

REQUEST FOR ADDITIONAL INFORMATION (RAI)
Volume 3—Postclosure Chapter 2.2.1.3.4—Radionuclide Release Rates and Solubility
Limits—1st Set (RAIs 1 through 16)
(DEPARTMENT OF ENERGY'S SAFETY ANALYSIS REPORT Section 2.3.7—Waste Form
Degradation and Mobilization and Engineered Barrier System and Flow and Transport)

Section 1. Waste Form Degradation

RAI #1

Provide bases for not considering (i) multiple-layer sorption, (ii) radiocolloid formation and sorption, or (iii) precipitation, of actinides (e.g., Pu-239) on the surface of groundwater colloids, iron oxide colloids, or other waste form colloids, during the fast dissolution of commercial spent nuclear fuel (SNF).

Basis: The applicant presented the model for radionuclide release from the commercial SNF matrix in SAR section 2.3.7.7.3.2. The model determines the fractional dissolution rate of the SNF matrix as a function of the effective surface area of the SNF, temperature, carbonate concentration in solution, oxygen partial pressure, and solution pH. To determine these parameter values, the applicant used results from flow-through tests and dripping tests (BSC, 2004a). The upper end of the range of dissolution rates was derived from flow-through tests in carbonate solutions. This portion of the range appears to have contributed to fast dissolution times for SNF (120 to 2,100 years). While fast dissolution rates could seem to be risk-conservative, not all consequent processes resulting from these dissolution rates appear to have been evaluated in the SAR. These consequent processes could increase overall release rates from the repository.

Fast matrix dissolution might result in (i) multiple-layer sorption, (ii) radiocolloid formation and sorption (Ahn, 1996), or (iii) precipitation, of actinides (e.g., Pu-239) on the surface of groundwater colloids, iron oxide colloids, or other waste form colloids (e.g., uranophane or zirconium oxide colloids). Iron oxide colloids could also form in the waste form domain from the corrosion of the steels used in the waste package internals and the transportation aging and disposal (TAD) canister. The applicant observed the colloid formation in solution and the precipitation of actinides on the surface of the SNF and test vessels in the tests of bare SNF at 25 and 85 °C in slow replenished J-13 well water (Wilson, 1990a, 1990b). Faster dissolutions at higher temperatures and in earlier test times resulted in more colloid formation and precipitation. The requested Information is needed to verify compliance with 10 CFR 63.21(c)(9), (12) and (15) and 63.114(b) and (g).

RAI #2

Describe the TSPA implementation of colloid formation (e.g., Pu-239) and radionuclide release (e.g., Tc-99) occurring from rapid mobilization (or dissolution) of (i) the commercial SNF and

HLW glass under igneous intrusive conditions and (ii) the DOE-owned SNF under nominal and igneous intrusive conditions.

Basis: The applicant presented the bounding assumption in the radionuclide release from the commercial SNF and HLW glass under igneous intrusive conditions in SAR 2.3.11.3.2.4. To avoid underestimating radionuclide mobility, the applicant assumed that all commercial SNF and HLW glass in the drifts is unprotected, instantaneously degraded, and the radionuclides are immediately available for mobilization by groundwater (SNL 2007, Section 8.1). Similarly, the applicant assumed in SAR 2.3.7.8.1 that the DOE-owned SNF degrades instantaneously upon waste package breach (BSC 2004b, Section 6.2 and 8.1). The applicant needs to describe how its bounding assumption addresses (i) colloid formation and precipitation of actinides such as Pu-239 and (ii) faster release of highly soluble radionuclides such as Tc-99 or I-129. The requested information is needed to verify compliance with 10 CFR 63.21(c)(9), (12) and (15) and 63.114(b) and (g).

RAI #3

Justify the assumption that crack surface areas for HLW glass in the seismic scenario and under conditions of glass alteration are the same as those for the nominal case scenarios.

Also, provide bases for the assumption that only half of the cracked surfaces would be exposed to water vapor during dissolution in the vapor-phase aqueous environment.

