

REVIEW BY THE U.S. NUCLEAR REGULATORY COMMISSION
OFFICE OF NUCLEAR MATERIAL SAFETY AND SAFEGUARDS
OF THE U.S. DEPARTMENT OF ENERGY AGREEMENT RESPONSES RELATED TO
THE POTENTIAL GEOLOGIC REPOSITORY AT YUCCA MOUNTAIN, NEVADA:
KEY TECHNICAL ISSUE AGREEMENTS CLST.3.03, CLST.3.03 AIN-1, CLST.3.04,
CLST.3.04 AIN-1, CLST.3.06, CLST.3.06 AIN-1, CLST.3.07, CLST.3.08, CLST.3.08 AIN-1,
CLST.3.09, CLST.3.09 AIN-1, ENFE.3.03, TSPAI.3.08, TSPAI.3.14, GEN.1.01 COMMENT 116,
GEN.1.01 COMMENT 124, DOE INITIAL RESPONSE TO GEN.1.01 COMMENT 124, and
GEN.1.01 COMMENT 126

1.0 INTRODUCTION

The U.S. Nuclear Regulatory Commission (NRC) issue resolution goal during this interim preclicensing period is to ensure the 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 NRC during preclicensing does not prevent anyone from raising any issue for the NRC staff consideration during the licensing proceedings. It is equally important to note that resolution of an issue by NRC during the preclicensing period does not prejudice the NRC staff evaluation of the issue during the licensing review. Issues are resolved by the NRC staff during preclicensing 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 a report titled Technical Basis Document No. 7: In-Package Environment and Waste Form Degradation and Solubility (Bechtel SAIC Company, LLC, 2004a) to satisfy the informational needs of numerous (KTI) agreement items reached with NRC pertaining to waste form degradation and radionuclide release at the potential repository at Yucca Mountain, Nevada. The information was requested by NRC during technical exchanges in September 2000, January 2001, August 2001, and September 2001. Specific agreements addressed in this NRC review of the information provided by DOE in the technical basis document include Container Life and Source Term (CLST).3.03, CLST.3.04, CLST.3.06, CLST.3.07, CLST.3.08, CLST.3.09 (Schlueter, 2000), Evolution of Near-Field Environment (ENFE) 3.03 (Reamer, 2001a), Total System Performance Assessment Integration (TSPAI).3.08, TSPAI.3.14 (Reamer, 2001b), GEN.1.01 Comment 116, GEN.1.01 Comment 124, and General (GEN).1.01 Comment 126 (Reamer, 2001c). An initial DOE response to GEN.1.01 Comment 124 was also published in Reamer (2001c). Additional information needs (AIN-1) were previously identified for agreements CLST.3.03, CLST.3.04, CLST.3.06, CLST.3.08, and CLST.3.09 (Schlueter, 2002).

2.0 AGREEMENTS

CLST.3.03

“Provide a more detailed calculation on the in-package chemistry effects of radiolysis. DOE stated that the calculations recently performed as discussed at the 9/12/00 Technical Exchange and preceding teleconferences are being documented. These calculations will be referenced

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and justified in the revision of the Summary of In-Package Chemistry for Waste Forms AMR, ANL-EBS-MD-000050 and will be available in January 2001.”

CLST.3.03 AIN-1

“Provide additional information on the dose rate calculation as a function of time for typical fuels in the waste package and the technical basis for 50 R/hr dose rate.”

CLST.3.04

“Need consistency between abstractions for incoming water and sensitivity studies conducted for in-package calculations, in particular, taking into account the interaction of engineered materials on the chemistry of water used for input to in-package abstractions. DOE stated that the revision of the Summary of In-Package Chemistry for Waste Forms AMR, ANL-EBS-MD-000050 will discuss the applicability of abstractions for incoming water, taking into account the revised Environment on the Surfaces of the Drip Shield and Waste Package Outer Barrier AMR. The revision will be available in January 2001.”

CLST.3.04 AIN-1

“Provide documentation, which shows consistency between abstractions for incoming water and sensitivity studies conducted for in-package calculations.”

CLST.3.06

“Provide additional technical basis for the failure rate and how the rate is affected by localized corrosion. DOE stated that the technical basis for local corrosion conditions will be added to by additional discussion of local chemistry in the Summary of In-package Chemistry for Waste Forms revision ANL-EBS-MD-000050 which will be available in January 2001. Current Clad Degradation Summary Abstraction AMR Section 6.3, ANL-WIS-MD-000007 and Clad Degradation—Local Corrosion of Zirconium and its Alloys Under Repository Conditions AMR, ANL-EBS-MD-000012 contain the overall technical basis.”

CLST.3.06 AIN-1

- “1. Provide the technical basis to define the chemical composition of the in-package water and its evolution with time.
2. Provide the technical basis for excluding the effect of localized corrosion in the form of pitting promoted by chloride.
3. Provide clarification on whether or not DOE assumes that acidic conditions will be present inside the waste packages and the pH range expected if acidic conditions prevail.
”

4. Provide additional information on the environmental conditions, in terms of chemical composition, prevailing inside breached waste packages.”

CLST.3.07

“Provide data to address chloride induced localized corrosion and SCC under the environment predicted by in-package chemistry modeling. DOE stated that the technical basis for the models used for localized corrosion and SCC will be expanded in future revisions of the Clad Degradation Summary Abstraction AMR, ANL–WIS–MD–000007, available by LA.”

CLST.3.08

“Provide the documentation on the distribution for cladding temperature and stress used for hydride embrittlement. DOE stated that the stresses are documented in the Initial Cladding Conditions AMR, ANL–EBS–MD–000048. CAL–UDC–ME–000001 contains the waste package internal temperatures. Waste package surface temperatures were provided within the TSPA model (ANL–EBS–HS–000003, Rev. 00 ICN 01 and ANL–EBS–MD–000049). The updated versions of these documents will be available in January 2001.”

