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YUCCA MOUNTAIN - REQUEST FOR ADDITIONAL INFORMATION- SAFETY EVALUATION REPORT, VOLUME 3—POSTCLOSURE CHAPTER 2.2.1.3.4—RADIONUCLIDE RELEASE RATES AND SOLUBILITY LIMITS, 3RD SET - (DEPARTMENT OF ENERGY'S SAFETY ANALYSIS REPORT SECTION 2.3.7)

Reference: Ltr, Cuadrado to Williams, dtd 8/26/2009, "Yucca Mountain – Request for Additional Information – Safety Evaluation Report, Volume 3 – Postclosure Chapter 2.2.1.3.4 – Radionuclide Release Rates and Solubility Limits, 3rd Set – (Department of Energy's Safety Analysis Report Section 2.3.7)"

The purpose of this letter is to transmit the U.S. Department of Energy's (DOE) responses to the Requests for Additional Information (RAIs) identified in the above-referenced letter. Each RAI response is provided as a separate enclosure to this letter (Enclosures 1 through 3). An optical storage media (OSM), provided as Enclosure 5, contains the results from the PHREEQC surface complexation model and all files in Data Tracking Number (DTN) SN0703PAEBSRTA.002, Rev 001, as requested by NRC in RAI Number 3.

A listing of the electronic files included on OSM is provided as Enclosure 4. The data files and computer model output files are required by NRC staff to evaluate DOE's responses and are not intended to be accessed through ADAMS. These electronic files will be made available to the public upon request.

The DOE references cited in the RAI responses have previously been provided with the License Application (LA) or as part of previous RAI responses. The references provided with previous RAI responses are identified within the enclosed responses' reference section.

There are no commitments in the enclosed RAI response. If you have any questions regarding this letter, please contact me at (202) 586-9620, or by email to jeff.williams@rw.doe.gov.

Jeffrey R. Williams, Supervisor
Licensing Interactions Branch
Regulatory Affairs Division
Office of Technical Management

OTM: CJM-1054

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Enclosures (5):

1. Response to RAI Volume 3, Chapter 2.2.1.3.4, Set 3, Number 1
2. Response to RAI Volume 3, Chapter 2.2.1.3.4, Set 3, Number 2
3. Response to RAI Volume 3, Chapter 2.2.1.3.4, Set 3, Number 3
4. Listing of files included on DVD provided as Enclosure 5
5. Optical Storage Media – DVD containing data requested in RAI Volume 3, Chapter 2.2.1.3.4, Set 3, Number 3; DTN: SN 0703PAEBSRTA.002, Rev 001.

cc w/Enclosures 1 through 4:
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RAI Volume 3, Chapter 2.2.1.3.4, Third Set, Number 1:

Describe and provide quantitative information on ^{242}Pu mass in and fluxes between the various EBS domains for commercial SNF packages (analogous to Figures 1.1-21 through 1.1-27 in the response to RAI 3.2.2.1.3.4-2-007) in a realization representative of the igneous intrusion case, in which Pu-rich zirconium oxide colloids are unstable in the waste form domain and iron-oxide colloids are stable in the corrosion products domain. The response should include information on partitioning among various immobile and mobile states (dissolved, precipitated, sorbed to mobile and settled colloids, sorbed to stationary corrosion products).

Basis: Fig. 1.1-26 in DOE's response to RAI: 3.2.2.1.3.4-2-007 shows that for a particular realization, ^{242}Pu mass is mostly associated with the settled SNF colloids. In this particular realization, stationary corrosion products do not sequester a significant fraction of ^{242}Pu and suspended iron-oxide colloids are not effective at transporting ^{242}Pu. According to the applicant, this type of scenario occurs in 38% of realizations (Supplemental Response to 3.2.2.1.3.4-2-007). Clarification is needed regarding the Pu release behavior and mass distribution on stationary and colloidal corrosion products for the 62% of realizations in which SNF waste form colloids are unstable in the waste form domain and, thus, ineffective at releasing Pu to the corrosion products domain.

1. RESPONSE

This response provides a general description of the movement and retention of ^{242}Pu mass in the Engineered Barrier System (EBS), supported by plots of time histories of ^{242}Pu mass flux and mass allocation in the EBS domains (waste form domain, corrosion products domain, and invert) for commercial spent nuclear fuel (SNF) waste packages. The information is provided for a single realization of the igneous intrusion modeling case where plutonium-rich zirconium oxide (waste form) colloid suspensions are unstable in the waste form domain and iron oxyhydroxide (corrosion product) colloid suspensions are stable in the corrosion products domain. The representative realization selected for this response is #525, which uses epistemic vector #53.

The information provided in this response is analogous to the information provided for epistemic vector #286 (realization #2855) in Section 1.1.3 and Figures 1.1-21 to 1.1-27 of the response to RAI 3.2.2.1.3.4-2-007, but realization #2855 represents conditions where plutonium-rich zirconium oxide colloid suspensions are stable in the waste form domain and become unstable and settle in the corrosion products domain As indicated in the first supplemental response to RAI 3.2.2.1.3.4-2-007, colloid stability in epistemic vector #53 is typical for the igneous intrusion modeling case for 62% of the epistemic sample elements. This single realization has an igneous intrusion event at 10,000 years and has a calculated late time plutonium solubility value of 0.156 mg/l in the waste form domain, which is about 30 times the value (0.0053 mg/l) in epistemic vector #286.

Figure 1 in the first supplemental response to RAI 3.2.2.1.3.4-2-007 is a time history plot showing the difference in ^{242}Pu mass that is sorbed onto corrosion products and ^{242}Pu mass that is settled in colloid suspensions in the corrosion products domain for all 300 epistemic uncertainty vectors, with epistemic vector #286 identified. Figure 1 of this response presents the same information with epistemic vector #53 identified.

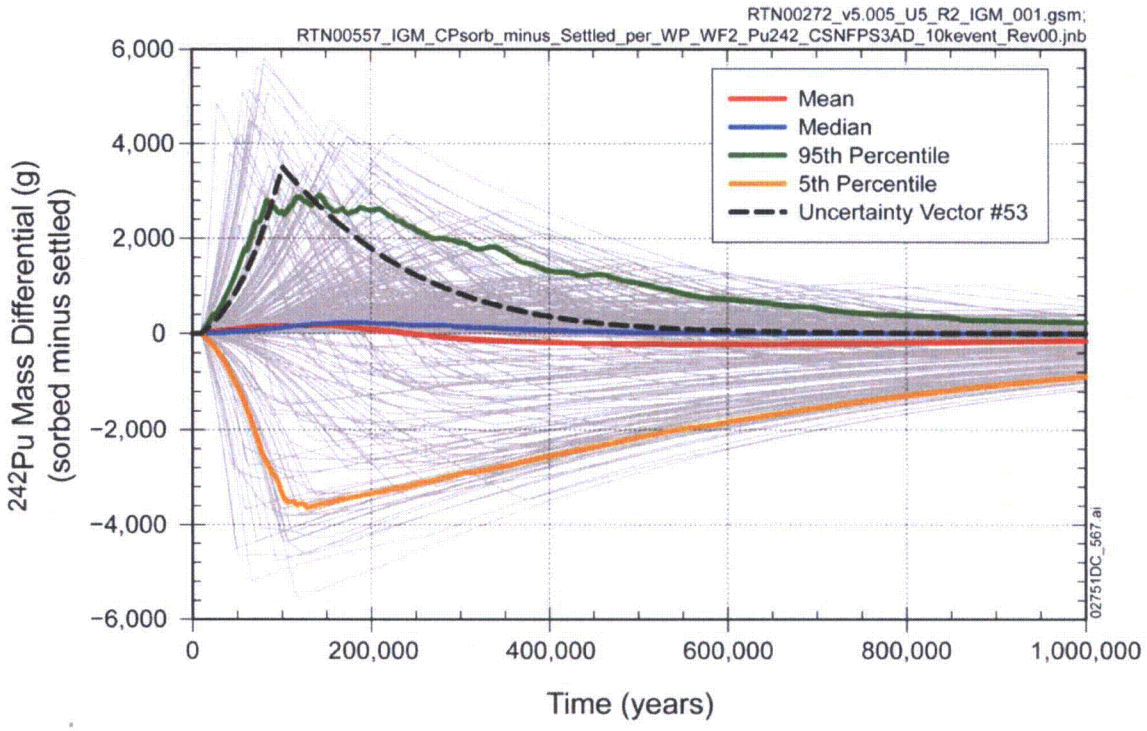


Figure 1. Difference in Mass of ^{242}Pu in the Corrosion Products Domain (mass sorbed onto stationary corrosion products minus mass irreversibly attached to settled waste form colloids) of a Commercial SNF Waste Package in a Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion at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

1.1 AVAILABLE MASS COMPARISION

In the response to RAI 3.2.2.1.3.4-2-007, time histories of plutonium mass flux between the EBS domains and plutonium mass allocation in the EBS domains are provided for epistemic vector #286 (realization #2855) of the igneous intrusion modeling case. The results are presented for commercial SNF waste packages in the seeping environment of percolation subregion 3 following an igneous intrusion event that occurs at 10,000 years. In realization #2855 the sampled parameters produce a seepage fraction of approximately 0.72 and an inventory uncertainty value of 1.11, so that there are 2,369 waste packages in the seeping environment of percolation subregion 3 with a total initial ^{242}Pu inventory of 1.44×10^7 grams.

In this response, time histories of plutonium mass flux between the EBS domains and plutonium mass allocation in the EBS domains are provided for epistemic vector #53 of the igneous

intrusion modeling case with a 10,000 year igneous intrusion event time. Similar to the response to RAI 3.2.2.1.3.4-2-007, the results for this response are presented for commercial SNF waste packages in the seeping environment of percolation subregion 3. As noted in Section 1.1 of the response to RAI 3.2.2.1.3.4-2-007, locations in percolation subregion 3 contain 40% of the waste packages and are associated with the middle 40% of the expected range of percolation values. Due to differences in the sampled values of the infiltration case and drift seepage uncertainty parameters (e.g., seepage uncertainty, capillary strength, and permeability) between epistemic vectors, the number of commercial SNF waste packages in the seeping environment of percolation subregion 3 for epistemic vectors #53 is 2,086. In addition, the sampled value for the inventory uncertainty in epistemic vectors #53 is 0.922, compared to the value of 1.11 in epistemic vector #286. As a result, the initial amount of plutonium in commercial SNF waste packages in the seeping environment of percolation subregion 3 is approximately 1.05×10^7 grams, which is 73% of the value in epistemic vector #286.

1.2 RELEASE RATE AND MASS DISTRIBUTION COMPARISONS

The figures and discussion in this section are comparable to Figures 1.1-21 to 1.1-27, and the associated discussion, in the response to RAI 3.2.2.1.3.4-2-007.

1.2.1 Waste Form Domain

In epistemic vector #53 of the igneous intrusion modeling case with a 10,000-year event time, the environmental conditions inside the failed commercial SNF waste packages result in unstable plutonium-rich zirconium oxide colloid suspensions in the waste form domain. Therefore, the release rate of total ^{242}Pu from the waste form domain, shown in Figure 2, is nearly equivalent to the release rate for ^{242}Pu that is dissolved or reversibly sorbed onto other colloids, shown in Figure 3, with very little ($<10^{-6}$ g/yr) contribution from plutonium-rich zirconium oxide colloids, shown in Figure 4. There is no release of ^{242}Pu associated with iron oxyhydroxide colloid suspensions from the waste form domain (Figure 5), because iron oxyhydroxide colloids are produced in the corrosion products domain, not in the waste form domain.

The advective release of ^{242}Pu from the waste form domain in epistemic vector #53 occurs over a 100,000-year period. The release rate of ^{242}Pu from the waste form domain is controlled by the solubility of plutonium in the waste form domain and the flow rate through the domain. The advective release rate is relatively high while a ^{242}Pu precipitate is present in the waste form domain, but is reduced sharply once the precipitate is depleted. The rate of both advection and diffusion from the waste form domain increases until ~100,000 years. During the same time period, the aqueous concentration of ^{242}Pu increases because of the increasing solubility, as discussed below. When the ^{242}Pu precipitate is depleted, the concentration of dissolved ^{242}Pu in the waste form domain drops below the concentration in the corrosion products domain and diffusion from the waste form domain to the corrosion products domain reverses. A negligible amount of plutonium on iron oxyhydroxide colloids diffuses from the corrosion products domain into the waste form domain and becomes immobile in the waste form domain.

Figure 6 indicates that most of the ^{242}Pu mass in the waste form domain in the first 100,000 years exists as precipitated mass. Following the igneous intrusion event at 10,000 years, the entire

^{242}Pu inventory from the 2,086 failed commercial SNF waste packages is exposed for potential dissolution in the available water in the waste form domain. However, for the 100,000 years, the dissolved concentration in the waste form domain is constrained by solubility limits, and precipitation occurs. During this time period, the mass that is reversibly sorbed to colloids is also controlled by the solubility.

In the first 100,000 years, the ^{242}Pu release rate from the waste form domain by advection (Figure 3) and the dissolved mass and mass reversibly sorbed on colloids (Figure 6) increase due to an increase in the solubility of plutonium in the waste form domain. The increase in the plutonium solubility is due (directly and indirectly through the pH) to increasing PCO_2. Once the ^{242}Pu precipitate in the waste form domain is redissolved (approximately 100,000 years), the concentration of ^{242}Pu in the waste form domain drops significantly, as does the release rate from the waste form domain. Accordingly, the dissolved mass and mass that is reversibly sorbed to colloids decreases proportionally. After 100,000 years, backwards diffusion from the corrosion products domain into the waste form domain is the source of ^{242}Pu in the waste form domain; however, this mass is readily transported back out of the waste form domain by advection (Figure 2).

1.2.2 Corrosion Products Domain

In epistemic vector #53 of the igneous intrusion modeling case with a 10,000-year event time, the environmental conditions inside the failed commercial SNF waste packages result in stable iron oxyhydroxide colloid suspensions in the corrosion products domain. Despite the favorable conditions for iron oxyhydroxide colloid suspensions, the release rate of ^{242}Pu associated with iron oxyhydroxide colloid suspensions from the corrosion products domain (Figure 5) is lower by two- to three-orders of magnitude than the release rate of dissolved ^{242}Pu from the corrosion products domain (Figure 3).

