

REVIEW BY THE U.S. NUCLEAR REGULATORY COMMISSION
OFFICE OF NUCLEAR MATERIAL SAFETY AND SAFEGUARDS
OF THE U.S. DEPARTMENT OF ENERGY RESPONSES RELATED TO THE
PROPOSED GEOLOGIC REPOSITORY AT YUCCA MOUNTAIN, NEVADA:
KEY TECHNICAL ISSUE AGREEMENTS CLST.3.02, ENFE.3.04, AND CLST.3.05

1.0 INTRODUCTION

The U.S. Nuclear Regulatory Commission (NRC) issue resolution goal during this interim pre-licensing period is to ensure that U.S. Department of Energy (DOE) has assembled sufficient information on a given issue for NRC to accept a potential license application for review. Resolution by the NRC staff during pre-licensing does not prevent anyone from raising any issue for NRC consideration during the licensing proceedings. Also, and just as important, resolution of an issue by NRC during pre-licensing does not prejudice the NRC staff evaluation of the issue during the licensing review. Issues are resolved by the NRC staff during pre-licensing when the staff have no further questions or comments about how DOE is addressing an issue. Pertinent new information could raise new questions or comments about a previously resolved issue. The NRC licensing decision will be based on information provided as part of a potential license application.

By letter dated July 30, 2004, DOE submitted Technical Basis Document No. 7 (TBD 7) (Bechtel SAIC Company, LLC, 2004), a report summarizing the technical basis and conceptual understanding for the in-package chemical environment and how this environment would affect waste form degradation and the mobility of released radionuclides inside a breached waste package. The technical basis document contained appendices with DOE responses to numerous Key Technical Issue agreement items about how the evolution of the chemical environment inside the waste package was represented by DOE model abstractions. The DOE responses to three of these agreement items are reviewed here. The agreement items addressed in this NRC review are Container Life and Source Term (CLST).3.02 (Schlueter, 2000) and CLST.3.02 Additional Information Need (AIN)-1 (Schlueter, 2002), Evolution of the Near-Field Environment (ENFE).3.04 (Reamer, 2001), and CLST.3.05 (Schlueter, 2000). CLST.3.02 AIN-1 was from a review of an earlier DOE agreement response (Bechtel SAIC Company, LLC, 2001).

2.0 AGREEMENTS

CLST.3.02

“In the revision to the ‘Summary of In-Package Chemistry for Waste Forms,’ Analysis Model Report (AMR), address specific NRC questions regarding radiolysis, incoming water, localized corrosion, corrosion products, transient effects, and a sensitivity study on differing dissolution rates of components. DOE stated that these specific questions are currently being addressed in the revision of the ‘Summary of In-Package Chemistry for Waste Forms’ AMR, ANL-EBS-MD-000050 and related AMRs and calculations. To be available in January 2001.”

Enclosure

CLST.3.02 AIN-1

1. Provide the technical basis for the corrosion rates and the likely corrosion mechanisms of internal waste package components.
2. Provide the technical justification and appropriate ranges for the model abstraction parameters for each internal component of the waste package.
3. Provide the results of sensitivity analyses using current values of the parameters included in the model abstractions.
4. Provide the technical basis for the in-package water chemistry, taking into account the formation of locally aggressive environments in crevices and tight spaces inside the waste package that can enhance the degradation of waste forms and the solubility of radionuclides.
5. Provide documentation which validates the use of EQ3/6 to predict the formation of secondary phases resulting from high-level waste glass dissolution.
6. Provide the technical bases for ignoring the role of halides in the dissolution process.”

ENFE.3.04

“Complete validation of in-package chemistry models. Agreement #5 for CLST Subissue 3 addresses testing plans. Model validation based on this testing and further analysis will be documented in an update to the Summary of In-Package Chemistry for Waste Forms AMR (ANL-EBS-MD-000050), expected to be available in fiscal year (FY) 2002.”

CLST.3.05

“Provide the plan for experiments demonstrating in-package chemistry, and take into account subsequent NRC comments, if any. DOE has stated that the current planning provides the analysis of additional in-package chemistry model support. This analysis will determine which parts of the model are amenable to additional support by testing, and which parts are more amenable to sensitivity analysis, or use of analogs. Based on these results, longer range testing will be considered. If the testing is determined to be appropriate, test plans will be written in FY 2001 and made available to the NRC.”