CLST.3.08 AIN-1

- “1. Provide additional information to justify that the temperature is not sufficiently high for dissolution of some of the precipitated hydrides.
2. Provide additional information regarding the accuracy and validity of the stress and temperature data.”

CLST.3.09

“Provide a technical basis for critical stress that is relevant for the environment in which external SCC takes place. DOE stated that critical stress from SCC experiments under more aggressive conditions will be cited in the Revision of the Cladding Degradation Summary Abstraction AMR, ANL–WIS–MD–000007, which will be available in January 2001.”

CLST.3.09 AIN–1

“Provide specific information on the critical stress required for the occurrence of stress corrosion cracking initiated from the external surface of the fuel cladding in the presence of the possible in-package environments.”

ENFE.3.03

“Provide analyses to verify that bulk-scale chemical processes dominate the in-package chemical environment. The DOE will provide analyses justifying the use of bulk chemistry as opposed to local chemistry for solubility and waste form degradation models. These analyses will be documented in an update to the Miscellaneous Waste-Form FEPs AMR

(ANL–WIS–MD–000009) or in an update to the Summary of In-Package Chemistry for Waste Forms AMR (ANL–EBS–MD–000050), expected to be available in FY 02.”

TSPA.3.08

“Provide the technical basis (quantification) for the abstraction of in-package chemistry and its implementation into the TSPA which will demonstrate that the utilization of the weighted-moving-average methodology will not result in an underestimation of risk (ENG3.1.3). DOE will provide the technical basis (quantification) for the abstraction of in-package chemistry and its implementation into the TSPA, which will demonstrate that the implementation methodology will not result in an underestimation of risk. The technical basis will be documented in TSPA-LA and is expected to be available in FY 2003.”

TSPA.3.14

“DOE should account for the full range of environmental conditions for the in-package chemistry model (ENG4.1.1). DOE will update the in-package chemistry model to account for scenarios and their associated uncertainties required by TSPA. This will be documented in the In-Package Chemistry AMR (ANL–EBS–MD–000056) expected to be available to NRC in FY 2003.”

GEN.1.01 Comment 116

“Page 10-22: What does the calculation that shows the rate of water consumption is 27 times greater than the diffusional inflow suggest about the chemical conditions inside the package? Are the simulations for in-package chemistry assuming a bathtub model appropriate?”

GEN.1.01 Comment 124

“In p. 7-74, Ferric Chloride generation is very remote spatially. Provide the basis for the hypothesis.”

“NRC Clarification: Correct page number is 7-64, Section 7.3.7.1.”

“Although useful in ranking a range of alloys, these standard tests utilize aggressive environments (in particular, a ferric chloride solution) that are not directly relevant to expected waste package surface environments because the potential for ferric chloride generation is very remote.”

“The NRC expressed a concern that DOE models might neglect the potential accelerating effect of ferric chloride on corrosion/degradation of waste package materials and fuel cladding materials. NRC requested clarification about DOE's intent to evaluate the potential role of ferric chloride in waste package and fuel cladding degradation.”

DOE Initial Response to GEN.1.01, Comment 124

“Electrochemical corrosion testing is determining the effect that minor constituents will have on the waste package corrosion processes. This will include the effects of ferric ion in the aqueous test solutions. Activities will also determine the extent that minor constituents can

concentrate in the aqueous solutions. This work is covered under existing KTI agreement CLST.1.1.”

“Per existing KTI agreement CLST.3.7, a ferric-chloride local clad corrosion model is being developed that will be documented in a future revision of the Clad Degradation Summary Abstraction AMR.”

GEN.1.01 Comment 126

“In Chapter 9, the assessment of in-package chemistry did not consider the spatial heterogeneity. Provide the rationales for the homogeneity.”

3.0 RELEVANCE TO OVERALL PERFORMANCE

The waste package, composed of metal containers and enclosed waste forms, is the primary engineered barrier controlling the release of radionuclides from spent nuclear fuel and high-level waste glass. Penetration of the waste package by corrosion or disruptive events may allow seepage water to enter and contact the waste forms. Because the release of radionuclides from the waste packages is dependent on the dissolution of cladding, spent nuclear fuel, and vitrified high-level waste, the chemistry of the aqueous environment in contact with waste forms needs to be evaluated to assess degradation, radionuclide release, solubility limits, and colloid stability. Important parameters include the composition, Eh, and pH of the aqueous solutions contacting the waste forms, temperature, and the formation of corrosion products from the internal components. The in-package chemistry abstraction should be adequate to estimate time-dependent waste form degradation rates and solubility constraints and to assess the stability and concentration of colloids.

In an NRC evaluation of the risk significance of postclosure performance assessment model abstractions (NRC, 2004), staff provided the basis for the importance of specific abstractions affecting radionuclide release rates and solubility limits. The conclusions of NRC (2004) reflected the relevance to performance of the in-package environment as discussed in the preceding paragraph. Four aspects of the abstractions that are relevant to Bechtel SAIC Company, LLC (2004a) were concluded to have medium significance to waste isolation: waste form degradation rate, cladding degradation, solubility limits, and the effect of colloids on waste package releases. The three risk insights assigned a rating of “low significance”—mode of release from waste package, invert flow and transport, and criticality—were not principal topics of Bechtel SAIC Company, LLC (2004a) and are not included in this evaluation. In addition, issues specific to colloid-facilitated release were addressed principally in a separate technical basis document (Bechtel SAIC Company, LLC, 2003) and are also not included in this evaluation.