Basis: The exposed surface area of the glass is important in assessing the radionuclide release from the glass dissolution. The applicant modeled surface area increase from cracking during vitrification in SAR 2.3.7.9.3 (and BSC 2004c, Section 6.6, Table 6-14). The exposure (i.e., increase) factor was represented by a triangular distribution with values from 4 to 17 (maximum probability at 4) in the nominal case (Table 2.4-11, SAR 2.4-400). The applicant estimated this distribution from the thermal cracking during the vitrification, the probability-weighted cracking from inadvertent handling, and the accessibility of water to tight cracks. However,

- (i) the applicant did not provide the exposure factor under seismic conditions. Under seismic conditions there could be additional cracking.
- (ii) under both nominal and seismic conditions, the applicant did not consider additional cracking due to potential volume increase of glass alteration products. The intact portion of glass may be subject to stress induced by the expansion of the alteration products during the glass dissolution (Abrajano, et al, 1990).

The bases for DOE's assumption that only half of the cracked surface would be exposed to vapor phase for inducing hydration and subsequent radionuclide release are unclear. Exposure to more than half the cracked surface area would seem to increase release rates; therefore, this assumption seems to be potentially non-conservative. It is also unclear whether more surface

area releases both high solubility radionuclides (e.g., dissolved Tc-99) and low solubility radionuclides (e.g., Pu-239 colloids). The requested information is needed to verify compliance with 10 CFR 63.21(c)(9), (12) and (15) and 63.114(b) and (g).

RAI #4

Justify the assumption that commercial SNF conditions will not be altered during transportation and/or interim storage.

Basis: In SAR 1.5.1.1.1 the applicant did not justify its assumptions that SNF conditions in the transportation, aging and disposal (TAD) canister would be unaltered at receipt. The applicant assumes that the conditions would not be altered during transportation and interim storage, or any alteration would not affect the commercial SNF performance in the repository.

During the transportation and interim storage, the commercial SNF matrix in the TAD canister may be hydrolyzed preferentially along grain boundaries (Finch, et al., 1999; Ahn and Mohanty, 2008) with residual moisture if there is no protection or even if only some cladding damage. This preferential hydration may lead to disintegration of the SNF matrix into grains. The grain-boundary hydration could be facilitated if the TAD canister is not leak-tight and allows more moisture during the transportation and interim storage. The canister may not be leak-tight because of improper welding or the deterioration of welds during transportation. Additionally, the applicant did not address the conditions of high burnup SNF above 60 – 65 GWd/MTU. The fine-grained rim structure, which is enriched with plutonium, on the outer surface of the commercial SNF (NRC, 2007) may be subject to fracturing from vibration during transportation. The requested information is needed to verify compliance with 10 CFR 63.21(c)(3), (9), (12) and (14) and 63.114(a), (b) and (g).

References

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Section 2. In-Package Chemical and Physical Environment

RAI #5

Assess the effects and significance on abstracted glass waste form pH limits (i.e., Cell 1a pH limits illustrated in SAR Figure 2.3.7-20) of using the 5-DHLW/DOE Long design to represent codisposal waste packages in the in-package chemistry model.

Basis: The applicant selected the 2-MCO/2-DHLW design to represent Cell 1a and Cell 1b of codisposal waste packages for developing in-package pH abstractions for TSPA (SAR Section 2.3.7.5.3). However, the most common codisposal waste package is the 5-DHLW/DOE Long, which contains 5 HLW glass canisters instead of the 2 HLW glass canisters in the 2-MCO/2-DHLW design. The applicant has identified dissolution of glass waste as a process capable of generating high pH inside waste packages (SAR Sections 2.3.7.5.1 and 2.3.7.5.3.1). The more abundant and glass-rich 5-DHLW/DOE Long design would provide greater glass surface area to react with incoming waters and, therefore, could potentially increase the maximum abstracted

pHs for Cell 1a, which in turn could increase release rates. The requested information is needed to verify compliance with 10 CFR 63.21(c)(3), (9), and (11)-(14) and 10 CFR 63.114(b) and (c).

RAI #6

Provide support to address predicted high ionic strength conditions in the waste form cells resulting from in-package chemistry model simulations.

