	Model Error Resolution Document <i>Complete only applicable items.</i>		QA: QA Page 1 of 25
	1. Document Number: ANL-EBS-GS-000002	2. Revision/Addendum: REV 01 AD 01	3. ERD: 01
4. Title: Geochemistry Model Validation Report: External Accumulation Model		5. No. of Pages Attached: 24	
6. Description of and Justification for Change (Identify affected pages, applicable CRs and TBVs):			
<p>1. Introduction</p> <p>This document addresses condition report (CR) 14019 and CR 14087, to correct errors in the external accumulation model results, and CR 13006, to qualify DTN: MO0604SPAPHR25.001 [DIRS 176868] for intended use within the document <i>Geochemistry Model Validation Report: External Accumulation Model</i>.</p> <p>1.1 Description of CR 14019 and CR 14087, Summary of Action Taken, and Impact</p> <p>CR 14019 and CR 14087 both identified errors in one of the cases (<i>FFTFIG1adEhdec</i>) in which the external accumulation model was executed. CR 14087 was closed to CR 14019 since the corrective action and the impacted documents are the same. To correct the errors, the <i>FFTFIG1adEhdec</i> source term was rerun with EQ6 and the external accumulation model was implemented using the new source term. Due to the new results for <i>FFTFIG1adEhdec</i>, the following sections were updated: the sensitivity analyses in Section 6.8.1.2, the adsorption validation in Section 7.2.2.2, the results in Section 6 and 8 of the parent report, and the results in Section 6[a] of the addendum. During the sensitivity analysis update, the adsorption in case <i>TMI_IG1</i> was found to be incorrect, and therefore, that correction was also made. The corrected values of uranium and plutonium accumulation for cases <i>FFTFIG1adEhdec</i> and <i>TMI_IG1</i> are still lower than the minimum critical mass limit for accumulation in the invert; therefore there is no impact on nuclear safety. However, the corrections do have an impact on the maximum observed accumulation for FFTF and TMI spent nuclear fuel in the igneous scenario, which are listed in the following documents: ANL-DS0-NU-000001 Rev. 00 (SNL 2008 [DIRS 173869], Tables 4.1-9 and 4.1-10), ANL-WIS-MD-000027 Rev. 00 (SNL 2008 [DIRS 183041], Table 2.1.14.26.0A-1 and Table 2.2.14.12.0A-1), and the License Application (Table 2.2-14). The following documents cite this report (or one of the revised output DTNs), but are not impacted by the changes: ANL-EBS-GS-000001 Rev. 02, ANL-EBS-NU-000009 Rev. 00, ANL-EBS-PA-000013 Rev. 00, ANL-EBS-PA-000014 Rev. 00, TDR-DS0-NU-000002 REV 01, ANL-WIS-PA-000001 REV 03. The FEIS and SEIS do not cite this report or the impacted DTNs and are therefore not impacted by the ERD.</p>			

	Printed Name	Signature	Date
7. Checker	David Mohr Wendy Mitcheltree	<i>David E. Mohr</i> <i>Wendy Mitcheltree</i>	2/3/10
8. QCS/QA Reviewer	Brian Mitcheltree	<i>Brian Mitcheltree</i>	2/3/10
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SCI-PRO-006.3-R2

(Continued from Block 6)

1.2 Description of CR 13006, Summary of Action Taken, and Impact

As documented in CR 13006, the DTN: MO0604SPAPHR25.001 [DIRS 176868], was identified as “qualified” in the TDMS; however it was not developed in an approved QA document, such as analysis/model or calculation report, addendum, or error resolution document (ERD). The DTN has been placed behind the firewall in TDMS. In the data qualification report that was written to resolve the issue for this DTN and five other DTNs (SNL 2009 [DIRS 186283], p. vii), it was recommended that DTN: MO0604SPAPHR25.001 [DIRS 176868] not be qualified for general use because an updated version of the PHREEQC database (addendum output DTN: MO0704PHREEQC25.000) is available for current analyses. When *Geochemistry Model Validation Report: External Accumulation Model* was written, the older PHREEQC database (DTN: MO0604SPAPHR25.001 [DIRS 176868]) was the current database to use. In this ERD, the older PHREEQC database (DTN: MO0604SPAPHR25.001 [DIRS 176868]) is qualified for intended use within Section 4.1.14.5[a] of the addendum. Qualification is conducted by a sensitivity analysis designed to assess the impact on the external accumulation model results. This is done by comparing the results generated by using the original database to the results generated by using the updated version of the database. The results of the sensitivity analysis (presented later in ERD in Table L-1[a]) showed no impact on the results or on any other documents.

2. Software Used and Inputs

To address the CRs, the following qualified software, baselined by the Lead Laboratory, was run on CPU#: S887211, using the virtual Windows 2000 operating system: EQ6 V7.2bLV (STN 10075-7.2bLV-02 [DIRS 159731]), ASPRIN V1.0 (STN 10487-1.0-00 [DIRS 179458]), PHREEQC V2.3 (STN 10068-2.3-01 [DIRS 157837]), PHREEQC_Post V1.1 (STN 10723-1.1-00 [DIRS 157839]), Acc_with_decay V1.2 (STN 10499-1.2-00 [DIRS 157838]), and MinAcc V1.0 (STN: 10724-1.0-00 [DIRS 157841]). The virtual Windows 2000 operating system was created using VMWare Player V2.02, which is considered exempt because it is commercially available software such as operating systems, system utilities, compilers and their associated libraries, word processors, spreadsheets, database managers, Email, and other types of automated office support systems (support software) in and of themselves are not required to be qualified as stated in IM-PRO-003.

No additional inputs were required for the ERD.

3. Results of Addressing CR 14019 and CR 14087

The source term for case *FFTFIG1adEhdec* was rerun using EQ6 V7.2bLV (STN 10075-7.2bLV-02 [DIRS 159731]) and ASPRIN V1.0 (STN 10487-1.0-00 [DIRS 179458]), while suppressing the aqueous species $N_2(aq)$, which was necessary to accurately simulate the aqueous concentrations in the reduced oxygen conditions of the case, as explained in CR 14087. The external accumulation model was implemented using the new source term. The steps involved in implementing the external accumulation model were (1) plotting the concentrations of uranium and plutonium versus time from the source term and identifying ten points for the external accumulation model implementation, (2) running PHREEQC V2.3 (STN 10068-2.3-01 [DIRS 157837]) for each of the ten points, (3) post-processing the PHREEQC results using qualified software PHREEQC_Post V1.1 (STN: 10723-1.1-00 [DIRS 157839]), Acc_with_decay V1.2

(STN 10499-1.2-00 [DIRS 157838]), and MinAcc V1.0 (STN: 10724-1.0-00 [DIRS 157841]), and (4) recalculating the adsorption on the tuff. The output files in the DTN that are changed are indicated in Table 1.

Table 1. Changes in Document and Output DTNs

Output DTN	Changes
MO0609SPAINOUT.002	Replace file <i>FFTFig1adEhdec.zip</i> (dated 7/28/2006) with new one (dated 10/30/2009) that contains updated folders with the new results from EQ6, ASPRIN, PHREEQC, PHREEQC_Post, Acc_with_decay and MinAcc: FFTFG1_Acc_with_decay FFTFG1_Asprin_Sourceterm FFTFG1_MinAcc FFTFG1_Phreeqc_and_Phreeqc_post Replace file <i>Dissolved U and Pu acc total with sensitivities.xls</i> (dated 7/27/2006) with updated version (dated 10/26/2009). Replace <i>adsorption_calc.xls</i> (dated 9/27/2006) with updated version (dated 10/21/2009) of results from <i>FFTFig1adEhdec</i> and <i>TMI_IG1</i> . Add new file <i>Database Sensitivity.zip</i> (dated 10/30/2009) that contains folders FFTFG1_Acc_with_decay_updated_db FFTFG1_Phreeqc_and_phreeqc_post_updated_db and contains files <i>Adsorption_calc_updated_db.xls</i> <i>Dissolved U and Pu acc total_updated_db.xls</i>
MO0609SPASENSI.003	Replace file <i>FFTFig1adEhdec_logK_sens_5.8049.zip</i> (dated 7/27/2006) with new one (dated 10/30/2009) Replace file <i>FFTFig1adEhdec_logK_sens_6.1249.zip</i> (dated 7/27/2006) with new one (dated 10/30/2009) Add new files <i>TMI_IG1_logK_sens_5.8049.zip</i> (dated 10/30/2009) <i>TMI_IG1_logK_sens_6.1249.zip</i> (dated 10/30/2009) Replace file <i>sens_adsorption_calc.xls</i> (dated 7/27/2006) with new one (dated 10/21/2009)
MO0607SPADSORP.000	Replace file <i>fftfG1_10.936k adsval.xls</i> (dated 7/12/2006) with new file <i>fftfG1_10.2k adsval.xls</i> (dated 10/21/2009)