4.0 RESULTS OF THE NRC REVIEW

Specific aspects of the in-package environment treated by the appendixes of Bechtel SAIC Company, LLC (2004a) include (i) the potential for conditions that could promote cladding corrosion and degradation (by localized corrosion, stress corrosion cracking, and hydride embrittlement), such as might be promoted by radiolysis, (ii) the range of chemistries of

seepage waters entering the waste package, (iii) the range of chemistries resulting from reaction with internal waste package components, and (iv) potential underestimation of exposure of engineered materials to corrosive waters in total system performance assessment.

4.1 Agreements Pertaining to In-Package Chemistry Environment

4.1.1 Agreement ENFE.3.03

The DOE response to agreement ENFE.3.03 was provided in Appendix A of Bechtel SAIC Company, LLC (2004a). The agreement is focused on the adequacy of the DOE approach that neglects local environments in favor of a bulk chemistry approach to modeling in-package chemical processes. The DOE in-package chemistry model was developed on the basis of two specified types of waste packages—a waste package containing commercial spent nuclear fuel and a codisposal waste package containing DOE spent nuclear fuel and high-level waste glass. These two package types were considered representative of all waste packages in the repository. By varying input parameters to account for a range of system properties, environmental properties, and material properties, the in-package chemistry model simulates interactions of water with the specified waste package components under different physical, hydrological, and chemical conditions. DOE stated the exact pathway infiltrating or condensing water would follow within the waste package is unknown, but that it is very unlikely that water would contact only the fuel and bypass the other materials inside the waste package.

DOE argued, although reactions with some of these individual waste package components may lead to local pH variations (e.g., pH <3 or >10), other and more abundant reactants in the waste package would tend to buffer the extreme pH toward more neutral values, regardless of the path followed by the water moving through the waste package. In particular, the secondary uranium mineral schoepite (a likely corrosion product of spent nuclear fuel) and iron corrosion products were identified as important solid phases that would resist deviations to high or low pH in the system. Although reaction of water with steel would alone lead to potentially large decreases in pH, the acidity could be buffered by sorption of hydrogen ions onto corrosion product surface sites [e.g., ferrihydrite ($5\text{Fe}_2\text{O}_3 \cdot 9\text{H}_2\text{O}$), or goethite ($\text{FeO} \cdot \text{OH}$)], and by dissolution of schoepite ($\text{UO}_3 \cdot 2\text{H}_2\text{O}$). Corrosion of waste glass might lead to high pH, but DOE argued this would also be buffered by schoepite dissolution. Details about these processes were provided by reference to a report on the in-package chemistry abstraction that was not available at the time of this evaluation and therefore could not be checked, and also by reference to a summary in the main body of the technical basis document (Bechtel SAIC Company, LLC, 2004a, Section 3).

DOE has provided sufficient information to support the argument that infiltrating or condensing water within the waste package will interact with all materials present, either by direct contact or by diffusion. On the timescale of the repository and for conditions where there is sufficient water present to allow transport out of the waste package, diffusion will tend to reduce local in-package chemistry variations. DOE has provided sufficient information on the mechanisms that may limit large changes in pH.

Although staff considers this agreement closed, DOE should note the following comments. Codisposal waste packages, in the in-package chemistry abstraction, were represented by a waste package design type that has only two high-level waste glass canisters and two DOE spent nuclear fuel canisters. This waste package type would account for only about 1 percent

of all waste packages in a potential repository. In contrast, two other design types of codisposal packages contain considerably greater proportions of high-level waste glass and together account for approximately 29 percent of all waste packages in a potential repository (Bechtel SAIC Company, LLC, 2004a, Table 2-1). 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 package, it is not clear why DOE has excluded the more abundant and glass-rich codisposal packages from consideration in the model. NRC staff will look for a technical basis for the use of only two waste package types to represent chemical processes in all types, particularly with respect to differences among the codisposal waste package types.

In addition, although the approach to modeling pH buffering by surface complexation on corrosion products appears sound, staff found small errors in representations of surface complexes in the chemical reactions presented (Bechtel SAIC Company, LLC, 2004a, Section 3.4.3). The unreleased revision to In-Package Chemistry Abstraction may provide the additional information necessary to evaluate how the model parameters related to surface site concentrations were derived.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of KTI agreement ENFE.3.03, and the agreement is closed.

4.1.2 Agreement TSPAI.3.14

The focus of agreement TSPAI.3.14 is to ensure DOE has considered the full range of environmental conditions and accounted for scenarios and their associated uncertainties in its total system performance assessment abstraction of in-package chemistry. In Appendix A and Section 3.4 of Bechtel SAIC Company, LLC (2004a), DOE indicated its updated in-package chemistry abstraction considers wide ranges in input fluid compositions and ambient gas (carbon dioxide and oxygen) partial pressures; the presence and absence of cladding; the effects of an igneous intrusion; degradation in the presence of condensed and seepage water; and wide ranges in the fluxes of both condensed and seepage water. A range of fluid compositions has also been considered that bounds the compositions of fluids likely to prevail inside the repository over long periods of time. DOE noted a pH range of 4.5–8.1 was found to adequately account for the full range of in-package conditions. A number of pH-buffering solid-solution reactions—including schoepite dissolution, steel corrosion, and proton uptake or release by corrosion products—tends to maintain local and bulk pH close to neutral. The DOE analyses indicated the assemblage of reactions has the effect of driving water chemistries toward a common end point, despite the possibly large variation in initial environmental conditions.

DOE in-package chemistry model used two approaches to evaluate uncertainty. One approach applied a factorial design to account for known large potential variations in model input (reactant combinations, water flux, fuel exposure, temperature, and seepage composition). The other approach used sensitivity analyses to evaluate the effect of more uncertain input variations (carbon dioxide fugacity, carbon steel Type 516 sulfur content, corrosion rates, and flux values).

