RAI Volume 3, Chapter 2.2.1.3.4, First Set, Number 2, Supplemental Question:

> It appears the modeling approach for waste form colloid generation, and plutonium and americium association with those colloids is the same in the nominal and igneous intrusive cases. It is not clear from the response why, in the igneous intrusive case, the model does not consider the potential for generation of different quantities of waste form colloids than in the nominal case.

1. SUPPLEMENTAL RESPONSE

The potential for generation of significantly different quantities of waste form colloids in the igneous intrusion modeling case than in the nominal modeling case has been considered. Some specific details regarding the three waste-form types, commercial spent nuclear fuel (SNF), DOE SNF, and high-level waste (HLW) glass are provided in Sections 1.1, 1.2, and 1.3 respectively. The colloid model uses the same waste-form colloid concentration distributions for all modeling cases, and because waste-form colloids are assumed to be created at fixed concentrations in the aqueous phase, more colloid mass is generated when greater volumes of water flow through failed waste packages. Consequently, the igneous intrusion modeling case generally has greater colloid mass than the nominal modeling case because the igneous intrusion modeling case has a higher rate of advection.

1.1. COMMERCIAL SPENT NUCLEAR FUEL

The same distributions of the parameters used to calculate the commercial SNF waste-form colloid concentration (when the commercial SNF colloids are stable) (SNL 2007 Section 6.3.2.4) are used in all modeling cases. The potential for generation of different quantities of commercial SNF waste form colloids in the igneous intrusion modeling case was considered in *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (SNL 2007) in Section 6.3.6.2, "Effects of Dry Oxidation of CSNF on Colloid Formation." Available data was examined to determine whether exposure of commercial SNF to "dry" air oxidizing conditions followed by exposure to humid air is likely to lead to formation of submicrometer particles that would be available for suspension as colloids when water contacts the oxidized fuel. It was concluded that dry oxidation at high temperatures would mainly produce particles larger than a micrometer, and that humid conditions following the dry oxidation would lead to increasing particle size, further decreasing the fraction of colloid-size oxidized material.

1.2. DOE SPENT NUCLEAR FUEL

The same sequence of DOE SNF degradation and secondary phase formation is expected in all modeling cases. DOE SNF is modeled as uranium metal fuel, which rapidly degrades in the presence of water and oxygen to produce initial colloid suspensions of 10-nm spheres of UO_2 (SNL 2007 Section 6.3.6) no matter what the modeling case. These suspensions are unstable, and the resulting flocculated UO_2 colloid particles are expected to behave like high-surface area commercial SNF, eventually forming secondary phases such as meta-schoepite and uranophane. It is these secondary phases that are modeled as reversible uranium-mineral colloids for both

DOE SNF and commercial SNF (SNL 2007 Section 6.3.2.6). The same distribution of uranium–mineral colloid concentration (SNL 2007, Table 6-8), and the same K_ds for sorption of radionuclides onto the uranium-mineral colloids (SNL 2007, Table 6-9), are used for all modeling cases.

1.3. HIGH-LEVEL WASTE GLASS

The same distributions of the parameters used to calculate the glass waste-form colloid concentration (SNL 2007, Tables 6-3 and 6-4) are used for all modeling cases. As discussed in Section 6.3.3 of *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (SNL 2007), waste-form colloids form by precipitation of radionuclide-bearing phases within the glass-weathering rind and spallation of this rind. This process will occur during vapor hydration of the glass as the temperature falls between 125°C and 100°C, and by aqueous degradation of the glass as the temperature falls below 100°C (BSC 2004, Section 8.1). No hydration of the glass is expected when the temperature is higher than 125°C and the relative humidity is below 44%. In the nominal modeling case, breach of the waste package occurs after the temperature falls below 100°C, so only aqueous degradation occurs. In the igneous intrusion modeling case, the intrusive event is modeled to breach the waste package and heat it above 125°C, so vapor hydration occurs as the waste cools below 125°C. However, the same secondary mineral phases are expected to form under higher temperature vapor and lower temperature aqueous conditions, so the same colloid formation model is applicable (BSC 2004, Section 6.5.3.3).

1.4. CONCLUSION

Because the igneous intrusion modeling case is dominated by advective releases, driven by the assumption that flow (through the waste form) is equal to the percolation rate, waste-form colloid generation and release rate will be higher for the igneous intrusion modeling case than the nominal modeling case. This is because waste-form colloids are assumed to be created at fixed concentrations in the aqueous phase, no matter what the modeling case. At the higher flow rates of the igneous intrusion modeling case, more colloid mass is generated compared to the nominal modeling case, although not through any intrinsically different generation mechanisms.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

BSC (Bechtel SAIC Company) 2004. *Defense HLW Glass Degradation Model.* ANL-EBS-MD-000016 REV 02. Las Vegas, Nevada: Bechtel SAIC Company. ACC: DOC.20041020.0015; DOC.20050922.0002; LLR.20080408.0271; DOC.20081021.0002.

SNL (Sandia National Laboratories) 2007. *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary.* MDL-EBS-PA-000004 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071018.0019.[a]

NOTE: [a] Provided as an enclosure to letter from Williams to Sulima dtd 03/03/2009. "Yucca Mountain – Request for Additional Information Re: License Application, Safety Evaluation Report Volume 3, Chapters 2.2.1.2.1, Second Set - Submittal of U.S. Department of Energy Reference Citations."