Since the boltwoodite-Na log K sensitivity study in Section 6.8.1.2 was based on the *FFTFig1adEhdec* source term, the sensitivity cases were rerun using the new source term. In the original case the uranium accumulation due to precipitation of boltwoodite-Na was 31.17 moles (Table 6-15). Increasing and decreasing the log K by one standard deviation resulted in $\pm 9.9\%$ and $+17.2\%$ differences in moles of uranium accumulated (Table 6-37). These same percentage differences were used to determine the uncertainty limits for the *TMI_IG1* source term in the file *Dissolved U and Pu acc total with sensitivities.xls* in output DTN MO0609SPAINOUT.002. The new results from the FFTF source term indicate the moles of uranium accumulated were only 0.708 moles (down from 31.17 moles), with a range from 0 to 5.97. This small range in moles is equivalent to large percentage differences of $\pm 100\%$ and $+743\%$. Rather than applying these percentage differences to TMI as was done earlier, the decision was made to proceed with a separate sensitivity analysis for the TMI case. Therefore, the log K for boltwoodite-Na was increased and decreased by one standard deviation, similar to

what was done for the FFTF case. The results for the TMI sensitivity cases showed a range of $\pm 87.6\%$ and $+119\%$ for increasing and decreasing the log K of boltwoodite by one standard deviation. The new results from the sensitivity cases are revised in Table 6-37 and revised pages 6-88 and 6-89 presented later in the ERD. The new files that were added to the DTN are listed in Table 1.

While calculating the adsorption for the TMI sensitivity cases, it was discovered that the adsorption values in the base case TMI case were in error. The adsorption values in the PHREEQC output file *tmi_10.933k.xls* did not match the values copied to the file *adsorption_calc.xls*. Once the correction was made, the adsorption was found to be 7.05 moles of uranium rather than 112 moles as reported in the original report (Table 6-15). The output files in the DTN that are changed are indicated in Table 1.

The quantitative evaluation (Section 7.2.2.2.2) of the validation of the adsorption model was based on source term *FFTFIG1adEhdec*, therefore the text and tables were updated to reflect the new results. The validation is not impacted because the numbers in the tables changed very little (see revised pages 7-25 and 7-26 and Tables 7-7 and 7-8 later in ERD).

The updated accumulation results for cases *FFTFIG1adEhdec* and *TMI_IG1* are presented for comparison with the previous values in Table 2. The updated values show lower uranium accumulation and an increase in plutonium accumulation from 6.34×10^{-3} kg to 2.49×10^{-2} kg. The new value of 2.49×10^{-2} kg plutonium is still below the minimum critical mass limit of 1.66 kg for plutonium accumulation in the invert, which is the lowest minimum critical mass limit calculated for accumulation outside of the waste package (invert, fractured tuff, etc.), as listed in Table 6.9-1[a] of the addendum to the report. Since the plutonium accumulation is still below the minimum critical mass limit and since all values of uranium accumulation are lower than the original values, the conclusions of the document are not impacted.

Table 2. Comparison of U and Pu Accumulation in Updated Results with Previous Results

Source Term	Pu Accumulation (kg)	Total U Accumulation (kg)			U-235 Accumulation (kg)		
		Low	Medium	High	Low	Medium	High
TMI_IG1	N/A	2.98E+01 <u>2.11</u>	3.01E+01 <u>5.13</u>	3.07E+01 <u>9.24</u>	7.83E-01 <u>5.56E-02</u>	7.92E-01 <u>1.38E-01</u>	8.08E-01 <u>2.50E-01</u>
FFTFIG1adEhdec	6.34E-03 <u>2.49E-02</u>	1.52E+01 <u>9.44</u>	1.59E+01 <u>9.61</u>	1.72E+01 <u>1.09E+01</u>	9.82E-01 <u>5.65E-01</u>	1.03 <u>5.73E-01</u>	1.12 <u>5.85E-01</u>

Source: Updated results (red underlined text): DTN: MO0609SPAINOUT.002, All PHREEQC runs, spreadsheet *Dissolved U and Pu acc total with sensitivities.xls*. Previous results (strikethrough): Table 8-2 of the parent report.

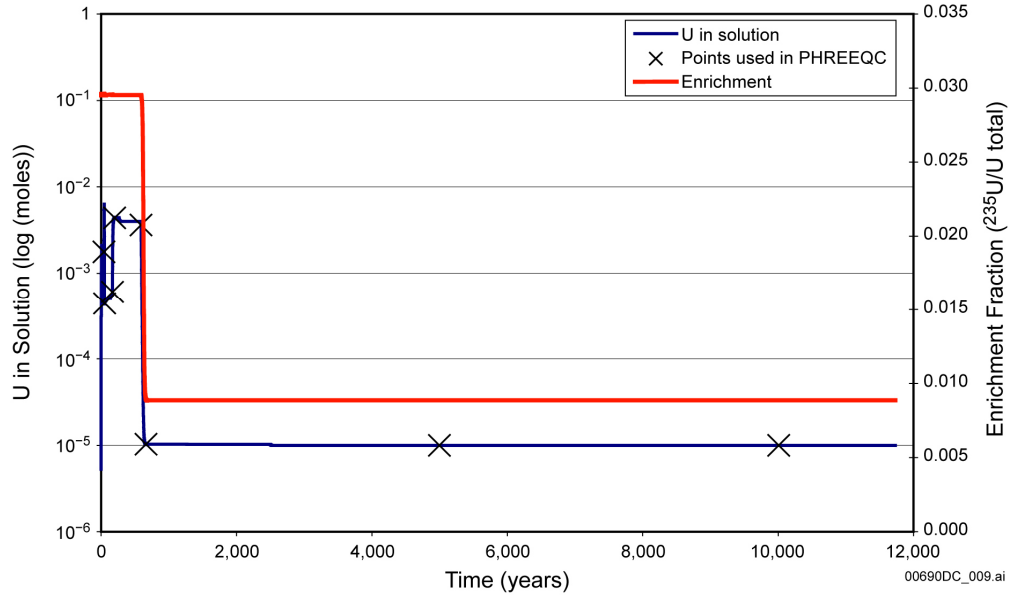
4. Results of Addressing CR 13006

The older PHREEQC database (DTN: MO0604SPAPHR25.001 [DIRS 176868]) is qualified for intended use within Section 4.1.14.5[a] of the addendum (presented later in ERD). The results of the sensitivity analysis, in which the results of using the older database are compared against the results of using the updated database (presented later in ERD in Table L-1[a]) showed no change in plutonium accumulation and a slightly lower accumulation of uranium. Since lower

accumulation indicates a lower likelihood of criticality, the original results are more conservative; therefore, there is no impact. The results show that the updates to the database had no impact on the external criticality conclusions. The output file *Database Sensitivity.zip* contains all the files for the sensitivity analysis and is added to the output DTN as listed in Table 1.

