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Sandia National Laboratories
Waste Isolation Pilot Plant

Results of the Calculations of Actinide Solubilities for The WIPP CRA-2009 PABC

Work Carried Out under Task 4 of the Analysis Plan for the Calculation of Actinide Solubilities for the WIPP PABC, AP-143, Rev. 0
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1 INTRODUCTION

This analysis report provides the results of our calculations of the speciation and solubilities of actinide elements for the Performance Assessment Baseline Calculation (PABC), a compliance-related calculation for the second Compliance Recertification Application (CRA-2009) for the U.S. Department of Energy's (DOE's) Waste Isolation Pilot Plant (WIPP) (U.S. DOE, 2009). Actinide solubilities (the sums of the concentrations of all of the dissolved actinide species in chemical equilibrium with actinide-bearing solid phases under the conditions expected in WIPP disposal rooms) and the concentrations of colloidal (suspended) actinides, which are calculated from these solubilities, together constitute the actinide source term used in WIPP performance assessment (PA) calculations. The analysis described in this report was carried out under Task 4 of the analysis plan (AP) for the actinide solubility calculations for the CRA-2009 PABC (Brush and Xiong, 2009a, Subsection 4.4).

These actinide solubilities will replace those used for the PA calculations for the CRA-2009 (CRA-2009 PA), which were established by Brush and Xiong (2005a; 2005b) and Brush (2005) for the PABC for the first WIPP CRA (CRA-2004 PABC). When approved by the U.S. Environmental Protection Agency (EPA), the solubilities provided by this report will become part of the new WIPP PA baseline.

These actinide solubilities incorporated the changes specified for the CRA-2009 PABC by the EPA (Cotsworth, 2009), based on its preliminary review of the CRA-2009 (U.S. DOE, 2009). Cotsworth (2009) specified that the solubilities calculated for the CRA-2009 PABC include new estimates of the masses of the organic ligands to be emplaced in the WIPP (Crawford et al., 2008). However, Cotsworth (2009) did not specify that a new estimate of the minimum volume of brine required for a direct brine release (Clayton, 2008), is also required for the CRA-2009 PABC. (The minimum brine volume is also necessary for the calculation of organic-ligand concentrations in WIPP brines.) There is currently no reference for this and the other EPA specifications described below, because the EPA provided them in recent discussions with the DOE's Carlsbad Field Office. Brush and Xiong (2009b) calculated these new organic-ligand concentrations under Task 3 of Brush and Xiong (2009a, Subsection 4.3). The results are summarized in Subsection 2.2 of this report (see below).

Brush and Xiong (2009a, Subsection 4.4) stated that, to carry out Task 4, "We will continue to use FMT in a manner identical to that used previously to calculate chemical conditions, and actinide speciation and solubilities for the CRA-2009 PABC. However, we will also use EQ3/6 and compare the results to those obtained with FMT, if the qualification of EQ3/6 for actinide-solubility calculations according to the SNL/WIPP software QA [quality assurance] requirements were completed in time (Wolery, 2008). If not, we will use only FMT." The EPA, however, specified that the DOE continue to use FMT (Babb and Nowak, 1997 and addenda; Wang, 1998) for the actinide-solubility calculations for the CRA-2009 PABC, instead of the EQ3/6 geochemical software package (Wolery and Jarek, 2003). The EPA also indicated that it would not review the qualification of EQ3/6 for actinide-solubility calculations prior to its

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second recertification of the WIPP. Furthermore, although qualification of EQ3/6 is under way (Wolery, 2008), it has not been completed. Therefore, we used only FMT for this analysis.

Table 1 defines the abbreviations, acronyms, initialisms, etc., used in this analysis report.

Table 1. Abbreviations, Acronyms, Initialisms, etc.

Abbreviation, Acronym, or Initialism	Definition
Ac, acetate	CH_3COO^- or $CH_3CO_2^-$
Am	americium, americium in the +III oxidation state
An	actinide
An(III)	actinide element(s) in the +III oxidation state
An(IV)	actinide element(s) in the +IV oxidation state
An(V)	actinide element(s) in the +V oxidation state
An(VI)	actinide element(s) in the +VI oxidation state
anhydrite	$CaSO_4$
AP	analysis plan
aq	aqueous (dissolved in an aqueous solution)
B	boron, boric acid
Br, Br^-	bromine, bromide ion
brucite	$Mg(OH)_2$
C	carbon
Ca, Ca^{2+}	calcium, calcium ion
CCA	(WIPP) Compliance Certification Application
calcite	$CaCO_3$
Cit, citrate	$(CH_2COO)_2C(OH)(COO)^{3-}$ or $(CH_2CO_2)_2C(OH)(CO_2)^{3-}$
Cl, Cl^-	chlorine, chloride ion
CMS	(Compaq Computer Corp., now a part of the Hewlett-Packard Co.) DECset Code Management System
CO_2	carbon dioxide
CO_3, CO_3^{2-}	carbonate, carbonate ion
CRA-2004	the first WIPP Compliance Recertification Application, submitted to the EPA in March 2004
CRA-2009	the second WIPP Compliance Recertification Application, submitted to the EPA in March 2009
DOE	(U.S.) Department of Energy