The advective release rate of ^{242}Pu from the corrosion products domain is initially much less than the advective release rate of ^{242}Pu from the waste form domain (Figure 2, cf. the red and green solid lines). Initially, the difference in the release rates is attributed to ^{242}Pu sorption onto corrosion products (Figure 7). ^{242}Pu that is released from the waste form domain sorbs onto the corrosion products, which reduces the aqueous concentration in the corrosion products domain and reduces the amount of mass that is transported out of the corrosion products domain by advection. Initially, the higher release rate from the waste form domain is maintained by the precipitate in the waste form domain; however, when the mass of precipitate in the waste form domain is depleted at approximately 100,000 years, the release rates from the waste form domain are reduced sharply. After 100,000 years, the release rates from the corrosion products domain decrease only gradually, despite the reduction in release from the waste form domain. Desorption from the corrosion products maintains the concentration in the aqueous phase so that the advective release rate from the corrosion products domain decreases gradually.

As shown in Figure 7, the mass of ^{242}Pu that is sorbed onto the corrosion products after 100,000 years accounts for more than 99.99% of the mass in the corrosion products domain and more than 95% of the mass retained by the EBS (i.e., 95% of the total mass summed over all three EBS domains). The mass of steel in the waste packages is completely converted to

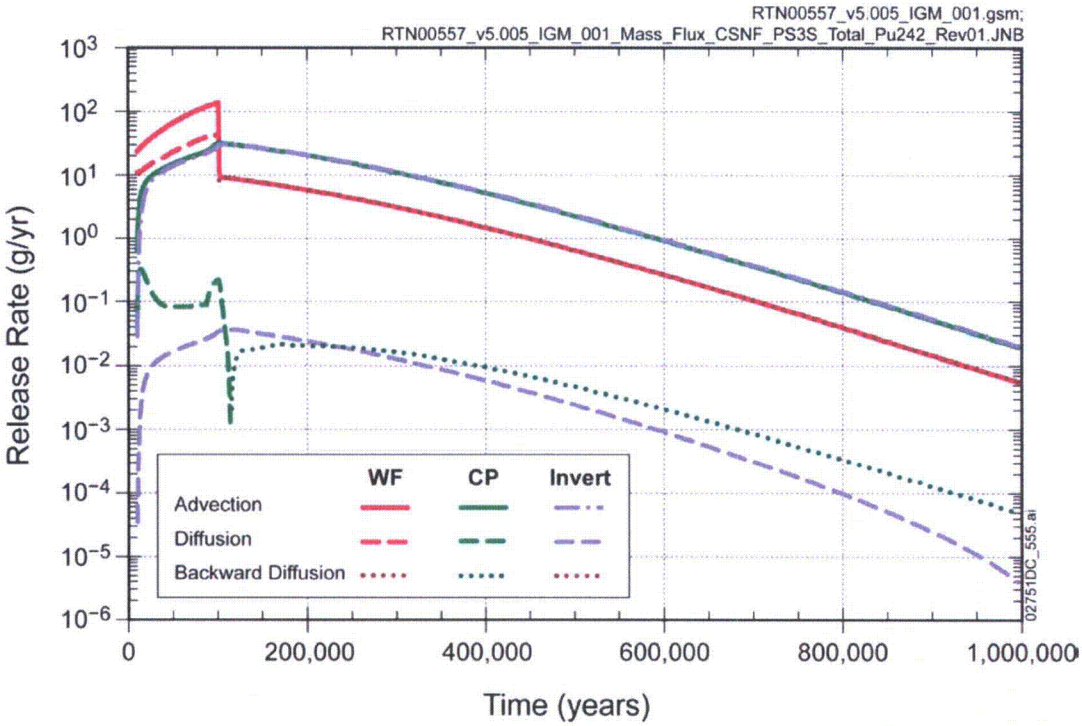
corrosion products by 93,000 years. During this time the dissolved ^{242}Pu mass in the corrosion products domain is kept low because sorption onto the increasing mass of corrosion products removes mass from the aqueous phase. Beyond 100,000 years, when the mass of corrosion products is constant but the upstream source is depleted, there is a continuous decline in both the sorbed and dissolved mass in the corrosion products domain due to slow desorption and radioactive decay. The aqueous concentration of ^{242}Pu in the corrosion products domain is below the solubility limit at all times.

1.2.3 Invert Domain

The release rate of ^{242}Pu from the invert domain by advection is nearly equivalent to the release rate from the corrosion products domain (Figure 2). As was the case in the upstream domains, the release of ^{242}Pu from the invert is predominantly dissolved ^{242}Pu (Figure 8). Similarly to the corrosion products domain, downward diffusion of ^{242}Pu from the invert occurs at a rate that is three orders of magnitude lower than the rate of advection (Figure 3). After 100,000 years, desorption from the corrosion products is a source for ^{242}Pu mass within the EBS and maintains a concentration in the corrosion products domain that is higher than the waste form domain and invert domain, and diffusion from the corrosion products domain into these domains occurs.

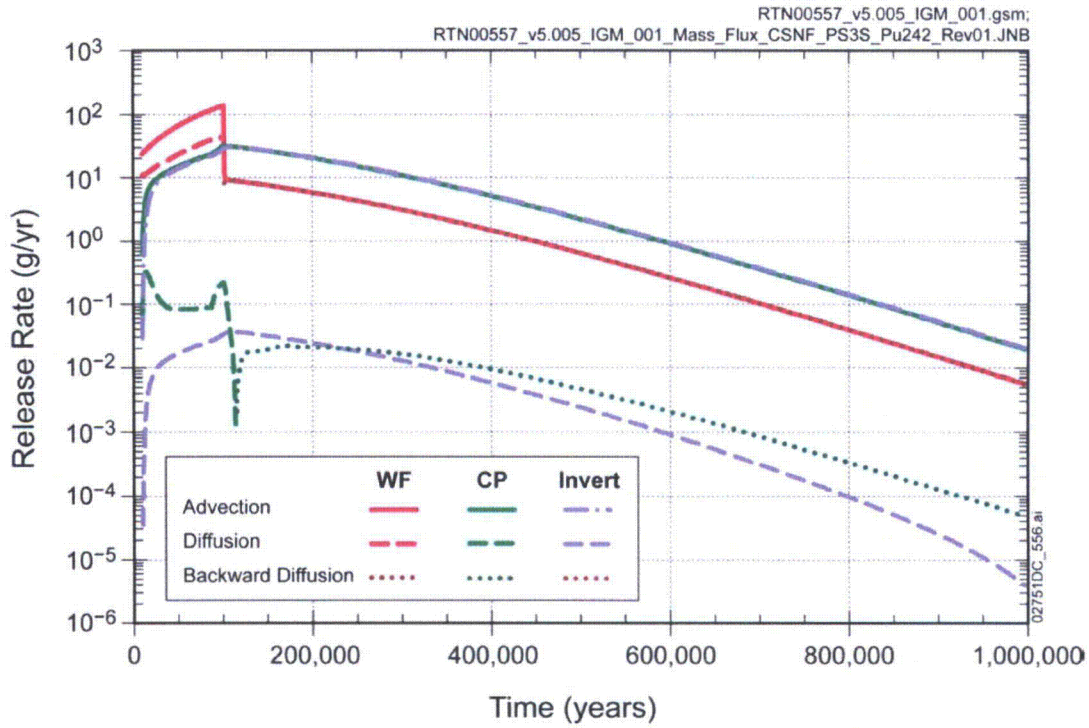
Sorption onto the invert tuff provides some delay in transport between the corrosion products domain and the unsaturated zone beneath the repository. As shown in Figure 8, after 100,000 years, about 99.8% of the ^{242}Pu mass in the invert is sorbed onto the stationary tuff, but this sorbed mass accounts for less than 5% of the ^{242}Pu mass retained in the EBS. Most of the retained ^{242}Pu mass is sorbed onto the stationary corrosion products. A small percentage of the ^{242}Pu mass in the invert is dissolved and even less ^{242}Pu is irreversibly sorbed to stable iron oxyhydroxide colloid suspensions or reversibly sorbed to groundwater and uranium mineral colloids.

In the invert, a very small amount (up to 30 grams distributed over 2,086 waste packages) of ^{242}Pu is associated with settled iron oxyhydroxide colloid suspensions in the first 170,000 years. This is a result of the numerical solution method in the GoldSim implementation, as described in the second supplemental response to RAI 3.2.2.1.3.4-2-007.



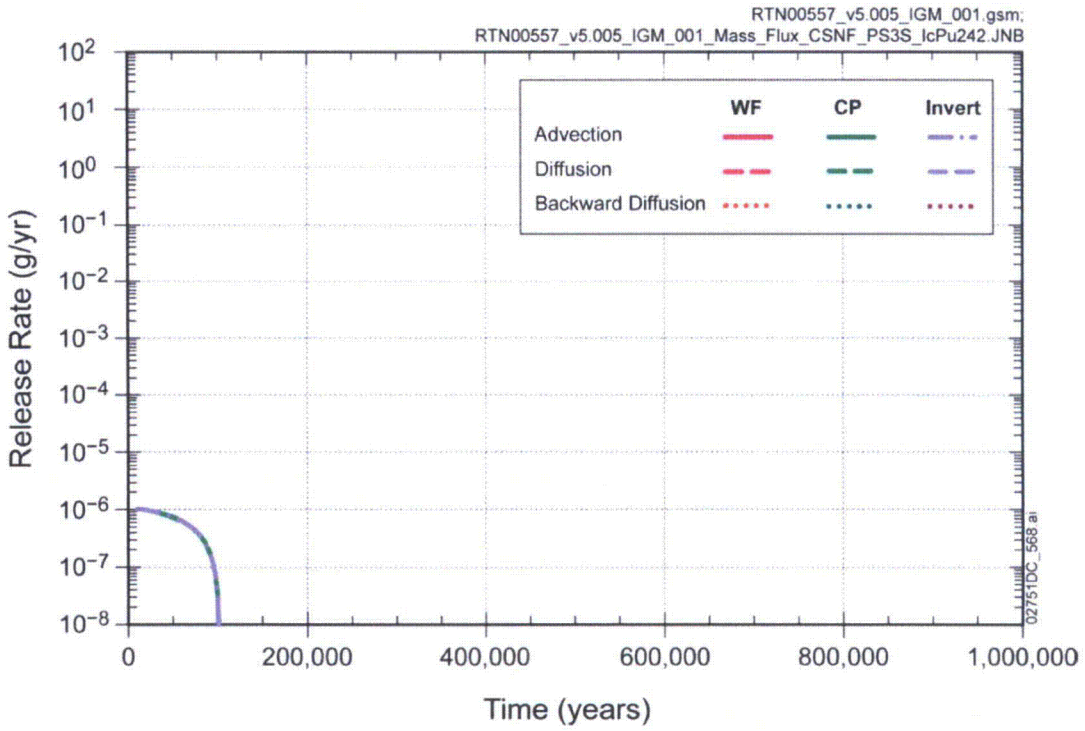
NOTE: Invert advection overlays corrosion product (CP) advection, and waste form (WF) advection overlays WF backward diffusion (after 100,000 years).

Figure 2. Release Rate of Total ^{242}Pu from Each EBS Domain from Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure



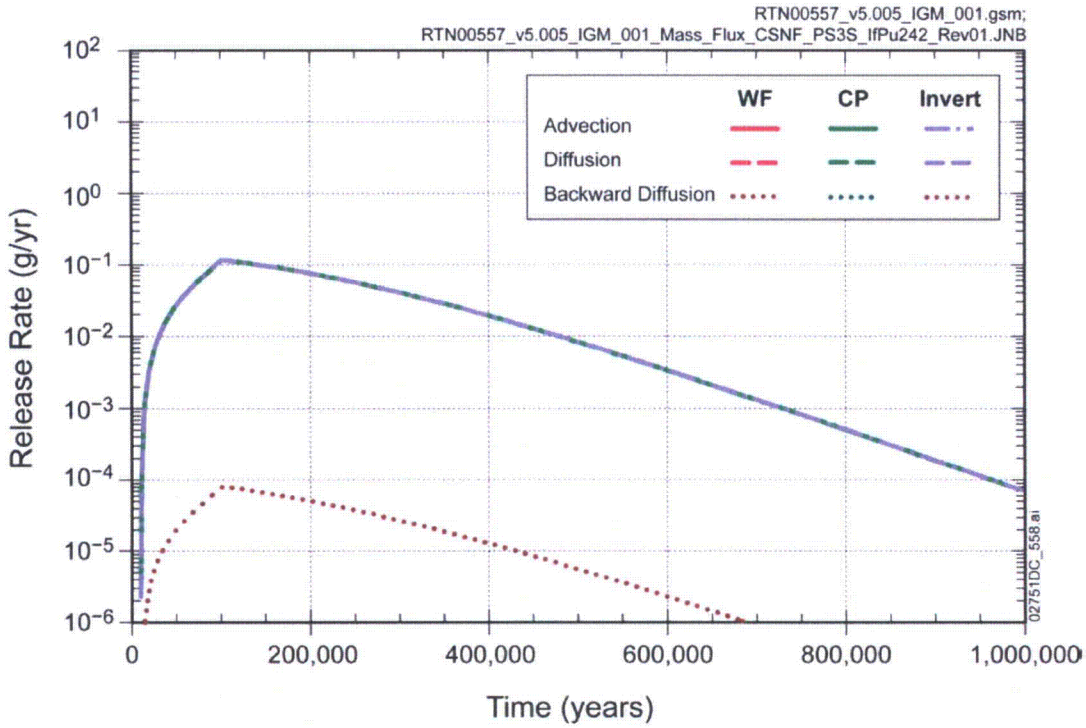
NOTE: Invert advection overlays corrosion product advection, and waste form advection overlays waste form backward diffusion (after 100,000 years). WF = waste form, CP = corrosion product.