RAI Volume 3, Chapter 2.2.1.3.4, First Set, Number 3, Supplemental Question:

> In this response DOE concluded that available experimental data indicate that glass corrosion, at the location of a crack, does not contribute to crack extension or additional glass fracturing. Did DOE consider whether the water chemical conditions in the cited experiments were relevant to the modeled in-package chemical environment?

1. SUPPLEMENTAL RESPONSE

Yes, DOE considered the following water chemical conditions in the cited experiments to be relevant to the modeled in-package chemical environment;

1. The important chemical parameters that influence the rate of corrosion of glass are pH and dissolved silicon concentration in the local solution inside the crack (BSC 2004, Section 6.3.1).
2. The pH and the dissolved silicon concentration in the local solution inside the crack are both quickly controlled by the glass dissolution under the high glass surface area to solution volume conditions of the crack (BSC 2004, Section 6.5.4).
3. Because the pH and dissolved silicon concentration inside the crack are both controlled by glass dissolution, they are not dependent on the crack's external chemical environment.

The cited experimental results (i.e., glass corrosion at the location of a crack does not contribute to crack extension or additional glass fracturing) are insensitive to the chemical conditions in the experiments. This insensitivity extends over the range of chemical conditions of the modeled in-package chemical environment. Therefore, the cited experimental results are relevant to the modeled in-package chemical environment.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

BSC (Bechtel SAIC Company) 2004. *Defense HLW Glass Degradation Model.* ANL-EBS-MD-000016 REV 02. Las Vegas, Nevada: Bechtel SAIC Company. ACC: DOC.20041020.0015; DOC.20050922.0002; LLR.20080408.0271; DOC.20081021.0002.

RAI Volume 3, Chapter 2.2.1.3.4, First Set, Number 8, Supplemental Question 1:

> Page 2 of that response states that a "strong correlation exists between the models for pH and [the waste form and corrosion products] domain." Does this statement refer to a mathematical correlation or is it simply a statement that there is a relationship because they both rely, in part, on CO_2 partial pressure? Also, please direct NRC staff to discussions of model support for the corrosion products pH abstraction.

1. SUPPLEMENTAL RESPONSE

The referenced statement is not meant to imply a direct statistical correlation embedded in the models, but simply that the pHs in the two domains are related through their dependence on CO_2 partial pressure. However, the observable relationship or correlation is relatively weak, as indicated in a scatterplot comparing the pH values in the two domains (Figure 1).

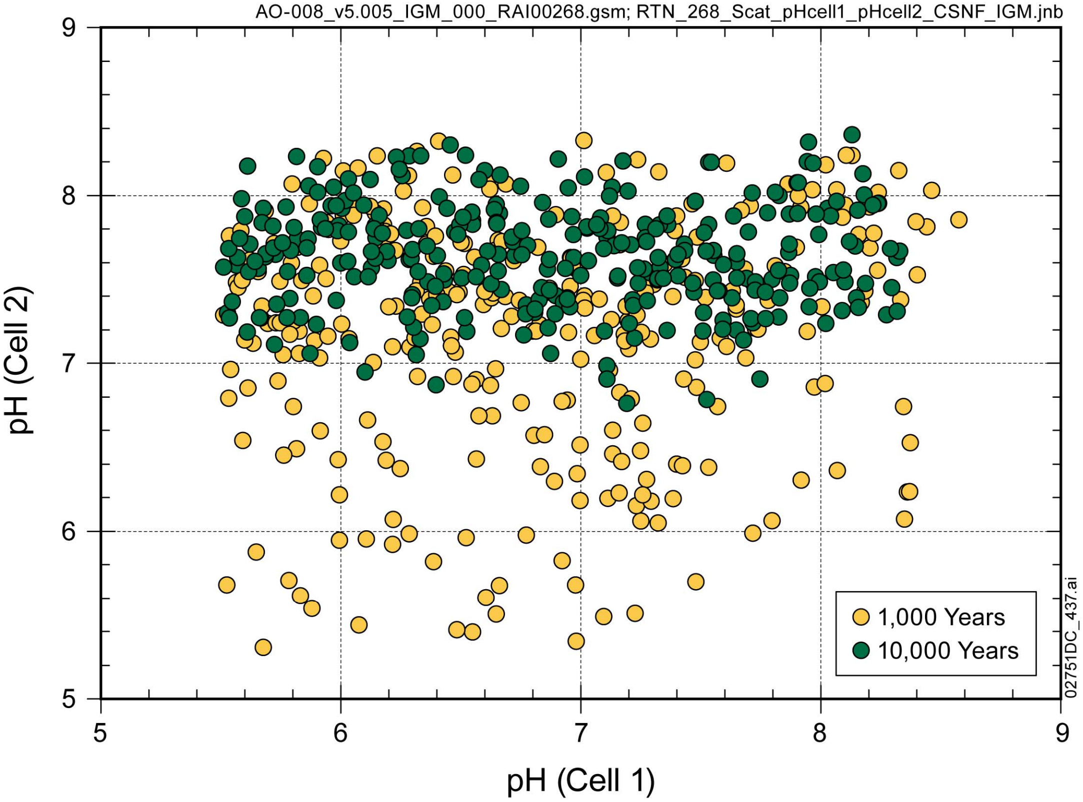


Figure 1. Scatterplot of pH in the Waste Form Domain (cell 1) and the Corrosion Product Domain (cell 2) for Commercial SNF Waste Packages in Percolation Bin 3 at 1,000 Years and 10,000 Years Postclosure after an Igneous Intrusion at 250 Years