3.0 RELEVANCE TO OVERALL PERFORMANCE

Agreements CLST.3.02, ENFE.3.04, and CLST.3.05 are all related to demonstrating the adequacy of the in-package chemistry model, including the technical basis and justification for the corrosion rates of waste package components and the expected ranges of model input parameters that are used to simulate the changes in solid and aqueous chemistry that would occur over time inside a breached waste package. The waste package is the primary engineered barrier controlling the release of radionuclides from the spent nuclear fuel and high-level waste glass waste forms. The chemical environment inside the waste package is important because the rates and processes of waste form degradation depend in large part on

the in-package water chemistry, which changes composition over time in response to reactions with waste package components, including reactions with the waste form itself. Moreover, degradation of the waste form leads to the release of radionuclides from the waste form, at which point the in-package water chemistry is important because it can either limit or promote the mobility of certain radionuclides after their release from the waste form. For example, a high-pH environment would increase the solubility of neptunium, a significant radionuclide due to its large inventory and long half-life, and it would promote the stability of radionuclide-bearing colloids in the waste package. Dissolved radionuclides and those mobilized as colloids provide the source term for the transport of radionuclides through the engineered barrier system and into the unsaturated zone. Therefore, an effective representation of in-package chemistry is important in characterizing overall repository performance.

NRC staff have performed a risk insights analysis that indicates radionuclide release and solubility limits have a medium significance with regard to waste isolation (NRC, 2004). However, within the context of developing an effective representation of radionuclide release and mobility inside the waste package, the contribution of Agreements CLST.3.02, ENFE.3.04 and CLST.3.05 to the in-package chemistry abstraction are considered to be of high significance (Travers, 2003).

4.0 RESULTS OF THE NRC REVIEW

4.1 Agreement CLST.3.02 and CLST.3.02 AIN-1

The DOE response to agreement items CLST.3.02 and CLST.3.02 AIN-1 was provided in Appendix A of Bechtel SAIC Company, LLC (2004). Agreement CLST.3.02 was reached to provide information about aspects of the conceptualization and construction of the DOE in-package chemistry model, such as the choice of mechanisms or rates for corrosion or dissolution of waste package components, the choice of materials and conditions used to define the waste package environment, and the sensitivity of the model to uncertainties in parameter values. The NRC review of this agreement item is presented in the following subsections consistent with the specific additional information items that were requested in CLST.3.02 AIN-1.

4.1.1 Technical Basis for Corrosion Rates and Corrosion Mechanisms

The focus of Item 1 of CLST.3.02 AIN-1 was to ensure that the DOE has considered an appropriate range of environmental conditions with respect to the selection of corrosion rates for materials inside the waste package. The corrosion rates used are presented in Table 3-4 of Bechtel SAIC Company, LLC (2004), where they are compared with measured corrosion rates under comparable environmental conditions. NRC has compared the tabulated corrosion rates with published values of corrosion rates for carbon steels (Matsushima, 2000; Heidersbach et al., 1987) aluminum alloys (Hollingsworth and Hunsicker, 1987; Ghali, 2000), and stainless steels (Malik et al. 1995, Jallerat et al., 1984). The information presented by DOE has satisfied the intent of this item.

4.1.2 Technical Justification and Appropriate Ranges for the Model Abstraction Parameters for Each Internal Component of the Waste Package

The focus of Item 2 of CLST.3.02 AIN-1 was to ensure that the DOE has provided the technical justification and the appropriate range for the model abstraction for each internal component of the waste package. The DOE response is sufficient to satisfy the request for information, especially with respect to the system risk perspective. However, NRC has several specific comments, as listed below, about how codisposal waste packages were included in the model. An effective representation of glass degradation is important because the glass waste form would make the most significant contribution to high-pH conditions in a breached waste package. If degradation of vitrified high-level waste is part of the in-package chemistry abstraction in a potential license application, NRC will carefully consider how DOE has represented this process in the model.

