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

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

1. SUPPLEMENTAL RESPONSE

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

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

Figure 1 shows that the plutonium mass difference in realization 2855 (epistemic uncertainty vector 286) is similar to the 5th percentile plutonium mass difference for the set of 300 epistemic uncertainty vectors, indicating that realization 2855 is near one extreme of the outcomes. However, as indicated by the mean (red) curve in Figure 1, the mean of the differences is approximately zero over the entire simulation time of 1,000,000 years. This indicates that, on average over the entire range of epistemic uncertainty, the plutonium mass settled as waste form colloids is approximately equal to the plutonium mass sorbed onto stationary corrosion products.
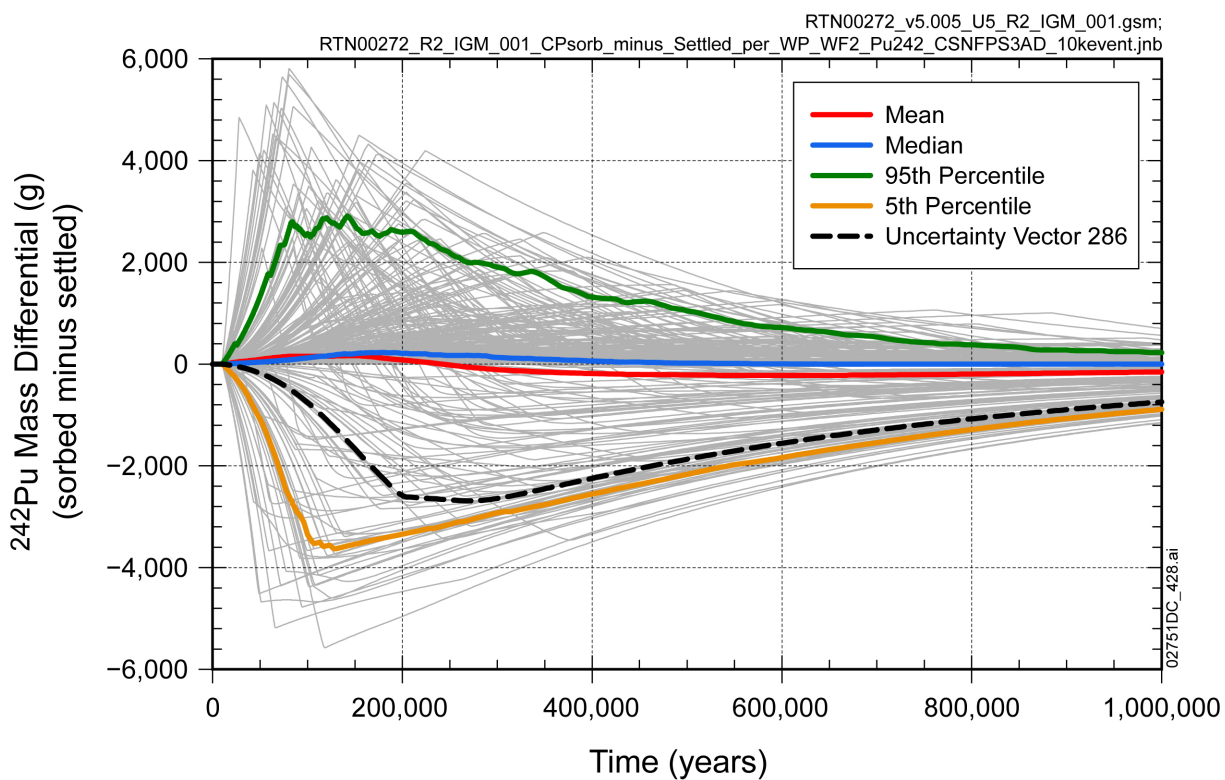
Figure 1.   Difference in Mass of ^{242}Pu in the Corrosion Products Domain (mass sorbed onto stationary corrosion products minus mass irreversibly attached to settled waste form colloids) of a Commercial SNF Waste Package in a Seeping Environment of Percolation Subregion 3 after an Igneous Intrusion at 10,000 Years after Repository Closure, for the 300 Epistemic Uncertainty Vectors

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.

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

> Describe why the iron oxide colloids associated with the plutonium mass would settle in the invert domain (Fig. 1.1-27 in the response to RAI 3.2.2.1.3.4-2-007) if their stability would be the same as in the upstream corrosion products domain for the igneous intrusion modeling case (DOE's response to RAI 3.2.2.1.3.4-2-007). Describe why the mass of plutonium associated with settled iron oxide colloids ends at about 250,000 years.