A summary of the sorption onto corrosion products model validation is presented in SAR Section 2.3.7 on pages 2.3.7-83 to 2.3.7-84. The detailed discussion of model support for the corrosion products pH abstraction is developed in Section 6.5.2.4 of *EBS Radionuclide Transport Abstraction* (SNL 2007), which describes the bases of the competitive surface complexation model (C-SCM). This competitive surface complexation model, from which the corrosion products pH is derived, is validated in Section 7.2.3 of that same report (SNL 2007), through a comparison of measured linear equilibrium sorption coefficients (K_ds) from the literature with the K_ds calculated from the C-SCM. These sorption coefficient comparisons are a function of pH.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

SNL (Sandia National Laboratories) 2007. *EBS Radionuclide Transport Abstraction.* ANL-WIS-PA-000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories.
ACC: DOC.20071004.0001.

RAI Volume 3, Chapter 2.2.1.3.4, First Set, Number 8, Supplemental Question 2:

> Please clarify whether, under flowing conditions, the pH abstraction for the invert domain assumes mixing between waste package effluent and seepage water as implied by the first paragraph in Section 1.3. The second paragraph suggests that mixing is not simulated but that invert water chemistry is the same as the waste package effluent under flowing conditions. Also, please indicate where in the LA and supporting materials the abstraction discussed in the second paragraph of Section 1.3, is described.

1. SUPPLEMENTAL RESPONSE

There are no conditions under which the pH abstraction for the invert domain assumes mixing between the waste package effluent and seepage water in the total system performance assessment (TSPA) Model. The description in the first paragraph of Section 1.3 of the original response to this RAI is simply to indicate that mixing or dilution of the waste package effluent with diverted seepage water is a likely physical process. However, a simplifying assumption is made that the waste package effluent chemistry will dominate the radionuclide transport pathway through the invert, partly because of the low buffering capacity of the invert rock.

The use of waste package effluent chemistry in the invert, under conditions of advective flow through the waste package, is reflected in the TSPA GoldSim model file (see model selector *pH_Invert_Inter*) and in Table 6.3.4-4 of *Total System Performance Assessment Model /Analysis for the License Application* (SNL 2008, Row 6). Table 6.3.4-4 (SNL 2008), which is based on Table 6.15-1 of *Engineered Barrier System: Physical and Chemical Environment* (SNL 2007a), states that in-package chemistry should be used to represent advective transport in the invert for conditions in which the drip shield and waste package have both failed. This is not an abstraction but an assumption that is part of the overall EBS chemical environment submodel in the TSPA model. This implementation is reflected in the SAR in the second bullet on page 2.3.5-143 and also intimated in portions of SAR 2.3.7.12. Also, in the context of Table 6.3.4-4 “in-package chemistry” means the chemistry that has equilibrated in Cell 2 (also called the corrosion products cell) of the waste package, which is the farthest downstream cell in the waste package; therefore, the chemistry of Cell 2 has a pH computed from the competitive surface complexation model (SNL 2007b, Section 6.5.2.4). When there is no advective flow through the waste package, the invert chemistry is the seepage composition equilibrated to the invert temperature, invert relative humidity, and P_{CO2}, as documented in Section 6.3.4 of *Total System Performance Assessment Model /Analysis for the License Application* (SNL 2008).

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

SNL (Sandia National Laboratories) 2007a. *Engineered Barrier System: Physical and Chemical Environment.* ANL-EBS-MD-000033 REV 06. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070907.0003.

SNL 2007b. *EBS Radionuclide Transport Abstraction.* ANL-WIS-PA-000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071004.0001.

SNL 2008. *Total System Performance Assessment Model /Analysis for the License Application.* MDL-WIS-PA-000005 REV 00 AD 01. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20080312.0001; LLR.20080414.0037; LLR.20080507.0002; LLR.20080522.0113; DOC.20080724.0005.

RAI Volume 3, Chapter 2.2.1.3.4, First Set, Numbers 9 and 10, Supplemental Question 6

> According to the response to RAI #9, the adjusted Eh model was fit to plutonium experimental data from Rai 1984, that were obtained below pH 5, yet the model is applied at pH up to about 9. In the response to RAI #10, it is stated that "the modeled plutonium solubilities using Equation 3 match experimental results very well for the range of pH of interest, including pH > 4.25." Clarify how this statement is consistent with the fact that, in Figure 2 of the RAI #9 response, most experimental data points lie well above the model curve.

1. SUPPLEMENTAL RESPONSE

The purpose of the quoted sentence from the response to RAI 3.2.2.1.3.4-010 was to further explain the anomalously low Eh measurements at pH values greater than 4.25, shown in Figure 6 of the article by Chen and Pearson (2008). For clarity, the sentence in question from the response to RAI 3.2.2.1.3.4-010 is expanded as follows:

> The explanation about poor poising of the Rai 1984 experiment is supported by the fact that using the measured Eh values for pH > 4.25 from the five sets of experimental data (shown in Figure 2 of RAI 3.2.2.1.3.4-009) would have produced modeled plutonium solubilities much lower than the measured plutonium solubilities from the same experiments. Thus, the adjusted-Eh equation (shown as Equation 3 of RAI 3.2.2.1.3.4-010) produces modeled plutonium solubilities closer to the experimentally measured solubilities (data shown in Figure 2 of RAI 3.2.2.1.3.4-009).

