This note book was prepared specifically to support the YM SAR review, and it is referenced in Volume 3, Chapter 7 of the SER. Therefore, it will need to be available in ADAMS before Volume 3 is published. NRC staff directed that the analysis contained in this notebook be removed from the SER and placed in this notebook.

SN 1052E Note.docx

SCIENTIFIC NOTEBOOK

SN 1052E

Title: Confirmatory Calculations of Radionuclide Release Rates and Colloid Transport Supporting Yucca Mountain Review

Project Number: 14002.01.354

by

David Pickett

Center for Nuclear Waste Regulatory Analyses
Southwest Research Institute
San Antonio, Texas

Table of Contents

Initial Entry

David Pickett
August 30, 2010

This notebook was created during final preparation of Yucca Mountain Safety Evaluation Report (SER), Volume 3, Chapter 7 – Radionuclide Release Rates and Solubility Limits. Nuclear Regulatory Commission (NRC) staff directed me to remove detailed discussions of confirmatory calculations from two locations within the chapter, record the calculations in a scientific notebook, and cite that notebook in the SER chapter to show where the detailed calculations could be viewed. This notebook serves that purpose and is now listed as a reference in Chapter 7. The text in this notebook is modified from what was removed from the draft SER chapter.

The entries in this notebook need to be considered in the context of the original calculations on which they are based, recorded in CNWRA notebooks SN 844E Volume 1 and 318E Volume 14.

The first set of calculations—Calculations of Plutonium Attachment to Colloids and Corrosion Products—is based on calculations originated by Hakan Basagaoglu and recorded in SN 844E. I also contributed to that notebook, and Section 5 of SN 844E shows my particular approach to the calculation which formed the basis for what is in this notebook. The purpose of these calculations is to evaluate how important iron oxyhydroxide (or iron oxide) colloids are, in TSPA, for plutonium release from the waste package.

The second set of calculations—Calculations of Estimated Radionuclide Release from Engineered Barrier System—are almost identical to those recorded by Scott Painter in SN 318E Volume 14. The purpose of these calculations is to estimate release rates of Tc-99, Pu-242, and Np-237 from the waste package, using simplified assumptions and equations, and DOE parameters. The purpose of these estimates is to compare them to TSPA outputs to get a sense of whether we understand what controls radionuclide release in TSPA. Starting from the Painter calculations, I have changed the value of one parameter, added more explanatory text, and added a solubility-limited calculation for neptunium.

Calculations of Plutonium Attachment to Colloids and Corrosion Products

David Pickett
August 30, 2010

This text is derived from a draft of the Yucca Mountain SER, Volume 3, Chapter 7; some of this text may remain in the chapter. The calculations are modified from those originally recorded by H. Basagaoglu and D. Pickett in SN 844E Volume 1. I acknowledge here H. Basagaoglu's contributions to the development of these calculations.

I report here independent, simplified, confirmatory calculations on the effectiveness of iron oxide colloids in facilitating Pu-242 releases in the igneous intrusion modeling case. The igneous intrusion modeling case was chosen for independent calculations because (i) chemical conditions and colloid stability remain unchanged throughout the entire simulation after a relatively short cooling period (less than 1,000 years), (ii) this modeling case dominates the long-term total mean annual dose, and (iii) that dose is dominated by Pu-242 after 200,000 years. According to SNL Equation 6.5.1.2-14 (2007aj), in the applicant's TSPA model, the rate of reversible, kinetic plutonium sorption from solution to stationary corrosion products (kg of plutonium per m^3 water per year) is

$$Q_{Pus}^{kinetic} = \rho_b s_{CP} k_{Puf} C_{Pu} \tag{1}$$

where ρ_b is the density of corrosion products on a bulk volume basis (kg /m^3), s_{CP} is the specific surface area of corrosion products (m^2/kg), k_{Puf} is the forward sorption rate constant (m/yr), and C_{Pu} is the concentration of plutonium in water on a water volume basis (kg/m^3).