Figure 3. Release Rate of ^{242}Pu Dissolved and Reversibly Sorbed from Each EBS Domain from Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure



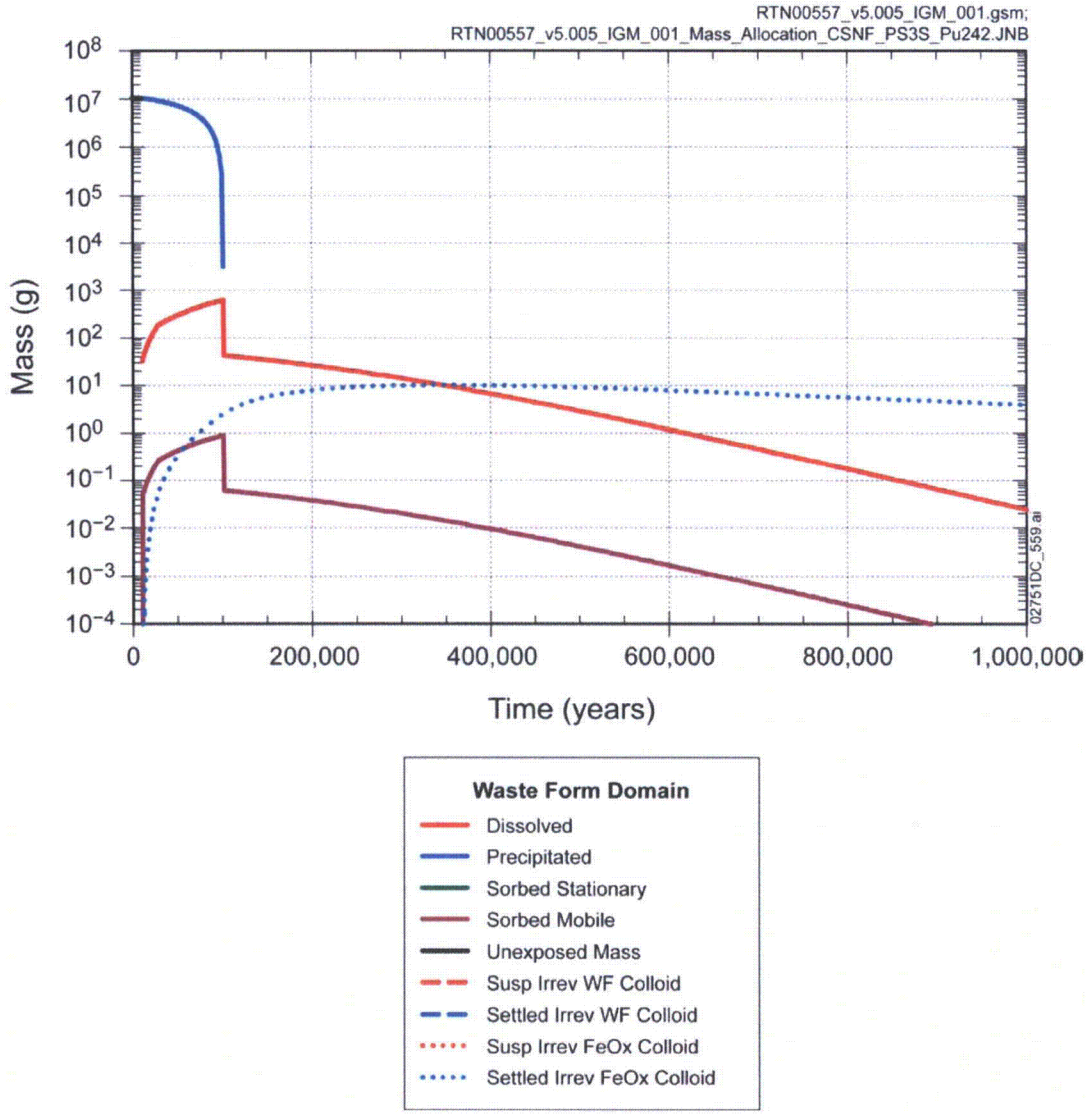
NOTE: Quantities that are identified in the legend but that do not appear in the figure have values that are below the plotted range. WF = waste form, CP = corrosion product.

Figure 4. Release Rate of ^{242}Pu Irreversibly Attached to Waste Form (Ic) Colloids from Each EBS Domain from Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure



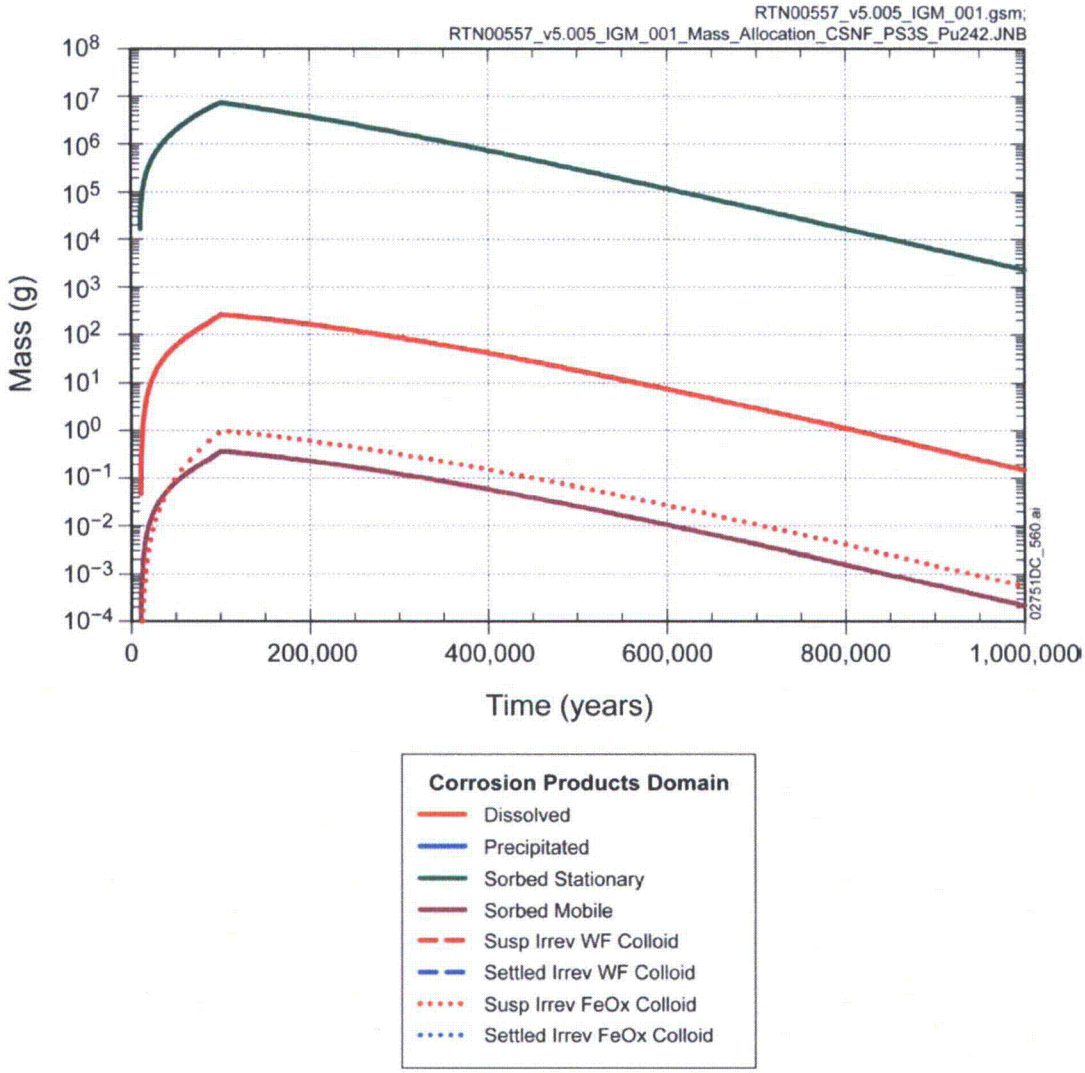
NOTE: Quantities that are identified in the legend but that do not appear in the figure have values that are below the plotted range.

Figure 5. Release Rate of ^{242}Pu Irreversibly Attached to Iron Oxyhydroxide (If) Colloids from Each EBS Domain from Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure



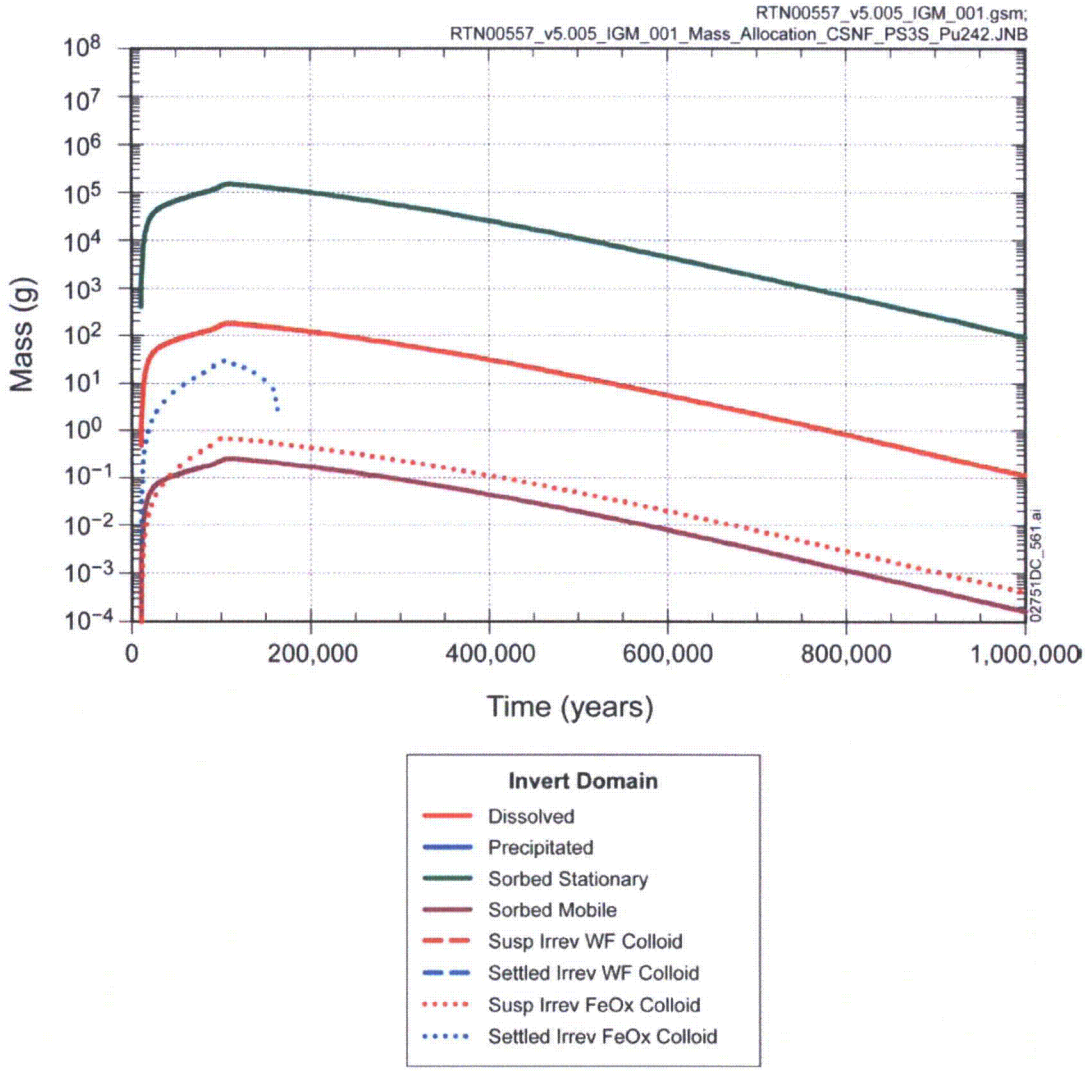
NOTE: Quantities that are identified in the legend but that do not appear in the figure have values that are below the plotted range. "Sorbed Mobile" indicates reversibly sorbed to colloids. "Irrev" indicates irreversibly attached to colloids. WF = waste form.

Figure 6. Mass of ^{242}Pu in the Commercial SNF Waste Form Domain of Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure



NOTE: Quantities that are identified in the legend but that do not appear in the figure have values that are below the plotted range. "Sorbed Mobile" indicates reversibly sorbed to colloids. "Irrev" indicates irreversibly attached to colloids. WF = waste form.

Figure 7. Mass of ^{242}Pu in the Corrosion Products Domain of Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure



NOTE: Quantities that are identified in the legend but that do not appear in the figure have values that are below the plotted range. "Sorbed Mobile" indicates reversibly sorbed to colloids. "Irrev" indicates irreversibly attached to colloids. WF = waste form.

Figure 8. Mass of ^{242}Pu in the Invert below Commercial SNF Waste Packages in a Seeping Environment of Percolation Subregion 3 for Realization #525 of the Igneous Intrusion Modeling Case for 1,000,000 Years after Repository Closure

1.3 SUMMARY

The response to RAI 3.2.2.1.3.4-2-007 presents and describes the transport of ^{242}Pu through the EBS following an igneous intrusion event at 10,000 years that results in conditions where plutonium-rich zirconium oxide colloid suspensions are stable in the waste form domain, but are unstable in the corrosion products domain. As a result, the significant releases of ^{242}Pu from the waste form domain that occur over the first 200,000 years are primarily associated with

zirconium-oxide colloids. At 200,000 years, about 90% of the mass in the corrosion products domain is associated with settled zirconium oxide colloids and only 10% of the mass is sorbed onto stationary corrosion products. Beyond 200,000 years, the percentage of the mass on the stationary corrosion products decreases as desorption from the corrosion products occurs, but zirconium oxide colloid suspensions remain unstable.

In this response, the transport of ^{242}Pu through the EBS is presented and described for a single realization of the igneous intrusion modeling case where zirconium oxide colloid suspensions are unstable in the waste form domain. Because zirconium oxide colloid suspensions are unstable in the waste form domain, ^{242}Pu is transported out of the waste form domain primarily in dissolved form, but its rate of movement is controlled by the plutonium solubility. In the corrosion products domain, mass that is transported from the waste form domain is either transported downstream to the invert, primarily by advection, or is sorbed onto the stationary corrosion products. Sorption onto corrosion products accounts for more than 99% of the ^{242}Pu mass distribution in the corrosion products domain.

The release of ^{242}Pu from the EBS in epistemic vectors #286 and #53 is predominantly by advection. When zirconium oxide colloid suspensions are stable in the waste form domain (epistemic vector #286), the release from the EBS increases gradually while a precipitate exists, then the release rate declines, and most of the ^{242}Pu mass in the EBS is in zirconium oxide colloid suspensions that are settled in the corrosion products domain. When zirconium oxide colloid suspensions are unstable in the waste form domain (epistemic vector #53), the release rates from the EBS are also controlled by the solubility in the waste form domain. In addition, the EBS release rates are primarily controlled by sorption onto or desorption from the corrosion products after the precipitate in the waste form domain is depleted.

When zirconium oxide colloid suspensions are stable in the waste form domain, the release of ^{242}Pu mass from the waste form domain is enhanced. In epistemic vector #286 the solubility of plutonium is 30 times smaller than that in epistemic vector #53, but early time release rates of total plutonium are comparable. As shown in Figures 1.1-22 and 1.1-23 of the response to RAI 3.2.2.1.3.4-2-007, the transport of plutonium from the waste form domain is enhanced by colloid suspensions. However, because these colloid suspensions are unstable in the corrosion products domain, EBS releases are reduced.