DOE has provided sufficient information on the in-package chemistry abstraction with respect to agreement TSPA.3.14. However, NRC staff may need to review Revision 3 to the analysis and model report In-Package Chemistry Abstraction when it becomes available to fully evaluate the abstraction, because Bechtel SAIC Company, LLC (2004a) contains only summary information. In addition, NRC staff may need to review Revision 1 to the analysis model report Igneous Intrusion Impacts on Waste Packages and Waste Forms when it becomes available to fully evaluate igneous impacts on the in-package chemical environment. NRC staff also may conduct independent calculations to verify the results of DOE in-package chemistry analyses and models.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of Key Technical Issue (KTI) agreement TSPA.3.14, and this agreement is closed.

4.1.3 Agreement GEN.1.01 Comment 116

Comment 116 of Agreement GEN.1.01 identified a potential inconsistency in hydrologically unsaturated models between the rate of water consumption by in-package chemical reactions and the amount of water available from diffusional inflow. In Appendix A of Bechtel SAIC Company, LLC (2004a), DOE indicated, previously, its model required more water than was available by diffusion. DOE stated the inconsistency was resolved in the two currently adopted unsaturated models, one invoking water film development from dripping into the waste package and the other invoking condensation of water films from the vapor phase. Both dripping and nondripping models have separate process models and separate abstraction models for reactions between water and waste package components, allowing distinct implementation in total system performance assessment consistent with the available water mass.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of KTI agreement GEN.1.01 Comment 116, and the agreement is closed.

4.1.4 Agreement GEN.1.01 Comment 126

Comment 126 of Agreement GEN.1.01, questioning the assumption of spatial homogeneity in the DOE in-package chemistry model for performance assessment, was addressed by DOE in Appendix A of Bechtel SAIC Company, LLC (2004a). DOE stated the dominant in-package processes—waste form degradation, solubility, and colloidal stability—are strongly dependent on variability and uncertainty in pH, which are captured by the total system performance assessment abstraction. Accounting for spatial heterogeneity is of secondary importance in performance assessment. The DOE model as summarized in Section 3 of Bechtel SAIC Company, LLC (2004a) does consider spatial heterogeneity by investigating how each component of the waste package reacts individually with water and by reaction-path modeling of the waste package as a “bulk mixture.”

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff

considers the DOE response has satisfied the intent of KTI agreement GEN.1.01 Comment 126, and the agreement is closed.

4.2 Agreements Pertaining to Effects of Radiolysis and Engineered Materials on In-Package Chemistry

4.2.1 Agreement CLST.3.03 and CLST.3.03 AIN-1

The DOE response to agreement CLST.3.03 and CLST.3.03 AIN-1 was provided in Appendix B of Bechtel SAIC Company, LLC (2004a). The focus of CLST.3.03 and CLST.3.03 AIN-1 is on the DOE analysis of in-package chemical effects of radiolysis and the justification for the dose rate used in the cladding degradation analysis. High concentrations of radiolysis products, particularly nitric acid, could promote corrosion of zirconium alloy and accelerate degradation of cladding material. DOE earlier assessment (Bechtel SAIC Company, LLC, 2001) considered high concentrations of nitric acid (e.g., 14.2 mol/L) are required to promote the corrosion of cladding materials. That report stated such high concentrations are not likely because the in-package radiation dose rate is expected to be low at the time waste package breach, and because nitric acid is removed by mechanisms such as escape through package breaches and flushing by dripping water. The calculation for the formation of nitric acid was based on a dose rate of 50 R/hr; however, the justification for that dose rate was not provided. In addition, radiolysis by long-lived alpha particles that may escape from perforated cladding was not considered in the earlier DOE assessment.

The DOE response to the agreement and the additional information need (Bechtel SAIC Company, LLC, 2004a, Appendix B) provided an analysis of dose rates at the outer surface of typical spent nuclear fuel rods. According to the DOE analysis, the gamma dose rate outside typical pressurized water reactor spent nuclear fuel nuclear rods (Babcock and Wilcox 15-by-15 assembly) will be approximately 25 R/hr at 350 years after removal from the reactor and 5 R/hr at 500 years. The gamma dose rate will remain below 5 R/hr after 500 years. Therefore, the dose rate of 50 R/hr used in the DOE calculation is bounding because the waste package is not expected to fail at 350 years after removal. The DOE response sufficiently addressed the intent of agreement CLST.3.03 and CLST.3.03 AIN-1.

Although staff considers this agreement closed, DOE should consider the following comments. The effect of alpha radiolysis on corrosion only applies to waste packages that contain cladding-perforated fuels. If no cladding is perforated, alpha particles will be confined to the inside of the cladding and will not produce detrimental chemical species that would cause corrosion to the cladding materials in the waste package. However, if there are cladding-damaged fuels in a waste package, the alpha effect on the corrosion of the cladding materials may be significant. An analysis of CANDU fuel with a burnup of 721 GJ/kg U [8.3 Gwd/MTU] as a function of time (Shoesmith, 1999) showed that the alpha dose rate in the water layer in contact with the fuel bundle during the first 1 million years will be approximately three orders of magnitude higher than the gamma dose rate at 300 years. The dose rate of 50 R/yr used in the DOE analysis corresponds to the gamma dose rate at 300 years after removal from the reactor (Bechtel SAIC Company, LLC, 2004a). Therefore, if the typical fuel rods to be stored at the potential repository are similar to the CANDU fuel, the alpha dose rate at the damaged fuel and water interface may be significantly higher than the rate used for the bounding calculation. Although the alpha effect is limited to only a very thin layer of water, if hydrogen peroxide is produced steadily (Christensen and Sunder, 2000) at sufficient concentrations, and if sufficient chloride is present,

the corrosion potential of the cladding materials may be maintained above the repassivation potential and may cause localized corrosion of the intact cladding materials. The DOE analysis of alpha radiolysis should include quantitative information on the alpha radiolysis effect or a basis for excluding the damaged fuel because of the low probability.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of KTI agreement CLST.3.03 and CLST.3.03 AIN-1, and this agreement is closed.