5. Revised Pages in Document

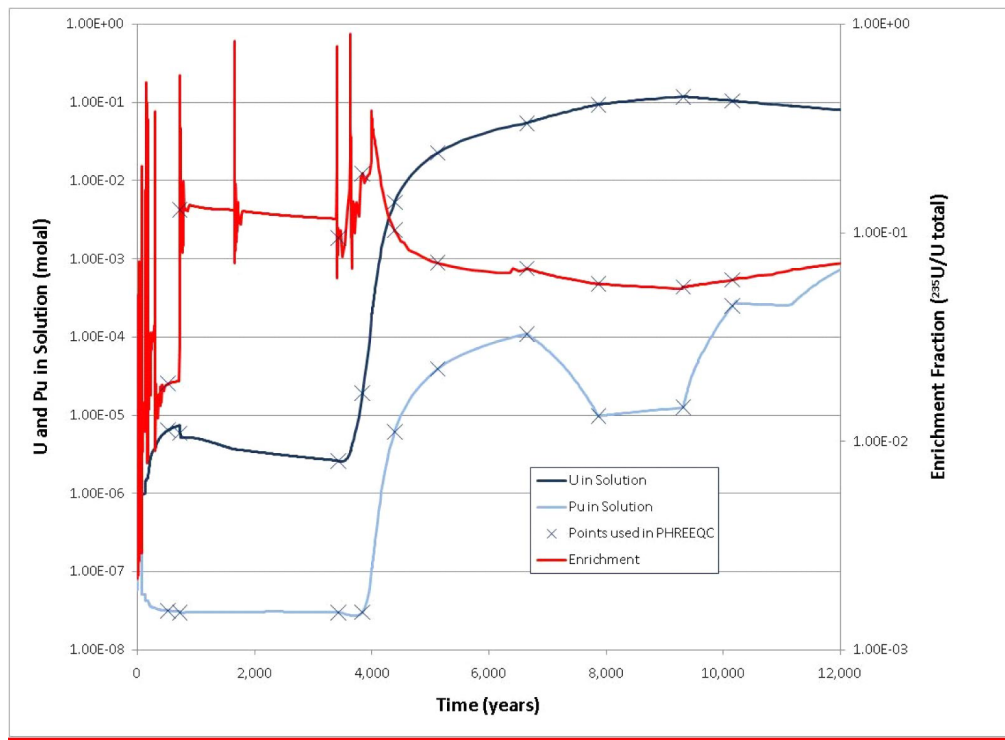
The following pages 6 through 14 of the ERD are marked-up pages of the original parent document. Pages 15 through 25 are marked-up pages of the addendum. The header gives the ERD document number, name, and page number. The footer indicates the page number in the original document.



Source: Output DTN: MO0609SPAINOUT.002, folders: TMI_IG2\TMI_IG2_Asprin, file: TMI_IG2_asprin_all.xls, tab: Chart

Source term description: TMI SNF, Igneous Scenario, 1,000 L/yr.

Figure 6-5. TMI_IG2 Source Term, Uranium in Solution Versus Time



Source: Output DTN: MO0609SPAINOUT.002, folders: FFTFIG1adEhdec \ FFTFIG1_Asprin_Sourceterm, file: FFTFIG1adEhdec.xls, tab: Chart1

Source term description: FFTF SNF, Igneous Scenario, 1 L/yr.

Figure 6-6. FFTFIG1adEhdec Source Term, Uranium and Plutonium in Solution Versus Time

Table 6-11. Average Volume of Minerals Accumulated at 10,000 Years for Source Term FFTFIG1adEhdec, Igneous Scenario, Flow at 1 L/yr

Type of Mineral	Mineral Name	Volume (cm ³ /kg of solution) at 10,000 Years
Uranium Minerals	Boltwoodite-Na	1.66
Plutonium Minerals	None	0
Other Minerals	Celadonite	46.66
	Hydroxylapatite	2.65
	Powellite	1.69
	Nontronite-Na	1.00
	Dawsonite	0.16
	Goethite	0.07
	Gibbsite	0.06
	Trevorite	0.02
TOTAL:		53.98

Source: Output DTN: MO0609SPAINOUT.002, Folder: FFTFIG1adEhdec/FFTFG1_MinAcc, file: *fftfG1_Vol_Summary.xls*, tab: Sorted Minerals, columns E and F.

NOTE: Only volumes greater than or equal to 0.01 Volume (cm³/kg of solution) for non-uranium and non-plutonium minerals are presented.

Table 6-12. Average Volume of Minerals Accumulated at 10,000 Years for Source Term FFTFIG2adEhdec, Igneous Scenario, Flow at 1,000 L/yr

Type of Mineral	Mineral Name	Volume (cm ³ /kg of solution) at 10,000 Years
Uranium Minerals	None	0
Plutonium Minerals	None	0
Other Minerals	Dawsonite	307.26
	Analcime	298.57
	Kaolinite	57.56
	Celadonite	2.19
	Erionite	1.33
	Montmorillonite-Na	0.09
	TOTAL:	

Source: Output DTN: MO0609SPAINOUT.002, folders: FFTFIG2adEhdec/FFTF2-Min_Acc, file: *fftfG2_Vol_Summary.xls*, tab: Sorted Minerals, columns E and F.

NOTE: Only volumes greater than or equal to 0.01 volume (cm³/kg of solution) for non-uranium and non-plutonium minerals are presented.

Table 6-15. Uranium and Plutonium Flushed from Various Waste Packages, Accumulated and Adsorbed at Approximately 10,000 Years after Waste Package Breach

Source Term	Waste Package Type, Scenario, and Conditions	²³⁹ Pu Moles			Total U Moles			²³⁵ U Moles		
		Flushed from Waste Package	Accumulated	Adsorbed	Flushed from Waste Package	Accumulated	Adsorbed	Flushed from Waste Package	Accumulated	Adsorbed
TMI_IG1	TMI Igneous 1 L/yr	N/A	N/A	N/A	1.15E+03	14.5	<u>7.05</u>	3.04E+01	0.4	<u>1.87E-01</u>
TMI_IG2	TMI Igneous 1,000 L/yr	N/A	N/A	N/A	1.97E+03	0.0	9.45E-04	5.64E+01	0.0	8.36E-06
CDSPIG2	N-Reactor Igneous 1,000 L/yr	N/A	N/A	N/A	4.24E+04	0.0	4.59E-01	5.38E+02	0.0	5.83E-03
FFTFIG1adEhdec	FFTF Igneous 1 L/yr Adjusted Eh Pu decay included	<u>2.23E-01</u>	<u>0.0</u>	<u>1.04E-01</u>	<u>3.49E+02</u>	<u>7.08E-01</u>	<u>3.97E+01</u>	<u>2.06E+01</u>	<u>3.97E-02</u>	<u>2.40</u>
FFTFIG2adEhdec	FFTF Igneous 1,000 L/yr Adjusted Eh Pu decay included	4.83	0.0	5.62E-05	8.23E+02	0.0	1.37E-03	5.57E+01	0.0	4.37E-04

Table 6-34. Total Uranium and Plutonium Accumulated for Source Term CSFlux9, Seismic Scenario, Flow at 1,000 L/yr, Method: Separate Simulations Using Two Separate Mixing Waters, Pore Water SD-9 and J-13 Water

Details	Uranium Accumulation (moles)	²³⁵ U Accumulation (moles)	Uranium Adsorbed (moles)	Plutonium Accumulation (moles)	Plutonium Adsorbed (moles)
Original Mixing Water SD-9 ^a	942.03	10.47	7.51E-04	0.00	5.11E-07
Sensitivity Mixing Water J-13 ^b	858.5	9.54	7.77E-04	0.00	5.11E-07

^a For SD-9 mixing water data, Output DTN: MO0609SPAINOUT.002, path: CSFlux9/CSFlux_Acc_with_decay/CSFlux9_Acc_with_decay.xls for accumulation; file *adsorption_calc.xls* for adsorption.

^b For J-13 mixing water data, Output DTN: MO0609SPASENSI.003, path: CSFlux9_J-13/CSFlux_J-13_Acc_with_decay/CSFlux9_J-13_Acc_with_decay.xls for accumulation; file *sens_adsorption_calc.xls* for adsorption.

6.8.1.2 Variation of the Values of log K for Uranium Minerals Uranophane and Boltwoodite-Na

Equilibrium thermodynamic-based geochemical codes like PHREEQC use log K to access the position of equilibrium for specific chemical reactions at specific conditions. In most cases, the uncertainty for the log K values is not reported in the databases. In order to evaluate the effects of log K uncertainty on precipitation and accumulation of uranium minerals, the log K's for uranophane and boltwoodite-Na were modified by the standard deviation. The log K values for uranophane and boltwoodite-Na are presented in Table 6-36. The log K value used in the PHREEQC thermodynamic database was previously qualified for the EQ 3/6 data0 databases in *Qualification of Thermodynamic Data for Geochemical Modeling of Mineral-Water Interactions in Dilute Systems* (BSC 2004 [DIRS 171916]). As discussed in Section 4.1.1, the EQ 3/6 data0.ymp.R4 database was translated for use in PHREEQC and presented in DTN: MO0604SPAPHR25.001 [DIRS 176868]. The sources of this log K value (Pérez et al. 2000 [DIRS 157910]; Nguyen et al. 1992 [DIRS 100809]) were also justified for intended use in this model in Section 4.1.14.3.