1. RESPONSE

Figure 1.1-27 of the DOE response to RAI 3.2.2.1.3.4-2-007 shows that a small amount (less than 20 grams) of ^{242}Pu is associated with settled iron oxyhydroxide colloid suspensions in the invert of commercial spent nuclear fuel (SNF) waste packages in the seeping environment of percolation subregion 3 after an igneous intrusion at 10,000 years. However, this amount is the total estimated mass for all commercial SNF waste packages (2,369) in this environment. Thus, the actual mass per waste package is much smaller (less than 0.01 g per waste package). In this particular realization (#2855), settled colloids are not due to a predicted change in colloid stability between the corrosion products domain and the invert domain (which are assumed to have the same chemical environment when advective flow through the waste packages occurs). Rather, it is the effect of iron oxyhydroxide colloid behavior as modeled in the total system performance assessment (TSPA) model.

Irreversible sorption of plutonium onto iron oxyhydroxide colloids occurs in the corrosion products domain of the TSPA model and is described by a kinetic sorption model (as discussed in SAR Section 2.3.7.12.1). Kinetic sorption reactions for plutonium onto iron oxyhydroxide colloids do not occur in the invert. The only effect the invert can have on the aqueous concentration of plutonium transported on iron oxyhydroxide colloids is through the stability of the colloid suspension, which is a function of pH and ionic strength. The solubility feature in GoldSim is used to constrain iron oxyhydroxide colloid suspension stability in the invert domain. This is accomplished by setting the solubility of plutonium and americium that is irreversibly sorbed to iron oxyhydroxide colloids (designated as separate "If" species in the Engineered Barrier System submodel of the TSPA model) equal to the concentration of ^{242}Pu associated with iron oxyhydroxide colloids in the upstream corrosion products domain times the ratio of the concentration of iron oxyhydroxide colloid particles in the invert domain to the concentration of iron oxyhydroxide colloid particles in the corrosion products domain. If colloids were to become unstable in the invert, this implementation will cause them to flocculate. The resulting concentration of plutonium and americium irreversibly sorbed to iron oxyhydroxide colloids in aqueous solution in the invert will reflect the very low colloid concentrations associated with unstable colloid suspensions.

This formulation was based on an assumption of steady-state conditions. Therefore, the formulation can produce slightly nonphysical behavior in transient conditions. Specifically, the phenomenon of slight settling of colloids in the invert, seen in Figure 1.1-27 of the DOE response to RAI 3.2.2.1.3.4-2-007, is a result of the numerical solution scheme in GoldSim, which is implicit in concentrations but explicit in other quantities such as solubilities or mass of

corrosion products. A semi-implicit method was implemented by setting the solubility (i.e., stability) of the iron oxyhydroxide colloids in the invert to the previous time value of the upstream concentration. As a result of the semi-implicit method, when the concentration of ^{242}Pu associated with iron oxyhydroxide colloids is increasing in the upstream corrosion products domain, this one-time step delay results in the maximum allowed concentration in the invert (i.e., the solubility or colloid stability limit) being lower than the current concentration in the corrosion products domain. Because the concentration of colloids in water exiting the corrosion products domain exceeds the maximum concentration permitted in the invert, some settling of the iron oxyhydroxide colloid suspension is observed. Conversely, when the concentration of ^{242}Pu associated with iron oxyhydroxide colloids in the corrosion products domain begins to decrease, the maximum concentration in the invert is greater than the concentration in the corrosion products domain, resulting in resuspension of settled colloid masses and subsequent transport from the invert, until the settled mass is exhausted, which happens at around 250,000 years. The red dotted line in Figure 1.1-26 of the DOE response to RAI 3.2.2.1.3.4-2-007 shows that the concentration of ^{242}Pu associated with iron oxyhydroxide colloids increases in the corrosion products domain until about 200,000 years, which results in the numerical settling phenomenon in the invert domain. After 200,000 years, the concentration decreases in the corrosion products domain because of the total depletion of ^{242}Pu mass in the waste form domain at 200,000 years, as seen on Figure 1.1-25 of the DOE response to RAI 3.2.2.1.3.4-2-007. As a result, the maximum concentration in the invert is greater than the concentration in the corrosion products domain, resulting in resuspension of settled colloid masses in the invert until the settled mass is exhausted.

The small amount of settled mass, however, is insignificant to the plutonium mass balance in the Engineered Barrier System. Specifically, as shown on Figures 1.1-21 and 1.1-22 of the DOE response to RAI 3.2.2.1.3.4-2-007, the mass of ^{242}Pu transported from the invert is dominated by dissolved and reversibly sorbed ^{242}Pu, with release rates of up to 10 g/yr. Thus, using a fully implicit method would not result in a significantly different mass of total ^{242}Pu exiting the invert, so the effect of using the semi-implicit method is negligible.

A minor clarification to the information presented in the NRC clarification call for ENG 4, Set 3 RAIs held on August 20, 2009, is needed. A verbal response to this supplemental question was presented. In that verbal response, the per package mass of ^{242}Pu that is associated with settled iron oxyhydroxide colloid suspensions in the invert of commercial SNF waste packages in the seeping environment of percolation subregion 3 was inadvertently reported to be 0.02 g per waste package. As presented above, the actual mass is less than 0.01 g per waste package.

2. COMMITMENTS TO NRC

None.

3. DESCRIPTION OF PROPOSED LA CHANGE

None.