4.2.2 Agreement CLST.3.04 and CLST.3.04 AIN-1

The DOE response to agreement CLST.3.04 and CLST.3.04 AIN-1 that concerns the effects of engineered materials on incoming water chemistry, was provided in Appendix B of Bechtel SAIC Company, LLC (2004a). DOE cited information documented in the unreleased Revision 3 of In-Package Chemistry Abstraction that showed the compositions of fluids resulting from in-package chemical reactions are largely insensitive to the initial composition of seepage water entering the package. A sensitivity study in the in-package chemistry abstraction report investigated three starting ambient water compositions: calcium-pore water, sodium-pore water, and J-13 Well water. The pore water values were taken from site data. The sensitivity study also employed three thermally perturbed water compositions representing a range of carbon dioxide fugacity and temperature conditions, abstracted from the unreleased Revision 2 of Engineered Barrier System: Physical and Chemical Environment Model. The results from the two sets of calculations—involving both ambient and thermally perturbed waters—showed that pH and ionic strength vary little with input water chemistry (e.g., Bechtel SAIC Company, LLC, 2004a, Figures B-1 and B-2). DOE neglected the potential effect of high-ionic-strength solutions that could develop due to evaporative concentration on the basis that evaporation would substantially lower the amount of seepage water that could enter the waste package while the waste package is hot.

Staff reviewed the summary DOE descriptions of chemical processes and couplings that might affect the in-package chemistry. In Bechtel SAIC Company, LLC (2004a), DOE provided the information requested in the agreement related to the evolution of in-package chemistry, taking into account the variability in composition of incoming water. Specifically, DOE analyses indicated that incoming water composition has little influence on the pH and ionic strength of in-package fluid. NRC staff agrees with the DOE conclusion that chemical interactions of water with in-package components will dominate the changes in chemistry of in-package water. NRC staff considers the DOE rationale for neglecting the potential effect of high-ionic-strength solutions to be appropriate.

DOE has provided sufficient information on the in-package chemistry abstraction with respect to agreement CLST.3.04. However, NRC staff may review Revision 3 to the analysis and model report In-Package Chemistry Abstraction when it becomes available to fully evaluate the abstraction. Bechtel SAIC Company, LLC (2004a) contains only summary information on the sensitivity study and the abstraction, whereas Revision 3 of In-Package Chemistry Abstraction may contain detailed information on how DOE accounts for the effects of engineered materials on incoming water chemistry.

The NRC staff may also need to review Revision 2 to the analysis and model report Engineered Barrier System: Physical and Chemical Environment Model when it becomes available to evaluate further the abstracted range of thermally perturbed water chemistries employed in the sensitivity study.

As necessary, the NRC staff may conduct independent calculations to (i) verify the results of DOE sensitivity analyses of incoming water compositions and (ii) gain confidence that evaporation may lead to quantities of more concentrated solutions insufficient to affect the in-package environment.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of KTI agreement CLST.3.04 and CLST.3.04 AIN-1, and this agreement is closed.

4.3 Agreements Pertaining to Localized Corrosion and Stress Corrosion Cracking in Cladding

4.3.1 Agreement CLST.3.06 and CLST.3.06 AIN-1

In Appendix D of Bechtel SAIC Company, LLC (2004a), DOE provided its responses to the four additional information needs requested by NRC to satisfy agreement CLST.3.06. In response to Item 1, DOE indicated the technical basis for the time-dependent chemical composition of the in-package water, summarized in Section 3.4 of Bechtel SAIC Company, LLC (2004a), is documented in detail in an unreleased revision to the analysis and model report In-Package Chemistry Abstraction. The in-package chemistry model simulates chemical interactions of water with the waste package materials and the waste form. The results from the in-package chemistry model include time histories of pH, Eh, ionic strength, aqueous concentrations, and mineralogical phase abundances. In response to Item 2, DOE stated acid conditions and high Fe^{3+} concentrations needed to cause localized corrosion of zirconium alloys cannot be achieved based on pH and Fe^{3+} concentrations predicted by the in-package chemistry model. In response to Item 3, DOE stated in-package pH levels are not expected to drop below 4.5, based on analyses documented in the unreleased analysis and model report. The absence of low pH indicates localized corrosion of cladding will not be an important factor inside breached waste packages. In response to Item 4, DOE again stated a summary of in-package chemistry (pH, Eh, ionic strength, fluoride concentration, chloride concentration, and total carbonate concentration) evolution is presented in Section 3.4 of Bechtel SAIC Company, LLC (2004a) and the technical basis for environmental conditions in the waste packages is found in the unreleased analysis and model report.

The focus of CLST.3.06 is to ensure DOE has adequate technical bases for the chemical composition of in-package water, evolution of in-package chemistry with time, and potential effects of in-package chemistry on radionuclide release from the waste form and waste package. In Section 3.4 of Bechtel SAIC Company, LLC (2004a), DOE provided a summary of its in-package chemistry model, which simulates the chemistry of water as it reacts with waste package components and that waste forms inside failed waste packages. The output from the in-package chemistry model includes values for pH, Eh, ionic strength, fluoride concentration, chloride concentration, and total carbonate concentration as a function of time. The results of the model appear to support the DOE conclusion that low pH and high Fe^{3+} levels necessary for localized corrosion of zirconium alloys are not likely. The model results also appear to support

the DOE conclusion that low pH necessary for localized corrosion will not be a likely condition inside a breached waste package. The DOE responses satisfy the intent of CLST.3.06 and the additional information needs requested by NRC. NRC staff may need to review Revision 3 of the document In-Package Chemistry Abstraction when it becomes available.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of KTI agreement CLST.3.06 and CLST.3.06 AIN-1, and this agreement is closed.

