional work that DOE has agreed to address. Issues that relate to the use of this information in the TSPA-SR model (e.g., the abstraction of information into TSPA-SR) are currently open and will be addressed in the technical exchange for the TSPA-KTI.

In the CLST KTI subissue 3, the NRC staff reviewed various aspects of the issues discussed here, including issues such as in-package chemistry, radionuclide solubilities, spent fuel dissolution rates, and the assumed oxidizing condition for near-field chemistry. Issues involving the implementation of the in-package chemistry model continue to be evaluated by the NRC and a number of agreements with DOE on the issue resolution process are intended to address these concerns. For example, the NRC staff has pursued the issue of Np solubility limits and the current range of values used by DOE appears to be "conservative." NRC has also concluded that, although a reducing state may occur soon after the waste package is breached, ultimately the waste package environment will be oxidizing. The staff concurs with DOE that the assumption of an oxidizing environment is "conservative." The staff believes that the diffusion release model is very "conservative," but at this time has not identified significant problems with the degree of "conservatism." However, it needs to be noted that the DOE's basis for meeting the dose standard within the regulatory compliance period is based on the performance of the waste package. In the absence of long waste package performance times, the "conservative" approaches to the release and transport of radionuclides would lead to doses exceeding the regulatory limits at much earlier time frames. It is not clear that the NRC staff has fully addressed all of the in-package issues identified above.

## **Radionuclide Transport in Near- and Far-Field Environment**

The TSPA-SR employs a sophisticated mathematical model of flow and radionuclide transport to predict doses as a function of time at a compliance point in the saturated zone that is 20 kilometers down gradient from the proposed repository. This model necessarily requires input assumptions concerning a wide range of chemical information. Given the long time frames involved, a good understanding of those assumptions to which the predictions are most sensitive is essential.

Radionuclides can be transported as “true colloids” or “pseudo colloids.” In the TSPA model the latter include both irreversibly bound colloids (IRBC) and reversibly bound colloids (RBC).<sup>xi</sup> In IRBCs, radionuclides are irreversibly associated with colloidal material (e.g., silicate particles from HLW glass degradation). In RBCs, radionuclides are reversibly attached to colloidal material (e.g., natural clays in ground water and iron oxyhydroxide corrosion products). In TSPA, the IRBCs are transported as anionic solutes that can be retarded through filtration processes but are not permitted to diffuse into and sorb onto the rock matrix in units with significant fracture flow. In RBC, the radionuclides partition between colloids (natural or waste form) and the aqueous dissolved phase. The partitioning is defined by a colloid partition coefficient,  $K_c$ . Unattached radionuclides are transported as dissolved solutes and can be attenuated through diffusion into and sorption onto the rock matrix in fracture-flow-controlled units. The NRC staff has evaluated many issues and concerns with regard to the approach used by DOE in modeling colloid formation

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<sup>xi</sup> True colloids are those formed by the radionuclide itself (e.g., PuO<sub>2</sub> colloids). The TSPA model considers pseudocolloids, where radionuclides become associated with other colloidal particles such as clays and iron oxyhydroxides.

and transport. These concerns and issues are described and discussed in the ENFE IRSR under subissue 3 and are addressed by agreements with DOE from the ENFE technical exchange<sup>19</sup>.

Radionuclide transport is driven by water flow through the subsurface, with incorporation of the pertinent retardation processes for each model unit. Some important subsurface processes include sorption of dissolved solutes on alluvial and rock matrix surfaces and filtration of IRBC. Laboratory and field tests have been used both to demonstrate the efficacy of attenuation processes, such as sorption, and to develop the appropriate parameters needed for the model. Laboratory studies with both site-specific materials and field testing (e.g., through the injection of tracers as was done with the C-wells complex) have been done. There are, however, issues that need to be resolved about how well the tracers used in these studies represent radionuclide behavior (e.g., the specific size ranges of polymer microspheres used to represent colloid behavior). Continuing investigations are being performed to address existing NRC concerns and to provide data to determine  $K_d$ s and other transport parameters for the alluvium.

Chemistry is included only indirectly in transport modeling through experimentally determined parameters such as  $K_d$ , a measure of the distribution of chemical species between liquid and solid phases. Chemistry considerations should also be included to support the appropriateness and degree of “conservatism” associated with the modeling assumptions that are made. For example, iodine may be expected to be present in a more reactive form as the iodate species ( $\text{IO}_3^-$ ); however, it is being modeled as if it were always present as the non-sorbing iodide ( $\text{I}^-$ ) anion, thus potentially overestimating its mobility.