Table 6-35. Log K values for Uranophane and Boltwoodite-Na

Mineral	Source	Temperature C	log K value
Uranophane	Pérez et al. 2000 [DIRS 157910], p. 606	25	11.7 ± 0.6
Boltwoodite-Na	Nguyen et al. 1992 [DIRS 100809], Table 6, p. 374	30	5.82 ± 0.16

Source term sensitivity simulations were done by changing the log K by ± the standard deviation for uranophane for CSFlux9 and boltwoodite-Na for FFTFIG1adEhdec and TMI_IG1. The input files were modified to override the log K value in the PHREEQC thermodynamic database. The results of these simulations are presented in Tables 6-37 and 6-38. By changing the log K value of boltwoodite-Na by adding and subtracting one standard deviation, which is 2.7% of the log K, the accumulation decreased by 100% and increased by 743% for the FFTF case and the accumulation decreased by 87.6% and increased by 119% for the TMI case. By changing the log

K value of uranophane by the standard deviation (both up and down), which is 5% of the log K, the accumulation either decreased by 24% or increased by 13%. The overall findings were that varying the log K value by the standard deviation did not impact the type of minerals that formed, but it did impact the amount of uranium accumulation.

Table 6-36. Comparison of Uranium Accumulation for CSFlux9 Source Term in the Invert with Changes in log K for Uranophane by Standard Deviation

Details	Uranophane log K Value	Total Uranium Accumulation (moles)	Moles of Uranium Adsorbed	Percentage Difference from Original Value of Total Uranium Accumulation
Original case	11.6981	942.03	7.51E-04	N/A
+0.6 (standard deviation)	12.2981	718.49	1.13E-03	-23.7%
-0.6 (standard deviation)	11.0981	1066.04	4.86E-04	+13.2%

Source: For original case, Output DTN: MO0609SPAINOUT.002, path: CSFlux9/CSFlux9_Acc_with_decay/CSFlux9_Acc_with_decay.xls for accumulation; file adsorption_calc.xls for adsorption.

For +0.6 sensitivity case, Output DTN: MO0609SPASENSI.003, path CSFlux9_logK_sens_12.2981/CSFlux9_logK_sens_12.2981_Acc_with_decay/CSFlux9_Acc_with_decay_logK_12.2981.zip for accumulation; file sens_adsorption_calc.xls for adsorption. For -0.6 sensitivity case, follow similar path.

Table 6-37. Comparison of Uranium Accumulation with Changes in log K for Boltwoodite-Na by Standard Deviation

Details	Boltwoodite -Na, log K value	FFTFIG1adEhdec			TMI_IG1		
		Uranium Accumulation (moles)	Moles of Uranium Adsorbed	Percentage Difference from Original Value	Uranium Accumulation (moles)	Moles of Uranium Adsorbed	Percentage Difference from Original Value
Original case	5.9649	<u>0.708</u>	<u>39.7</u>	N/A	<u>14.5</u>	<u>7.05</u>	<u>N/A</u>
+0.16 (standard deviation)	6.1249	<u>0.0</u>	<u>39.7</u>	<u>-100%</u>	<u>1.8</u>	<u>7.10</u>	<u>-87.6%</u>
-0.16 (standard deviation)	5.8049	<u>5.97</u>	<u>39.7</u>	<u>+743%</u>	<u>31.8</u>	<u>6.99</u>	<u>+119%</u>

Source: For original case, Output DTN: MO0609SPAINOUT.002, path: FFTFIG1adEhdec / FFTFIG1adEhdec_Acc_with_decay/ FFTFIG1adEhdec_Acc_with_decay.xls for accumulation; file adsorption_calc.xls for adsorption. For TMI, follow a similar path.

For +0.16 sensitivity case, Output DTN: MO0609SPASENSI.003, path FFTFIG1adEhdec_logK_sens_6.1249/ FFTFIG1adEhdec_logK_sens_6.1249_Acc_with_decay/ FFTFIG1adEhdec_logK_sens_6.1249_Acc_with_decay.zip for accumulation; file sens_adsorption_calc.xls for adsorption. For -0.16 sensitivity case and for TMI case, follow similar paths.

The PHREEQC simulations described here comply with Equations 7.2.2-1 through 7.2.2-5 and thus maintain the relationships portrayed in Figures 7-9 and 7-10. The implementation is more complicated, however, because reactions have to be defined for each of the dominant aqueous U and Pu species, and the activities of these aqueous species must be converted to concentrations. Section 6.4.3.2 explains how the adsorption model is implemented in PHREEQC.

To verify that the PHREEQC approach obeys the Langmuir competitive model, a set of PHREEQC results was compared to calculations from the competitive adsorption equations. The selected set of PHREEQC model calculations is presented in Table 7-7. It is taken from simulation *fftfG110.936k.xls* (output DTN: MO0606SPAINOUT.001, folder: FFTFIg1adEhdec, folder FFTFG1_Phreeqc_Post, folder: fftfG1_10.2, file: *fftfG1_10.2k.xls*) at 10 years and includes cells 1 through 10. The concentrations of U and Pu are highest in the first several cells. These high concentrations result in Langmuir competitive effects on K_d values. As shown in Table 7-7, the U K_d decreases from 2 mg/L at low aqueous U concentrations (cells 5 through 10) to 0.30 mL/g at the highest aqueous U concentration (cell 1). For the same cells, the Pu K_d decreases from 200 mL/g to 30 mL/g.

Calculations from the Langmuir competitive adsorption model equations (Equations 7.2.2-4 and 7.2.2-5) are presented in Table 7-8 for the same conditions as Table 7-7. These calculations corroborate the PHREEQC calculations, exceeding them by 2% or less. Considering the large uncertainty in U and Pu K_d values (Table 6.5), the RPD values are very small and verify that the prescribed adsorption model is functioning as intended in the PHREEQC model and easily within the limits of K_d uncertainty.

Table 7-7. PHREEQC Calculations from *fftfG1_10.2k.xls* Output File at 10 years.

Cell	U (aq) (molal)	Pu (aq) (molal)	SorU (molal)	SorPu (molal)	U K_d (mL/g)	Pu K_d (mL/g)
1	9.8E-02	8.2E-06	2.8E-01	2.3E-03	0.30	30
2	8.2E-02	8.7E-08	2.7E-01	2.9E-05	0.35	35
3	1.4E-02	1.1E-10	1.4E-01	1.1E-07	1.12	112
4	5.6E-04	3.4E-14	1.0E-02	6.2E-11	1.94	194
5	1.8E-05	9.2E-18	3.4E-04	1.7E-14	2.00	200
6	4.6E-07	2.0E-19	8.8E-06	3.8E-16	2.00	200
7	9.3E-09	2.0E-19	1.8E-07	3.8E-16	2.00	200
8	1.3E-10	2.0E-19	2.5E-09	3.8E-16	2.00	200
9	1.2E-12	2.0E-19	2.3E-11	3.8E-16	2.00	200
10	5.2E-15	2.0E-19	9.8E-14	3.8E-16	2.00	200

Source: Output DTN: MO0609SPAINOUT.002, folder: FFTFIg1adEhdec, folder FFTFG1_phreeqc_post, folder: fftfG1_10.2, file: *fftfG1_10.2k.xls*.

Table 7-8. Comparison of Langmuir Competitive Adsorption Equation Calculations to PHREEQC Calculations from *ffffG1_10.2k.xls* Output File at 10 Years

Cell	U (aq) (molal)	Pu (aq) (molal)	SorU (molal) ^a	SorPu (molal) ^b	SorU (RPD) ^c	SorPu (RPD) ^c
1	<u>9.8E-02</u>	<u>8.2E-06</u>	<u>2.8E-01</u>	<u>2.3E-03</u>	0.3%	<u>0.3%</u>
2	<u>8.2E-02</u>	<u>8.7E-08</u>	<u>2.7E-01</u>	<u>2.9E-05</u>	<u>0.3%</u>	<u>0.4%</u>
3	<u>1.4E-02</u>	<u>1.1E-10</u>	<u>1.5E-01</u>	<u>1.1E-07</u>	<u>1.1%</u>	<u>1.1%</u>
4	<u>5.6E-04</u>	<u>3.4E-14</u>	<u>1.0E-02</u>	<u>6.3E-11</u>	<u>1.9%</u>	<u>1.9%</u>
5	<u>1.8E-05</u>	<u>9.2E-18</u>	<u>3.4E-04</u>	<u>1.8E-14</u>	2.0%	2.0%
6	<u>4.6E-07</u>	2.0E-19	<u>9.0E-06</u>	3.9E-16	2.0%	2.0%
7	<u>9.3E-09</u>	2.0E-19	<u>1.8E-07</u>	3.9E-16	2.0%	2.0%
8	<u>1.3E-10</u>	2.0E-19	<u>2.6E-09</u>	3.9E-16	2.0%	2.0%
9	<u>1.2E-12</u>	2.0E-19	<u>2.4E-11</u>	3.9E-16	2.0%	2.0%
10	<u>5.2E-15</u>	2.0E-19	<u>1.0E-13</u>	3.9E-16	2.0%	2.0%

Source: Output DTN: MO0607SPADSORP.000; File: *ffffG1_10.2k adsva1.xls*.