According to SNL Equation 6.5.1.2-20 (2007aj), in the TSPA model, the rate of irreversible, kinetic plutonium sorption from solution to iron oxide colloids (kg of plutonium per m^3 water per year) is

$$Q_{Pucm}^{kinetic} = \theta_w C_{cFe0} s_{cFe0} k_{Puf} C_{Pu} \tag{2}$$

where θ_w is the volumetric water content (dimensionless), C_{cFeO} is the concentration of iron oxide colloids on a water volume basis (kg/m^3), and s_{cFeO} is the specific surface area of iron oxide colloids (m^2/kg). It is reasonable that the forward sorption rate constant k_{Puf} is the same for both stationary corrosion products and corrosion-product-derived iron oxide colloids (SNL, 2007aj, page 6-152).

The ratio between the rate of irreversible sorption to iron oxide colloids and the forward sorption rate onto stationary corrosion products, assuming the specific surface areas of stationary corrosion products and iron oxide colloids are the same (SNL, 2007aj), is

$$\frac{Q_{Pucm}^{kinetic}}{Q_{Pus}^{kinetic}} = \frac{\theta_w C_{cFe0} s_{cFe0} k_{Puf} C_{Pu}}{\rho_b s_{CP} k_{Puf} C_{Pu}} = \frac{\theta_w C_{cFe0}}{\rho_b} \tag{3}$$

Assuming a water-saturated system, a reasonable value for the volumetric water content θ_w is 0.4, according to SNL p. 6-93 (2007aj). The mean value for the iron oxide colloid concentration C_{cFeO} in the corrosion products domain for the igneous intrusion modeling case is 0.001 kg/m^3 [1 ppm], according to DOE Enclosure 3, Page 5 (2009ay). As for bulk density ρ_b, the corrosion products are made up of goethite (45 to 80 percent) and hydrous ferric oxide (20 to 55 percent) (SNL, 2007aj, Table 6.5-7). Grain densities for these are, respectively, 4,260 and 3,960 kg/m^3 [266 and 247 lb/ft^3] according to SNL Table 6.5-6 (2007aj). Using the midpoints of the two mineral abundance ranges yields a combined corrosion product grain density of 4,150 kg/m^3 [259 lb/ft^3] and, multiplying by the 0.6 volume fraction, a bulk density ρ_b of 2,490 kg/m^3 [155 lb/ft^3]. Using these values in Equation 3 yields a ratio of sorption rates of 1.6×10^{-7}. This calculation shows that the rate of plutonium attachment to stationary corrosion products is much higher than the rate of attachment to iron oxide colloids, in terms of mass per water volume.

The applicant showed in DOE Enclosure 8 (2009ay) that reversible, kinetic plutonium sorption onto stationary corrosion products can be approximated as an equilibrium process. The rate of plutonium desorption from the stationary corrosion products is, therefore, approximately equal to the rate of sorption (see SN 844E Volume 1 Section 5). This observation, combined with the calculation in the previous paragraph, leads to the conclusion that the rate of irreversible plutonium sorption to iron oxide colloids is many orders of magnitude slower than the rate of plutonium desorption from stationary corrosion products to solution. Therefore, any transfer of dissolved plutonium to iron oxide colloids will be, essentially, instantaneously compensated by desorption from the stationary corrosion products—which contain the vast majority of plutonium mass in the corrosion products domain—to maintain the quasi-equilibrium relationship. It is clear from this calculation that irreversible sorption of plutonium to iron oxide colloids cannot substantially deplete dissolved plutonium.

For codisposal packages in disruptive scenarios, I compare the plutonium release effectiveness of HLW glass colloids against dissolved plutonium. Results of a representative igneous intrusion modeling case realization in DOE Enclosure 1, Figures 1.1-29 and 1.1-30 (2009dc) showed dissolved Pu-242 being released from the engineered barrier system more than ten times faster than Pu-242 associated with HLW glass colloids. In addition, the mean plutonium solubility limit in the corrosion products domain in TSPA, estimated as 10^{-5} g/L [0.01 ppm] from applicant data presented in SNL Table 6.5-1 (2007ah) (this value was determined in SN 318E Volume 14, and I confirmed it myself), is about 10 times higher than the mean concentration of plutonium associated with HLW glass colloids, calculated as 1.2×10^{-6} g/l [0.0012 ppm], which is the mean of the distribution shown in SNL Table 6-4 (2008ak). (Note that the units in Table 6-4 are mol/L – not the mL/g shown in the Table title.)