Approximately 84% of the initial ^{242}Pu mass (half-life = 375,000 years) will decay after 1,000,000 years. At 1,000,000 years, 12% of the initial ^{242}Pu mass (77% of the undecayed ^{242}Pu mass) is still in the EBS in epistemic vector #286, mostly retained as settled waste form colloids in the corrosion product domain (Figure 1.1-26 of the response to RAI 3.2.2.1.3.4-2-007). Because the colloid suspensions remain unstable in the corrosion product domain, the mass ^{242}Pu in the colloid suspensions in the corrosion products domain decreases primarily by radioactive decay. By comparison, in epistemic vector #53, the zirconium oxide colloid suspensions are unstable in the waste form domain. Although most of the ^{242}Pu mass that remains within the EBS at 1,000,000 years is still in the corrosion product domain (Figure 7), there is desorption of ^{242}Pu mass from the corrosion products. After 1,000,000 years only 0.13% of the initial ^{242}Pu mass (0.85% of the undecayed ^{242}Pu mass) is still in the EBS. As indicated in the first

supplemental response to RAI 3.2.2.1.3.4-2-007, the latter conditions are typical in the igneous intrusion modeling case for 62% of the epistemic sample elements.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

RAI Volume 3, Chapter 2.2.1.3.4, Third Set, Number 2:

Provide information that supports the predicted pH range in the corrosion products domain. In addition, explain the key chemical processes that lead to this limited range.

Basis: The response to RAI 3.2.2.1.3.4-2-003 suggests that, for multiple realizations in both igneous intrusive and seismic ground motion scenarios, and for both waste package types, the pH in the corrosion products domain will be restricted to the relatively narrow range of 7.0 to 8.4. This range favors CSNF waste form colloid instability, actinide sorption, and low actinide solubility. All these processes tend to reduce releases of actinides such as neptunium and plutonium from the corrosion products domain. Clarification is needed regarding model support for the pH predictions for the corrosion products domain by the competitive surface complexation model. [10 CFR 63.21(c)(15) and 10 CFR 63.101(a)(2)]

1. RESPONSE

Two lines of support for the relatively narrow pH range in the corrosion products domain caused by the buffering effect of surface complexation reactions at the surface of iron oxyhydroxide corrosion products are: (1) the self-consistent set of surface complexation reactions in the competitive surface complexation model (SCM) and the literature bases for this reaction network and (2) a comparison of the pH predictions of the competitive SCM with pH predictions from simpler models and analyses done in support of the in-package chemistry (IPC) abstraction (SNL 2007a).

1.1 KEY INPUTS FOR COMPETITIVE SCM DEVELOPMENT

The pH range used in the corrosion products domain is a direct output from the competitive SCM, which is based on a consistent set of equilibrium constants for the underlying surface complexation reactions. The basis for this set of equilibrium constants and other key inputs (such as CO_2 partial pressure) used in the development of the competitive SCM are presented in the model report *EBS Radionuclide Transport Abstraction* (SNL 2007b). The complete list of key input parameters and the location of their discussion within the radionuclide transport abstraction (RTA) report is:

1. The literature data that provide the surface complexation chemical reactions and constants (SNL 2007b, Table 6.5-12 and Appendix J, "Qualification of Thermodynamic Data for Geochemical Modeling of Adsorption of Radionuclides by Stationary Corrosion Products and Corrosion Product Colloids")

2. The thermodynamic database *phreeqcDATA025bdotCr3az.dat* (Section 4.1.1)

3. The range of possible element concentrations for U, Pu, Np, Am, Th, and Ni (1 to 10^{-8} times the maximum solubility) (Section 6.5.2.4.1)

4. The range of CO_2 partial pressure (10^{-4} to 10^{-2} bar) (Section 6.5.2.4.1)

5. The range of moles sorption sites per liter of water (0.4 to 52; DTN: SN0703PAEBSRTA.002), which was based on the ratio of mass of corrosion products to water volume (1 to 50 kg/L water), described in Section 6.5.2.2.1, and the properties of waste package corrosion products, described in Section 6.3.4.2.3.1.

Of these inputs, the first is the most important to the discussion of corrosion product pH values. The surface complexation chemical reactions and constants shown in Table 6.5-12 and qualified in Appendix J of the RTA report (SNL 2007b) are the primary basis of the competitive SCM. Appendix J of the RTA report (SNL 2007b) describes an initial set of surface complexation constants used in calculations in a surface complexation sorption model for performance assessment and qualifies the surface complexation constants used in calculations of radionuclide sorption by stationary corrosion products and corrosion product colloids. Criteria for selection of recommended surface complexation constants are outlined in Section J.6.2 of the RTA report (SNL 2007b), which also discusses the internal consistency of the thermodynamic data used in the competitive SCM; Section J.6.5 discusses the surface complexation constants used in the competitive SCM.

The process of compiling an internally self-consistent database consists of several steps: (1) compilation of equilibrium constants for reactions involving the element of interest, (2) extrapolation of equilibrium constants to reference conditions (usually zero ionic strength and 25°C), and (3) calculation of property values such as free energies of formation of the products through reaction networks. Surface complexation constants are obtained through fits of sorption data (sorption as a function of sorbate concentration or pH). The approach outlined in Appendix J of the RTA report (SNL 2007b) is to compile an internally self-consistent network of constants. The largest self-consistent network within the literature was identified and additional data were used to fill in the gaps. The compilations of Wang et al. (2001) and Turner and Sassman (1996) comprise the best reaction network upon which to base the set of surface complexation constants. These authors fit experimental data from several sources to a single-site diffuse layer model using a standard set of acidity constants for goethite. The additional data were based on oxidation-state analogues or on using analogous data for other ferric oxyhydroxide substrates.

Appendix J of the RTA report (SNL 2007b) also discusses several other aspects of the development of the competitive SCM that further support the modeling methods chosen in the competitive SCM.

1.2 SUPPORT FOR THE COMPETITIVE SCM pH RANGE

This section explains how the competitive SCM pH results, which control pH in the corrosion products domain, are supported by in-package chemistry modeling of pH completed in 2005 (SNL 2007a, Sections 6.7.1 and 6.8). The surface complexation models from 2005 were developed using methods that are different and independent of the 2007 competitive SCM (SNL 2007b), yet predict pH ranges similar to the competitive SCM.

The water within the corrosion products domain (SNL 2007a) flows from the waste form domain(s). In the corrosion products domain, the waste-form-reacted water further reacts with the corrosion products. For this reason, the chemistry within both the waste form domains (also known as cells 1, 1a, and 1b) and the corrosion products domain (also known as cell 2) is examined in the following discussion. Table 1 shows the pH ranges calculated for the waste form and corrosion products domains in 2005 and 2007. Table 1 distinguishes the valid range of the model from the output range used in the total system performance assessment (TSPA). For example, the competitive SCM defines a pH regression space that ranges from 6.25 to 9.16, but the output of the TSPA model ranges from 7 to 8.4. This is because the regression model was built from equilibrium calculations where the CO_2 partial pressure ranged from 10^{-4} to 10^{-2} bar and the uranium concentration ranged from 10^{-11} to 3 moles per liter, but the actualized CO_2 partial pressure and uranium concentration ranges in TSPA were much narrower. For example, by 50,000 years in the igneous intrusion modeling case of igneous intrusion at 250 years (see figures in response to RAI 3.2.2.1.3.4-2-003), the CO_2 partial pressure range narrows to $10^{-2.88}$ to $10^{-3.01}$ bar, and the uranium concentration range narrows to $10^{-7.7}$ to 10^{-4} moles/L within the corrosion products domain. This narrow CO_2 partial pressure and uranium concentration range results in the narrow TSPA output range.

Table 1. Ranges of pH Calculated for the Waste Form and Corrosion Products Domains

Model, Domain	Valid Model pH Range	TSPA Actual Range, IGM[a]	TSPA Actual Range, SMM[b]
2005 IPC, Combined Domain	4.5 to 8.5[c]		
2005 Analytic SCM, Combined Domain	6.8 to 8.1[c]	7.5 to 7.7[d]	
2005 SCM Demonstration, Combined Domain	6.6 to 9.05[c]	7.16 to 8.24[d]	
2007 IPC, Commercial SNF Waste Form Domain	4.65 to 9.07[e]	5.5 to 8.4[f]	5.0 to 8.2[f]
2007 IPC, DOE SNF Waste Form Domain	4.63 to 9.06[e]	5.6 to 8.2[f]	5.0 to 8.2[f]
2007 IPC, Glass Waste Form Domain	4.45 to 10.98[e]	5.7 to 8.8[f]	5.2 to 9.9[f]
2007 Competitive SCM, Corrosion Products Domain	6.25 to 9.16[g]	7.0 to 8.4[f]	7.0 to 8.3[f]

[a] IGM = million year igneous intrusion modeling case with intrusion at 250 years.
[b] SMM = million year seismic ground motion modeling case with aleatory vector 4641.
[c] SNL 2007a, Sections 6.10.1.1 and 6.10.1.2, Table 6-20, Table 6-18.
[d] Range for CO_2 partial pressure = 10^{-3} bar, see also footnote c.
[e] SNL 2007a.
[f] pH values read from figures in response to RAI 3.2.2.1.3.4-2-003.
[g] SNL 2007b, output DTN: SN0703PAEBSRTA.002, workbook, *Final_Calc_Results_Surf_Complx_Data_1.xls*, worksheet Raw Output, column B.

NOTE: SNF = spent nuclear fuel.

1.2.1 2005 IPC Model

The 2005 IPC model (SNL 2007a, Section 6.10.1) calculated the in-package chemistry considering the entire waste package and waste form as a single mixing cell. Separate abstractions were provided for water flow rates of less than 150 L/yr and greater than 150 L/yr, for commercial SNF waste packages and codisposal waste packages, and for different time periods. The low end of the pH range for commercial SNF and codisposal waste packages was conservatively set to 4.5 based on both the long-term minimum for high temperature runs and the

Cr(VI) sensitivity analyses, even though neither of these conditions, long-term high temperature and Cr(VI) production, are likely to happen (see SNL 2007a, Section 6.6.3).

The analytic SCM that was used to define the upper pH range for the first 600 years for the commercial SNF waste packages provided higher pH values ranging from 6.8 to 8.1 (SNL 2007a, Table 6-18), depending on the CO_2 partial pressure and the moles surface sites available for sorption. This model could have also been used for the lower pH limit, but the lower pH range was chosen to be conservatively low, because degradation rates of waste forms, stability of commercial SNF colloids, and solubilities of actinides are higher at low pH values. For commercial SNF waste packages after 600 years and for codisposal waste packages at all times, the upper pH range is defined to be 7 to 8.5, depending on the flow rate, based on EQ6 reaction path calculations—with the highest pH seen in the high flow rate runs.

In summary, the 2005 IPC model pH ranges from 4.5 to 8.5 (Table 1) but includes low pH values resulting from carbon steel degradation, which are unlikely to occur at early times due to pH buffering of iron corrosion products, and which will not occur at later times when the carbon steel is fully degraded. There is no carbon steel in the 2007 IPC model for commercial SNF waste packages as a result of a design change.

1.2.2 2007 IPC Model

The 2007 IPC model considered two spatial domains: (1) the waste form domain and (2) the corrosion products domain. Most of the steels in the engineered barrier system (EBS) were partitioned to the corrosion products domain, with only the waste forms and the internal structures of the waste package included in the "in-package" or waste form domain. The surface complexation model was not used in the calculation of pH range in the waste form domain because there is a much lower abundance of iron alteration products in the waste form cells. In addition, there was no need to account for the effects of surface complexation on the pH range because rigorous and conservative pH ranges can be established based on the pH buffering capacities of the degradation products (SNL 2007a, Section 6.3.4[a]). The established pH limits are defined by pH buffer reactions that cannot be overcome by potential sources of acid or base. These reactions in turn define the acid neutralizing capacity (ANC) and base neutralizing capacity (BNC) of the waste form cells (SNL 2007a, Section 6.3.4.1[a]).

As discussed in Section 6.3.4[a] of the IPC report (SNL 2007a), the degradation of fuels and structural materials in the waste form cells generates ANC and BNC primarily in the form of secondary minerals such as schoepite ($UO_3 \cdot 2H_2O$), from dissolution of the uranium oxide waste form, and trevorite ($NiFe_2O_4$), from corrosion of the metal alloys. The pH range in a waste form cell is determined by titrating the contents of a waste form cell with acid and base. These titrations are used to identify the pH buffering minerals in the cells, define the pH limits, and quantify the buffer capacities. Titrations were performed for a range of ionic strength and CO_2 partial pressure. Schoepite and trevorite are the primary mineral buffers that limit the pH range in the commercial SNF and DOE SNF waste form domains. For the glass waste form domain, the corresponding buffers are carbon dioxide and trevorite. As discussed in Section 6.10.1.1 of the IPC report (SNL 2007a), the ANC and BNC accumulation rates are far in excess of the rates of any reasonable external source of acid or base. The largest internal sources of acid and base

are the degradation reactions themselves, but even when their rates are increased to maximum values, they also cannot overcome the rates of ANC and BNC accumulation. Corrosion of stainless steel by itself produces some acidity (as shown in Figure 6-22 of SNL 2007a). However, it will not cause the pH to fall below the stability field of trevorite because the corrosion reaction produces trevorite. Thus, any increase in the stainless steel corrosion rate will be accompanied by an increase in the production of trevorite. In this way, the corrosion of stainless steel is self-buffering.

The 2007 IPC model included the buffering effects of CO_2, schoepite, and trevorite but did not include the buffering effects of the corrosion products surfaces. Tables 6.3.8 and 6.3.9 of the RTA report (SNL 2007b) indicate that while most iron is in the corrosion products domain, significant iron is still in the waste form domains. For example, 28% of the iron in the 21-PWR TAD (without shield plug) resides within the waste form domain, while 14.5% of the iron in the codisposal waste package is in the glass waste form domain and 5% is in the DOE SNF waste form domain. Because the IPC did not consider the buffering effect of the corrosion products surfaces, the pH range predicted by the 2007 IPC model is conservatively broad. This is because of the conservative nature of the titrations with acid and base, and the lack of surface complexation.