4.3.2 Agreement CLST.3.07

The DOE response to agreement CLST.3.07 is contained in Appendix D of Bechtel SAIC Company, LLC (2004a). The focus of CLST.3.07 was the effect of chloride on localized corrosion and stress corrosion cracking of the cladding material under the environment predicted by the in-package chemistry models. Zirconium alloys are known to be susceptible to pitting corrosion in chloride solutions containing oxidizing species such as ferric ions or at potentials higher than the repassivation potential (Brossia, et al., 2002). It was reported that the repassivation potential of Zr-4 material is linearly related to the logarithm of chloride concentration (Brossia, et al., 2002). Based on literature data obtained from as-polished specimens, DOE developed a zirconium-pitting model (Bechtel SAIC Company, LLC, 2004a,b). Considering a range of in-package chemical conditions, including radiolytic production of nitric acid and hydrogen peroxide, the model predicted that the corrosion potentials of as-polished specimens are significantly lower than the repassivation potential (by approximately 500 mV). Pitting corrosion was not predicted to occur even when acid production from radiolysis was increased by a factor of 10. The NRC staff considers this approach appropriate. The DOE model did not consider the corrosion potential measured from oxide-covered specimens. The basis for this consideration was that no localized corrosion was observed on oxide-covered specimens even in a highly corrosive 5-mol/L FeCl₃ solution. The NRC staff agrees with this consideration because oxide must first break down for pitting corrosion of the oxide-covered zirconium alloy to initiate.

Stress corrosion cracking occurs only when both the environmental conditions necessary for localized corrosion and high stress are present. As stated previously, the DOE pitting model predicted that the corrosion potentials of the as-polished specimens are significantly lower than the repassivation potential; therefore, the localized corrosion condition does not exist. In addition, the DOE analysis indicated that the stresses and stress intensities are too low for stress corrosion cracking to occur even if an aggressive chemical environment exists in waste packages. During the times of interest, the stress intensity factor varies from a mean value of 0.47 MPa \cdot m^{1/2} [0.42 ksi \cdot m^{1/2}] to a maximum of 2.73 MPa \cdot m^{1/2} [2.48 ksi \cdot m^{1/2}]. The lowest threshold value for cracks to propagate in various chemical solutions and temperatures that are relevant to in-package conditions is 4 MPa \cdot m^{1/2} [3.6 ksi \cdot m^{1/2}]. Therefore, DOE concluded that stress corrosion cracking is not expected (Bechtel SAIC Company, LLC, 2004a). The DOE response sufficiently addressed agreement CLST.3.0.7.

Although staff considers this agreement closed, DOE should consider the following comments. For oxide-covered specimens, corrosion potentials significantly higher than the repassivation potential have been measured (Brossia, et al., 2002; Yau and Maguire, 1990). If high corrosion potentials are maintained, pitting corrosion of the oxide-covered zirconium alloy should not be ruled out. The DOE analysis (Bechtel SAIC Company, LLC, 2004b), concludes that $E_{corr} > E_{rp}$ is a

necessary but not sufficient condition for pitting does not provide a basis to exclude pitting corrosion. The observations by Brossia, et al. (2002) and Yau and Maguire (1990) that showed no pitting corrosion when E_{corr} was higher than E_{rp} were from short-term experiments. Brossia, et al. (2002) have shown that when the potential of the specimen was held slightly above the repassivation potential, it took approximately 100 hours for a 1.7-Fm [6.7×10^{-5} -in] oxide-covered specimen to initiate, and 1,000 hours for a 3.4-Fm [1.3×10^{-4} -in] oxide-covered specimen to initiate pitting corrosion. The high corrosion potential is probably due to the extremely low anodic current on the oxide-covered zirconium alloy. In the event of the oxide film breakdown, the corrosion potential may be driven to values below the repassivation potential by the increased anodic current from the oxide-damaged sites because of the poor conductivity of the oxide film. This hypothesis should be demonstrated or included in the DOE analysis.

During handling or transportation, the oxide on small areas of the spent nuclear fuel cladding may be damaged. The oxide-damaged areas of the cladding are essentially electrically coupled with the areas covered by the intact oxide film. If the corrosion potential of the oxide-covered cladding is high, the coupling potential may rise beyond the repassivation potential so that the oxide-damaged cladding may be susceptible to pitting corrosion. DOE may need to demonstrate that the high potential observed on the oxide-covered zirconium alloy is not stable and cannot be maintained if there are small areas damaged. DOE may also elect to evaluate the effect on the coupling potential of the large amount of active materials, including stainless steel, surrounding the spent nuclear fuel rods. After breach of the waste package and ingress of water, the stainless steel structures will be in contact with the same solution as the cladding materials. The low potential of these materials, even with the presence of ferric chloride,¹ may significantly lower the coupling potential so that the effective potential of the fuel rods would be well below the repassivation potential.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, the NRC staff considers the DOE response has satisfied the intent of KTI agreement CLST.3.07, and the agreement is closed.

4.3.3 Agreement CLST.3.08 and CLST.3.08 AIN-1

The focus of agreement CLST.3.08 was the cladding temperature and stress for hydride embrittlement. The DOE response provided in Appendix D of Bechtel SAIC Company, LLC (2004a) indicates that the maximum cladding temperature was estimated to be 268 EC [514 EF]. At this peak temperature, the saturation limit for hydrogen is about 44 ppm (Bechtel SAIC Company, LLC, 2004b). For all the existing circumferential hydrides to dissolve at this temperature and for hydride reorientation to occur, the initial hydrogen concentration must be below 44 ppm. Because less than 5 percent of the fuel cladding has an average hydrogen concentration this low, dissolution of circumferential hydrides is not expected. In addition, the minimum stress for hydride reorientation to occur was estimated to be 174 MPa [25.2 ksi] (Bechtel SAIC Company, LLC, 2004b). Only 0.45 percent of the fuel rods have stresses higher than this value. Cladding failure as a result of hydride reorientation was therefore excluded by DOE on the basis of low consequence.