References

DOE. 2009ay. "Yucca Mountain—Response to Request for Additional Information Regarding License Application (Safety Analysis Report Section 2.3.7), Safety Evaluation Report, Vol. 3, Chapter 2.2.1.3.4, Set 2." Letter (May 12) J.R. Williams to J.H. Sulima (NRC). ML 091330282. Washington, DC: DOE, Office of Technical Management.

DOE. 2009dc. "Yucca Mountain—Response to Request for Additional Information Regarding License Application (Safety Analysis Report Section 2.3.7), Safety Evaluation Report Vol. 3, Chapter 2.2.1.3.4, Set 2." Letter (May 28) J.R. Williams to J.H. Sulima (NRC). ML091480752. Washington, DC: DOE, Office of Technical Management.

SNL. 2008ak. "Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary." MDL–EBS–PA–000004. Rev. 03. ERD 01. Las Vegas, Nevada: Sandia National Laboratories.

SNL. 2007ah. "Dissolved Concentration Limits of Elements with Radioactive Isotopes." ANL–WIS–MD–000010. Rev. 06. Las Vegas, Nevada: Sandia National Laboratories.

SNL. 2007aj. "EBS Radionuclide Transport Abstraction." ANL–WIS–PA–000001. Rev. 03. ERD 01. Las Vegas, Nevada: Sandia National Laboratories.

Calculations of Estimated Radionuclide Release from Engineered Barrier System

David Pickett
August 30, 2010

This text is derived from a draft of the Yucca Mountain SER, Volume 3, Chapter 7; some of this text may remain in the chapter. The calculations are modified from those originally recorded by S. Painter in SN 318E Volume 14. I acknowledge here S. Painter's origination and development of these calculations and point the reader to that notebook for further background information. I use a different value for one parameter that yields different results, as noted below. These calculations are estimates only, using parameters and probabilities the applicant provided in the SAR, supporting reports, and associated RAI responses. Because simplifications are involved, precise agreement between these calculations and the applicant's results is not expected; rather, reasonable agreement of the release rates within an order of magnitude is evaluated.

We perform simplified confirmatory analyses to assess whether the applicant's TSPA results for the engineered barrier system radionuclide releases are consistent with the applicant's abstractions. As detailed in the following paragraphs, we use hand calculations to estimate peak expected total-repository engineered barrier system release rates for Tc-99, Pu-242, and Np-237 using applicant-provided information, and then compare these releases with applicant-provided values. The calculations focus on the igneous intrusion and seismic ground motion modeling cases because these cases result in the largest release rates from the engineered barrier system. These estimates are based on advection and diffusion of dissolved radionuclides and neglect transport of radionuclides associated with colloids. The calculations are presented here only for selected cases for which representative applicant results were available for comparison. Results are presented for waste package release of Pu-242 and Np-237 in the igneous intrusion modeling case, on the basis of control by solubility or corrosion product sorption, and of Tc-99 in the seismic ground motion modeling case, on the basis of control by waste package failure rate.