^a Calculated from Equation 7.2.2-4.

^b Calculated from Equation 7.2.2-5.

^c RPD is relative percent difference (i.e., the difference between the equation value and PHREEQC output value divided by the PHREEQC output value).

7.2.3 Method B: Types of Minerals Accumulated Corroborated with Natural Analogues and Experimental Work

7.2.3.1 Introduction

As mentioned above, Method B model validation will corroborate the PHREEQC modeling results with information published from natural analogues and experimental data. In particular, the corroboration will be successful if the types of minerals that form (such as clays and Fe-oxides) and radionuclide-bearing phases that are estimated to form in the model are corroborated with natural analogues or experimental work published in peer-reviewed or industrial literature or both.

The minerals that are estimated to form by the model are controlled by the thermodynamic database. Whereas not all minerals that are known to form in nature are included in the database, a representative set of uranium minerals is included. The database includes single end member compositions (such as boltwoodite-Na) and does not include some intermediate, metastable phases. As an example, compreignacite (as seen at the Shinkolobwe Mine, Zaire) is a K-rich uranyl phase and would not be stable at Yucca Mountain due to differing rock major element chemistry (lack of potentially mobile K in the source rock). Another phase, becquerelite, is shown to be unstable in the experimental work of Wronkiewicz et al. (1996 [DIRS 102047]) and is described as such in Section B.4.1 of this report. It is also depicted as unstable in the long term experiments plotted in Figure B-1.

calculated. Table 8-2 provides the U and Pu accumulation due to precipitation and adsorption on to the crushed tuff of the invert or in the fractures of the host rock for the igneous scenario. The low and high values are a result of the log K sensitivity results for boltwoodite-Na, the major uranium mineral forming in the DOE SNF waste package simulations (Section 6.8.1.2; DTN: MO0608SPASENSI.002). The cases in Table 8-2 that show no variation from low to high (e.g., TMI_IG2) are those cases that only had adsorption and no mineral precipitation.

Table 8-2. U and Pu Accumulated in the Igneous Scenario (10,000 years)

Source Term	Pu Accumulation (kg)	Total U Accumulation (kg)			U-235 Accumulation (kg)		
		Low	Medium	High	Low	Medium	High
TMI_IG1	N/A	2.11	5.13	9.24	5.56E-02	1.38E-01	2.50E-01
TMI_IG2	N/A	2.25E-04	2.25E-04	2.25E-04	1.96E-06	1.96E-06	1.96E-06
FFTFIG1adEhdec	2.49E-02	9.44	9.61	1.09E+01	5.65E-01	5.73E-01	5.85E-01
FFTFIG2adEhdec	1.34E-05	3.26E-04	3.26E-04	3.26E-04	1.03E-04	1.03E-04	1.03E-04
CDSPIG2	N/A	1.09E-01	1.09E-01	1.09E-01	1.37E-03	1.37E-03	1.37E-03

Source: DTN: MO0609SPAINOUT.002, All PHREEQC runs, spreadsheet *Dissolved U and Pu acc total with sensitivities.xls*.

The accumulation results presented in Table 8-2 give a total amount, but it does not give a geometry of accumulation within the invert or host rock. The location within the invert where the accumulation occurs is presented in Table 8-3, based on Section 6.4.7 and developed in DTN: SN0607T0504506.002 and DTN: MO0605SPAINVRT.000. The geometry of the accumulation within the fractures and lithophysae of the host rock should be based on the properties presented in Section 6.4.8.

Table 8-3. Location of Accumulation within the Invert for Igneous Scenario

Source Term	Case	Accumulation in Poorly-Sorted Invert			Accumulation in Well-Sorted Invert		
		Percent within Invert	Location within Invert	Percent along Bottom of Invert	Percent within Invert	Location within Invert	Percent along Bottom of Invert
TMI_IG1	1	100%	Figure 6-36	0	100%	Figure 6-39	0
TMI_IG2	3	100%	Figure 6-38	0	55.6%	Figure 6-41	44.4%
FFTFIG1adEhdec	1	100%	Figure 6-36	0	100%	Figure 6-39	0
FFTFIG2adEhdec	3	100%	Figure 6-38	0	55.6%	Figure 6-41	44.4%
CDSPIG2	3	100%	Figure 6-38	0	55.6%	Figure 6-41	44.4%

NOTE: Case numbers corresponds to the cases mentioned in Sections 6.4.6 and 6.4.7.

The masses of corrosion products in the igneous case listed in Table 4-6 were extracted and tabulated in Table 6-31 and in output DTN: MO0607SPASOLID.001. These values represent mass of material that could flow out of the waste package as a slurry if the bottom of the waste package failed.

172712 SN0410T0510404.002. Thermodynamic Database Input File for EQ3/6 - DATA0.YMP.R4. Submittal date: 11/01/2004.

9.4 OUTPUT DATA, LISTED BY DATA TRACKING NUMBER

MO0609SPAINOUT.002. PHREEQC Modeling Inputs and Outputs for Geochemistry Model Validation Report: External Accumulation Model. Submittal Date: [01/21/2010](#).

MO0604SPANOMIN.000. Nominal Case Diffusive Releases. Submittal Date: 04/26/2006.

MO0609SPASENSI.003. Sensitivity Analyses for PHREEQC Modeling for Geochemistry Model Validation Report: External Accumulation Model. Submittal Date: [01/21/2010](#).

MO0608SPASOLID.002. Solid Releases for all Scenarios. Submittal Date: 08/14/2006.

MO0605SPAINVRT.000. Accumulation in Invert. Submittal date: 05/02/2006.

SN0607T0504506.002. Modeling of Mixing in the Invert. Submittal date: 08/15/2006.

9.5 OUTPUT DATA FOR MODEL VALIDATION, LISTED BY DATA TRACKING NUMBER

MO0608SPACONFI.001. Model Validation – Confidence Building by Corroboration of PHREEQC and EQ3/6 Model Outputs. Submittal Date: 08/11/2006

MO0604SPANUMER.000. Model Validation – Validation of a Numerical Model for Mixing in Invert with an Analytical Model. Submittal Date: 04/25/2006.

MO0604SPAPREDI.000. Model Validation –PHREEQC Prediction of the Accumulation of Uranyl Materials and Leachate Compositions Observed in the Argonne UO2 Drip Test. Submittal Date: 04/25/2006.

MO0607SPADSORP.000. Model Validation- Comparasion of PHREEQC results to calculations from the competitive adsorption studies. Submittal Date: [01/21/2010](#).

9.6 SOFTWARE CODES

155712 BSC 2001. *Software Code: ASPRIN*. V1.0. 10487-1.0-00.

157838 BSC 2002. *Software Code: Acc_with_decay*. V1.2. PC. 10499-1.2-00.

159731 BSC 2002. *Software Code: EQ6. 7.2bLV*. PC. 10075-7.2bLV-02. Windows NT, 2000.

Morgan 1996 [DIRS 125332], Equations 129 and 130). (The inputs from Palandri and Kharaka 2000 [DIRS 175261] and Parrington et al. 1996 [DIRS 103896] are classified as “Established fact” because they are sources that scientists would use in their normal work practices, containing a compilation of rate parameters and numerical constants.) The values of the activation energies and the calculations are contained in *activation_energy_multipliers.xls* (tab: “Tuff mineral rates 50,” output DTN: MO0705PHREEMOD.000). The resulting dissolution rates of the tuff minerals at 50°C are tabulated in Section 6.4-2[a].