Table 1 continued on next page

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Table 1. Abbreviations, Acronyms, Initialisms, etc. (continued).

Abbreviation, Acronym, or Initialism	Definition
EDTA	ethylenediaminetetraacetate $(CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)^{4-}$ or $(CH_2CO_2)_2N(CH_2)_2N(CH_2CO_2)^{4-}$. Note that, although the Am(III) and Th(IV) models include only one EDTA complex each ($AmEDTA^-$ and $ThEDTA_4(aq)$, respectively), the Np(V) model includes three ($NpO_2H_2EDTA^-$, NpO_2HEDTA^{2-}, and NpO_2EDTA^{3-})
EQ3/6	a geochemical software package for speciation and solubility calculations (EQ3NR) and reaction-path calculations (EQ6)
EPA	(U.S.) Environmental Protection Agency
eq.	equilibration
ERDA-6	Energy Research and Development Administration (WIPP Well) 6, a synthetic brine representative of fluids in Castile brine reservoirs
f_{CO_2}	fugacity (similar to the partial pressure) of CO_2
Fm.	formation
FMT	Fracture-Matrix Transport, a geochemical speciation and solubility code (the component that calculates transport is not used)
GWB	Generic Weep Brine, a synthetic brine representative of intergranular Salado brines
H, H^+	hydrogen, hydrogen ion
halite	NaCl
hydromagnesite	$Mg_4(CO_3)_3(OH)_2 \cdot 3H_2O$ or $Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$
I	ionic strength, defined by $I = \frac{1}{2} \times \sum_i M_i z_i^2$, in which M and z are the molarity and charge of species i
K, K^+	potassium, potassium ion
kg	kilogram(s)
M	molar
m	meter(s) or molal, depending on context
magnesite	$MgCO_3$
Mg, Mg^{2+}	magnesium, magnesium ion
mg	milligram(s)
MgO	magnesium oxide, used to refer to the WIPP engineered barrier, which includes periclase as the primary constituent and various concentrations of impurities
mM	millimolar
N	nitrogen
Na or Na^+	sodium, sodium ion

Table 1 continued on next page

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Table 1. Abbreviations, Acronyms, Initialisms, etc. (continued).

Abbreviation, Acronym, or Initialism	Definition
nesquehonite	$MgCO_3 \cdot 3H_2O$
Np, Np(IV), Np(V)	neptunium, neptunium in the +IV oxidation state , neptunium in the +V oxidation state
O	oxygen
OH or OH^-	hydroxide or hydroxide ion
Ox, oxalate	$(COO)^{2-}$ or $C_2O_4^{2-}$
ρ	density
PA	performance assessment
PABC	(WIPP) Performance Assessment Baseline Calculations, carried out in 2005 and 2009
PAVT	(WIPP) Performance Assessment Verification Test, conducted in 1997
periclase	pure, crystalline MgO, the primary constituent of the WIPP engineered barrier
pcH	the negative, common logarithm of the molar concentration of H^+
pH	the negative, common logarithm of the activity of H^+
pmH	the negative, common logarithm of the molal concentration of H^+
phase 3	$Mg_2Cl(OH)_3 \cdot 4H_2O$
phase 5	$Mg_3(OH)_5Cl \cdot 4H_2O$
Pu, Pu(III), Pu(IV)	plutonium, plutonium in the +III oxidation state, plutonium in the +VI oxidation state
Rev.	revision
RH	relative humidity
SO_4, SO_4^{2-}	sulfate, sulfate ion
SPC	Salado Primary Constituents (brine), a synthetic solution similar to Brine A, but without the minor constituents
Th, Th(IV)	thorium, thorium in the +IV oxidation state
TIC	total inorganic C (the sum of the dissolved species of inorganic C)
U, U(IV), U(VI)	uranium, uranium in the +IV oxidation state, uranium in the +VI oxidation state
WIPP	(U.S. DOE) Waste Isolation Pilot Plant

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2 METHODS

This section provides a brief summary of the methods we used to calculate the solubilities of actinide elements in the +III, +IV, and +V oxidation states (An(III), An(IV), and An(V)) for the CRA-2009 PABC actinide source term. Brush and Xiong (2009a) described these methods in detail.