Basis: The applicant provided support for the in-package chemistry model by showing that predicted secondary phases and pH ranges are consistent with observations from natural soils and groundwater, natural analogues, and/or laboratory experiments (SAR Section 2.3.7.5.3.2). However, SAR Section 2.3.7.5.3.2 does not include support for predicted ionic strength conditions. Ionic strength abstractions generated from the results of in-package chemistry model simulations are used in TSPA to determine the stability of colloids (e.g., colloid suspensions are unstable at high ionic strengths). High ionic strengths, predicted in the waste form cells based on in-package chemistry model simulations (SAR Figures 2.3.7-22 through 2.3.7-25), weaken repulsive forces between colloids, causing colloidal suspensions to become unstable and settle out of solution, decreasing overall release rates. Therefore, it is important that model support for predicted high ionic strength conditions resulting from in-package chemistry model simulations are addressed in the SAR. The requested information is needed to verify compliance with 10 CFR 63.21(c)(15) and 10 CFR 63.114(g).

RAI #7

Provide justification for excluding available pH data from glass dissolution experiments to support predicted HLW glass waste form pH ranges resulting from in-package chemistry model simulations.

Basis: The applicant provided support for the in-package chemistry model in SAR Section 2.3.7.5.3.2. The applicant supported abstracted pH ranges for the HLW glass waste form (i.e., Cell 1a represented by 2-DHLW) generated by in-package model simulations by comparison to pH ranges observed in groundwater in contact with basalt and basalt glass. However, abstracted pH ranges for the HLW glass waste form were not compared to available data from glass dissolution experiments (Bates et al., 1994; Jantzen et al., 1994). The pHs reported in these glass dissolution experiments had relatively high pHs (pH > 10). Because high pH contributes to processes that are risk significant to waste isolation (i.e., neptunium solubility), it is important that predicted pH ranges in the HLW glass waste form cell be adequately addressed and justified in the model. The requested information to verify compliance with 10 CFR 63.21(c)(15) and 10 CFR 63.114(g).

References:

Bates, J.K., C.R. Bradley, E.C. Buck, J.C. Cunnane, W.L. Ebert, X. Feng, J.J. Mazer, D.J. Wronkiewicz, J. Sproull, W. L. Bourcier, B. P. McGrail, and M.K. Altenhofen 1994. "High-Level Waste Borosilicate Glass A Compendium of Corrosion Characteristics Volume 2." DOE-EM-0177 Vol. 2 of 3. Washington, DC: Department of Energy.

Jantzen, C.M., N.E. Bibler, D.C. Beam, C.L. Crawford, and M.A. Pickett 1993. "Characterization of the Defense Waste Processing Facility (DWPF) Environmental Assessment (EA) Glass Standard Reference Material (U)." WSRC-TR-92-346, Revision 1. Aiken SC: Westinghouse Savannah River Company.

Section 3. Concentration Limits

RAI #8

Provide justification for the approach in which pH is computed in the three EBS radionuclide transport domains (waste form, corrosion products, and invert) using unrelated abstractions. Provide justification that this approach does not potentially lead to underestimating EBS release rates.

Basis: Within the EBS component of TSPA, pH is calculated separately in Cell 1 (waste form domain), Cell 2 (corrosion products domain), and the invert. In Cell 1, pH is sampled from a range that depends on (i) package type, (ii) vapor or liquid influx conditions, (iii) ionic strength, and (iv) CO₂ partial pressure (SNL, 2008, Section 6.3.7.2.2). In Cell 2, pH is calculated from a formula that involves CO₂ partial pressure and the aqueous U concentration (SAR Section 2.3.7.5.3.1). The invert pH is obtained from the seepage evaporation abstraction (SAR Section 2.3.5.5). There is no integration of abstracted pH as water flows from the waste package to the invert. Therefore, concentration limits and colloid stabilities, some of which depend strongly on pH, can vary markedly from one cell to another within a single realization. The requested information is needed to verify compliance with 10 CFR 63.21(c)(9), (11)-(15) and 63.114(a), (b), and (g).

RAI #9

Justify the use of high-temperature Pu solubility experimental data in fitting the adjusted-Eh Pu solubility abstraction as it applies to all repository thermal conditions.