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

Chen, Y. and Pearson, F.J. 2008. "Modeling Plutonium Solubility for Yucca Mountain Performance Assessment." *Radiochimica Acta, 96,* pp. 521-526. München, Germany: Oldenbourg Wissenschaftsverlag.

Rai D. 1984. "Solubility Product of Pu(IV) Hydrous Oxide and Equilibrium Constants of Pu (IV)/Pu (V), Pu (IV)/Pu (VI), and Pu (V)/Pu (VI) Couples." *Radiochimica Acta*, 35, 97-106. München, Germany: R. Oldenbourg Verlag.

RAI Volume 3, Chapter 2.2.1.3.4, First Set, Number 10, Supplemental Question 1:

> The response states that "experiments used to support the adjusted Eh model were conducted under open-to-air conditions" (page 5 of response) and that the conditions for the experiment by Nitsche et al. (1993, 1994) and Efurd et al. (1998) "were representative of open-to-air conditions" (page 4). Explain how these statements are consistent with the fact that the Nitsche et al. and Efurd et al. experiments were conducted under oxygen free, Argon - CO_2 gas conditions.

1. SUPPLEMENTAL RESPONSE

The quoted sentence from the original response to RAI 3.2.2.1.3.4-010 (Section 1.4, p. 5) is expanded to, "experiments used to support the adjusted Eh model were conducted under *conditions similar to or representative of* open-to-air conditions." To clarify, the original RAI response states that the plutonium redox conditions in experiments reported by Nitsche et al. (1993, 1994) and Efurd et al. (1998) are similar to those conducted by Rai et al. (2001) and Rai (1984) because Pu(V) is found to be the dominant species in these experiments, indicating a similar redox state and a similar solubility. Thus, these experiments lend support to the primary intent of the adjusted-Eh solubility model, which is a conservative estimate of plutonium solubility under repository conditions.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

Efurd, D.W.; Runde, W.; Banar, J.C.; Janecky, D.R.; Kaszuba, J.P.; Palmer, P.D.; Roensch, F.R.; and Tait, C.D. 1998. "Neptunium and Plutonium Solubilities in a Yucca Mountain Groundwater." Environmental Science & Technology, 32, (24), 3893-3900. Easton, Pennsylvania: American Chemical Society.

Nitsche, H.; Gatti, R.C.; Standifer, E.M.; Lee, S.C.; Müller, A.; Prussin, T.; Deinhammer, R.S.; Maurer, H.; Becraft, K.; Leung, S.; and Carpenter, S.A. 1993. *Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region.* LA-12562-MS. Los Alamos, New Mexico: Los Alamos National Laboratory. ACC: NNA.19930507.0136.

Nitsche, H.; Roberts, K.; Prussin, T.; Muller, A.; Becraft, K.; Keeney, D.; Carpenter, S.A.; and Gatti, R.C. 1994. *Measured Solubilities and Speciations from Oversaturation Experiments of Neptunium, Plutonium, and Americium in UE-25P #1 Well Water from the Yucca Mountain Region Milestone Report 3329-WBS1.2.3.4.1.3.1.* LA-12563-MS. Los Alamos, New Mexico: Los Alamos National Laboratory.

Rai, D. 1984. "Solubility Product of Pu(IV) Hydrous Oxide and Equilibrium Constants of Pu (IV)/Pu (V), Pu (IV)/Pu (VI), and Pu (V)/Pu (VI) Couples." *Radiochimica Acta, 35*, 97-106. München, Germany: R. Oldenbourg Verlag.

Rai, D.; Moore, D.A.; Felmy, A.R.; Choppin, G.R.; and Moore, R.C. 2001. "Thermodynamics of the PuO_2^+-Na^+-OH^--Cl^--ClO_4^--H_2O System: Use of NpO_2^+ Pitzer Parameters for PuO_2^+." *Radiochimica Acta, 89*, ([8]), 491-498. München, Germany: Oldenbourg Wissenschaftsverlag.

RAI Volume 3, Chapter 2.2.1.3.4, First Set, Number 10, Supplemental Question 2:

> Please clarify the last paragraph, Section 1.3, p. 5, which asserts that the redox conditions in Yucca Mountain groundwater do not affect their applicability as corroborative data. In particular, the 3rd sentence is not clear. Clarify why you conclude that these data corroborate the adjusted Eh model when the water geochemical conditions appear to be quite different from the modeled conditions.

