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Wagner, Steve Steve Wagner 10/11/16

Subject: FW: [EXTERNAL] Changes to actinide references used in the calculation of actinide solubility uncertainty

Attachments: Actinide Solubility Uncertainty references Sept 30 2016.docx

From: Peake, Tom [mailto:Peake.Tom@epa.gov]
Sent: Friday, September 30, 2016 1:03 PM
To: Sean Dunagan (Sean.Dunagan@cbfo.doe.gov); George Basabilvazo; Russ Patterson; Alton Harris
Cc: Economy, Kathleen; Werner, Jacqueline; Perrin, Alan; Schultheisz, Daniel; Janet Schramke; Dave Back; Charlie Wilson; Shoemaker, Paul E; Camphouse, Russell Chris; Reed, Donald T. (LANL); Leigh, Christi D; Lee, Raymond; Betsy Forinash (Betsy.Forinash@EM.Doe.Gov)
Subject: [EXTERNAL] Changes to actinide references used in the calculation of actinide solubility uncertainty

Hello,
As identified in the September 1, 2016 technical exchange, attached is the list of references/data we believe need to be excluded and included in the development of the actinide solubility uncertainty distributions/ranges.

If you have any specific technical questions, please contact Kathy Economy (202-343-9844).

Thank you.

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WIPP: 4.2.1.PA.NQ:563151

Reference and Data Selection for the CRA-2014 PABC Actinide Solubility Uncertainty Distributions

1.0 Introduction for Data Selection

Actinide solubility in the WIPP repository is uncertain. DOE addresses this uncertainty by reviewing experimental solubility data for different actinides and surrogates, then develops probability distributions around an expected solubility for each actinide and oxidation state. The solubility for a particular computer run is sampled from within the probability distribution and used in the calculations. EPA has accepted this method in addressing actinide solubility uncertainty in performance assessments.

Development of the uncertainty distributions for the CRA-2014 performance assessment (PA) actinide solubilities is described by Brush and Domski (2013b). Am(III), Nd(III), Cm(III) and Th(IV) solubility studies were evaluated for calculating actinide solubility uncertainty distributions for Am(III) and Th(IV). A Np(V) solubility uncertainty distribution was not developed because this uncertainty is not sampled for WIPP PA due to neptunium's insignificant effects on long-term WIPP performance. An uncertainty distribution was not developed for the U(VI) solubility because a WIPP solubility model has not been developed for U(VI) and DOE instead uses a constant, upper-limit U(VI) concentration of 10^{-3} M for WIPP PA.

The EPA's review of DOE's CRA-2014 PA selection criteria has identified areas of disagreement with what DOE has done and with some of the rationale used. This has been noted in the Agency's review of the CRA-2009 PABC (EPA 2010). Based on the EPA's CRA-2009 PABC review, the Agency has modified the DOE criteria to be used in selecting the references for use in the actinide solubility uncertainty distributions. The original DOE criteria and the EPA modified criteria are included in the Table 3 given at the end of this document.

References, and selected data within those references, to be used in deriving the actinide solubility uncertainty distributions are provided in the following sections. For comparison sake these tables also list the references used in the CRA-2014 PA, but with text struck out.

2.0 Th(IV) Uncertainty Data Selection

Table 1. Th(IV) Data Evaluation

Solubility Study	CRA-2014 PA	CRA-2014 PABC
Felmy et al. (1991)	~~Include 18 samples in 3 M NaCl, 8.0 ≤ pcH ≤ 11.2 (Appendix)~~	Include 18 samples in 3 M NaCl, 8.0 ≤ pcH ≤ 11.2 (Appendix)
Altmaier et al. (2004)	~~Include 6 uncentrifuged samples, two each in 5 M NaCl, 2.5 M $MgCl_2$ and 4.5 M $MgCl_2$ (Figure 2)~~	Include ultracentrifuged samples, five in 5 M NaCl and five in 2.5 M $MgCl_2$ (Figure 2)
Altmaier et al. (2006)	~~Exclude all samples~~	Include 12 samples in 4 M NaCl with 0.02 M TIC (Figure 2)
Altmaier et al. (2008)	~~Exclude all samples~~	Include 4 samples in 5.26 M $CaCl_2$ with pcH < 10 (Figure 5)
Borkowski et al. (2012), Borkowski (2012)	~~40~~	Exclude all samples
Total Samples	64[1]	44