4.1.8[a] Invert Properties

No changes.

4.1.9[a] Adsorption Coefficients

No changes.

4.1.10[a] Characteristics of Fractures, Matrix, and Lithophysae

In addition to the references listed in Table 4-18 of the parent report, DTN: GS991108314224.015 [DIRS 151042] provided the sizes of lithophysae (5 cm to greater than 100 cm) and the void percentage of the lithophysae clusters (3% to 30%). The maximum value of infill thickness in lithophysae within the repository was observed to be 50-mm (DTN: GS980308315215.008 [DIRS 107355], values in column “Fracture Infill Thickness (cm)” for those entries with “Lithophysal Cavities” in column “Type”). The lithophysae data are used in Section 6.9[a].

4.1.11[a] Atomic Weights

No changes.

4.1.12[a] Waste Package Dimensions

No changes.

4.1.13[a] Log K Values Used in Sensitivity Analyses for Uncertainty

No changes.

4.1.14[a] Justification and Qualifications of External Sources and Project Data

4.1.14.5[a] Qualification of PHREEQC Database Used in Parent Report

The project data contained in DTN: MO0604SPAPHR25.001 [DIRS 176868] is qualified using Method 2, Corroborating Data, as outlined in Attachment 2 of SCI-PRO-001, *Qualification of Unqualified Data*. This method was chosen because corroborating data is available by recalculating external accumulation results using the qualified PHREEQC database (output DTN: MO0704PHREEQ25.000).

Description of Data: The data contained in DTN: MO0604SPAPHR25.001 [DIRS 176868] represents a thermodynamic database for use with the geochemical equilibrium simulation code PHREEQC v2.3 (STN: 10068-2.3-01) [DIRS 157837]. The database was developed by translating the qualified EQ3/6 database *data0.ymp.R4* (DTN: SN0410T0510404.002 [DIRS 172712]) to a format suitable for PHREEQC using the qualified code TRANSL v2.0 (STN: 10251-2.0-00 [DIRS 155029]). The database was used for the external accumulation calculations in the parent report (Section 4.1.1 of parent report). After the parent report was completed, an updated version of the EQ3/6 database, *data0.ymp.R5* (DTN: SN0612T0502404.014, [DIRS 178850], was developed, in part, to address errors that had been noted in the *data0.ymp.R4* database, as explained in the data qualification report for the database (SNL 2007 [DIRS 177409], Section 7.2). The updated version of the PHREEQC database (output DTN: MO0704PHREEQ25.000 was translated from *data0.ymp.R5*, as described in Section 6.4.5.1[a] and used in the addendum calculations (Table 4-2[a]).

Qualification Team: The data qualification team consisted of Susan LeStrange, chairperson, (M.S. chemical engineering, Ph.D. agricultural engineering, with expertise in geochemistry) and Clinton Lum (M.S. geology, Ph.D. geology).

Corroborating data: A sensitivity analysis was performed to determine if the changes made to the database would have an impact on the external accumulation model results for one of the cases presented in the parent report. The results of the sensitivity analysis, presented in Table L-1[a] of Appendix L[a], showed no impact.

Qualification status: Based on the assessment, Data Qualification Method 2 has been satisfied and the data contained in DTN: MO0604SPAPHR25.001 [DIRS 176868] is qualified for use as direct input for the analysis presented in the parent report.

4.1.15[a] Seepage Rates

The range of drift seepage flux expected for the seismic and igneous scenarios is presented in Figures 4-1[a] and 4-2[a]. The values represent locations in the repository with the lowest seepage (PS1) and the highest seepage (PS5). The drift seepage represents the water that enters the area defined by the diameter of the emplacement drift and the waste package length. The values shown are for the CSNF waste packages. Data is also available for codisposal waste

59 0.9
999 0.1
MIX 60
60 0.9
999 0.1

The INCREMENTAL_REACTIONS data block is used in this example to implement the same mixing ratios for each batch reaction step defined in the KINETICS data block.

```
INCREMENTAL_REACTIONS false  
END
```

Table 6.4-7[a] contains the moles of uranium and plutonium released from the waste package, moles precipitated within the crushed tuff of the invert or the host rock, and moles adsorbed onto the tuff. The moles of precipitated and adsorbed radionuclides were summed for total accumulation and converted to units of mass in *Mass accumulated.xls* (output DTN: MO0705PHREEMOD.000), using molecular weights from Audi and Wapstra (1995 [DIRS 149625], p. 60). (The inputs from Audi and Wapstra 1995 [DIRS 149625] are considered “established fact” since it is a source that scientists and engineers would use in their normal work practices, containing tables of the atomic mass for radioisotopes of the chemical elements.) The results are presented in Sections 8.1.2[a] and 8.1.3[a].

Both sets of results presented in Table 6.4-7[a] show no plutonium precipitation and very small amounts of adsorbed plutonium, which is similar to all of the cases analyzed in the parent report (Table 6-15). The results presented in Table 6.4-7[a] also show that about 80% of the uranium released from the waste package was precipitated, with very little adsorption. This is similar to the previous results for CSNF (Table 6-15 of the parent report), but differs for the previous FFTF case (Table 6-15 of the parent report, case *FFTFIG1adEhdec*) in which **less than 1%** precipitated and 10% was adsorbed. The differences are explained by the different compositions of the mixing waters used for the accumulation calculations. For the FFTF case from the parent report, the scenario is an igneous scenario in which the source term was mixed with basalt water with a pH of 9.02 (Table 4-1 of the parent report), whereas, for the FFTF case in the addendum, the scenario is a seismic scenario in which the mixing water was J-13 well water with a pH of 8.36 (Table 4-3[a]). When a mixing water with a higher pH (such as the basalt water) is combined with a source term, the resultant solution has a higher pH than the resultant solution would have if a lower pH solution (such as the J-13 well water) was mixed with the source term. The higher pH solution has a higher uranium solubility, which leads to less material precipitating. In addition, the higher uranium concentration of the higher pH solution leads to higher adsorption. Therefore, though some of the results presented in the addendum and the parent report are different, the differences are based on the different scenarios being modeled.

ways. Large lithophysae (> 200 mm, second heterogeneity) were treated as spherical voids filled with a fissile waste and seepage water mixture reflected by saturated tuff. In this case, a single lithophysa was treated as a separate entity. Small lithophysae (< 200 mm, third heterogeneity) were treated as an array of voids filled with a similar fissile waste/seepage water mixture (Figure 6.9-1[a], diagram (c)). The break point between large and small was chosen as 200 mm. Above 200 mm, the neutrons in one void are not able to communicate with adjacent voids, and the volume in the void is large enough to hold a critical mass for certain enrichments and fissile waste concentrations. Below 200 mm, the neutrons created in one void are able to communicate with fissile material in another void. To obtain a critical configuration with lithophysae below 200 mm in diameter, there must be more than one, and they need to be close together. While the arrays are random in nature, they were modeled as a regular arrangement. This simplification of the modeling captures the salient behavior of the system. Lithophysae that are located close together will have more interaction with each other, while those at greater separation will have less interaction, but in the aggregate, the effects would average out. The values of porosity of the rock containing the lithophysae and the lithophysae sizes were taken from DTN: GS991108314224.015 [DIRS 151042].

The main inputs to the external criticality calculations are the compositions of the rock matrix, the seepage water, the fissile material from the waste package effluent, and their mixing ratios. The rock matrix composition used for all of the external criticality calculations was Topopah Spring Tuff (Section 4.1.6 of the parent report). For the seismic cases, J-13 well water (Table 4-4[a]) filled the voids and pores in both the invert and the host rock. A sensitivity case using the concentration of SD-9 pore water gave nearly identical results to the J-13 water (output DTN: MO0705PHREEMOD.000, file: *CSNF.xls*, tab: "SSN5", columns M, N, O). For the igneous cases, basaltic water (Table 4-3[a]) filled the voids and pores in both the invert and the host rock. The uranium concentration for the seismic scenario was set equal to 1.21×10^{-4} moles/liter, the peak concentration observed in the CSNF seismic scenario from the parent document (output DTN: MO0609SPAINOUT.002, folder: CSFlux9\CSFlux9_Asprin, file: *Aqueous_Species_CSFlux9.xls*, column AJ). The uranium concentration for the all igneous scenarios (except TMI) was based on the peak concentration observed in the CSNF igneous scenario (5.17×10^{-5} molal) from the addendum (output DTN MO0705PHREEMOD.000, folder: CSNF-Igneous\asprin, file: *CSIGAdEh U Pu.xls*, tab: "phreeqc points, U Pu released"). The actual value used (5.43×10^{-5} molal) was taken from a preliminary simulation and was retained because it only slightly overestimates the uranium concentration and would only slightly overestimate the likelihood of a criticality. For the TMI igneous scenario, the highest uranium concentration (1.32×10^{-1} molal) predicted during the first 10,000 years for the TMI case in the parent report was used (parent report output DTN: MO0609SPAINOUT.002, folder: TMI_IG1\TMI_IG1_Asprin\TMI_IG1_Asprin, file: *TMI_IG1A.xls*).