1.2.3 2005 Analytical SCM

In 2005, two SCMs were developed. The "analytical surface complexation model" was used in defining the upper pH range in the 2005 IPC model and is documented in Section 6.7.1 of the IPC report (SNL 2007a). In addition the "surface complexation demonstration model" was developed to support the analytic SCM (see Section 1.2.4).

The results presented in Table 6-18 of the IPC report (SNL 2007a) for the analytic SCM indicate a pH of 7.5 to 7.7 at a CO_2 partial pressure of 10^{-3} and 0.01 to 0.1 mole sites/mole Fe, respectively, for Carbon Steel Type A516. The pH was found to be insensitive to the number of mole sites per liter of water as long as there are at least 0.1 moles of steel per liter of water (approximately 0.01 mole sites per liter of water). (Note that this is a much smaller concentration of surface sites than the competitive SCM range of 0.4 to 52 moles sites per liter of water in the corrosion products domain.) To simplify the calculation, adsorption of phosphate, uranium, nickel, chromate, sulfate, and other sorbates was neglected. Under favorable conditions, these sorbates, if abundant, could have a noticeable impact on the final calculated pH values. However, as a check on the potential effects of these sorbates, surface complexation reactions involving the entire database are performed for the "surface complexation demonstration model," as discussed below.

1.2.4 2005 Surface Complexation Demonstration Model

The buffering effect of corrosion product surfaces on waste form domain waters was demonstrated in the 2005 "surface complexation demonstration model," which is documented in Section 6.8 of the IPC report (SNL 2007a). The effects of ferric oxyhydroxide surfaces on pH, as defined in Section 6.7 of the IPC report (SNL 2007a), are confirmed in the "surface complexation demonstration model" using PHREEQC. The purpose of the PHREEQC surface

complexation calculations is to build confidence in the analytical SCM. Additional complexity in surface complexation calculations is afforded by PHREEQC due to its ability to include effects of surface charge, ionic strength, and a much larger assortment of surface complexation reactions. However, as the calculations demonstrate, this increased complexity does not result in pH predictions largely different from those made by the simplified analytical model.

The particular surface complexation model chosen for this demonstration is from Dzombak and Morel's (1990, Chapter 2) generalized two-layer model, which is based on the Gouy-Chapman theory of the electrical double layer. In this model, there is one layer associated with the surface charge (affected by site-specific adsorption) and a diffuse layer between the surface layer and the bulk solution that contains counter-charged ions.

The surface complexation calculations require several inputs. These inputs include, as presented in the IPC report (SNL 2007a), a thermodynamic database (Section 6.8.1), surface properties (Section 6.8.2), and initial water and gas compositions (Section 6.8.3). The initial chemistry of the waters prior to surface complexation was taken from the EQ3/6 calculations of groundwater or distilled water reacted with the internal components of the commercial SNF and codisposal waste packages. The chemistry of these waters was taken before 100 years of EQ6 calculation time, at a time when degradation of carbon steel is modeled to release sulfuric acid. Prior to surface complexation, the pH ranged from 3.29 to 4.70 (SNL 2007a, Table 6-19). After reaction with 1.2 mole sites per liter of water of corrosion products surfaces, the surface complexation demonstration model predicts a pH of 7.16 to 8.24 at a CO_2 partial pressure of 10^{-3} bar (SNL 2007a, Table 6-20), showing the very large buffering effect of iron oxyhydroxide surfaces.

1.2.5 2007 Competitive SCM

The abstraction of the competitive SCM is a regression fit that predicts pH and sorbed concentrations for uranium, plutonium, neptunium, americium, thorium, and nickel from the dissolved concentrations of these elements, the CO_2 partial pressure, and the number of sorption sites per liter of water as implemented in the TSPA. The development of the model is qualified and documented in the RTA report (SNL 2007b).

Unlike the surface complexation demonstration model, which is based on a narrow range of acids and bases released during degradation of the waste forms and steels, the competitive SCM encompasses a wide range of potential EBS environmental conditions in the TSPA for the following input variables: the aqueous concentrations for uranium, plutonium, neptunium, americium, thorium, and nickel; the range in CO_2 partial pressure (10^{-4} to 10^{-2} bar); and the number of surface sorption sites per liter of water (SNL 2007b, Section 6.5.2.4.1). The concentrations of uranium, plutonium, neptunium, americium, thorium, and CO_2 were fixed by increasing or decreasing the solubility limits that are contained in the database using the saturation index feature of PHREEQC. By sampling a large range of actinide and CO_2 concentrations, the pH range from the first PHREEQC equilibration is quite large (6.38 to 8.97 DTN: SN0703PAEBSRTA.002). This range does not go as low as the pre-sorption equilibrated pH in the surface complexation demonstration model (3.2 to 4.7, SNL 2007a, Table 6-19) but is similar to the final surface equilibrated pH (6.6 to 9.05, SNL 2007a, Table 6-20) in the surface complexation demonstration model.

In the competitive SCM (SNL 2007b), Latin hypercube sampling was conducted on the potential uncertainty ranges of the water composition variables (i.e., aqueous actinide concentrations and CO_2 partial pressure) to produce 100 realizations of the starting water (i.e., the water composition prior to reacting with the iron oxyhydroxide surfaces). Of these 100 realizations, only 96 converged in PHREEQC because four of the combinations were not physically realistic. These 96 resulting water compositions were then reacted in a second set of PHREEQC calculations with 50 sampled realizations of sorption sites per liter of water to generate 4,800 realizations of surface-complexation-reacted solutions. In this second set of PHREEQC calculations, the pH was allowed to vary and the surface sites were allowed to populate to equilibrium. This resulted in a slightly larger pH range of 6.25 to 9.16 (for the full range of CO_2 partial pressures) compared to the starting water range (6.38 to 8.97). With these 4,800 fully equilibrated solutions, regression models were built for pH and for the sorbed concentration of each actinide, and it was found that the pH was significantly correlated with CO_2 partial pressure and uranium concentration. The moles sites per liter were not found to be correlated to pH, most likely because the range of 0.4 to 52 moles sites per liter is well above the concentration of sites needed to buffer the pH. The selected regression model for pH is given in Equation 1 (SNL 2007b, Section 6.5.2.4.6). This model has an R^2 of 0.6281, indicating an acceptable predictive capability with some unexplained variance. The unexplained variance was accounted for by adding the error term to the regression model.

$$\mathrm{pH} = 4.5342 + 0.6132\,(p\mathrm{CO_2}) - 0.3805\,\log_{10}[U] - 0.0254(\log_{10}[U])^2 + E \qquad \text{(Eq. 1)}$$

where $p\mathrm{CO_2}$ is the negative log of the in-drift CO_2 partial pressure (bars), $[U]$ is the dissolved concentration of uranium in mol/L, and E is the error term (pH_Cell_2_Regression_Error) defined by a normal distribution with mean of zero and standard deviation of 0.32 truncated at ±2 standard deviations.

1.2.6 Summary of the Support for the 2007 Competitive SCM

In the 2005 IPC model (documented in SNL2007a), the lower pH limit in the combined waste form-corrosion products domain was due to acids produced from the degradation of the steel, specifically sulfuric, phosphoric, molybdenum (VI), and chromium (VI) acids. In 2007, the IPC model titrated in acid and base to obtain the pH values that would occur at the limit of the buffering capacity of the corrosion products within the waste form domains. In both these cases, the lower pH limit was chosen to be conservatively low, because degradation rates of waste forms, stability of commercial SNF colloids, and solubilities of actinides are higher at low pH values. This low pH, however, is unlikely to be achieved within either the waste form or corrosion products domains where surface complexation will be occurring.

A waste form domain pH range less conservative than the 2007 IPC would include no chromium(VI) production (see SNL 2007a, Section 6.6.3) and include some SCM buffering effects. The 2005 SCM demonstration for the combined domain provides the best indication of what pH range such a TSPA model would predict, 7.16 to 8.24. Thus the pH of the waters entering the corrosion products domain would be nearly equal to that of the surface-reacted waters calculated by the 2007 competitive SCM of 7 to 8.4 for the corrosion products domain.

The 2005 SCM demonstration model was built for the combined domain using EQ3/6 calculations of waste package pH followed by PREEQC surface complexation, which is quite different from the method of development of the 2007 competitive SCM for the corrosion products domain, yet these two models provide similar pH ranges. This similarity provides support for the 2007 competitive SCM.

1.3 KEY CHEMICAL PROCESSES

Key chemical processes that limit the pH range in the corrosion products domain include the protonation and deprotonation surface reactions, as well as dissolution reactions that affect aqueous hydrogen ion concentration. The protonation and deprotonation surface reactions that tend to buffer pH in the near-neutral range are:

$$HfssOH_2^+ \leftrightarrow HfssOH + H^+ \qquad K_1 = [HfssOH][H^+]/[HfssOH_2^+] = 10^{-7.35}$$

$$HfssOH \leftrightarrow HfssO^- + H^+ \qquad K_2 = [HfssO^-][H^+]/[HfssOH] = 10^{-9.17}$$

where Hfss represents the iron oxyhyroxide surface (SNL 2007b, Table 6.5-12). Dissolution reactions that buffer the pH in the fluid within the waste form and corrosion products domains are (SNL 2007a, Section 6.3.4.1[a]:

Base Neutralizing Capacity:

$$CO_2 + H_2O \rightarrow HCO_3^- + H^+ \leftrightarrow CO_3^{2-} + 2H^+$$

$$UO_3 \cdot 2H_2O + 3CO_2 \rightarrow UO_2(CO_3)_3^{4-} + 4\,H^+$$

Acid Neutralizing Capacity:

$$NiFe_2O_4 + 2H^+ \rightarrow Ni^{2+} + 2FeOOH.$$

1.4 CONCLUSION

The pH range used in the corrosion products domain is a direct output from the competitive SCM, which is based on a consistent set of equilibrium constants for the underlying surface complexation reactions. The information that supports the predicted pH in the corrosion products domain includes: (1) the literature data that supports the surface complexation constants, and (2) the in-package-chemistry modeling of 2005 and 2007, especially the surface complexation modeling of 2005. The key chemical processes that lead to the limited pH range include the protonation and deprotonation reactions of the corrosion products surface sites, as well as the buffering effects of schoepite and trevorite dissolution.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

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NOTE: [a]Provided as an enclosure to letter from Williams to Sulima dtd 02/17/2009. "Yucca Mountain – Request for Additional Information Re: License Application (Safety Analysis Report Section 2.1), Safety Evaluation Report Volume 3 – Postclosure Chapters 2.2.1.1 and 2.2.1.3.7 – Submittal of Department of Energy Reference Citations."

RAI Volume 3, Chapter 2.2.1.3.4, Third Set, Number 3:

Provide the results from the PHREEQC surface complexation models and related files in Data Tracking Number (DTN) SN0703PAEBSRTA.002 – "Surface Complexation Modeling Results, Actinide Kd abstractions and pH abstraction" and the zip file: EBS-RTA-SURFCPLX_DTN.ZIP that support the actinide K_d distributions for the stationary corrosion products. In particular, provide the surface complexation model results and the support for the ranges and combinations of parameter values that produced the actinide K_d values at the upper end of the distribution.

Basis: K_d values adopted for actinides in the corrosion products domain have important effects on the release rates of dose-significant radionuclides, since greater K_d values in the corrosion product domain limit releases. In addition, pH in the corrosion products domain strongly affects colloid stabilities, another mechanism that has a potentially strong effect on plutonium release.

1. RESPONSE

This response provides the requested results from the PHREEQC surface complexation model and all files in DTN: SN0703PAEBSRTA.002, "Surface Complexation Modeling Results, Actinide K_d Abstractions and pH Abstraction." The specific file requested, *EBS-RTA-SURFCPLX_DTN.ZIP*, is the previous name for the file that contained all of the files in the DTN. Its name was changed to *SN0703PAEBSRTA.002.zip* when it was placed in the Technical Data Management System. The contents of this file, which includes all of the results of the surface complexation model, are provided in the enclosed DVD (Section 5).

A summary of the development and use of the surface complexation model is provided below. In particular, support is provided for the ranges and combinations of parameter values used to produce the actinide K_d response surfaces. The K_d response surfaces are not intended to indicate the frequency with which the K_d values are realized in the total system performance assessment (TSPA) model simulations. Rather, they are intended to cover large ranges of combinations of parameter values to ensure that they will encompass the combinations realized in the TSPA simulations.

The ranges of the K_d response surfaces are compared to the ranges of K_d values realized in TSPA simulations for the seeping environment of percolation subregion 3 after an igneous intrusion event at 100,000 years. The igneous intrusion modeling case is one of the primary contributors to the overall mean annual dose (SAR Figure 2.4-18). This case was chosen over the seismic ground motion modeling case because no waste packages are intact after the igneous intrusion, allowing for a full set of 300 K_d realizations from the time of the modeled igneous intrusion event. This comparison indicates that the maximum K_d values of the response surfaces are approximately three to ten times higher than the maximum values realized. This difference arises because the combinations of actinide concentrations, corrosion product properties, and partial pressures of carbon dioxide that generated the highest values in the K_d response surfaces do not occur in the TSPA model realizations.

1.1 SURFACE COMPLEXATION MODEL

In the TSPA model, actinide sorption to stationary corrosion products and iron oxide colloids is represented by a distribution coefficient (K_d), the equilibrium ratio of a component's sorbed concentration to its aqueous concentration. The value of this ratio for a given actinide depends on the composition of the solution and the surface properties of the sorbent.

To evaluate the effects of aqueous composition and corrosion product surface properties and concentrations on actinide K_d values for the TSPA model, a surface complexation model was developed. This model was used to calculate K_d values for americium, neptunium, plutonium, thorium, and uranium over a broad range of conditions. These conditions included broad ranges and combinations of: (1) aqueous actinide concentrations, (2) nickel concentrations, (3) partial pressures of carbon dioxide (P_{CO2}), (4) pH values, and (5) concentrations of sorption sites per volume of water. The results of this model were subsequently abstracted by regression analysis to generate functions used in TSPA to calculate sorbed concentrations. Details of this model can be found in Sections 6.5.2.4 and 7.2.3 and Appendix J of *EBS Radionuclide Transport Abstraction* (SNL 2007a).

1.1.1 Surface Complexation Reactions

The surface complexation model required a set of surface complexation reactions to simulate equilibrium sorption of aqueous components and to simulate the acid-base behavior of the surface sites. The specific sorption model chosen was the single-site diffuse layer model. The set of sorption reactions selected included one or more sorbed complexes for nickel, hydrogen ion, carbonate, and each actinide (americium, neptunium, plutonium, thorium, and uranium). The development and qualification of this database and the sorption model chosen are described in Appendix J of *EBS Radionuclide Transport Abstraction* (SNL 2007a).