Altmaier et al. (2006, Figure 2) investigated the solubility of amorphous $Th(OH)_4(s)$ or $ThO_2 \cdot xH_2O(s)$ in $NaHCO_3$-Na_2CO_3-NaCl solutions and reported twelve solubilities with TIC = 0.02 M and I = 4.0 M. Brush and Domski (2013b) excluded these data because the $Th(OH)_y(CO_3)_z^{4-y-z}$ complexes found to be important by Altmaier et al. (2006) are not in the WIPP Th(IV) model. However, based on EPA's revised Criteria G9 and G11, listed in Table 3, these solubilities are to be included in the calculation of the Th(IV) solubility uncertainty distribution.

Altmaier et al. (2008, Figure 5) reported four solubility results for amorphous $ThO_2 \cdot xH_2O(s)$ at pcH < 10 in 5.26 M $CaCl_2$. Brush and Domski (2013b) excluded the results based on the use of ultracentrifugation for phase separation. Altmaier et al. (2008) performed both ultracentrifugation and ultrafiltration and reported no difference between the two phase-separation methods. Consequently, EPA has determined that the four solubilities at pcH < 10 in 5.26 M $CaCl_2$ are to be included in the Th(IV) uncertainty distribution calculations.

[1] DOE (2014) stated in the caption of Figure SOTERM-31 that 45 measured and predicted solubilities were compared for Th(IV); however, the histogram in Figure SOTERM-31 contains 64 values, which is consistent with the number of measured and predicted solubilities reported by Brush and Domski (2013b).

3.0 Am(III) Uncertainty Data Selection

Table 2. Am(III), Nd(III) and Cm(III) Data Evaluation

Solubility Study	CRA-2014 PA	CRA-2014 PABC
Khalili et al. (1994)	~~Exclude all samples~~	Include 24 samples in G-Seep brine at pcH 8.4 (Figure 1)
Runde and Kim (1994)	~~Include six samples in carbonate-free 5 M NaCl~~	Include ~10 samples in carbonate-free 5 M NaCl (Figure 5.17)
Rao et al. (1999)	~~Include one sample in ERDA-6 brine~~	Include 14 samples in ERDA-6 brine from pcH 8.05 to 9.55 (Figure 6a)
Borkowski et al. (2009), Borkowski (2010)	~~Include 100 samples in 5 M NaCl, six samples in ERDA-6~~	Exclude because of absence of post-test solids characterization
Neck et al. (2009)	~~Include eight samples in 5 M NaCl, eight samples in 1 M $MgCl_2$, eight samples in 3 M $MgCl_2$, nine samples in 1 M $CaCl_2$, and 23 samples in 3 M $CaCl_2$~~	Include eight samples in 5 M NaCl (Figure 3); eight samples in 1 M $MgCl_2$, eight samples in 3 M $MgCl_2$, nine samples in 1 M $CaCl_2$, and 23 samples in 3 M $CaCl_2$ (Figure 4)
Borkowski et al. (2010), Borkowski (2012)	~~Include three samples in 3 – 5 M NaCl with $Na_2B_4O_7$~~	Exclude because of absence of post-test solids characterization
Total Samples	172	~ 104

Brush and Domski (2013b. Table 7) summarize the results of DOE's screening of available Am(III), Nd(III) and Cm(III) solubility data for calculation of the CRA-2014 PA Am(III) solubility uncertainty distribution. The Agency concurs with DOE's assessment of many of the excluded studies based on EPA's revised criteria provided in Table 3. The following discussion provides an explanation of data to be included in the CRA-2014 PABC calculation of the Am(III) solubility uncertainty distribution and identifies data included by Brush and Domski (2013b) in the CRA-2014 PA calculation that are to be excluded from the CRA-2014 PABC calculation.