1. SUPPLEMENTAL RESPONSE

The last paragraph of Section 1.3 of the response to RAI 3.2.2.1.3.4-010 is stating that the adjusted-Eh solubility model is shown to be bounding irrespective of the miniature waste package data and the Yucca Mountain Project groundwater measurements. This is because the adjusted-Eh solubility model corresponds to Eh conditions greater than the empirical upper boundary Eh model for natural waters in contact with the atmosphere (SNL 2007, Figure V-6). However, independent of this conclusion, the redox conditions of the Yucca Mountain Project groundwater measurements and the miniature waste package experiments (discussed in SNL 2007, Sections V.3 and V.4, respectively) show that the adjusted-Eh equation is conservative when applied to in-package conditions with plutonium present. Figure V-6 of *Dissolved Concentration Limits of Elements with Radioactive Isotopes* (SNL 2007) shows that all the Eh–pH measurements made at Yucca Mountain are below the empirical boundary for Eh-pH conditions in natural waters, which is in turn below the Eh-pH boundary computed with the adjusted-Eh model. Furthermore, experiments with miniature waste packages support the hypothesis that a reducing environment would form within the waste package, which would produce much lower Eh conditions and therefore much lower plutonium solubilities inside the waste packages. Thus, the geochemical conditions found in the Yucca Mountain groundwater and the miniature waste packages support the conservative application of the adjusted-Eh equation to the plutonium solubility model.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

SNL (Sandia National Laboratories) 2007. *Dissolved Concentration Limits of Elements with Radioactive Isotopes*. ANL-WIS-MD-000010 REV 06. Las Vegas, Nevada: Sandia National Laboratory. ACC: DOC.20070918.0010.

RAI Volume 3, Chapter 2.2.1.3.4, Second Set, Number 3, Supplemental Question:

> Clarify why disruptive modeling cases (high dose) are chosen over the nominal case (lower dose) for elaborating processes that limit colloid release and transport in the EBS. Clarify which processes controlling the aqueous chemical characteristics governing colloid stability discussed for the disruptive cases are also applicable to the nominal case and whether iron oxide and/or SNF colloids are typically unstable in the corrosion products domain also for the nominal case.

1. SUPPLEMENTAL RESPONSE

1.1 DISRUPTIVE CASES CHOSEN VERSUS NOMINAL CASE

Disruptive modeling cases (high dose) were chosen over the nominal case (lower dose) for elaborating processes that limit colloid release and transport in the Engineered Barrier System (EBS) because the disruptive cases are the most risk-significant cases and because the separate nominal modeling case is not included in the post-10,000-year dose calculations used to assess compliance with the dose limits in 10 CFR 63.311 (see SAR Section 2.4.2.1, Equation 2.4-21). Rather, the post-10,000-year seismic ground motion modeling case includes nominal corrosion processes and the dose arising from them.

1.2 PROCESSES THAT LIMIT COLLOID RELEASE AND TRANSPORT

The same processes that limit colloid release and transport in the EBS for the seismic ground motion and igneous intrusion modeling cases, discussed in the response to this RAI, also apply to the nominal modeling case. These processes include the colloid suspension stability and the low diffusion rate of colloids compared to dissolved species. With respect to these two processes, the nominal case at early times is similar to the seismic ground motion modeling case. In both cases the early releases are by diffusion through cracks, a situation that results in high ionic strengths, low colloid suspension stabilities, and low diffusive colloid releases. After general corrosion of the waste package allows advection through open patches in the waste package, the nominal modeling case has processes similar to the igneous intrusion modeling case. As in the igneous intrusion modeling case, advection results in low ionic strengths, greater colloid suspension stabilities, and advective release of colloids.

1.3 PROCESSES CONTROLLING THE AQUEOUS CHEMICAL CHARACTERISTICS GOVERNING COLLOID SUSPENSION STABILITY

The controlling chemical characteristics governing colloid suspension stability are ionic strength and pH. Any process that affects these characteristics will affect colloid suspension stability. The processes that influence ionic strength include: (1) water flux through the waste package, and (2) processes affecting the relative humidity within the package such as rubble fill of the drift in the seismic ground motion modeling case. The processes that influence pH include: (1) water flux through the waste package, (2) degradation rate of the waste forms and waste package materials, (3) buffering capacity of the waste form and waste package degradation products, and (4) influx of CO_2. These processes occur in most modeling cases, but they occur at different times and to differing extents.