Based on the results of the external accumulation model, the dominant uranium compounds that appear to form in the invert are uranophane and Na-boltwoodite (Tables 6-9, 6-11, and 6-14 of the parent report). ~~The mineral $(\text{UO}_2)_3(\text{PO}_4)_2 \cdot \text{H}_2\text{O}$ formed for the FFTF igneous scenario at low flow rates (Table 6-11), but was not used in the criticality calculations because FFTF is only a concern for plutonium accumulation due to the low uranium enrichment in the fuel (Table 6.4-1[a]).~~ Schoepite is found in spent fuel degradation experiments and could eventually form in the external environment (Section 7.2.3 of the parent report). Therefore, for the invert criticality calculations, schoepite was considered in addition to the primary minerals uranophane

drops below 1 kg, and the representations will need to be refined to give a truly realistic estimate. Since the 0.20-m-diameter void is at the upper end of the range considered for an array, array data were not calculated. The minimum masses for an array should be bounded by the minimum mass for a single lithophysa. An array model is not relevant when a single element can produce the bounding k_{eff} .

6.9.3[a] Summary

To summarize all of the above calculations and the predicted releases to the invert and the far-field host rock, a direct comparison is provided in Table 6.9-1[a]. None of the cases showed an accumulated mass higher than the mass required to achieve $k_{eff} = 0.96$. The DOE SNF addressed in this report (N-Reactor, TMI, and FFTF) make up approximately 90% of the mass of heavy metal in the DOE SNF inventory expected to be stored in the repository. Some of the other DOE SNF with high enrichments, such as LWBR and Fort St. Vrain, are also not expected to be a concern due to the corrosion resistance of the waste form (see Section 6.9.2.1[a]).

Table 6.9-1[a]. Summary of External Criticality Results—Minimum Mass for $k_{eff} = 0.96$

Scenario	Waste Package Type	Calculated Accumulation or Mass Released from Waste Package		Mass of U or Pu (for FFTF) required to achieve $k_{eff} = 0.96$			
		Uranium Mass, Unless Otherwise Noted (kg)	Location of Value	Invert (kg)	Fractured Tuff	Lithophysae Array	Large Lithophysa
Seismic	N-Reactor	Not calc ^a	Not calc	266,000	Inf ^b	Not calc	Not calc
	TMI Fuel	Not calc	Not calc	350	Inf	Not calc	Not calc
	CSNF	90.3 ^c	Folder: CSNF-Seismic, File: <i>U released.xls</i> (output DTN: MO0705PHR EEMOD.000)	126	Inf	Not calc	Not calc
	FFTF (Plutonium mass)	0	Table 8-2[a]	1.66	4.3	Not calc	Not calc
Igneous	N-Reactor	0.109	Parent report, Table 8-2, CDSPIG2	Inf	Inf	Inf	Inf
	TMI	9.24	Parent report, Table 8-2, TMI IG1	538	Inf	Inf	Inf
	CSNF	74.8	Table 8-1[a]	159	Inf	1390	Inf
	FFTF (Plutonium mass)	2.49×10^{-2}	Parent report, Table 8-2, FFTFIG1adEhdec	1.66	4.3	4.0	2.2

^a “Not calc” means that this scenario was of little interest given that it was bounded by another scenario. In most cases, this simply meant that, if CSNF waste was very sub-critical, then TMI and N-Reactor had to be also.

^b “Inf” means that an infinite amount of fissile waste released in this model will not produce an arrangement that can reach $k_{eff} = 0.96$ ($k_{\infty} < 0.96$).

^c Maximum mass released from the waste package. This value supersedes the value in the parent report (Table 8-4) due to the new CSNF waste package design.

Source: The mass required to achieve $k_{eff} = 0.96$ is found in Output DTN: MO0705SCALEGEO.000.

- 177409 [SNL 2007. *Qualification of Thermodynamic Data for Geochemical Modeling of Mineral-Water Interactions in Dilute Systems*. ANL-WIS-GS-000003 REV 01. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070619.0007; DOC.20090303.0002.](#)
- 178869 SNL 2007. *Technical Work Plan for: Postclosure Criticality*. TWP-EBS-MD-000018 REV 01. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070206.0003.
- 179567 SNL 2007. *Total System Performance Assessment Data Input Package for Requirements Analysis for DOE SNF/HLW and Navy SNF Waste Package Overpack Physical Attributes Basis for Performance Assessment*. TDR-TDIP-ES-000009 REV 00. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070921.0009.
- 179354 SNL 2007. *Total System Performance Assessment Data Input Package for Requirements Analysis for Engineered Barrier System In-Drift Configuration*. TDR-TDIP-ES-000010 REV 00. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070921.0008.
- 179394 SNL 2007. *Total System Performance Assessment Data Input Package for Requirements Analysis for TAD Canister and Related Waste Package Overpack Physical Attributes Basis for Performance Assessment*. TDR-TDIP-ES-000006 REV 00. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070918.0005.
- 175177 SNL 2007. *UZ Flow Models and Submodels*. MDL-NBS-HS-000006 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20070907.0001.
- 183041 [SNL 2008. *Features, Events, and Processes for the Total System Performance Assessment: Analyses*. ANL-WIS-MD-000027 REV 00. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20080307.0003; DOC.20080407.0009; LLR.20080522.0166; DOC.20080722.0002; DOC.20090130.0001; DOC.20090312.0002; DOC.20090414.0002.](#)
- 173869 [SNL 2008. *Screening Analysis of Criticality Features, Events, and Processes for License Application*. ANL-DS0-NU-000001 REV 00. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20080208.0001; DOC.20080317.0008; LLR.20080401.0255; LLR.20080423.0161; DOC.20090302.0002; DOC.20090429.0003; DOC.20090824.0002.](#)
- 186283 [SNL 2009. *Data Qualification of Selected Data Tracking Numbers with Incorrect Acquisition/Development Methods*. TDR-MGR-GS-000004 REV 00. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20090429.0002.](#)
- 125332 Stumm, W. and Morgan, J.J. 1996. *Aquatic Chemistry, Chemical Equilibria and Rates in Natural Waters*. 3rd Edition. New York, New York: John Wiley & Sons. TIC: 246296.

- 154726 Taylor, L.L. 2001. *Fort Saint Vrain HTGR (Th/U Carbide) Fuel Characteristics for Disposal Criticality Analysis*. DOE/SNF/REP-060, Rev. 0. Washington, DC: U.S. Department of Energy, Office of Environmental Management. TIC: 249783. ACC: DOC.20030905.0002.
- 128733 Weast, R.C., ed. 1978. *CRC Handbook of Chemistry and Physics*. 59th Edition. West Palm Beach, Florida: CRC Press. TIC: 246814.
- 102047 Wronkiewicz, D.J.; Bates, J.K.; Wolf, S.F.; and Buck, E.C. 1996. "Ten-Year Results from Unsaturated Drip Tests with UO₂ at 90°C: Implications for the Corrosion of Spent Nuclear Fuel." *Journal of Nuclear Materials*, 238, (1), 78-95. Amsterdam, The Netherlands: North-Holland. TIC: 243361.
- 165505 YMP (Yucca Mountain Site Characterization Project) 2003. *Disposal Criticality Analysis Methodology Topical Report*. YMP/TR-004Q, Rev. 02. Las Vegas, Nevada: Yucca Mountain Site Characterization Office. ACC: DOC.20031110.0005.

9.2[a] CODES, STANDARDS, REGULATIONS, AND PROCEDURES

IM-PRO-002, *Control of the Electronic Management of Information*.

IM-PRO-003, *Software Management*.