2.1 Predictions of Long-Term Chemical Conditions

Prediction of long-term chemical conditions in WIPP disposal rooms included: (1) use of Generic Weep Brine (GWB) (Krumhansl et al., 1991; Snider, 2003) and Energy Research and Development Administration (WIPP Well) 6 (ERDA-6) (Popielak et al., 1983) to simulate brines from the Salado Fm. and the Castile Fm., respectively; (2) the assumption that instantaneous, reversible equilibria among these brines and several of the important solids in WIPP disposal rooms will control chemical conditions; and (3) use of FMT (Babb and Nowak, 1997 and addenda; Wang, 1998) to calculate chemical conditions. Brush and Xiong (2009a, Subsection 2.1) describe the prediction of long-term chemical conditions in detail.

In the case of GWB, we included the major Salado minerals halite (NaCl) and anhydrite ($CaSO_4$), the MgO hydration and carbonation products brucite ($Mg(OH)_2$) and hydromagnesite ($Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$), respectively, and the actinide-bearing solids hydrous, amorphous ThO_2, $KNpO_2CO_3$ and $Am(OH)_3$ in the FMT input files; and FMT predicted that the solids phase 3 ($Mg_2Cl(OH)_3 \cdot 4H_2O$) and whewellite (Ca oxalate hydrate, or $CaC_2O_4 \cdot H_2O$), will precipitate from GWB. For ERDA-6, we included the same solids in the input files, but FMT predicted that glauberite ($Na_2Ca(SO_4)_2$) will precipitate instead of phase 3. These results are consistent with all the DOE's previous results (Novak et al. 1996; U.S. DOE, 1996, Appendix SOTERM; Novak, 1997; Brush and Xiong, 2003d; Brush, 2005).

In addition to these runs, which we carried out for the CRA-2009 PABC actinide source term, we also carried out several additional runs to quantify the sensitivity of the long-term chemical conditions predicted in the WIPP to the carbonate-bearing solid produced by carbonation of brucite. In each calculation, we included calcite ($CaCO_3$), magnesite ($MgCO_3$), hydromagnesite, or nesquehonite ($MgCO_3 \cdot 3H_2O$). Brush and Xiong (2009a, Subsection 2.1.2) discuss the possible formation of these carbonate minerals in the WIPP. We also conducted additional runs to quantify the sensitivity of long-term chemical conditions to phase 5 ($Mg_3(OH)_5Cl \cdot 4H_2O$) in the FMT thermodynamic database (Brush and Xiong (2009a, Subsection 2.1.2 and Subsection 2.2.4).

2.2 Calculations of Actinide Solubilities

Our calculations of actinide solubilities under the long-term chemical conditions expected in the WIPP included: (1) use of thermodynamic speciation and solubility models, based on the Pitzer activity-coefficient model, for Th(IV), Np(V), and Am(III); (2) inclusion of the effects

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of acetate, citrate, EDTA, and oxalate - the organic ligands in the TRU waste to be emplaced in the WIPP - on Th(IV), Np(V), and Am(III) speciation and solubilities; (3) use of the code FMT to calculate the speciation and solubilities of Th(IV), Np(V), and Am(III); (4) use of the FMT thermodynamic database FMT_050405.CHEMDAT (Xiong, 2005), which includes phase 3 but not phase 5, for most of the calculations, including the runs used for the CRA-2009 PABC; and use of FMT_090720.CHEMDAT (Xiong et al., 2009; Xiong, 2009), which includes both phase 3 and phase 5, to assess the effects of the later phase, (5) development of new uncertainty ranges and probability distributions for Th(IV) and Am(III) solubility predictions (this analysis will be described in a separate analysis report); (6) redox speciation of Th, U, Np, Pu, and Am entirely as Th(IV), U(IV), Np(IV), Pu(III), and Am(III); or Th(IV), U(VI), Np(V), Pu(IV), and Am(III); (7) use of the oxidation-state analogy to apply the solubilities calculated for Th(IV) to U(IV), Np(IV), and Pu(IV); and to apply those calculated for Am(III) to Pu(III) (the model developed for Np(V) was used only for Np(V)); and (8) use of a solubility estimate for U(VI). Because the DOE uses the oxidation-state analogy for other actinide elements in the +III and +IV oxidation states, the Th(IV) model is often referred to as the "An(IV) model and the Am(III) model is often referred to as the "An(III) model." Furthermore, the Np(V) model is often referred to as the "An(V) model" even though Np is the only actinide element included in WIPP PA that is expected to speciate in the +V oxidation state.