1.1.2 Input Parameter Ranges

The combinations of input values for surface properties, actinide concentrations, and carbon dioxide partial pressure were determined by Latin hypercube sampling. Surface site parameters were sampled across established ranges, actinide concentrations were sampled across eight orders of magnitude, and the partial pressure of carbon dioxide was sampled across two orders of magnitude. The Latin hypercube sampling, described in more detail in Section 1.1.3, produced a set of realizations as input to a set of surface complexation model simulations. These simulations, in turn, were used to calculate actinide K_d response surfaces that correspond with the broad ranges of sampled input parameters.

1.1.2.1 Surface Properties

The surface properties of the stationary corrosion products are defined in the surface complexation model by the bulk surface site concentration (moles per liter of water), the bulk specific surface area (area per mass of corrosion products), and the mass concentration (mass of corrosion products per liter of water). These values are determined from a sampling of: (1) the relative proportion of goethite and hydrous ferric oxide (HFO) in the stationary corrosion

products, (2) the specific surface areas of goethite and HFO, (3) the site densities of goethite and HFO (number of sites per surface area), and (4) the corrosion products mass concentration range.

For the surface complexation model, the composition of the iron oxide corrosion products was modeled as a mixture of goethite and HFO. The goethite fraction of the goethite-HFO mixture was approximated using a uniform distribution ranging from 0.45 to 0.8. This distribution was based on studies cited in Section 6.3.4.2.3.1 of *EBS Radionuclide Transport Abstraction* (SNL 2007a) that indicate that HFO can convert to the more stable goethite phase fairly rapidly under laboratory conditions, but that a large fraction of amorphous ferric oxyhydroxide material can persist for long periods of time on corroded ancient artifacts under atmospheric conditions.

The distributions of the specific surface areas of goethite and HFO are represented by lognormal truncated distributions with ranges of 14.7 to 110 m^2/g and 68 to 600 m^2/g, respectively. These distributions are based on the specific surface area measurements compiled in Tables 4.1-12 and 4.1-14 of *EBS Radionuclide Transport Abstraction* (SNL 2007a).

The distributions of the site densities of goethite and HFO are defined by the cumulative probability distributions in Tables 6.3-5 and 6.3-6 of *EBS Radionuclide Transport Abstraction* (SNL 2007a). These distributions are derived from a compilation of literature measurements presented in Tables 4.1-12 and 4.1-14 of the same report.

The mass concentration range for iron oxide corrosion products, C_{CP}, is represented by a uniform distribution between 1 and 50 kg per liter of water. The model range for this parameter is constrained primarily by the porosity of the corrosion products and the water saturation. C_{CP} is calculated from the following equation:

$$C_{CP} = \frac{\rho_{CP}(1-\theta)}{\phi_w \theta} \qquad \text{(Eq. 1)}$$

where ρ_{CP} is the weighted average of the densities of goethite and HFO in the corrosion products domain as determined by the sampled proportion of goethite in the goethite-HFO mixture (mass corrosion products per volume of corrosion products), θ is the porosity (volume pore space per total volume), and ϕ_w is the water saturation (volume water per volume pore space). The densities of goethite and HFO are approximately 4.26 kg/L and 3.96 kg/L, respectively (SNL 2007a, Tables 4.1-10 and 4.1-11). Thus, for the modeled porosity of the corrosion products domain, which is 0.4 (SNL 2007a, Section 6.3.4.3.4.2), a water-saturated domain (ϕ_w=1) would have an iron corrosion products mass concentration of approximately 6 kg per liter of water. This is not the minimum mass concentration, however, because the presence of other corrosion products and minerals in the domain reduces this concentration in proportion to the fractional volume of solids that are iron oxide corrosion products. To ensure that a sufficient range of mass concentrations was included in the surface complexation model simulations, the lower bound was set at 1 kg per liter of water. The upper bound of 50 kg per liter of water corresponds to a water saturation of approximately 0.1, assuming no other corrosion products or minerals are present. This upper bound is expected to occur only infrequently and only at early times because the relative humidity in the waste package (SNL 2008a, Figure 6.3-53) will almost

always be high enough for water saturations greater than 0.1 (SNL 2007a, Figures 6.5-4 and 6.5-5).

The bulk surface site concentration (moles of sites per volume of water) of the iron oxide corrosion products (C_s), used as input to the surface complexation model simulations, is calculated from the following equation:

$$C_s = C_{CP} \frac{N_{CP} \bar{s}_{CP}}{N_A} \qquad \text{(Eq. 2)}$$

where N_{CP} is the bulk site density of the iron oxide corrosion products (number of sites per surface area), $\bar{s}_{CP}$ is the bulk specific surface area (surface area per mass), and N_A is Avogadro's number (6.02×10^{23}). The bulk values of N_{CP} and $\bar{s}_{CP}$ are weighted averages of the sampled site densities and specific surface areas of goethite and HFO.

1.1.2.2 Aqueous Composition

The sorbed concentrations of actinides are highly dependent on the aqueous concentrations of actinides and other sorbing species due in part to the competition of these species for the limited number of sorption sites. In addition to the actinides, nickel is expected to sorb readily and to be present in significant quantities due to its abundance in steel. To predict sorption over a broad range of combinations of actinide and nickel concentrations, broad ranges of these parameters were included in the Latin hypercube sampling of these parameters.

For the actinides, the maximum concentrations were determined based on the results of the solubility calculations documented in *Dissolved Concentration Limits of Elements with Radioactive Isotopes* (SNL 2007b). As described in Section 6.5.2.4.1 of *EBS Radionuclide Transport Abstraction* (SNL 2007a), the maximum actinide concentrations were set at the mean aqueous solubility plus the maximum uncertainty. For nickel, the maximum value was set at its solubility with no additional uncertainty. To cover the lower ends of the concentration ranges for these actinides and nickel, minimum values were set at 10^{-8} of the maximum values, and log uniform distributions were assigned.

1.1.2.3 Carbon Dioxide

The carbon dioxide partial pressure (P_{CO2}) range represented in the set of surface complexation model simulations is 10^{-4} to 10^{-2} bar. The distribution of this range is treated as log uniform. An example of the distribution of realized P_{CO2} values in the drift over time for the nominal modeling case is provided in Figure K4.3-7 of *Total System Performance Assessment Model /Analysis for the License Application* (SNL 2008b). Excursions outside the 10^{-4} to 10^{-2} bar range are minor and infrequent and are therefore not warranted in the set of surface complexation model simulations.

1.1.3 Latin Hypercube Sampling and PHREEQC Simulations

Latin hypercube sampling of the surface complexation model inputs was performed using the GoldSim software. The sampling was divided into two parts: (1) the surface properties and mass concentrations of the iron oxide corrosion products, and (2) the water composition as affected by the sampled carbon dioxide partial pressure. Specifically, the sampling was set up to produce 50 realizations of the surface properties and 100 realizations of the aqueous inputs (actinide concentrations, nickel concentration, and P_{CO2}). The complete aqueous compositions were further defined by setting the major ion concentrations equal to that of average J-13 well water. These solutions were equilibrated with the sampled P_{CO2} and charge balanced on pH using PHREEQC, an aqueous geochemical code with surface complexation model capability. Four of the 100 aqueous solution realizations were non-physical and were eliminated. Thus, a total of 96 aqueous solutions were generated.

For the surface complexation model simulations, PHREEQC was used to equilibrate each of the 96 water compositions with each of the 50 realizations of the surface properties of the iron oxide corrosion products. These simulations produced 4,800 calculations for the sorbed concentrations of each of the actinides over the wide ranges of sampled input parameters and across a pH range expected to encompass the full range of the domain (6.25 to 9.16) (SNL 2007a, Section 7.2.3). These calculations effectively produced K_d response surfaces for each actinide. The minimum and maximum K_d values calculated for each radionuclide are listed in Table 1. These response surfaces are displayed in two dimensions in the figures of Section 7.2.3 of *EBS Radionuclide Transport Abstraction* (SNL 2007a). The results were subsequently analyzed by multiple regression to produce functions that were used in the TSPA model to calculate actinide sorption as a function of the primary predictors: P_{CO2}, surface site concentration, and aqueous actinide concentrations (SNL 2007a, Section 6.5.2.4.5 and Table 6.5-14).

Table 1. Minimum and Maximum Values of the Actinide K_d Response Surface Calculations

Actinide	Minimum (mL/g)	Maximum (mL/g)
Americium	2.0×10^{-1}	2.9×10^{7}
Neptunium	5.6×10^{-1}	1.0×10^{6}
Plutonium	6.5×10^{-1}	3.5×10^{5}
Thorium	1.4×10^{0}	7.4×10^{5}
Uranium	9.2×10^{-2}	9.6×10^{5}

Source: DTN: SN0703PAEBSRTA.002, file: *Final_Calc_Results_Surf_Complx_Data_1.xls.*

1.2 K_d REALIZATIONS IN TSPA

The K_d response surfaces calculated by the surface complexation model simulations do not indicate the frequencies or ranges of K_d values realized in the TSPA model simulations. For comparison, a set of K_d realizations was calculated from the TSPA 1,000,000-year igneous intrusion modeling case for an igneous intrusion at 100,000 years.

To calculate the realized K_d values, the aqueous and sorbed concentrations of each sorbing actinide in the corrosion products domain for both commercial spent nuclear fuel (SNF) and

codisposal waste package types were extracted. Sorbed concentrations were calculated from the regression equations represented in Table 6.5-14 of *EBS Radionuclide Transport Abstraction* (SNL 2007a) based on the actinide concentrations, sorption site concentration, and P_{CO2} in the corrosion products domain at each point in time. These sorbed concentrations were then divided by the corresponding aqueous concentrations to calculate the K_d histories for each of 300 realizations for each actinide and each waste package type. The results are plotted in Figures 1 to 5 along with statistic curves for the median, mean, and 5th and 95th percentiles.

Figures 1 to 5 indicate that the ranges of the realized K_d distributions are much smaller than the ranges of K_d values calculated by the PHREEQC surface complexation model simulations (Table 1). Maximum values calculated by the PHREEQC simulations are approximately three to ten times greater than the maximum values realized. The main reason for this difference is that the combinations of actinide concentrations, corrosion product properties, and partial pressures of carbon dioxide that generated the highest K_d values of the response surfaces do not occur in the TSPA model simulations. Although the maximum realized K_d values are within or above the maximum EPA soil K_d values, iron oxides are expected to have higher K_d values than soils, as explained in the response to RAI 3.2.2.1.3.4-2-10.

1.3 SUMMARY

The surface complexation model was used to calculate equilibrium actinide sorption over a broad range of possible combinations of concentrations of surface sites and aqueous species in the corrosion products domain. The model inputs included a thermodynamic surface complexation reaction database, the properties and concentrations of surface adsorption sites, the aqueous composition, and the partial pressure of carbon dioxide. The thermodynamic database and the ranges of carbon dioxide partial pressure and concentrations of surface sites and aqueous species used in the model are supported by literature data, available data, and the conceptual model. Each input and output file of this model and all other files related to this model are documented in DTN: SN0703PAEBSRTA.002 and provided in the enclosed DVD (Section 5).

The resulting K_d response surfaces, while covering the full range of potential parameter combinations in TSPA, do not by themselves indicate the frequency with which the K_d values are realized in the TSPA model simulations. Therefore, comparisons were made to the distributions of realized actinide K_d values for a specific representative modeling scenario and location – the seeping environment of percolation subregion 3 after an igneous intrusion event at 100,000 years. These realizations show that a much smaller range of K_d values are realized and that the maximum values calculated by the surface complexation model simulations are approximately three to ten times greater than the maximum values realized in this TSPA modeling case, due mainly to a narrower range of realized combinations of actinide concentrations, corrosion product properties, and partial pressures of carbon dioxide.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

4. REFERENCES

SNL (Sandia National Laboratories) 2007a. *EBS Radionuclide Transport Abstraction.* ANL-WIS-PA-000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071004.0001; LLR.20080414.0023.

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

SNL 2008a. *Multiscale Thermohydrologic Model.* ANL-EBS-MD-000049 REV 03 AD 02. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20080201.0003; LLR.20080403.0162; LLR.20080617.0077.

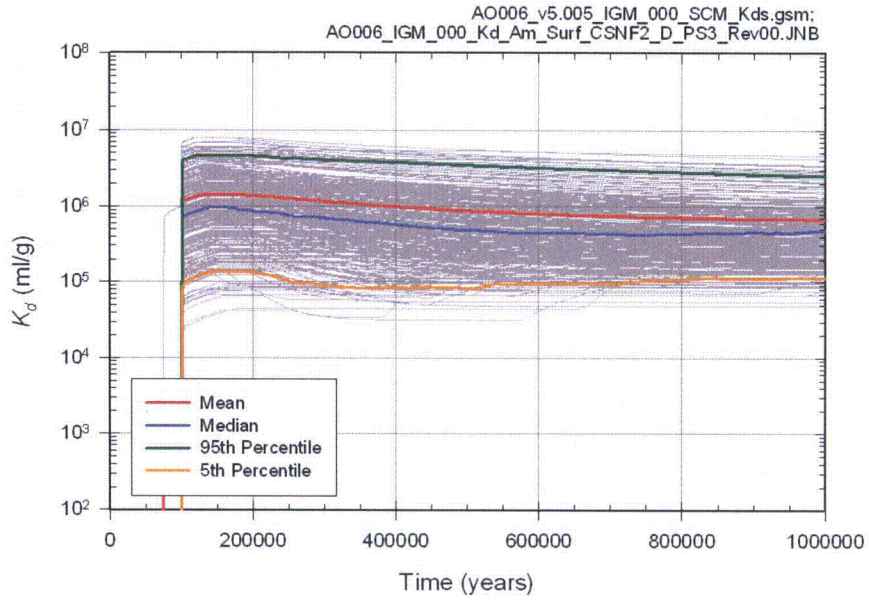
SNL 2008b. *Total System Performance Assessment Model /Analysis for the License Application.* MDL-WIS-PA-000005 REV 00 AD 01. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20080312.0001; LLR.20080414.0037; LLR.20080507.0002; LLR.20080522.0113; DOC.20080724.0005; DOC.20090106.0001[a].

NOTE: [a]Provided as an enclosure to letter from Williams to Sulima dtd 02/17/2009. "Yucca Mountain – Request for Additional Information Re: License Application (Safety Analysis Report Section 2.1), Safety Evaluation Report Volume 3 – Postclosure Chapters 2.2.1.1 and 2.2.1.3.7 – Submittal of Department of Energy Reference Citations."