- Among the conditions and properties that were established as starting assumptions for the in-package chemistry model, DOE chose two general types of waste packages – commercial spent nuclear fuel waste packages and codisposal waste packages containing DOE spent nuclear fuel and high-level vitrified wastes – to be representative of all waste packages. The selected codisposal package type is not representative of the majority of the codisposal packages in a potential repository. In the in-package chemistry model, all codisposal waste packages are represented by a waste package with 2 DOE spent nuclear fuel canisters and 2 high-level waste glass canisters. This type of codisposal waste package represents only about 1 percent of all waste packages and less than 1 percent of the metric tons of heavy metal in a potential repository (Bechtel SAIC Company, LLC, 2004, Table 2-1). In contrast, there are two other design types of codisposal packages that together account for about 29 percent of all waste packages in a repository. These latter two design types contain proportionately greater amounts of high-level waste glass (5 glass canisters and 1 DOE spent nuclear fuel canister per package) than the codisposal package type selected for the in-package chemistry abstraction. Given that DOE has identified the dissolution of glass as a process that is potentially capable of generating a high-pH environment inside the waste packages, a waste package with 5 high-level waste glass canisters would be likely to release a larger quantity of alkalis to solution than a waste package with only 2 high-level waste glass canisters. It is not clear why DOE has excluded these other types of codisposal waste packages from the model, given that they are abundant and have high proportions of high-level waste glass to spent nuclear fuel.
- Surface area is an important property used in modeling degradation of the glass waste form, but it is not clear how DOE has selected the range of values for this parameter. The DOE range was based in part on data from fracture studies of French high-level waste glass, but DOE did not account for differences in processing technologies that would result in lower surface areas for the French glass waste forms. Among details of the glass degradation model (CRWMS M&O, 2000a; Bechtel SAIC Company, LLC, 2003), NRC notes that the validation of the range of values used by DOE was based in part on comparison with published European studies of full-scale samples of non-radioactive R7T7 glass (Sene et al. 1999). However, the range of exposed glass surface area values proposed by DOE is not supported by the processing method for high-level waste glass used in the United States. Unlike the French high-level waste glass waste form, which is annealed to provide mechanical strength after the glass is

poured into a canister, the high-level waste glass produced in the United States is not annealed and so would shatter more easily. A typical high-level waste glass canister has a void volume of 10 to 15 percent. Given the fragility of the unannealed glass, vibrations during handling and transportation could easily result in enough fracturing to fill the void volume in the canister and significantly increase the exposed glass surface area. DOE has proposed a value of 4 times the geometric surface area as a lower limit and also as a most probable value for the estimated exposed surface area of the glass. The technical justification of this estimate, which does not account for a lack of annealing and additional fracturing associated with transportation and handling, needs to be provided.

- The glass degradation model also includes an estimate of 17 percent porosity for the glass rind. DOE has not provided a justification for selection of this porosity value. Compared to the dissolution rate that was provided in CRWMS M&O (2000a), the combination of 17 percent porosity and a multiplication factor of 4 for the most probable surface area, instead of a factor of 20 as was used in CRWMS M&O (2000a), has reduced the dissolution rate of high-level waste glass per day by a factor of approximately 25 without explanation.
- For codisposal waste packages, the conceptual model assumes that the defense spent nuclear fuel dissolves immediately after a waste package breach. Such conservatism could result in masking the contributions of other waste package internals. In particular, such rapid degradation of the fuel immediately makes available a large quantity of schoepite, which moderates elevated pH values that otherwise would result from the degradation of the high-level glass waste form. The effect of this assumption should be examined by a sensitivity analysis that demonstrates how in-package solution chemistry in a codisposal package would vary in response to slower corrosion rates for defense spent nuclear fuel.

4.1.3 Sensitivity Analyses Using Current Values of the Parameters Included in the Model Abstractions

The focus of Item 3 of CLST.3.02 AIN-1 was to ensure that the DOE has provided results of the sensitivity analysis using values of the parameters as included in the model abstraction. In the response to this item, the DOE presented results of in-package sensitivity analyses performed to determine the uncertainty range for ionic strength and for expanded ranges of temperature and CO₂ partial pressures. Other analyses demonstrated how differences in the waste package configuration and in seepage composition affected the model output. To the extent that the sensitivity analyses have been based on current values of parameters in the model, DOE has satisfied the intent of this item

4.1.4 Technical Basis for the In-Package Water Chemistry, Taking into Account the Formation of Locally Aggressive Environments in Crevices and Tight Spaces

The in-package chemistry model results support the DOE conclusion that extreme low values of pH, as results of localized corrosion, will not be an important factor inside a breached waste package. A review of the information provided for the response to a related Key Technical

Issue agreement, CLST.3.06, further supports the DOE conclusion that low pH and high Fe³⁺ levels are not likely to develop inside the waste package (Bechtel SAIC Company, LLC, 2004, Appendix D). The DOE responses satisfy the intent of Item 4 of the CLST 3.04 AIN-1.