1. Brush and Domski (2013b) had excluded the Khalili et al. (1994, Figure 1) amorphous $Nd(OH)_3 \cdot xH_2O(s)$ solubility results obtained in WIPP G-Seep brine at pcH 8.4 because of possible Nd(III) complexation by borate at this pcH. However, based on EPA revised Criterion G9, these data are to be included in the CRA-2014 PABC calculation of the Am(III) solubility uncertainty distribution (Table 2).

2. Brush and Domski (2013b) included six samples from Runde and Kim (1994) experiments with $Am(OH)_3(c)$ in 5 M NaCl. Examination of Runde and Kim (1994, Figure 5.17) shows that there were approximately 10 samples from the 5 M NaCl experiments within the appropriate pcH range (8 – 11.2). Brush and Domski (2013b) did not reference a specific figure or explain the selection process for the Runde and Kim

(1994) data. The samples in Runde and Kim (1994, Figure 5.17) appear to meet the revised criteria (Section 1.2). Therefore, the 10 data points (approximately) from Runde and Kim (1994, Figure 5.17) are to be included in the CRA-2014 PABC calculation of the Am(III) solubility uncertainty distribution (Table 2).

3. Brush and Domski (2013b) included one sample from $NaNd(CO_3)_2 \cdot xH_2O(c)$ solubility experiments reported by Rao et al. (1999), with all other samples excluded because of high TIC (Criterion G11) or because the pcH in the G-Seep brine (similar to GWB) or ERDA-6 brine exceeded the estimated pcH limit established to eliminate possible borate complexation of Am(III) (Criterion A1). Using the EPA revised criteria, 14 samples from solubility experiments in ERDA-6 brine were carried out with $P_{CO2} = 10^{-3.5}$ atm (Figure 6a) and had TIC ≤ 0.02 M, therefore, these samples are to be included in deriving the CRA-2014 PABC Am(III) solubility uncertainty distribution calculations.

4. Brush and Domski (2013b) included 106 samples from Nd(III) experiments in 5 M NaCl, GWB and ERDA-6 brines (Borkowski et al. 2009, Borkowski 2010) and three samples from Nd(III) solubility experiments in 3 – 5 M NaCl solutions with sodium tetraborate. All of these samples are not included in EPA set of references to derive the CRA-2014 PABC Am(III) solubility uncertainty distribution because the post-test solid phases were not characterized per revised Criterion G7.

5. Brush and Domski (2013b) selected a total of 56 samples from the $Nd(OH)_3(c)$ solubility experiments carried out by Neck et al. (2009) in concentrated NaCl, $MgCl_2$ and $CaCl_2$ solutions. These solubility data meet the EPA revised criteria and are included in the Am(III) solubility uncertainty distribution calculation for the CRA-2014 PABC.

6. Exclusion of the Borkowski et al. (2009, 2010, and 2012) data in the CRA-2014 PABC solubility uncertainty distribution is due to these experiments not reporting post-test solid phase characterization per revised Criterion G7.

4.0 Data Selection Criteria

Table 3. Data and Reference Selection Criteria Used in Developing Actinide Solubility Uncertainty Distributions

DOE Criteria	EPA Revised Criteria For PABC Reference Selection
G1: Include only results from experimental studies published from January 1, 1990, through October 31, 2011. Brush and Domski (2013b) deviated from this criterion by including the results of Borkowski et al. (2012) and Borkowski (2012). However, it is documented in Brush and Domski (2013b) that results beyond the October 31, 2011 cutoff date were to be include in the uncertainty distributions.	**G1 (revised):** Include only results from experimental studies published by December 31, 2012.
EPA issues with Criterion G1. This criteria establishes the range of publication dates for the peer-reviewed literature and reports considered for the Am(III) and Th(IV) solubility uncertainty distributions. The Agency noted during its review of the CRA-2009 PABC that establishing January 1, 1990 as the starting date for a literature search may be practical, but using an arbitrarily selected date to remove previously identified solubility data from the uncertainty distribution evaluation is unjustified (EPA 2010). The Agency and DOE agreed to include only studies published by December 31, 2012 in the evaluation of Am(III) and Th(IV) solubility uncertainties for the CRA-2014 PABC. Consequently, this Criterion G1 has been was revised in screening references to be used in the uncertainty distribution.	
EPA issues with Criterion G2 – G10 and S1. These criteria were previously used for the CRA-2009 PABC and the Agency, for the most part, accepted them (EPA 2010) but identified areas where the screening logic was not justified and are described below. The reasons for excluding of +III actinide solubility data in WIPP brines above pcH 7.4 (GWB) and above pcH 8.1 (ERDA-6) was not accepted by the Agency during review of the CRA-2009 PABC. Accepting that the WIPP thermodynamic database cannot be used to model higher-pcH brines containing borate would be an admission that the database cannot be used to adequately model WIPP repository brines, which contain 0.0623 M (ERDA-6) to 0.186 M (GWB) borate and are predicted to have pcH values of 9.5 to 9.7 (Brush and Domski 2013a). The +III actinide solubilities obtained in higher-pcH experiments with borate-containing brine should	