### **Comments on NRC Staff Approach to Issue Resolution**

The NRC staff has documented its approach to issue resolution in the Radionuclide Transport IRSR<sup>27</sup> and has established agreements with DOE in the radionuclide transport technical exchange for three subissue areas: radionuclide transport through porous rock, alluvium, and fractured rock.<sup>28</sup> While the issue resolution process has resolved many technical issues, a few “ongoing issues” appear to be worthy of mention. The length of the flow path in alluvium is important from the standpoint of radionuclide attenuation (sorption). Additional data from the ongoing Nye County work should help here. While the efficacies of the major attenuation processes have been demonstrated (e.g., matrix diffusion in fractured rock), there are concerns about limitations to the attenuation capacity of the system.

Much of the issue resolution process is focused on process model issues, resulting in a concern on our part for potential problems at critical interfaces, e.g., waste package/engineered barrier/unsaturated zone and unsaturated zone/saturated zone. Information which is produced by DOE to resolve an issue may or may not be accompanied by additional information that is necessary to enable deeper understanding of the importance of the issue to the total system performance. Thus there is merit to more comprehensive documents which include the interfaces. To a large degree this is the role of the TSPA-KTI and the integrated subissues in the NRC's framework and the TSPA-SR in the DOE approach.

## **CONCLUSIONS**

1. NRC's Issue Resolution Status Reports in the chemistry area are well documented. No important issues or questions with regards to corrosion appear to have been overlooked by the staff.
2. The NRC staff appears to understand what would be required to deal with possible future repository design changes, but resources may be inadequate to deal with some potential scenarios.
3. Alloy 22 and 316L stainless steel will likely corrode more slowly than predicted by DOE unless an unexpected catalytic effect by trace elements occurs. Studies are underway to investigate this possibility.
4. The effects of SCC and localized corrosion on the degradation of welds are uncertain and staff has required more information on the impacts of weld treatments from DOE.
5. The usefulness of DOE's sensitivity and importance analyses in TSPA-SR to identify and understand the most significant contributors to overall risk is questionable.
6. The central role of the waste packages and the drip shields in meeting the dose limits in the 10,000 year compliance period could lead to the erroneous conclusion that their slow corrosion is all that is protecting the public. Other important protective chemical processes may significantly limit the release and transport of radionuclides, such as secondary-phase formation and retardation.

7. The importance of THC processes during the 10,000-year compliance period is uncertain, but chemical reactions, including corrosion, could be significantly accelerated if liquid water heated by radioactive decay contacts the barriers or waste form materials.
8. In TSPA-SR, J-13 well water has been used as a surrogate for water contacting waste packages, drip shields, and waste forms. However, it probably is unrepresentative of water in the near field and does not provide bounding chemical characteristics. The importance to performance of differing water chemistries and the impacts of coupled interactions of water with emplaced materials are not well understood at this time. DOE has agreed to conduct experiments with a variety of water chemistries to evaluate impacts on material degradation.
9. The in-package chemistry model assumes a fully saturated waste package and does not address potentially important chemistry issues such as radionuclide speciation in the SNF, solubility controlling reactions, or redox reactions. DOE needs to more clearly link the simplifying assumptions used in the model to experimental and analog evidence and also to address varying moisture conditions.
10. The abstractions of process model chemical phenomena into TSPA-SR does not appear to capture important uncertainties in chemistry (e.g., pH), which control the release and transport of the important radionuclides.
11. Diffusive releases of radionuclides in the TSPA-SR model are significant. DOE assumes that diffusion can occur along thin moisture films. However, the release model actually

employed in TSPA-SR is not adequately described. Apparently data and assumptions based on fully saturated conditions are used in situations where no liquid water can exist in the waste package. According to DOE, this "non-mechanistic release scenario" in TSPA-SR will be changed to a more realistic approach.

12. Potentially important colloid chemistry complexities may not be addressed in transport experiments, which are performed using organic polymeric microspheres to represent the behavior of colloidal radionuclides.
13. Chemical processes governing radionuclide transport are represented in a gross sense in the TSPA-SR model by the use of experimentally measured parameters ( $K_d$ s) that give little insight into rate controlling chemical mechanisms, thus limiting testing of model predictive capability and understanding.
14. A general area of concern that emerged in our review is that by striving always for conservative assumptions in a chain of events one can obtain unreasonable answers. The cumulative effect of overly conservative assumptions may obscure the most important components of the repository system in terms of performance and risk. Moreover, overly conservative assumptions in a specific process model abstraction may not be conservative with respect to the overall risk.

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