4.1.5 Validate the Use of EQ3/6 to Predict the Formation of Secondary Phases Resulting from High-level Waste Glass Dissolution

In the response to this item, DOE states that altered volcanic glass is a suitable natural analogue for a degraded high-level glass waste form. Validation of the use of EQ3/6 is provided by a published study that compared secondary natural alteration products, identified in basaltic glass, with secondary phases predicted by EQ3/6 modeling (Grambow et al. 1986). The documentation that has been supplied is sufficient to satisfy the intent of the agreement.

4.1.6 Technical Bases for Ignoring the Role of Halides in the Dissolution Process

The DOE has provided sufficient information to satisfy the intent of the agreement item. The role of metal halides has been evaluated and excluded from further consideration on the basis that the necessary low-pH conditions are unlikely to exist.

4.1.7 Status of CLST.3.02 and CLST.3.02 AIN-1

The information provided by DOE has satisfied the intent of Key Technical Issue agreement CLST.3.02 and CLST.3.02 AIN-1. In a potential future license application, NRC staff would look carefully at how the DOE has represented codisposal waste packages in the in-package chemistry abstraction, particularly regarding the contribution of glass degradation to the pH of the effluent inside the waste package.

4.2 Agreement ENFE.3.04

The DOE response to agreement item ENFE.3.04 was provided in Appendix C of Bechtel SAIC Company, LLC (2004). The intent of agreement ENFE.3.04 was for NRC to better understand how DOE intends to complete the validation of the in-package chemistry models. In developing the in-package chemistry abstraction, DOE used two modeling approaches to simulate chemical changes over time inside a waste package—a physical chemistry model and a surface complexation model. In the response to ENFE.3.04, DOE addressed the validation of both modeling approaches. According to the response, more details about validation of the models are provided in an unreleased revision of a report on the in-package chemistry abstraction, but the report was not available for evaluation at the time of this review.

The main modeling approach in the in-package chemistry abstraction was a physical chemistry model, applied in two stages. First, the model was used to simulate kinetic reactions between incoming water and individual waste package components, calculating the contribution of each to waste package chemistry. Second, the model was applied to a more complex environment, in which it simulated kinetic reactions between incoming water and a combination of waste package components, including the waste forms themselves, to determine how coupled reactions affected the evolution of the in-package chemistry. The physical chemistry model was implemented with the reaction-path software code EQ3/6 (Wolery and Daveler 1992) for a range of specified conditions inside a waste package. The input parameters that were varied included system properties (water distribution and mode of occurrence inside the waste

package), environmental properties (temperature, carbon dioxide gas partial pressure, water flux rates), and material properties (composition of waste package components and of the incoming water, degradation and corrosion rates, masses and surface areas of reactants). Among the values calculated by EQ3/6 were the effluent pH, ionic strength, redox conditions, and the concentration of dissolved species such as carbonate and fluoride. These properties are important because they provide direct or indirect input to the submodels that simulate waste form degradation, radionuclide solubility limits, and colloid concentrations.

The second modeling approach used was a surface complexation model, described as an analytic derivation of pH, that was applied separately from the physical chemistry model. The physical chemistry model predicted that under certain conditions in the presence of sulfur, the corrosion of steel could result in very low pH values less than 3 inside the waste package, but such low values are not indicated by experiments. For example, a study by Zarrabi, et al. (2003) in which a miniature steel waste package with carbon steel internals was subjected to corrosion in a J-13 water found that the resulting pH of the in-package solution remained steady at a value of about 8. In developing the in-package chemistry abstraction, DOE addressed this difference in pH values by also modeling surface complexation reactions (protonation) involving steel corrosion products, which would tend to buffer any acidic pH conditions to a more moderate pH range of about 7 to 8. Due to a limitation of the EQ3/6 software, surface complexation processes could not be coupled to the other reactions in the physical chemistry model. Instead, the surface complexation model was applied separately to predict the in-package pH during the first 600 years after waste package breach, when corrosion of Carbon Steel Type 516A would largely dominate in-package chemistry.