The applicant provided information showing that engineered barrier system releases of low-solubility, sorbing radionuclides (e.g., plutonium and neptunium) are mainly controlled by processes within the corrosion products domain because waste form dissolution and invert transport processes are fast relative to transport within the corrosion products domain. In the TSPA analyses, the important dose contributions from plutonium and neptunium isotopes result from the igneous intrusion modeling case (SAR Section 2.4.2.2.1.1.3), in which all waste packages fail and releases of these radionuclides are controlled by advection modified by sorption and precipitation of radionuclide-bearing minerals. We therefore perform confirmatory Pu-242 and Np-237 engineered barrier system release calculations only for the igneous intrusion modeling case. For these two radionuclides, information provided in DOE (2009dc) showed that igneous intrusion case releases from commercial SNF packages dominated over those from codisposal packages. We therefore perform Pu-242 and Np-237 confirmatory calculations only for commercial SNF packages. These results are considered to be estimates of peak mean releases because (i) we use mean values for important parameters such as solubility limit and sorption coefficient; (ii) we use peak waste package inventory values for both radionuclides, with no accounting for radioactive decay or waste depletion; and (iii) the entire radionuclide inventory was assumed available for sorption in the corrosion products domain.

For Pu-242 calculations in the igneous intrusion modeling case, we assume that engineered barrier system release rates are controlled by advection and either (i) sorption onto corrosion products or (ii) precipitation of solubility-limiting minerals in the corrosion products domain. On

the basis of the applicant's mixing cell abstraction, the sorption-limited release rate Q_{Sorp} (g/yr) is approximated by (SN 318E, Volume 14, page 7)

$$Q_{Sorp} = \frac{N_{fp} q M}{K_d M_{CP}} \quad (4)$$

where N_{fp} is the number of failed waste packages experiencing flow, q is the water flow rate per package (L/yr), M is the Pu-242 mass per waste package (g), K_d is the sorption coefficient (L/g), and M_{CP} is the mass of corrosion products per waste package. (Recall that plutonium sorption in the corrosion products domain can be treated as an equilibrium process.) Painter, in SN 318E Volume 14, described parameter development for these calculations using applicant-presented data; this development is recounted here. All packages experience flow in the igneous intrusion case, so a value of 8,213 is taken from the total CSNF and naval spent fuel packages in SNL Table 6.3.7-1 (2008ag) and multiplied by the 1.7 percent igneous event probability (over 1 million years) to arrive at 140 packages for N_{fp}. Flow rate q is the post-10,000-year percolation flux of 890 L/yr [240 gal/yr] as reported in DOE Enclosure 1, Table 1 (2010ai). The Pu-242 inventory per CSNF package M is 5,460 g [12.04 lb], according to SNL Table 6.3.7-5 (2008ag). (No decay correction is applied but, if it were, it would have a large effect only after several hundred thousand years.)

A mean plutonium K_d is estimated from information provided in DOE Enclosure 3 (2009da). This was done using the Microsoft Excel spreadsheet "Final_Calc_Results_Surf_Complx_Data_1.xls," which was provided with the RAI 3.2.2.1.3.4-3-003 response. The worksheet in that file called "Results_Surf_Complx_Model" tabulates the surface complexation model outputs of log K_d for plutonium on corrosion products – there are 4800 values. I calculated the average of these in the spreadsheet and obtained 3.384. $10^{3.384}$ = 2421 mL/g or 2.42 L/g for the plutonium K_d.

Once all steel has corroded, the mass of corrosion products in a package M_{CP} is 3.54×10^7 g [7.80×10^4 lb], according to SNL Table 6.3-8 (2007aj). This last value differs from what is in SN 318E Volume 14 because the mass of uncorroded steel was used there. Using these values in Equation 4 yields a peak mean repositorywide, sorption-limited plutonium release rate of 7.9 g/yr [0.017 lb/yr]. This differs from the value of 11.4 g/yr reported in SN 318E Volume 14 because of the different M_{CP} used.

The solubility-limited release rate Q_{Solub} (g/yr) is represented by (SN 318E, Volume 14, page 7)

$$Q_{Solub} = N_{fp} q C_{SL} \quad (5)$$

where C_{SL} is the plutonium solubility limit, a mean value for which is 10^{-5} g/L [0.01 ppm], estimated from applicant data presented in SNL Table 6.5-1 (2007ah). (This approach assumes that Pu-242 dominates the plutonium inventory, which is true for long times.) Values for N_{fp} and q are the same as used in the Equation 4 calculation. The peak mean repositorywide release rate so estimated from Equation 5 is 1.2 g/yr [0.0026 lb/yr], which is lower than the sorption-limited rate and is, therefore, taken as the estimated value.