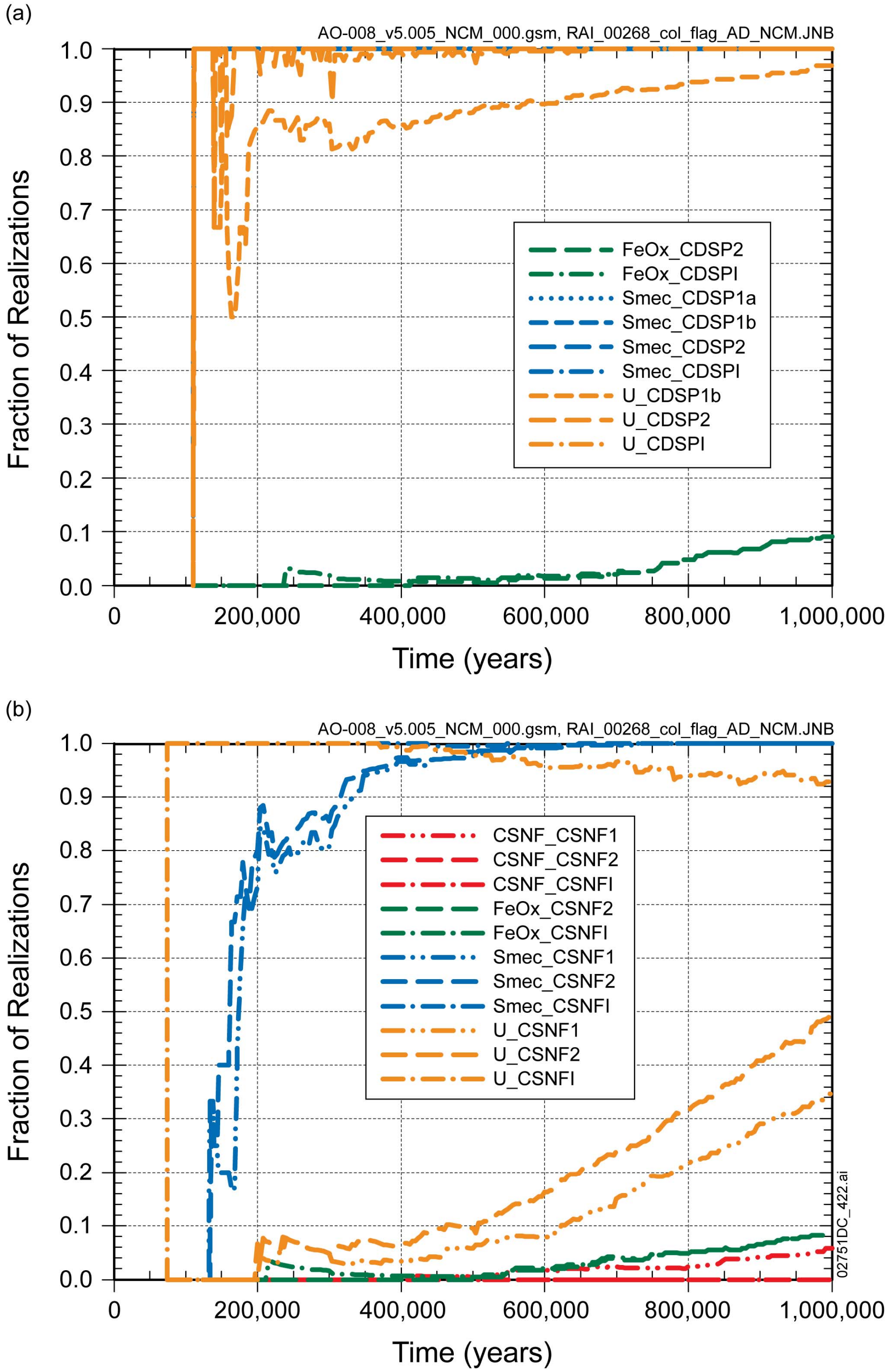
The nominal modeling case is most like the seismic ground motion modeling case, because both cases have nominal general corrosion rates of the waste packages which result in no water flow through the package until corrosion patches open. This is one of the reasons why colloid suspension stability plots 27 through 30 were provided for the seismic ground motion modeling case in the response to this RAI at both 300,000 years (which is before patch failures which are due to nominal processes) and at 1,000,000 years (which is subsequent to patch failures).

Like the seismic ground motion modeling case, in the nominal modeling case commercial spent nuclear fuel (SNF) colloid suspensions are unstable in the corrosion products domain, and only those realizations with advective flow have stable FeO_x colloid suspensions in the corrosion products domain. Thus, the green and red lines in Figure 1 of this supplemental response (nominal) are similar to those in Figure 15 of the initial response to this RAI (seismic). (see Section 1.3 of the initial response to this RAI for an explanation of the figure legends.)

However, there are minor differences between the nominal and seismic ground motion modeling cases related to the behavior of smectite and uranium-mineral colloids, resulting from drift collapse in the seismic ground motion modeling case, which causes a change in relative humidity in the drift. This produces a difference in the evolution of ionic strength in the corrosion products domain in the seismic case compared to the nominal case, which can be seen by comparing Figure 2 (nominal) of this supplemental response to Figure 18b (seismic) of the initial response to this RAI. The drift collapse occurs between 300,000 and 400,000 years in the seismic ground motion modeling case, which causes a drop in ionic strength during that time (Figure 18b). This drop in ionic strength is absent in the nominal modeling case (Figure 2), which means a significant difference in the fraction of realizations with stable uranium-mineral colloid suspensions within the commercial SNF packages in the nominal case compared to the seismic case. The much lower fraction in the nominal case can be seen by comparing Figure 1b (nominal) to Figure 15b (seismic) of the original RAI response.

1.4 CONCLUSION

Disruptive modeling cases are chosen over the nominal modeling case for elaborating processes that limit colloid release and transport in the EBS because the disruptive cases are the most risk-significant cases and because the separate nominal modeling case is not part of the post-10,000-year total dose calculations presented in the SAR (Section 2.4). Except for rubble fill of the drift, the processes controlling the aqueous chemical characteristics governing colloid stability discussed for the disruptive cases are also applicable to the nominal modeling case. Iron oxide and commercial SNF waste form colloid suspensions would typically be unstable in both the corrosion products domain for the nominal modeling case and the seismic ground motion modeling case that includes nominal corrosion processes.



NOTE: Some blue lines are obscured by orange lines in both plots.