4.2.1 Validation of In-package Chemistry Models

For the physical chemistry model, the DOE validation was based on a broad comparison of the modeled predictions with analogous systems for which similar conditions and processes have been observed. This validation procedure was used to confirm that the model predicted the formation of a realistic set of secondary solids by corrosion of steel and other metals in the waste package, and it was used in a more general sense to assess that the range of pH values predicted by the model compared appropriately with natural waters and a few laboratory experiments.

For the surface complexation model, the DOE response indicated the model was validated by an independent expert technical review (unidentified) which concluded that the approach was reasonable. The aforementioned work by Zarrabi et al. (2003) appears to support the approach taken with the surface complexation model to represent in-package pH variations associated with the corrosion of Carbon Steel Type 516A.

The information provided by DOE about the methods used to validate the in-package chemistry models is sufficient to satisfy the intent of agreement ENFE.3.04. However, staff have several comments that DOE should consider for consistency. These comments are:

- The model validation examples that were presented in the response to ENFE.3.04 dealt only with the calculated ranges of pH values. A thorough model validation extends to other important outputs of the EQ3/6 calculations, such as (i) the calculated ranges of ionic strength and of carbonate and fluoride concentrations, and (ii) the role of schoepite dissolution in buffering pH values.

- The pH values calculated by EQ3/6 in association with degradation of the glass waste form seem low compared with higher values (pH >10) that are reported in glass dissolution experiments (e.g., Bates, et al., 1994; Jantzen, et al., 1993). The DOE response to ENFE.3.04 does not indicate whether or not the EQ3/6 pH calculations were validated by comparison with any data from glass dissolution experiments.

4.2.2 Status of ENFE 3.04

The information provided by DOE has satisfied the intent of Key Technical issue agreement ENFE 3.04. In a potential future license application, NRC staff would look carefully at how the DOE has represented the details of the model validation, in the context of the comments above and information that will be provided in the revised in-package chemistry abstraction after it becomes available for review.

4.3 Agreement CLST.3.05

The DOE response to agreement item CLST.3.05 was provided in Appendix C of Bechtel SAIC Company, LLC (2004). The intent of CLST.3.05 was to ensure that the DOE has adequately considered the use of experiments to support the evolution of in-package chemistry. In response to CLST.3.05, DOE stated that future testing was considered unnecessary based on the results of a large set of additional sensitivity analyses that were completed subsequent to the establishment of CLST.3.05. According to the DOE response, the sensitivity analyses encompassed a more diverse set of waste package conditions and provided a more complete picture of in-package chemistry than could be investigated practically by experiments.

4.3.1 Status of CLST.3.05

DOE considered performing long-range experiments but chose not to conduct any, so no plans for experiments were produced. On that basis, and notwithstanding new information that could raise new questions or comments, NRC staff finds that agreement item CLST.3.05 is closed.

5.0 SUMMARY

NRC staff reviewed the DOE responses to Key Technical Issue agreement items CLST.3.02, CLST.3.05 and ENFE.3.04 to determine whether any important aspects of the agreement items were excluded from the response.

On the basis of the information provided in the DOE response, NRC's pre-licensing activities associated with agreement items CLST.3.02, CLST.3.05 and ENFE.3.04 can be considered closed. Sufficient information has been provided to document the methodology used to develop the in-package chemistry abstraction. However, DOE should consider the justification for several parts of the model, particularly with regard to (i) representation of processes involving glass waste forms (CLST.3.02 and CLST.3.05) and (ii) detailed validation of the physical chemistry model and the surface complexation model (ENFE.3.04).

6.0 STATUS OF THE AGREEMENTS

On the basis of this review, NRC staff finds that the information provided by DOE for Agreements CLST.3.02, CLST.3.05 and ENFE.304 is sufficient to close the items.

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