The applicant provided representative commercial SNF package release results for percolation subarea 3 (which includes approximately 40 percent of the waste packages) and a single

realization from the igneous intrusion case in DOE Enclosure 1, Figure 2 (2009da). For Pu-242, the peak corrosion products domain release from subarea 3 is approximately 30 g/yr [0.07 lb/yr] at 100,000 years, conditional on an igneous event having occurred at 10,000 years. Scaling that value to the full repository (i.e., dividing by 0.4) and multiplying by the probability of having a single igneous intrusion event in 1 million years (1.7 percent or 0.017) gives a value of 1.3 g/yr [0.0029 lb/yr]. This applicant result agrees well with our confirmatory estimate of 1.2 g/yr [0.0026 lb/yr].

A repositorywide, sorption-limited release rate for Np-237 is also calculated using Equation 4, as described in SN 318E Volume 14. The mass M of Np-237 in a commercial SNF waste package is calculated by taking the initial inventory of 5,380 g [11.86 lb] the applicant reported in SNL Table 6.3.7-5 (2008ag) and increasing it to 15,580 g [34.35 lb] to account for decay of all initial Am-241. (The Am-241 half-life is 433 years; it will, therefore, be essentially totally decayed in a few thousand years. No decay correction is applied to Np-237, which has a half-life of more than 2 million years.)

A mean neptunium K_d is estimated from information provided in DOE Enclosure 3 (2009da). This was done using the Microsoft Excel spreadsheet "Final_Calc_Results_Surf_Complx_Data_1.xls," which was provided with the RAI 3.2.2.1.3.4-3-003 response. The worksheet in that file called "Results_Surf_Complx_Model" tabulates the surface complexation model outputs of log K_d for neptunium on corrosion products – there are 4800 values. I calculated the average of these in the spreadsheet and obtained 4.267. $10^{4.267}$ = 18,490 mL/g or 18.5 L/g for the neptunium K_d.

Values for N_{fp}, q, and M_{CP} are the same as were used for plutonium. Using these values in Equation 4, we calculate a peak mean repositorywide neptunium release rate of 3.0 g/yr [0.0066 lb/yr] for the igneous intrusion modeling case. This differs from the value of 4.3 g/yr reported in SN 318E Volume 14 because of the different M_{CP} used.

The applicant provided representative results for Np-237 release from the corrosion products domain in percolation subarea 3 and a single realization of the igneous intrusion modeling case in DOE Figure 1.1-12 (2009dc). That realization assumes that an igneous intrusion event occurs. Thus, the applicant's calculated peak release rate of approximately 200 g/yr [0.4 lb/yr] from that figure must be weighted by the probability of an igneous event in 1 million years (1.7 percent or 0.017) and scaled to the full repository (i.e., divided by 0.4) to compare with our peak mean estimate. The result of that calculation is 8.5 g/yr [0.019 lb/yr] for Np-237 release for the igneous intrusion modeling case. This applicant result is in reasonable agreement with our estimate of 3.0 g/yr [0.0066 lb/yr].

We calculate a solubility-limited neptunium engineered barrier system release rate using Equation 5 and a mean solubility limit of 10^{-4} g/L [0.1 ppm], estimated from the NpO_2 abstraction in SNL Figure 6.6-1 (2007ah). The calculated rate of 12 g/yr [0.026 lb/yr] is higher than the sorption-limited rate and is therefore not used for comparison; a rate calculated using the Np_2O_5 solubility model (applicable after all in-package steel has corroded) would be even higher and so would also not be used. (Note that this calculation was not performed in SN 318E Volume 14.)