¹Yang, L., N. Sridhar, C.S. Brossia, and D.Dunn. "Evaluation of the Coupled Multielectrode Array Sensor as a Real Time Corrosion Monitor." *Journal of Corrosion Science*. Accepted for Publication. 2004.

Delayed hydride cracking, a particular form of hydride embrittlement, is a time-dependent crack propagation process. Although delayed hydride cracking was not addressed in the new analysis of temperature and stress (Bechtel SAIC Company, LLC, 2004b), it is not expected to occur, based on a previous DOE analysis (CRWMS M&O, 2000a). The important conclusions regarding the effect of temperature and stress on hydride reorientation in Bechtel SAIC Company, LLC (2004a) are supported by the data provided.

Although staff considers this agreement closed, DOE should consider the following comments. In the postclosure period, it is possible that drift degradation could result in natural backfill. The peak waste package temperature under drift degradation conditions is estimated to be 233 EC [451 EF] (Manepally, et al., 2004). In the case of 100-percent engineered backfill, the estimated maximum waste package temperature is approximately 350 EC [662 EF] (Mohanty, et al., 2004). This would in turn increase the cladding temperature and, as a result, the pressure of the pellet-cladding gap. Increased cladding stress caused by drift degradation and natural backfill could affect cladding life.

Spent nuclear fuel considered in the DOE model has burnup up to 75 MWd/kgU (Bechtel SAIC Company, LLC, 2004b). The gap pressure is expected to be higher in high burnup fuels (O'Donnell, et al., 2001), but detailed stress analyses under repository conditions have not been conducted, and related data for the high burnup fuels were not presented.

The critical stress value for hydride reorientation to occur is not well defined because it depends, among many other factors, on temperature, alloy composition, and microstructure. A high level of confidence in the estimation of the critical stress value for hydride reorientation and its possible variation for the range of fuel rod conditions expected in the repository would further support the screening argument.

The saturation limit for hydrogen in Zircaloy solid solution may be different from the average hydrogen concentration in fuel cladding because hydrogen is present as circumferential hydrides. For example, total hydrogen concentrations of 330 and 679 ppm were reported in fuel cladding at burnups of 40 and 65 MWd/kgU, respectively (CRWMS M&O, 1998). These concentrations are significantly higher than the hydrogen saturation limit of 44 ppm. Because of the higher hydrogen content, along with an increase in temperature of the cladding during the storage and loading operations of spent nuclear fuel, the probability of occurrence of hydride reorientation and subsequent cladding failure is higher.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, the NRC staff considers the DOE response has satisfied the intent of KTI agreement CLST.3.08 and CLST.3.08 AIN-1, and this agreement is closed.

4.3.4 Agreement CLST.3.09 and CLST.3.09 AIN-1

The focus of agreement CLST.3.09 was the critical stress for initiating stress corrosion cracking of cladding. The DOE response provided in Appendix D of Bechtel SAIC Company, LLC (2004a) indicated that the chemical environment in the waste package is not sufficiently aggressive for the occurrence of stress corrosion cracking on the outer fuel cladding surface. Even if an aggressive chemical environment existed in waste packages, stresses and stress intensities would be too low for stress corrosion cracking to occur. During the times of interest (when temperature or helium pressure is highest), the stress intensity factor varies from a mean

value of 0.47 MPa \cdot m^{1/2} [0.42 ksi \cdot in^{1/2}] to a maximum of 2.73 MPa \cdot m^{1/2} [2.48 ksi \cdot in^{1/2}]. The threshold values for various chemical solutions and temperatures are greater than or equal to 4 MPa \cdot m^{1/2} [3.6 ksi \cdot in^{1/2}], and that for moist chlorine is 28 MPa \cdot m^{1/2} [26 ksi \cdot in^{1/2}]. The important conclusions regarding the effect of stress on stress corrosion cracking of cladding are supported by the data provided.

Although staff considers this agreement closed, DOE should consider the following comments. The fuel burnup has a significant influence on the cladding hoop stress due to the internal pressure of the fuel rods. Related data for the high burnup fuels under repository conditions were not presented. A technical basis for the modeling of stress corrosion cracking for high burnup fuel cladding should be provided.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, the NRC staff considers the DOE response has satisfied the intent of KTI agreement CLST.3.09 and CLST.3.09 AIN-1, and this agreement is closed.

4.3.5 Agreement GEN.1.01, Comment 124

DOE responded to agreement GEN.1.01 Comment 124 (and agreement CLST.3.07; see Section 4.3.2), which focused on the potentially accelerating effect of ferric chloride on cladding corrosion, in Appendix D of Bechtel SAIC Company, LLC (2004a). Ferric chloride is a strong oxidizer and can raise the corrosion potential of Zircaloy material significantly beyond the repassivation potential (Brossia, et al., 2002). Therefore, Zircaloy cladding materials are susceptible to pitting corrosion in chloride solutions containing ferric ions. According to the DOE analysis (Bechtel SAIC Company, LLC, 2004a,b), the concentration of ferric ions is extremely low under the predicted in-package chemistry conditions. The ferric ion concentration is limited by solubility at pH at and above 3.5 and its effect on pitting corrosion of cladding materials is not likely to be significant. The DOE response sufficiently addressed agreement GEN 1.01 Comment 124.