5. LIST OF ATTACHMENTS

One DVD containing all files from DTN: SN0703PAEBSRTA.002, "Surface Complexation Modeling Results, Actinide K_d Abstractions and pH Abstraction" provided as Enclosure 5 to the transmittal letter.

(a)



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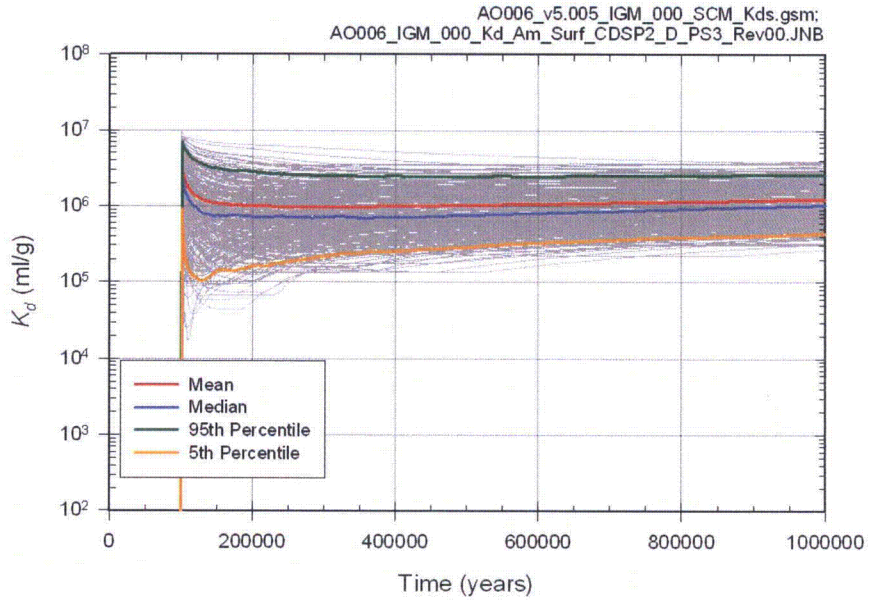
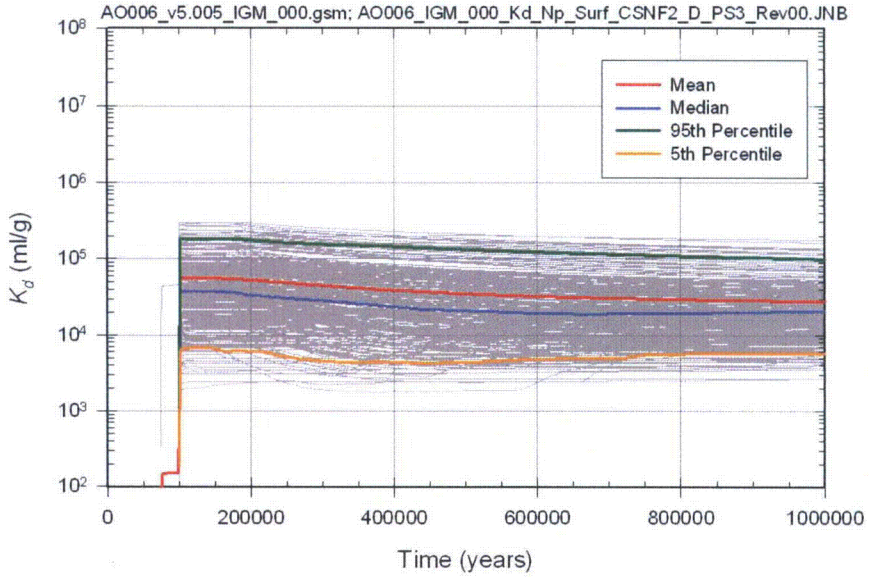


NOTE: The release prior to 100,000 years observed for the commercial SNF waste package is due to a general corrosion failure prior to the igneous event.

Figure 1. Americium K_d Value in the Corrosion Products Domain in the Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion Event at 100,000 Years for (a) Commercial SNF and (b) Codisposal Waste Packages

(a)



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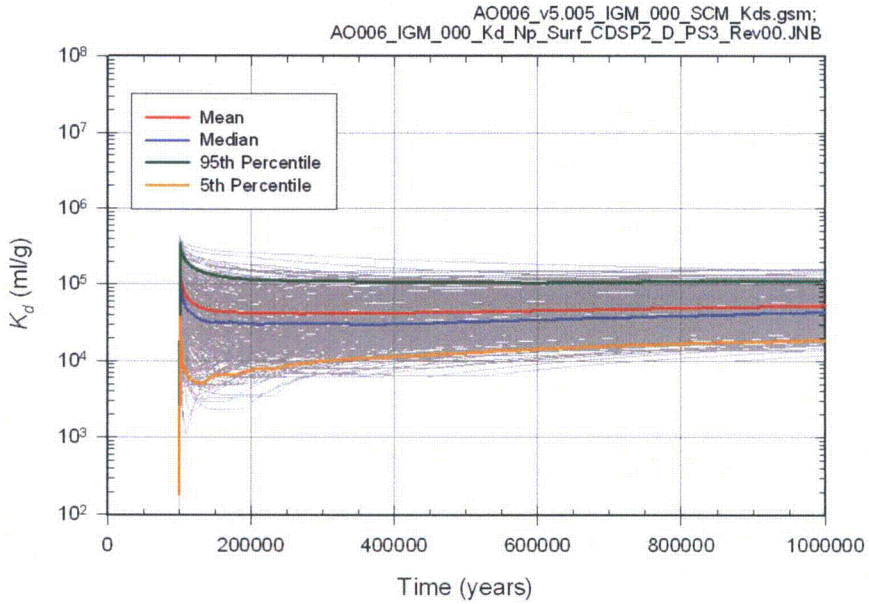
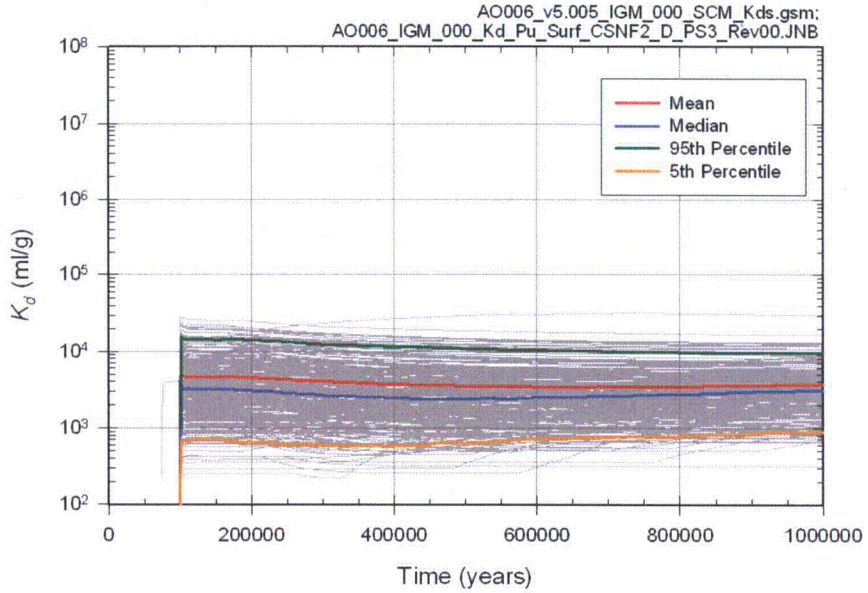


NOTE: The release prior to 100,000 years observed for the commercial SNF waste package is due to a general corrosion failure prior to the igneous event.

Figure 2. Neptunium K_d Value in the Corrosion Products Domain in the Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion Event at 100,000 Years for (a) Commercial SNF and (b) Codisposal Waste Packages

(a)



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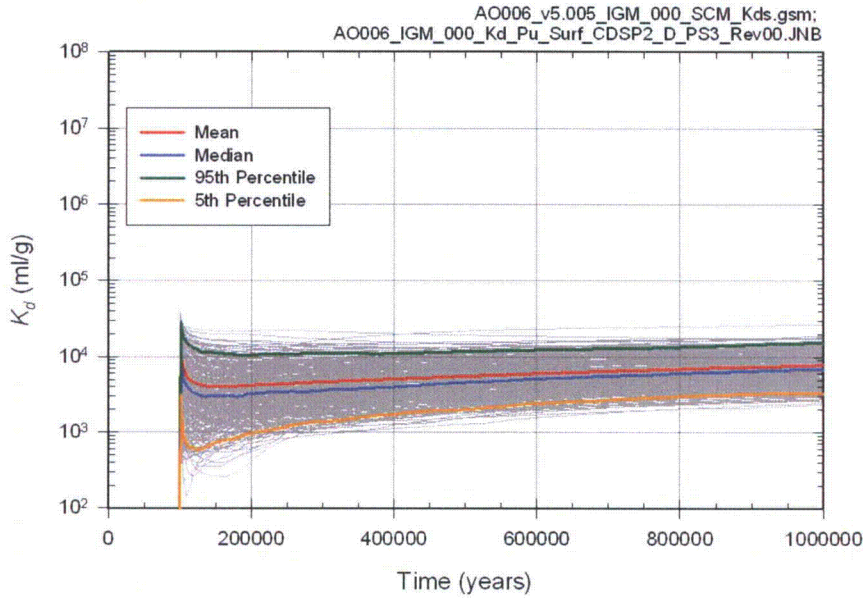
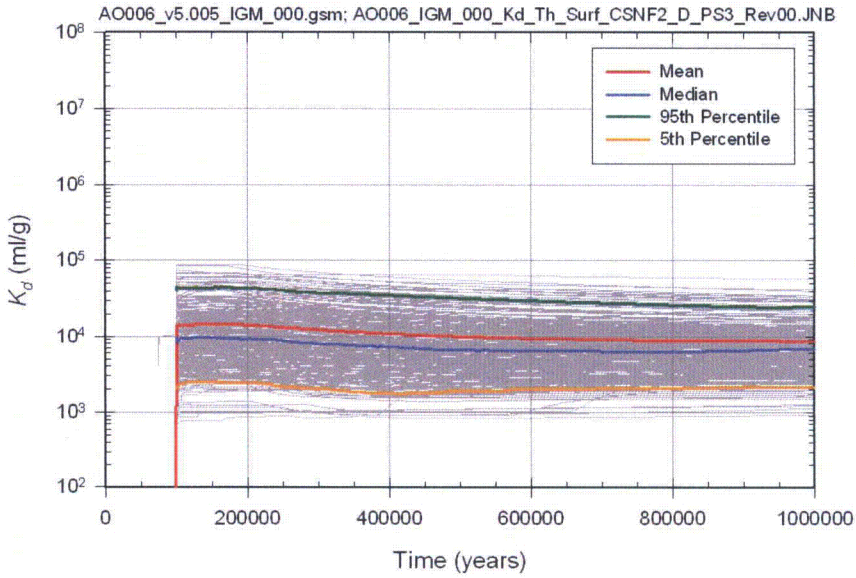


NOTE: The release prior to 100,000 years observed for the commercial SNF waste package is due to a general corrosion failure prior to the igneous event.

Figure 3. Plutonium K_d Value in the Corrosion Products Domain in the Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion Event at 100,000 Years for (a) Commercial SNF and (b) Codisposal Waste Packages

(a)



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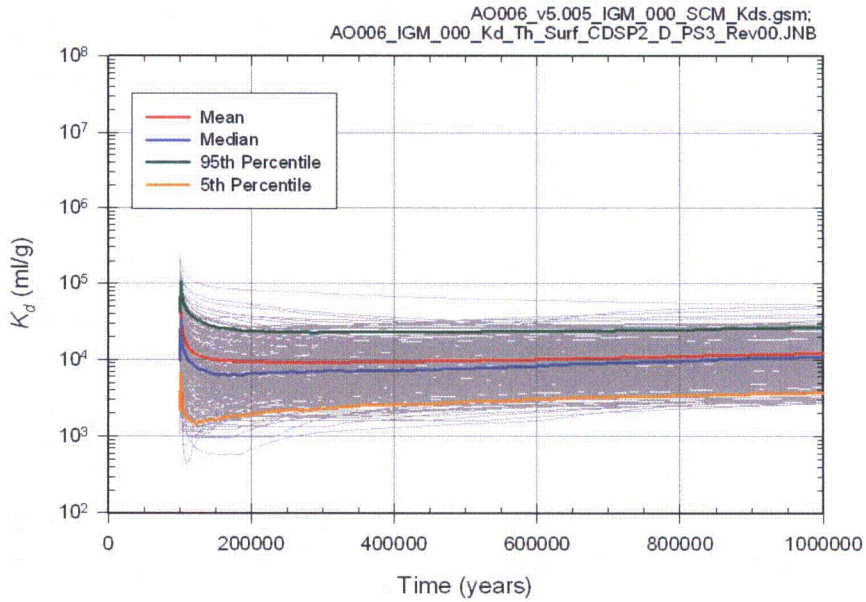
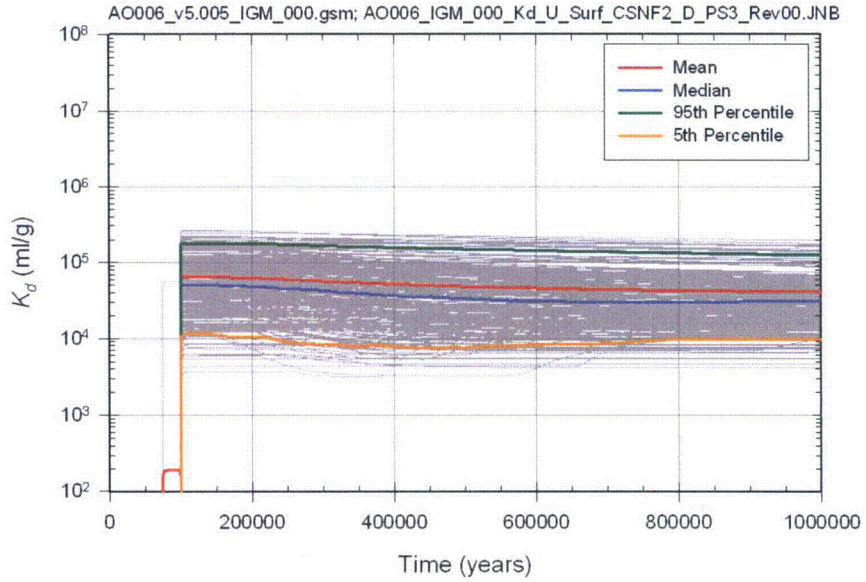


NOTE: The release prior to 100,000 years observed for the commercial SNF waste package is due to a general corrosion failure prior to the igneous event.