SCI-PRO-001, *Qualification of Unqualified Data*.

SCI-PRO-003, *Document Review*.

SCI-PRO-004, *Managing Technical Product Inputs*.

SCI-PRO-006, *Models*.

9.3[a] SOURCE DATA, LISTED BY DATA TRACKING NUMBER

- 107355 GS980308315215.008. Line Survey Information from the Exploratory Studies Facility Obtained to Estimate Secondary Mineral Abundance. Submittal date: 03/24/1998.
- 151042 GS991108314224.015. Geology of the ECRB Cross Drift: Tabular Data. Submittal date: 11/05/1999.
- 176868 MO0604SPAPHR25.001. PHREEQC Data 0 Thermodynamic Database for 25 Degrees C - File: PHREEQC DATA025.DAT. Submittal date: 04/10/2006.
- 181798 MO0705GEOMODEL.000. Input Files and Model Output Runs: Geochemistry Model Validation Report: Material Degradation and Release Model. Submittal date: 05/23/2007.

- 172712 SN0410T0510404.002. Thermodynamic Database Input File for EQ3/6 - DATA0.YMP.R4. Submittal date: 11/01/2004.
- 178850 SN0612T0502404.014. Thermodynamic Database Input File for EQ3/6 - DATA0.YMP.R5. Submittal date: 12/15/2006.

9.4[a] OUTPUT DATA

MO0705PHREEMOD.000. PHREEQC Input and Output Files for Geochemistry Model Validation Report: External Accumulation Model in Support of Criticality. Submittal date: 05/30/2007.

MO0705SCALEGEO.000. SCALE Input and Output Files for Geochemistry Model Validation Report: External Accumulation Model in Support of Criticality Calculations. Submittal date: 05/30/2007.

MO0704PHREEQ25.000. PHREEQC Thermodynamic Database from DATA0 REV. 05 for 25 C. Submittal date: 05/16/2007.

MO0704PHREEQ50.000. PHREEQC Thermodynamic Database for 50 C (323.15K) from DATA0 REV. 05. Submittal date: 05/16/2007.

MO0709PHREEQ90.000. PHREEQC DATA0R590.DAT. Submittal
date: 09/25/2007.

9.5[a] OUTPUT DATA FOR MODEL VALIDATION, LISTED BY DATA TRACKING NUMBER

No changes.

9.6[a] SOFTWARE CODES

157838 Acc_with_decay. V1.2. PC. 10499-1.2-00.
179458 ASPRIN V. 1.0. 2004. Windows 2000. STN: 10487-1.0-00.
159731 EQ6 V. 7.2bLV. 2002. Windows 2000. STN: 10075-7.2bLV-02.
176889 EQ3/6 V. 8.1. 2005. WINDOWS 2000. STN: 10813-8.1-00.
157840 GetEqPhases V. 1.0. 2002. PC. 10725-1.0-00.
157841 MinAcc V. 1.00. 2002. PC. 10724-1.0-00.
157837 PHREEQC V. 2.3. 2002. PC. 10068-2.3-01.
157839 PHREEQC_Post V. 1.1. 2002. PC. 10723-1.1-00.
181249 SCALE V. 5.1. 2007. WINDOWS XP. STN: 10129-5.1-00.
155029 transl V. 2.0. 2001. PC Windows98. 10251-2.0-00.

APPENDIX L[a]—DATABASE SENSITIVITY

As explained in Section 4.1.14.5[a], a sensitivity case was run to determine if PHREEQC cases modeled with the updated thermodynamic database (output DTN: MO0704PHREEQ25.000) would have an impact on the external accumulation model results (presented in Table 8-2 of the parent report). The case chosen for the sensitivity run (identified as *FFTFIG1adEhdec* in Table 8-2) models the accumulation of uranium and plutonium outside of the waste package, in the invert or fractured tuff of the repository, from a degrading co-disposal waste package containing FFTF SNF. A plot of the uranium and plutonium concentrations in the source term is provided in Figure 6-6 of the parent document. FFTF SNF was chosen because it is an SNF that contains both uranium and plutonium; and some of the changes to the database might have had impacts on both uranium and plutonium accumulation. The PHREEQC input files for the FFTF case (output DTN: MO0609SPAINOUT.002, file: *FFTFIG1adEhdec.zip*, file: *FFTFG1_Phreeqc_runs.zip*, files: *.in) were revised for use with the updated PHREEQC database. The revisions to the input files involved (1) limiting the oxidation state of chromium to Cr(III), rather than Cr(VI), to be consistent with the original case (Section 4.1.1 of the parent document) and (2) revising the mineral phases included in the input files to be consistent with the new database. For the first change to the input file, the following text was added to the input files to limit the oxidation state of chromium to Cr(III):

SOLUTION MASTER SPECIES #added to input file rather than changing database

Cr	Cr+3	0.0	Cr	51.9961
Cr(+6)	CrO4-2	0.0	Cr	
Cr(+2)	Cr+2	0.0	Cr	
Cr(+3)	Cr+3	0.0	Cr	
Cr(+5)	CrO4-3	0.0	Cr	

SOLUTION SPECIES # change to cr+3; 25C values

Cr+3 = Cr+3

log k	0.000	
-gamma	4.00	0.0410

Cr+3 + 0.5 H2O = Cr+2 + 0.25 O2 + H+1

log k	-28.7576	
-gamma	4.50	0.0410

Cr+3 + 2.5 H2O + 0.75 O2 = CrO4-2 + 5. H+1

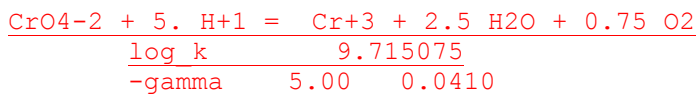
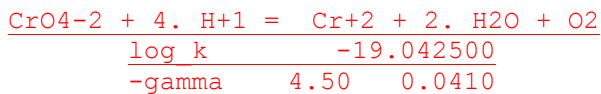
log k	-500.0	
-gamma	5.00	0.0410

0.5 O2 + Cr+3 + 3. H2O = CrO4-3 + 6. H+1

log k	-500	
-gamma	4.00	0.0410

The numerical values were taken from the updated PHREEQC database (output DTN: MO0704PHREEQ25.000). The second equation under "SOLUTION_SPECIES" (the conversion of Cr+3 to Cr+2) and the log_k were calculated from the combination of the

following two equations from the updated PHREEQC database (output DTN: MO0704PHREEQ25.000):



The second update to the PHREEQC input files required a revision to the list of minerals allowed to precipitate during a simulation. In the updated database, the formula for the mineral GdPO₄·10H₂O was changed to GdPO₄·2H₂O; therefore the formula in the input file was changed. Once the input files were updated, PHREEQC V.2.3 (STN: 10068-2.3-01 [DIRS 157837]) and the post-processors PHREEQC_Post V.1.1 (STN: 10723-1.1-00 [DIRS 157839]) and Acc_with_decay V.1.2 (STN: 10499-1.2-00 [DIRS 157838]) were run to determine the amount of uranium and plutonium accumulation.

Table L-1[a] shows that the values of uranium and uranium-235 (U-235) accumulation are slightly lower using the updated PHREEQC database compared to the older PHREEQC database. Since lower accumulation indicates a lower likelihood of criticality, the original results are more conservative; therefore, there is no impact. For plutonium, the accumulation remained the same using the updated database. The results show that the updates to the database had no impact on the external criticality conclusions and the use of the database within the AMR is considered qualified for intended use. All files are contained in output DTN (parent report): MO0609SPAINOUT.002, file *Database_sensitivity.zip*.

Table L-1[a]. Comparison of Results Using PHREEQC databases

Database Used	Uranium accumulated (kg)	U-235 accumulated (kg)	Plutonium accumulated (kg)
Older PHREEQC Database DTN: MO0604SPAPHR25.001 [DIRS 176868]	9.61	5.73 x 10 ⁻¹	2.49 x 10 ⁻²
Updated PHREEQC Database Output DTN: MO0704PHREEQ25.000	9.44	5.64 x 10 ⁻¹	2.49 x 10 ⁻²

Source: output DTN: MO0609SPAINOUT.002. For older database results, see file: *Dissolved U and Pu acc total with sensitivities.xls*, "medium" accumulation. For updated database results, see folder: *Database_sensitivity*, file: *Dissolved U and Pu acc total updated db.xls*, "medium" accumulation.