Basis: The Eh-pH equation used in the adjusted-Eh abstraction for the Pu concentration limit was selected so that the resulting curve of Pu concentration versus pH passes through a set of published Pu solubility experimental results (SAR Figure 2.3.7-37). Some of the data used in SAR Figure 2.3.7-37, however, were obtained at temperatures of 60° and 90° C (Nitsche, et al., 1993, 1994; Efurud, et al. 1998), which are higher than long-term conditions in the waste package. For Pu, higher temperatures result in lower solubility limits (SNL, 2007a, Section

6.3.3.3) and therefore lower release rates. The high-temperature data, therefore, may inappropriately skew the range of experimental data to lower values. Without the 60° and 90° C results, nearly every data point in SAR Figure 2.3.7-37 falls above the mean curve that was apparently fit through the data. The requested information is needed to verify compliance with 10 CFR 63.21(c)(11)-(15) and 63.114(a) and (b).

RAI #10

For the adjusted-Eh Pu solubility limit model, provide the basis for (i) the applicability of the oxidation-reduction conditions under which supporting Pu solubility data were obtained, and (ii) the applicability of Yucca Mountain region groundwaters and miniature waste package experiments in supporting the adjusted-Eh equation. Demonstrate that the adjusted-Eh abstraction does not result in underestimation of reasonable Pu concentration limits.

Basis: The adjusted-Eh Pu model is intended to apply to conditions inside a breached waste package that is likely to have access to atmospheric O₂ concentrations. The data to which the model was fitted included experimental Pu solubility experiments conducted in closed vessels originally under air (Rai, 1984; Rai, et al., 2001), but also experiments conducted in the absence of O₂ (Nitsche, et al., 1993, 1994; Efurud, et al. 1998) – counter to a implication on SAR page 2.3.7-55 that all experiments were “originally open to the air.” None of the experiments allowed free exchange with air, as would potentially be the case in a breached package as assumed in the model abstraction and which would facilitate increased release rates.

In addition, the Eh-pH conditions measured in all sets of experiments were well below the adjusted-Eh model line. It is not clear that the experimental conditions were all “comparable to the modeled conditions” (SNL, 2007a, p 6-57). Figure V-6 of SNL (2007a) shows that the adjusted-Eh model line lies above all Yucca Mountain groundwater measurements; these data, however, were obtained from the saturated zone and therefore may have limited applicability in the case of the interior of a breached waste package in the unsaturated zone. Appendix V of SNL (2007a) also argues that redox conditions inside miniature waste packages—as constrained by the formation of magnetite—in experiments by Zarrabi, et al. (2003) were well below the adjusted-Eh line. These experiments, however, also contained ferric oxyhydroxides that indicate more oxidizing conditions than the hematite-magnetite buffer. In addition, the Zarrabi, et al. (2003) experiments employed carbon steel, which is now excluded from CSNF waste packages. The requested information is needed to verify compliance with 10 CFR 63.21(c)(11)-(15) and 63.114(a), (b), and (g).

RAI #11

Provide the basis for using the adjusted-Eh redox conditions in modeling NaNpO₂CO₃ solubility for the Np concentration limits abstraction. Provide justification that use of adjusted-Eh conditions for this Np solid phase will not unreasonably lead to underestimation of dissolved Np concentration.

Basis: According to SNL (2007a, page 6-67), the adjusted-Eh model was used to calculate $\text{NaNpO}_2\text{CO}_3$ solubilities that apply to the Np concentration limit abstraction at certain pH and CO_2 partial pressure conditions. The adjusted-Eh model was developed for the Pu abstraction by fitting Pu solubility models to Pu experimental data. Its applicability to the Np abstraction is not clear. As discussed in RAI #10, that the adjusted-Eh model is more oxidizing than Yucca Mountain groundwaters is not a sufficient basis for the model's applicability. The requested information is needed to verify compliance with 10 CFR 63.21(c)(11)-(15) and 63.114(a), (b), and (g).

RAI #12

Clarify whether or not Np incorporation into uranyl minerals is included in any aspects of the Np concentration limits conceptual model. Provide justification that addresses the inconsistencies between the conceptual and alternative conceptual models described in the basis below.

Basis: As discussed in Appendix IV of SNL (2007a), DOE concluded that an Np concentration limit model that included incorporation into uranyl minerals did not have sufficient basis for inclusion in the base case. On page IV-6 of that same appendix, however, DOE relies on the conceptual model of Np co-precipitation in uranyl minerals to help support the use of an NpO_2 solubility model for CDSP packages in the nominal scenario and CSNF packages in the igneous intrusion scenario. This reliance on Np incorporation appears to be at odds with DOE's rejection of the alternative conceptual model. The requested information is needed to verify compliance with 10 CFR 63.21(c)(9), (11)-(15) and 63.114(b), (c), and (g).