Table 2 provides the concentrations of acetate, citrate, EDTA, and oxalate calculated by Brush and Xiong (2009b) for the CRA-2009 PABC. Table 2 also compares the organic-ligand concentrations used for the CRA-2009 PABC with all of the previously estimated concentrations. Table 2 shows that the dissolved concentration of acetate increased by a factor of 1.83; the concentration of citrate increased by a factor of 2.95; the concentration of EDTA increased by a factor of 7.95; and the concentration of oxalate decreased by a factor of 2.63 since the CRA-2004 PABC and the CRA-2009 PA. The reason that the concentrations of acetate citrate, and EDTA increased, and that the concentration of oxalate decreased is that the masses of these organic ligands in the WIPP TRU waste inventory changed since the CRA-2004 PABC and the CRA-2009 PA (compare the masses of these organic ligands estimated for the CRA-2004 PABC by Crawford, 2003; Crawford and Leigh, 2003; and Leigh, 2003; 2005a; 2005b; with those estimated for the CRA-2009 PABC by Crawford et al., 2009). In addition to the runs with these organic ligands that we carried out for the CRA-2009 PABC actinide source term, we also carried out several additional runs without any organic ligands to quantify the sensitivity of our calculated actinide solubilities to organic ligands.

Table 3 provides information on FMT (Babb and Nowak, 1997 and addenda; Wang, 1998), the software used for this analysis.

Tables 4, 5, and 6 list all of the FMT runs carried out for the CRA-2009 PABC. Runs 5 and 13 provided the solubilities actually used in the CRA-2009 PABC calculations; the other 22 runs were conducted for the sensitivity analyses described above.

Table 7 provides run-control information for the FMT calculations carried out for this analysis.

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The DOE has not developed a thermodynamic speciation and solubility model for actinides in the +VI oxidation state (U(VI)). Therefore, we will continue to use a fixed value of 1×10^{-3} M for the solubility of U(VI). The EPA specified this solubility for U(VI) for the CRA-2004 PABC (U.S. EPA, 2006, pp. 55-58). Brush and Xiong (2009a, Subsection 2.2.8) discussed the reasons why the DOE has not developed a U(VI) model, and reviewed the estimates of the solubility of U(VI) that have been used in WIPP PA.

Table 2. Comparisons of Organic-Ligand Concentrations Used to Calculate Actinide Solubilities for WIPP Certification- or Recertification-Related PA Calculations. From Brush and Xiong (2009b, Table 6).

Organic Ligand	CCA[A] and PAVT (m)	CRA-2004 PA[B] (M)	CRA-2004 PABC[C] and CRA-2009 PA (M)	CRA-2009 PABC[D] (M)
Total Acetate	1.1×10^{-3}	5.05×10^{-3}	1.06×10^{-2}	1.94×10^{-2}
Total Citrate	7.4×10^{-3}	3.83×10^{-4}	8.06×10^{-4}	2.38×10^{-3}
Total EDTA (ligands)	4.2×10^{-6}	3.87×10^{-6}	8.14×10^{-6}	6.47×10^{-5}
Total Oxalate	4.7×10^{-4}	2.16×10^{-2}	4.55×10^{-2}	1.73×10^{-2}

[A]U.S. DOE (1996, Appendix SOTERM, Table SOTERM-4, column labeled "Organic Concentration (scaled)."
[B]Brush and Xiong (2003, Table 4, column labeled "CRA Concentration (M)").
[C]Brush and Xiong (2005b, Table 4, column labeled "PABC (M)").
[D]Brush and Xiong (2009b).

Table 3. Software Used for This Analysis.

Code	Version	Executable	Build Date	CMS Library	CMS Class
FMT	2.4	FMT_QB0204.EXE	09-03-98	LIBFMT	QB0204

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Table 4. FMT Runs Carried Out for the CRA-2009 PABC with GWB and FMT_050405.CHEMDAT. (This database contains only phase 3.) All runs archived in the CMS Analysis Directory PAA:[ANALYSIS.PABC09.FMT], Library LIBPABC09_FMT, Class PABC09-0.