Figure 4. Thorium K_d Value in the Corrosion Products Domain in the Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion Event at 100,000 Years for (a) Commercial SNF and (b) Codisposal Waste Packages

(a)



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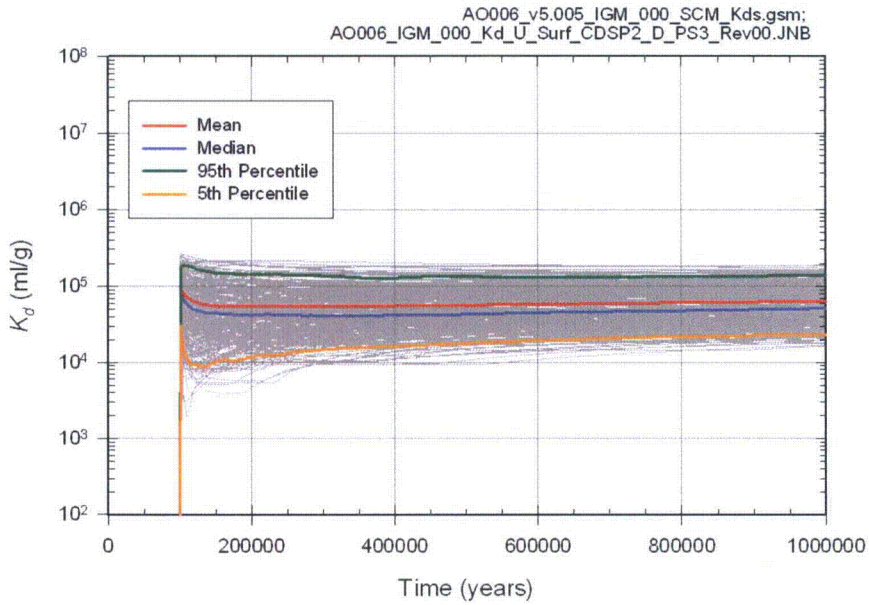

NOTE: The release prior to 100,000 years observed for the commercial SNF waste package is due to a general corrosion failure prior to the igneous event.

Figure 5. Uranium K_d Value in the Corrosion Products Domain in the Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion Event at 100,000 Years for (a) Commercial SNF and (b) Codisposal Waste Packages

RAI Volume 3, Chapter 2.2.1.3.4, Third Set, Number 3:

Provide the results from the PHREEQC surface complexation models and related files in Data Tracking Number (DTN) SN0703PAEBSRTA.002 - "Surface Complexation Modeling Results, Actinide Kd abstractions and pH abstraction" and the zip file: EBS-RTA-SURFCPLX_DTN.ZIP that support the actinide K_d distributions for the stationary corrosion products. In particular, provide the surface complexation model results and the support for the ranges and combinations of parameter values that produced the actinide K_d values at the upper end of the distribution.

LISTING OF THE ELECTRONIC FILES INCLUDED ON OPTICAL STORAGE MEDIA

This enclosure contains the listing of the electronic files included on optical storage media (OSM) that accompany the response to RAI 3.2.2.1.3.4-3-003 provided in Enclosure 3. An OSM, provided as Enclosure 5, contains the results from the PHREEQC surface complexation model and all files in Data Tracking Number (DTN) SN0703PAEBSRTA.002, Rev 001.

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08/21/2009  03:24 PM            20,375 Aqueo_comp.xls
08/21/2009  03:26 PM        10,065,913 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:26 PM            27,939 Stat_col_ZPC_05.xls
               4 File(s)     10,164,294 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\06
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:27 PM            20,375 Aqueo_comp.xls
08/21/2009  03:29 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:29 PM            27,939 Stat_col_ZPC_06.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\07
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:31 PM            20,375 Aqueo_comp.xls
08/21/2009  03:33 PM        10,066,631 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:33 PM            27,939 Stat_col_ZPC_07.xls
               4 File(s)     10,165,012 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\08
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:34 PM            20,375 Aqueo_comp.xls
08/21/2009  03:36 PM        10,065,913 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:36 PM            27,939 Stat_col_ZPC_08.xls
               4 File(s)     10,164,294 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\09
```

```
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:38 PM            20,375 Aqueo_comp.xls
08/21/2009  03:40 PM        10,066,465 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:40 PM            27,939 Stat_col_ZPC_09.xls
               4 File(s)     10,164,846 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\10
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:41 PM            20,375 Aqueo_comp.xls
08/21/2009  03:43 PM        10,065,913 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:43 PM            27,939 Stat_col_ZPC_10.xls
               4 File(s)     10,164,294 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\11
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:44 PM            20,375 Aqueo_comp.xls
08/21/2009  03:46 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:46 PM            27,939 Stat_col_ZPC_11.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\12
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:48 PM            20,375 Aqueo_comp.xls
08/21/2009  03:50 PM        10,066,916 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:50 PM            27,939 Stat_col_ZPC_12.xls
               4 File(s)     10,165,297 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\13
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:51 PM            20,375 Aqueo_comp.xls
08/21/2009  03:53 PM        10,065,545 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  03:53 PM            27,939 Stat_col_ZPC_13.xls
               4 File(s)     10,163,926 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\14
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:54 PM            20,375 Aqueo_comp.xls
08/21/2009  03:56 PM        10,066,181 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,068 Stat_col_ZPC.pqi
```

```
08/21/2009  03:56 PM            27,939 Stat_col_ZPC_14.xls
               4 File(s)     10,164,563 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\15
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  03:58 PM            20,375 Aqueo_comp.xls
08/21/2009  04:00 PM        10,066,465 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:00 PM            27,939 Stat_col_ZPC_15.xls
               4 File(s)     10,164,846 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\16
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:01 PM            20,375 Aqueo_comp.xls
08/21/2009  04:03 PM        10,065,361 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:03 PM            27,939 Stat_col_ZPC_16.xls
               4 File(s)     10,163,742 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\17
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:04 PM            20,375 Aqueo_comp.xls
08/21/2009  04:07 PM        10,066,650 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,068 Stat_col_ZPC.pqi
08/21/2009  04:07 PM            27,939 Stat_col_ZPC_17.xls
               4 File(s)     10,165,032 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\18
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:08 PM            20,375 Aqueo_comp.xls
08/21/2009  04:10 PM        10,065,544 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,066 Stat_col_ZPC.pqi
08/21/2009  04:10 PM            27,939 Stat_col_ZPC_18.xls
               4 File(s)     10,163,924 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\19
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:11 PM            20,375 Aqueo_comp.xls
08/21/2009  04:12 PM        10,065,545 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:12 PM            27,939 Stat_col_ZPC_19.xls
               4 File(s)     10,163,926 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\20
```

```
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:13 PM            20,375 Aqueo_comp.xls
08/21/2009  04:15 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:15 PM            27,939 Stat_col_ZPC_20.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\21
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:16 PM            20,375 Aqueo_comp.xls
08/21/2009  04:18 PM        10,066,096 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,066 Stat_col_ZPC.pqi
08/21/2009  04:18 PM            27,939 Stat_col_ZPC_21.xls
               4 File(s)     10,164,476 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\22
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:19 PM            20,375 Aqueo_comp.xls
08/21/2009  04:20 PM        10,065,545 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:20 PM            27,939 Stat_col_ZPC_22.xls
               4 File(s)     10,163,926 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\23
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:21 PM            20,375 Aqueo_comp.xls
08/21/2009  04:23 PM        10,065,730 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,068 Stat_col_ZPC.pqi
08/21/2009  04:23 PM            27,939 Stat_col_ZPC_23.xls
               4 File(s)     10,164,112 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\24
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:24 PM            20,375 Aqueo_comp.xls
08/21/2009  04:26 PM        10,065,707 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:26 PM            27,939 Stat_col_ZPC_24.xls
               4 File(s)     10,164,088 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\25
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:27 PM            20,375 Aqueo_comp.xls
08/21/2009  04:28 PM        10,065,360 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,066 Stat_col_ZPC.pqi
```

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08/21/2009  04:28 PM            27,939 Stat_col_ZPC_25.xls
               4 File(s)     10,163,740 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\26
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:29 PM            20,375 Aqueo_comp.xls
08/21/2009  04:31 PM        10,066,833 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:31 PM            27,939 Stat_col_ZPC_26.xls
               4 File(s)     10,165,214 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\27
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:32 PM            20,375 Aqueo_comp.xls
08/21/2009  04:34 PM        10,066,097 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:34 PM            27,939 Stat_col_ZPC_27.xls
               4 File(s)     10,164,478 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\28
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:35 PM            20,375 Aqueo_comp.xls
08/21/2009  04:37 PM        10,066,281 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:37 PM            27,939 Stat_col_ZPC_28.xls
               4 File(s)     10,164,662 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\29
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:38 PM            20,375 Aqueo_comp.xls
08/21/2009  04:39 PM        10,065,545 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:39 PM            27,939 Stat_col_ZPC_29.xls
               4 File(s)     10,163,926 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\30
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:41 PM            20,375 Aqueo_comp.xls
08/21/2009  04:42 PM        10,066,259 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:42 PM            27,939 Stat_col_ZPC_30.xls
               4 File(s)     10,164,640 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\31
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```
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:43 PM            20,375 Aqueo_comp.xls
08/21/2009  04:45 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:45 PM            27,939 Stat_col_ZPC_31.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\32
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:46 PM            20,375 Aqueo_comp.xls
08/21/2009  04:47 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:47 PM            27,939 Stat_col_ZPC_32.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\33
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:48 PM            20,375 Aqueo_comp.xls
08/21/2009  04:50 PM        10,065,361 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:50 PM            27,939 Stat_col_ZPC_33.xls
               4 File(s)     10,163,742 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\34
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:51 PM            20,375 Aqueo_comp.xls
08/21/2009  04:53 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:53 PM            27,939 Stat_col_ZPC_34.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\35
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:54 PM            20,375 Aqueo_comp.xls
08/21/2009  04:55 PM        10,066,079 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  04:55 PM            27,939 Stat_col_ZPC_35.xls
               4 File(s)     10,164,460 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\36
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:56 PM            20,375 Aqueo_comp.xls
08/21/2009  04:58 PM        10,065,544 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,066 Stat_col_ZPC.pqi
```

```
08/21/2009  04:58 PM            27,939 Stat_col_ZPC_36.xls
               4 File(s)     10,163,924 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\37
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  04:59 PM            20,375 Aqueo_comp.xls
08/21/2009  05:00 PM        10,065,728 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,066 Stat_col_ZPC.pqi
08/21/2009  05:00 PM            27,939 Stat_col_ZPC_37.xls
               4 File(s)     10,164,108 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\38
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:01 PM            20,375 Aqueo_comp.xls
08/21/2009  05:03 PM        10,066,834 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,068 Stat_col_ZPC.pqi
08/21/2009  05:03 PM            27,939 Stat_col_ZPC_38.xls
               4 File(s)     10,165,216 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\39
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:04 PM            20,375 Aqueo_comp.xls
08/21/2009  05:06 PM        10,065,545 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:06 PM            27,939 Stat_col_ZPC_39.xls
               4 File(s)     10,163,926 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\40
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:07 PM            20,375 Aqueo_comp.xls
08/21/2009  05:08 PM        10,066,815 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:08 PM            27,939 Stat_col_ZPC_40.xls
               4 File(s)     10,165,196 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\41
09/04/2009  10:22 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:09 PM            20,375 Aqueo_comp.xls
08/21/2009  05:11 PM        10,065,546 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,068 Stat_col_ZPC.pqi
08/21/2009  05:11 PM            27,939 Stat_col_ZPC_41.xls
               4 File(s)     10,163,928 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\42
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```
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:12 PM            20,375 Aqueo_comp.xls
08/21/2009  05:14 PM        10,065,361 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:14 PM            27,939 Stat_col_ZPC_42.xls
               4 File(s)     10,163,742 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\43
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:15 PM            20,375 Aqueo_comp.xls
08/21/2009  05:16 PM        10,065,361 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:16 PM            27,939 Stat_col_ZPC_43.xls
               4 File(s)     10,163,742 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\44
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:17 PM            20,375 Aqueo_comp.xls
08/21/2009  05:19 PM        10,065,913 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:19 PM            27,939 Stat_col_ZPC_44.xls
               4 File(s)     10,164,294 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\45
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:20 PM            20,375 Aqueo_comp.xls
08/21/2009  05:21 PM        10,065,913 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:21 PM            27,939 Stat_col_ZPC_45.xls
               4 File(s)     10,164,294 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\46
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:22 PM            20,375 Aqueo_comp.xls
08/21/2009  05:24 PM        10,065,359 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,065 Stat_col_ZPC.pqi
08/21/2009  05:24 PM            27,939 Stat_col_ZPC_46.xls
               4 File(s)     10,163,738 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\47
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:25 PM            20,375 Aqueo_comp.xls
08/21/2009  05:27 PM        10,065,913 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
```

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08/21/2009  05:27 PM            27,939 Stat_col_ZPC_47.xls
               4 File(s)     10,164,294 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\48
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:28 PM            20,375 Aqueo_comp.xls
08/21/2009  05:29 PM        10,065,728 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,066 Stat_col_ZPC.pqi
08/21/2009  05:29 PM            27,939 Stat_col_ZPC_48.xls
               4 File(s)     10,164,108 bytes
               2 Dir(s)               0 bytes free

 Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\49
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:30 PM            20,375 Aqueo_comp.xls
08/21/2009  05:32 PM        10,066,097 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:32 PM            27,939 Stat_col_ZPC_49.xls
               4 File(s)     10,164,478 bytes
               2 Dir(s)               0 bytes free

Directory of D:\DTN SN0703PAEBSRTA.002 Rev 001\EBS_RTA_SURFCPLX_DTN\Runs\50
09/04/2009  10:21 AM    <DIR>          .
09/04/2009  10:22 AM    <DIR>          ..
08/21/2009  05:33 PM            20,375 Aqueo_comp.xls
08/21/2009  05:34 PM        10,065,729 Stat_col_ZPC.out
08/21/2009  03:03 PM            50,067 Stat_col_ZPC.pqi
08/21/2009  05:34 PM            27,939 Stat_col_ZPC_50.xls
               4 File(s)     10,164,110 bytes
               2 Dir(s)               0 bytes free

     Total Files Listed:
             220 File(s)    545,439,138 bytes
              54 Dir(s)
```