RAI #13

Compare the Np concentration limit abstractions against available laboratory data on Np pure-phase solubility limits. Provide justification that these data are either not applicable or support the adopted abstraction.

Basis: In developing and validating the Np concentration limits abstraction, DOE did not refer to laboratory measurements of Np concentrations in pure-phase solubility experiments (e.g., summary by Neck, 2006), including data obtained by the Yucca Mountain Project (Nitsche, et al., 1993, 1994; Efurd, et al., 1998). At a given pH, the Np concentrations measured in these studies are 0.5 to 3 log units higher than the higher-Np Np_2O_5 DOE abstraction curve (SAR Figure 2.3.7-39). A higher Np concentration limit could result in higher Np release rates from the EBS. Neither the SAR nor the supporting AMR (SNL, 2007a) discussed these concentration results. Appendix IV of SNL (2007a) does discuss the Np solid phase identifications in Nitsche, et al. (1993, 1994) and Efurd, et al. (1998) and argues that the solids are not representative of long-term repository conditions. The appendix (SNL, 2007a, page IV-15) also compares the Efurd, et al. (1998) solubility constant with the abstraction value for Np_2O_5 and argues that the Efurd, et al. (1998) value, though within the abstraction uncertainty bounds, is too high because it reflects hydrated Np_2O_5 . However, DOE has not transparently described available solubility

data, including project data, that are relevant to the Np abstraction and has not evaluated whether the data are suitable for supporting the model abstraction. The requested information is needed to verify compliance with 10 CFR 63.21(c)(11)-(15) and 63.114(b) and (g).

RAI #14

Provide the basis for confidence that the U and Np concentration limit abstractions are appropriate with respect to uncertainty in the dissolved Na concentration.

Basis: Over portions of the ranges of chemical conditions in their respective concentration limits abstractions, U will be controlled by the solubility of sodium boltwoodite ($\text{NaUO}_2\text{SiO}_3\text{OH}\cdot 1.5\text{H}_2\text{O}$) or $\text{Na}_4\text{UO}_2(\text{CO}_3)_3$, and Np by the solubility of $\text{NaNpO}_2\text{CO}_3$ (SAR Section 2.3.7.10.3). Under solubility control by these solids, the U or Np aqueous concentration will be directly related to the Na aqueous concentration by the solubility reaction equilibrium constant. The U and Np concentration limits and release rates, therefore, could strongly depend on the dissolved Na content in the waste form domain, corrosion products domain, and the invert. The requested information is needed to verify compliance with 10 CFR 63.21(c)(11)-(15) and 63.114(b).

RAI #15

Development of the NpO_2 solubility model and the adjusted-Eh Pu model assumed the presence of carbon steel in the CSNF waste packages (Appendices IV and V of SNL, 2007a). Provide the technical bases to justify the use of these models given that carbon steel is no longer incorporated into the CSNF waste package design. Provide the technical basis in terms of (i) the quantitative/qualitative impact on these models and release rates and (ii) propagation of uncertainty from these models through the TSPA-LA.

Basis: The requested information is needed to verify compliance with 10 CFR 63.21(c)(3), (9), and (11)-(15) and 63.114(a), (b), and (g).

RAI #16

Clarify which thermodynamic databases, or modifications of same, were used for solubility calculations for each radioelement. Provide justification that the multiple thermodynamic databases used in calculating radioelement solubility are appropriately applied in a way consistent with the potential repository conditions. Provide information on the internal consistency of each modification of the thermodynamic database.

Basis: SNL (2007b) describes the development of three DOE databases developed for use in supporting the license application: data0.ymp.r2, data0.ymp.r4, and data0.ymp.r5. Because development of these databases continued during preparation of analysis model reports supporting SAR Section 2.3.7, it is not always clear which database was used for calculations for a given element. In addition, mention is made in SNL (2007a) of modifications made to the

databases for particular purposes. The use of multiple databases should be consistent with expected conditions and processes in the EBS. In addition, each database must maintain internal consistency among thermodynamic parameters. The requested information is needed to verify compliance with 10 CFR 63.21(c)(11)-(15) and 63.114(b).

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