instead be included in the actinide solubility uncertainty calculation to account for the possible underestimation of +III actinide solubilities due to the lack of Am(III)-borate species in the WIPP thermodynamic database.	
G2: Include results from papers published in peer-reviewed journals and from unpublished reports (e.g., officially released reports from government laboratories such as Los Alamos National Laboratory, Lawrence Livermore National Laboratory, Pacific Northwest National Laboratory, etc.).	G2: Include results from papers published in peer-reviewed journals and from unpublished reports (e.g., officially released reports from government laboratories such as Los Alamos National Laboratory, Lawrence Livermore National Laboratory, Pacific Northwest National Laboratory, etc.).
G3: Include only results from solubility studies. Exclude other studies that do not provide solubilities (e.g., studies of corrosion, leaching, sorption, or transport).	G3: Include only results from solubility studies. Exclude other studies that do not provide solubilities (e.g., studies of corrosion, leaching, sorption, or transport).
G4: Include only results from studies in which water was the solvent. Exclude studies in which other solvents were used (e.g., solids, molten metal or salts, or organic liquids).	G4: Include only results from studies in which water was the solvent. Exclude studies in which other solvents were used (e.g., solids, molten metal or salts, or organic liquids).
G5: Include only results obtained from studies at pressures at or close to atmospheric, at temperatures at, or close to, those expected in the WIPP (i.e., 20-30°C), and with post-test phase-separation methods similar to those used for the WIPP. These temperatures and pressure were selected because they were the conditions and phase-separation methods used to parameterize the Th(IV) and Am(III) speciation and solubility models for WIPP compliance-related PA calculations.	G5: Include only results obtained from studies at pressures at or close to atmospheric, at temperatures at, or close to, those expected in the WIPP (i.e., 20-30°C), and with post-test phase-separation methods similar to those used for the WIPP. These temperatures and pressure were selected because they were the conditions and phase-separation methods used to parameterize the Th(IV) and Am(III) speciation and solubility models for WIPP compliance-related PA calculations.
G6: Include only results from studies of Th(IV), Nd(III), Am(III), and Cm(III) because these elements were used to parameterize the WIPP	G6: Include only results from studies of Th(IV), Nd(III), Am(III), and Cm(III) because these elements were used to