Figure 1. Fraction of Realizations with Breached (a) Codisposal Waste Packages and (b) Commercial SNF Waste Packages that have Stable Colloid Suspensions, in Percolation Subregion 3, Drip Environment, for an Epistemic Sample of Size 300 for the Nominal Modeling Case for 1,000,000 Years

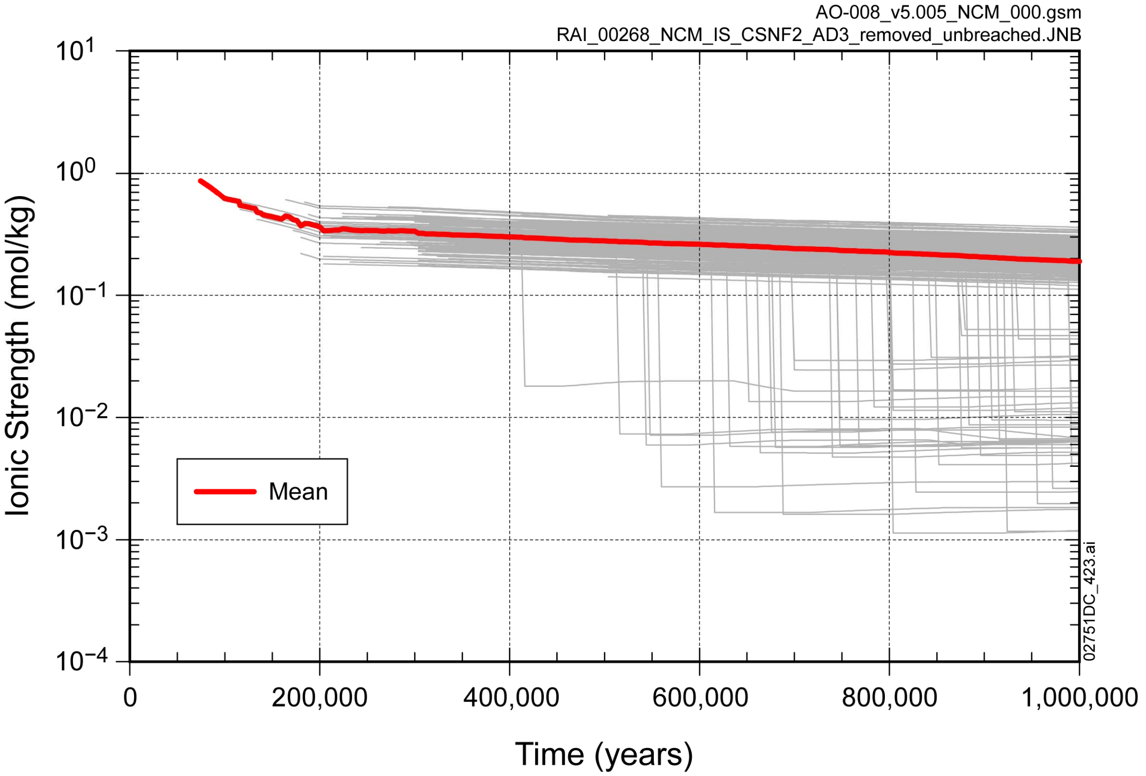


Figure 2. Ionic Strength in the Corrosion Products Domain of Commercial SNF Waste Packages, in Percolation Subregion 3, Drip Environment, for an Epistemic Sample of Size 300, for 1,000,000 Years, Nominal Modeling Case

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

RAI Volume 3, Chapter 2.2.1.3.4, Second Set, Number 7, Supplemental Question:

> In the response to RAI 3.2.2.1.3.4-2-007, Figure 1.1-26 shows that, for realization 2855, the vast majority of ^{242}Pu mass in the corrosion products domain is associated with settled CSNF waste form colloids. Clarify whether the result from this realization is representative of the plutonium behavior in the corrosion products domain.

1. SUPPLEMENTAL RESPONSE

The behavior shown in Figure 1.1-26 of the response to RAI 3.2.2.1.3.4-2-007 is observed in approximately 38% of realizations of the igneous intrusion modeling case in the TSPA-LA model. This can be seen in Figure 1b of the response to RAI 3.2.2.2.2.4-2-003, which shows that commercial spent nuclear fuel (SNF) waste form colloids are stable in the waste form domain (curve CSNF_CSNF1 in the figure) in about 38% of realizations (epistemic sample elements); but are not stable in the corrosion products domain (curve CSNF_CSNF2 in the figure) in any realization. The change in stability between domains results from an increase and narrowing of the pH range, caused by the buffering capability of the iron-oxyhydroxide corrosion products, as is shown in Figure 6 of the response to RAI 3.2.2.2.2.4-2-003.

Because of the threshold behavior of colloid stability in the chemical environment of the waste package and the associated uncertainty in the relevant environmental parameters, a wide range of behavior is observed in the plutonium budget from commercial SNF waste forms in the corrosion products domain. The range is defined by two extremes: (1) realizations similar to 2855, in which a significant mass of plutonium is released as irreversibly attached to waste form colloids from the waste form domain, and then flocculates (settles) in the corrosion products domain, and (2) realizations where waste form colloids are unstable in the waste form domain, in which plutonium is released mainly in the dissolved state from the waste form domain, and then sorbs onto stationary corrosion products. The comparison between plutonium mass sorbed onto stationary corrosion products and mass irreversibly attached to settled waste form colloids is illustrated in Figure 1, which shows the difference between these quantities for all 300 epistemic sample elements.

Figure 1 shows that the plutonium mass difference in realization 2855 (epistemic uncertainty vector 286) is similar to the 5th percentile plutonium mass difference for the set of 300 epistemic uncertainty vectors, indicating that realization 2855 is near one extreme of the outcomes. However, as indicated by the mean (red) curve in Figure 1, the mean of the differences is approximately zero over the entire simulation time of 1,000,000 years. This indicates that, on average over the entire range of epistemic uncertainty, the plutonium mass settled as waste form colloids is approximately equal to the plutonium mass sorbed onto stationary corrosion products.