For high-solubility radionuclides that are weakly sorbing or nonsorbing, radionuclide-specific engineered barrier system releases in the seismic ground motion and nominal modeling cases are controlled primarily by the waste package failure rate because dissolution rates of the waste form and transport of these radionuclides through the engineered barrier system are sufficiently

fast compared with typical intervals between package failures. As documented in SN 318E Volume 14, we calculate a peak mean release rate for Tc-99, which is representative of mobile high-solubility radionuclides, in the seismic ground motion case using the following equation

$$Q_{Tc} = NM_{Tc}r_f \quad (6)$$

where N is the number of waste packages, M_{Tc} is the Tc-99 mass in each package (g), and r_f is the fractional waste package failure rate (yr^{-1}). SAR Figure 2.1-24 showed that, in TSPA, mean Tc-99 releases from the engineered barrier system in the seismic ground motion modeling case occur relatively early. These releases are dominated by codisposal packages because few CSNF packages have failed prior to 100,000 years in the seismic ground motion case (SAR Figure 2.1-12). There are 3,416 codisposal packages in the modeled repository (N), according to SNL Table 6.3.7-1 (2008ag), each containing 1,168 g [2.575 lb] of Tc-99 (M_{Tc}), according to SNL Table 6.3.7-5 (2008ag). (In the context of these calculations, Tc-99 decay is not significant over 100,000 years, so no correction is applied.) For the time prior to 100,000 years, we estimate a maximum package failure rate r_f of 5.6×10^{-6} yr^{-1} by measuring the slope of the mean plot of waste package breaches in SAR Figure 2.1-12c. The calculated rate is considered a peak mean rate because the highest slope from the mean package failure history curve is used. Using these values in Equation 6, we calculate a peak mean Tc-99 release rate of 22 g/yr [0.049 lb/yr] for the seismic ground motion modeling case. The applicant provided plots of mean cumulative Tc-99 engineered barrier system release in SAR Figure 2.1-24. From the slopes of these plots, we estimate that the release rate peaks at approximately 12 g/yr [0.026 lb/yr] for the period between 10,000 and 100,000 years. This peak mean rate estimated from the applicant's Tc-99 release information is in reasonable agreement with our estimate of 22 g/yr [0.049 lb/yr].

References

DOE. 2010ai. "Yucca Mountain—Supplemental Response to Request for Additional Information Regarding License Application (Safety Analysis Report Section 2.3.1), Safety Evaluation Report Vol. 3, Chapter 2.2.1.3.5, Set 1 and (Safety Analysis Report Sections 2.3.2 and 2.3.3), Safety Evaluation Report Vol. 3, Chapter 2.2.1.3.6, Set 1." Letter (February 2) J.R. Williams to J.H. Sulima (NRC). ML100340034. Washington, DC: DOE, Office of Technical Management.

DOE. 2009da. "Yucca Mountain—Response to Request for Additional Information Regarding License Application (Safety Analysis Report Section 2.3.7), Safety Evaluation Report Vol. 3, Chapter 2.2.1.3.4, Set 3." Letter (September 11) J.R. Williams to J.H. Sulima (NRC). ML092600883. Washington, DC: DOE, Office of Technical Management.

DOE. 2009dc. "Yucca Mountain—Response to Request for Additional Information Regarding License Application (Safety Analysis Report Section 2.3.7), Safety Evaluation Report Vol. 3, Chapter 2.2.1.3.4, Set 2." Letter (May 28) J.R. Williams to J.H. Sulima (NRC). ML091480752. Washington, DC: DOE, Office of Technical Management.

SNL. 2008ag. "Total System Performance Assessment Model/Analysis for the License Application." MDL–WIS–PA–000005. Rev. 00. AD 01, ERD 01, ERD 02, ERD 03, ERD 04. Las Vegas, Nevada: Sandia National Laboratories.

SNL. 2007ah. "Dissolved Concentration Limits of Elements with Radioactive Isotopes." ANL–WIS–MD–000010. Rev. 06. Las Vegas, Nevada: Sandia National Laboratories.

SNL. 2007aj. "EBS Radionuclide Transport Abstraction." ANL–WIS–PA–000001. Rev. 03. ERD 01. Las Vegas, Nevada: Sandia National Laboratories.