Th(IV) and Am(III) solubility models. Exclude studies of U(IV), Np(IV), Pu(IV) and Pu(III) for the following reasons; 1) there could be systematic differences between the solubilities of these elements and those of their oxidation-state analogs used to parameterize the models, and 2) there are difficulties inherent in maintaining these elements in these oxidation states that could introduce experimental artifacts in the results obtained using these elements.	parameterize the WIPP Th(IV) and Am(III) solubility models. Exclude studies of U(IV), Np(IV), Pu(IV) and Pu(III) for the following reasons; 1) there could be systematic differences between the solubilities of these elements and those of their oxidation-state analogs used to parameterize the models, and 2) there are difficulties inherent in maintaining these elements in these oxidation states that could introduce experimental artifacts in the results obtained using these elements.
G7: Include only results from studies with a characterized solubility-controlling solid for which the value of the dimensionless standard chemical potential $(\mu^0/RT)^2$ is in the WIPP Th(IV) or Am(III) model (i.e., in the EPA-certified thermodynamic database), and in which the quantity of solid initially present was sufficient to prevent complete dissolution of this solid during the experiments.	**G7 (revised):** Include only results from studies with a characterized solubility-controlling solid for which solubility data is in the WIPP Th(IV) or Am(III) model (i.e., in the EPA-certified thermodynamic database), and in which the quantity of solid initially present was sufficient to prevent complete dissolution of this solid during the experiments.
G8: Include only results from studies with aqueous solutions of known composition. Exclude studies performed with groundwaters, sedimentary pore waters, and soil solutions that may contain unknown quantities of species that can be complexants or adsorbents (e.g., humic acids or other dissolved organic compounds, microbial colloids, or pseudocolloids).	G8: Include only results from studies with aqueous solutions of known composition. Exclude studies performed with groundwaters, sedimentary pore waters, and soil solutions that may contain unknown quantities of species that can be complexants or adsorbents (e.g., humic acids or other dissolved organic compounds, microbial colloids, or pseudocolloids).
G9: Include only results from studies with dissolved elements or species for which values of μ^0/RT^1 and Pitzer ion-interaction parameters are in WIPP models. Exclude studies with dissolved	**G9 (revised):** Include results from studies with dissolved elements or species that are present in WIPP brines. Exclude studies with dissolved elements or species that are absent in

[2] The statements regarding the dimensionless standard chemical potential in Criteria G7 and G9 are relevant to the geochemical modeling code (FMT) used in earlier WIPP performance assessments. The current geochemical modeling code (EQ3/6) uses solubility constants and association or dissociation constants instead of the dimensionless standard chemical potential for solid phases and aqueous species.

elements or species for which our models do not include values of μ^0/RT or Pitzer parameters.	WIPP repository brines and for which our models do not include association/dissociation constants or Pitzer parameters.
Brush and Domski (2013b) used three additional criteria during their data selection process that they linked to Criterion G9	
A1: Include only results from GWB brine experiments with pcH ≤ 7.4 and include only results from ERDA-6 brine experiments with pcH ≤ 8.1 because Borkowski et al. (2009) determined that significant Nd(III) complexation by borate can occur at higher pcH in WIPP brines.	EPA Omitted Criterion A1
EPA notes that because borate is present in WIPP brines and DOE has not incorporated Am(III)-borate complexation into the CRA-2014 thermodynamic database (DATA0.FM1), all +III actinide solubility data in WIPP brines with pcH 8.0 – 11.2 that meet the other criteria should be included in the uncertainty evaluation. Accordingly, Criterion A1 needs to be omitted from the data selection process and Criterion G9 should be revised as described in Section 1.2.	
A2: Include only +III actinide solubility results from concentrated $CaCl_2$ brine experiments with pcH < 10. This pcH limit is included because Neck et al. (2009) found that Cm(III) forms significant $Ca_x[Cm(OH)_y]^{2x+3-y}$ aqueous species at pcH > 10. Significant concentrations of these species are unlikely to form in WIPP repository brines because of the low calcium concentrations maintained by anhydrite precipitation.	A2: Include only +III actinide solubility results from concentrated $CaCl_2$ brine experiments with pcH < 10. This pcH limit is included because Neck et al. (2009) found that Cm(III) forms significant $Ca_x[Cm(OH)_y]^{2x+3-y}$ aqueous species at pcH > 10. Significant concentrations of these species are unlikely to form in WIPP repository brines because of the low calcium concentrations maintained by anhydrite precipitation.
A3: Include only Th(IV) actinide solubility results from concentrated $CaCl_2$ brine experiments with pcH < 10. This limit is included because Brendebach et al. (2007) and Altmaier et al. (2008) found that significant concentrations of the $Ca_4[Th(OH)_8]^{4+}$ form in concentrated $CaCl_2$ solutions at pcH > 10. Significant concentrations of this species are unlikely to form in WIPP repository brines because of the low calcium concentrations maintained by anhydrite precipitation	A3: Include only Th(IV) actinide solubility results from concentrated $CaCl_2$ brine experiments with pcH < 10. This limit is included because Brendebach et al. (2007) and Altmaier et al. (2008) found that significant concentrations of the $Ca_4[Th(OH)_8]^{4+}$ form in concentrated $CaCl_2$ solutions at pcH > 10. Significant concentrations of this species are unlikely to form in WIPP repository brines because of the low calcium concentrations maintained by anhydrite precipitation