Although staff considers this agreement closed, DOE should consider the following comments. The combined effect of both radiolysis and ferric ion on the localized corrosion of cladding material was not considered in the DOE response. Although the DOE analysis concluded that the formation of 14.2 mol/L nitric acid by radiolysis is unlikely, concentrations of nitric acid that are significantly lower than 14.2 mol/L may be sufficient to drive the solution pH below 3.5. In addition, at the time the waste package breaches, the relative humidity may be high and the surface of the fuel rods that are not submerged in liquid would be covered by a thin layer of condensed water. A small amount of nitric acid may significantly change the pH of the water film. However, spent nuclear fuel cladding is surrounded by a large amount of carbon steel and stainless steel materials. After breach of the waste package and ingress of water, the carbon steel and stainless steel structures will corrode first. Because the corrosion potentials of these materials in ferric chloride solutions are significantly lower than that of the cladding material,² these more active materials will act as anodes to lower the potential of the cladding materials and thus cathodically protect the cladding material from potential pitting corrosion. This cathodic protection process should be considered in the DOE cladding corrosion model.

²Yang, L., N. Sridhar, C.S. Brossia, and D.Dunn. "Evaluation of the Coupled Multielectrode Array Sensor as a Real Time Corrosion Monitor." *Journal of Corrosion Science*. Accepted for Publication. 2004.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), preview NRC staff review (Reamer, 2004), and notwithstanding new information that could raise new questions or comments, the NRC staff consider the DOE response has satisfied the intent of KTI agreement GEN.1.01 Comment 124, and this agreement is closed.

4.4 Agreement Pertaining to Total System Performance Assessment Implementation of In-Package Chemistry

4.4.1 Agreement TSPA1.3.08

The DOE response to KTI agreement TSPA1.3.08, concerned with potential risk dilution by the weighted-moving-average method in total system performance assessment, was presented in Appendix E of Bechtel SAIC Company, LLC (2004a). DOE acknowledged that the use of a weighted-moving-average could result in an underestimation of risk if the early in-package chemistry (after waste package failure and contact by seepage water) is significantly different from the late in-package chemistry (after the waste package internal components have been fully degraded). In the total system performance assessment for site recommendation model, significant differences between the early and late in-package chemical conditions were suggested (CRWMS M&O, 2000b). However, DOE reports revised studies that are not yet available suggest less significant differences between those periods; the revised model will be documented in the unreleased Revision 3 of the In-Package Chemistry Abstraction analysis and model report. Therefore, risk dilution is not expected to occur from the use of the weighted-moving-average methodology.

The weighted-moving-average is a technique for the definition of an average exposure time, for a set of failed waste packages, to select appropriate in-package chemical conditions as a function of the time of spent nuclear fuel exposure to a humid or aqueous environment. At any timestep, the average exposure time is defined as

$$t_{av}^{i+1} = t_{av}^i + \Delta t \frac{WP_{fail}^i}{WP_{fail}^{i+1}} \quad (1)$$

where

t_{av}^i, t_{av}^{i+1}	—	average exposure time at the i and $i + 1$ timesteps
Δt	—	timestep duration
$WP_{fail}^i, WP_{fail}^{i+1}$	—	number of waste packages failed at the i and $i + 1$ timesteps

Depending on the value of the average exposure time, t_{av}^{i+1} , in-package chemical conditions are selected for the set of failed waste packages at the $i + 1$ timestep. The DOE total system performance assessment model tracks groups of packages rather than individual waste packages. Because waste packages within a group can fail at different times, an average exposure time is used. The DOE total system performance assessment abstraction is designed so that a unique in-package environment can be applied to all of the failed waste packages in that particular group at a given timestep. Therefore, the average exposure time [Eq. (1)] is used to select an in-package environment for the computation of waste form dissolution rates from all of the failed waste packages in the group at a timestep.

In the total system performance assessment model for the site recommendation, the pH in a breached waste package dropped to very low values within about 50 years after breach (CRWMS M&O, 2000b). For example, if a second waste package takes 60 years to fail after failure of the first waste package, the average exposure time after failure of the second waste package, computed using Eq. (1), yields a value greater than 60 years. This average exposure time is used to select the appropriate in-package chemistry to apply to the two failed waste packages. Because the average exposure time exceeds 60 years, the pH of the in-package solution is selected in an alkaline regime in the model. Therefore, in this model implementation, the second waste package would not experience the expected early acidic conditions after breaching.

Although the average exposure time concept could lead to risk dilution, NRC staff agrees with DOE that the possibility of risk dilution is not significant if the early and late in-package pH ranges are similar. In Appendix E, DOE has presented a summary of arguments for the similitude of these pH ranges in the revised in-package chemistry abstraction.

On the basis of information presented in Bechtel SAIC Company, LLC (2004a), and notwithstanding new information that could raise new questions or comments, NRC staff considers the DOE response has satisfied the intent of KTI agreement TSPA1.3.08, and the agreement is closed.

5.0 SUMMARY

NRC reviewed the DOE KTI agreement responses within Bechtel SAIC Company, LLC (2004a) to determine whether any important aspect of agreements CLST.3.03, CLST.3.04, CLST.3.06, CLST.3.07, CLST.3.08, CLST.3.09, ENFE.3.03, TSPA1.3.08, TSPA1.3.14, GEN.1.01 Comment 116, GEN.1.01 Comment 124, and GEN.1.01 Comment 126 (and associated AIN) were excluded. In addition, NRC performed an independent assessment to determine if the information provided would support submission of a potential license application for a geologic repository. Notwithstanding new information that could raise new questions or comments concerning these agreements, the information provided satisfies the intent of the agreements.

6.0 STATUS OF THE AGREEMENTS

Based on the preceding review, NRC considers agreements CLST.3.03, CLST.3.04, CLST.3.06, CLST.3.07, CLST.3.08, CLST.3.09, ENFE.3.03, TSPA1.3.08, TSPA1.3.14, GEN.1.01 Comment 116, GEN.1.01 Comment 124, and GEN.1.01 Comment 126 to be closed.

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