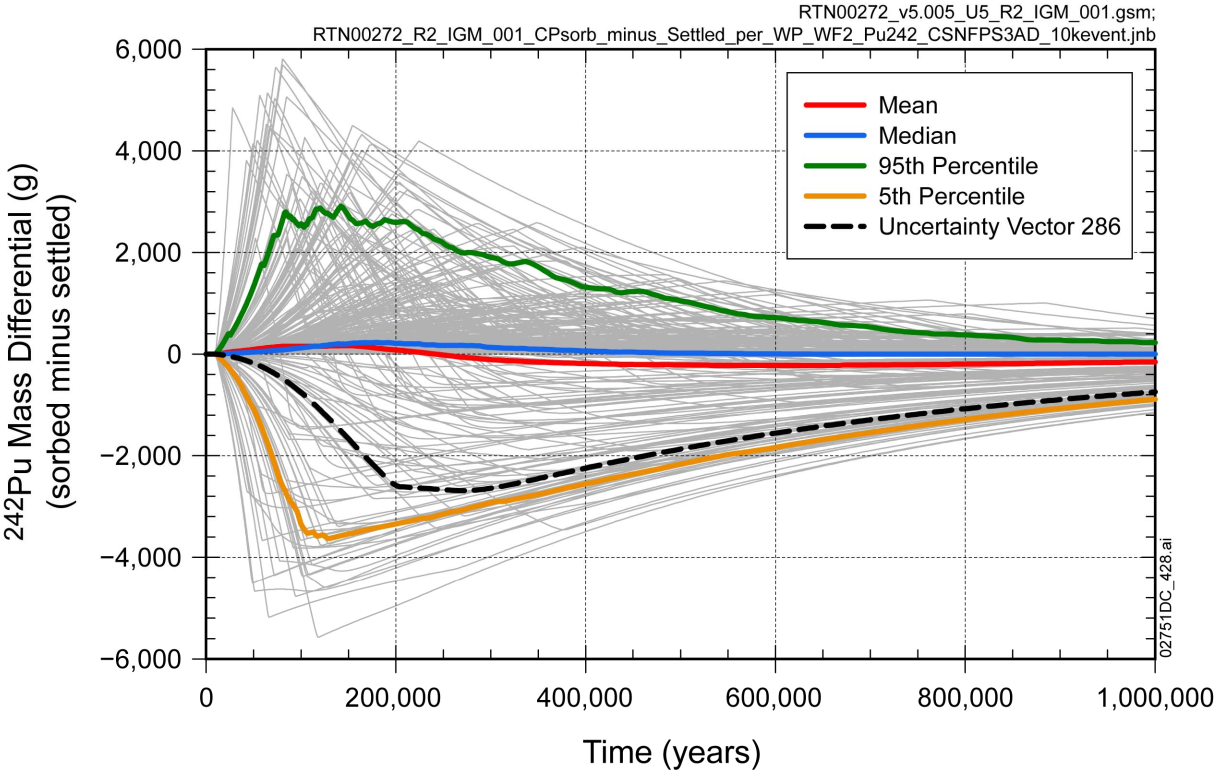


Figure 1. Difference in Mass of ^{242}Pu in the Corrosion Products Domain (mass sorbed onto stationary corrosion products minus mass irreversibly attached to settled waste form colloids) of a Commercial SNF Waste Package in a Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion at 10,000 Years after Repository Closure, for the 300 Epistemic Uncertainty Vectors

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

RAI Volume 3, Chapter 2.2.1.3.4, Second Set, Numbers 8 and 9, Supplemental Question:

> These 2 responses indicate that, effectively, equilibrium conditions apply for plutonium sorption on stationary corrosion products which implies that plutonium release should be independent of the desorption rate used in the kinetic model. Clarify how this is consistent with information in the SAR pages 2.1-72 and 2.1-76, and in previous RAI responses, that suggests that release of plutonium from the EBS is controlled by rates of desorption from stationary corrosion products.

1. SUPPLEMENTAL RESPONSE

Because a kinetic model, rather than an equilibrium sorption coefficient (K_d) model, is implemented for sorption/desorption of plutonium and americium on stationary corrosion products, the rate of release of plutonium from the corrosion products is always dependent on the desorption rate, which is consistent with that presented by the SAR Section 2.1 (pp. 2.1-72 and 2.1-76).

In responses to RAIs 3.2.2.1.3.4-2-009 and 3.2.2.1.3.4-2-008, the dimensionless ratio of the rate of reaction to the rate of transport, or Damköhler number, was used to demonstrate that local equilibrium conditions are approximated for plutonium sorption onto stationary corrosion products. Despite the fact that the high Damköhler numbers indicate conditions approximating local equilibrium, or even if an equilibrium K_d model had been used instead of a kinetic model, the relatively high K_d of plutonium results in a plutonium release rate (i.e., the movement of the reaction wave front or desorption wave front) that manifests as a relatively slow depletion of plutonium from the waste package, as implied on SAR page 2.1-72 (Section 2.1). In other words, because the Damköhler numbers indicate local equilibrium, the calculated K_d is more important than the forward rate or backward rate constants in controlling plutonium releases from the EBS.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.