EPA notes criteria A2 and A3 are adequately supported based on the currently available data showing that the $Ca_x[Cm(OH)_y]^{2x+3-y}$ and $Ca_4(Th(OH)_8)^{4+}$ species are unlikely to form in WIPP repository brines because of relatively low calcium concentrations (Altmaier et al. 2008, Neck et al. 2009).	
G10: Include only results from studies for which the investigators provided a complete description of their experiments and the original solubilities. Exclude literature reviews and summaries, and studies in which the authors only provided average dissolved concentrations or solubility products, thus necessitating back calculation of the solubilities. S1: Include only results from Th(IV) experiments carried out with solutions with ionic strength (I) ≥ 3 M or m.	G10: Include only results from studies for which the investigators provided a complete description of their experiments and the original solubilities. Exclude literature reviews and summaries, and studies in which the authors only provided average dissolved concentrations or solubility products, thus necessitating back calculation of the solubilities.
EPA notes that Criterion S1 is redundant, because Criterion G11 addresses both the +III and +IV actinides.	
G11. Include only results from experimental studies carried out under conditions at or close to those predicted for WIPP disposal rooms. Specifically, include only results from experiments in which: (1) I ≥ 3 m or M, (2) pcH = 8.0 – 11.2, and (3) total inorganic carbon (TIC) = 0 – 0.014 M.	**G11 (revised).** Include only results from experimental studies carried out under conditions at or close to those predicted for WIPP disposal rooms. Specifically, include only results from experiments in which: (1) I ≥ 3 m or M, (2) pcH = 8.0 – 11.2, and (3) total inorganic carbon (TIC) = 0 – 0.02 M.
EPA notes that Criterion G11 was not included in the CRA-2009 PABC calculation of actinide solubility uncertainties. Criterion G11 specifies that only experiments conducted under ionic strength, pcH and TIC conditions reasonably similar to WIPP repository brines should be included in the calculation of the actinide solubility uncertainty distributions. Limiting the selected solubility studies to those with conditions similar to anticipated WIPP repository brine conditions is likely to provide a more reasonable assessment of the actinide solubility uncertainty distributions. For example, during review of the CRA-2009 PABC, the Agency noted that some measured +III actinide solubilities at relatively low pcH values were systematically less than the corresponding calculated solubilities; including the lower-pcH data with these consistent differences between measured and predicted solubilities would tend to underestimate the mean actinide solubility uncertainty.	

The Criterion G11 pcH and ionic strength limitations appear reasonable and do not **systematically** exclude specific categories of data. However, because TIC was limited to 0 – 0.014 M, no Th(IV) solubilities measured with any carbonate in solution were included in the calculation of the CRA-2014 PA Th(IV) solubility uncertainty distribution (Brush and Domski 2013b). In addition, only a single solubility measurement in a solution containing carbonate from Rao et al. (1999) was included in the Am(III) solubility uncertainty distribution calculation because of the limits on pcH for solubility measurements in WIPP brines (Criterion A1). Although TIC will be constrained to low concentrations by reaction with the MgO backfill, carbonate and bicarbonate are predicted to be present in WIPP brines at concentrations ranging from 3.79×10^{-4} M to 4.75×10^{-4} M (Brush and Domski 2013a). At these TIC concentrations, dissolved Th(IV) is predicted by CRA-2014 PA calculations to be present as 25 – 34% of total aqueous Th(IV) in the form of $Th(OH)_3CO_3^-$. The Agency previously noted that Th(IV) solubility data measured in 4 M NaCl solutions with TIC of 0.02 M (Altmaier et al. 2006) had measured solubilities that were consistently higher than the concentrations predicted by WIPP geochemical modeling calculations. Excluding the Altmaier et al. (2006) data excludes all available, high-ionic-strength Th(IV) solubility data obtained with nonzero TIC concentrations.

For the purposes of this evaluation, the upper limit on TIC for both the +III and +IV actinide solubility uncertainty determination should be revised upward to 0.02 M.

4.0 References

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