FMT Run #	Abbreviated Run #	Brine	CO_2 Buffer	Organics
FMT_PABC09_GWB_CAL_ORGS_001	FMT Run 1	GWB	Brucite-calcite[A]	Yes
FMT_PABC09_GWB_CAL_NOORGS_002	FMT Run 2	GWB	Brucite- calcite[A]	No
FMT_PABC09_GWB_MAG_ORGS_003	FMT Run 3	GWB	Brucite-magnesite[B]	Yes
FMT_PABC09_GWB_MAG_NOORGS_004	FMT Run 4	GWB	Brucite-magnesite[B]	No
FMT_PABC09_GWB_HMAG_ORGS_005	FMT Run 5	GWB	Brucite-hydromagnesite[C]	Yes
FMT_PABC09_GWB_HMAG_NOORGS_006	FMT Run 6	GWB	Brucite-hydromagnesite[C]	No
FMT_PABC09_GWB_NES_ORGS_007	FMT Run 7	GWB	Brucite-nesquehonite[D]	Yes
FMT_PABC09_GWB_NES_NOORGS_008	FMT Run 8	GWB	Brucite-nesquehonite[D]	No

[A] f_{CO_2} buffered by the brucite-calcite carbonation reaction (Brush and Xiong, 2009a, Reaction 4).
[B] f_{CO_2} buffered by the brucite-magnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 3).
[C] f_{CO_2} buffered by the brucite-hydromagnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 1).
[D] f_{CO_2} buffered by the brucite-nesquehonite carbonation reaction (U.S. DOE, 2009, Appendix MgO, Reaction MgO.11).

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Table 5. FMT Runs Carried Out for the CRA-2009 PABC with ERDA-6 and FMT_050405.CHEMDAT. (This database contains only phase 3.) All runs archived in the CMS Analysis Directory PAA:[ANALYSIS.PABC09.FMT], Library LIBPABC09_FMT, Class PABC09-0.

FMT Run #	Abbreviated Run #	Brine	CO_2 Buffer	Organics
FMT_PABC09_E6_CAL_ORGS_009	FMT Run 9	ERDA-6	Brucite-calcite[A]	Yes
FMT_PABC09_E6_CAL_NOORGS_010	FMT Run 10	ERDA-6	Brucite- calcite[A]	No
FMT_PABC09_E6_MAG_ORGS_011	FMT Run 11	ERDA-6	Brucite-magnesite[B]	Yes
FMT_PABC09_E6_MAG_NOORGS_012	FMT Run 12	ERDA-6	Brucite-magnesite[B]	No
FMT_PABC09_E6_HMAG_ORGS_013	FMT Run 13	ERDA-6	Brucite-hydromagnesite[C]	Yes
FMT_PABC09_E6_HMAG_NOORGS_014	FMT Run 14	ERDA-6	Brucite-hydromagnesite[C]	No
FMT_PABC09_E6_NES_ORGS_015	FMT Run 15	ERDA-6	Brucite-nesquehonite[D]	Yes
FMT_PABC09_E6_NES_NOORGS_016	FMT Run 16	ERDA-6	Brucite-nesquehonite[D]	No

[A] f_{CO_2} buffered by the brucite-calcite carbonation reaction (Brush and Xiong, 2009a, Reaction 4).
[B] f_{CO_2} buffered by the brucite-magnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 3).
[C] f_{CO_2} buffered by the brucite-hydromagnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 1).
[D] f_{CO_2} buffered by the brucite-nesquehonite carbonation reaction (U.S. DOE, 2009, Appendix MgO, Reaction MgO.11).

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Table 6. FMT Runs Carried Out for the CRA-2009 PABC with GWB or ERDA-6 and FMT_090720.CHEMDAT. (This database contains both phase 3 and Phase 5.) All runs archived in the CMS Analysis Directory PAA:[ANALYSIS.PABC09.FMT], Library LIBPABC09_FMT, Class PABC09-0.

FMT Run #	Abbreviated Run #	Brine	CO_2 Buffer	Organics
FMT_PABC09_GWB_CAL_ORGS_017	FMT Run 17	GWB	Brucite-calcite[A]	Yes
FMT_PABC09_GWB_MAG_ORGS_018	FMT Run 18	GWB	Brucite-magnesite[B]	Yes
FMT_PABC09_GWB_HMAG_ORGS_019	FMT Run 19	GWB	Brucite-hydromagnesite[C]	Yes
FMT_PABC09_GWB_NES_ORGS_020	FMT Run 20	GWB	Brucite-nesquehonite[D]	Yes
FMT_PABC09_E6_CAL_ORGS_021	FMT Run 21	ERDA-6	Brucite- calcite[A]	Yes
FMT_PABC09_E6_MAG_ORGS_022	FMT Run 22	ERDA-6	Brucite-magnesite[B]	Yes
FMT_PABC09_E6_HMAG_ORGS_023	FMT Run 23	ERDA-6	Brucite-hydromagnesite[C]	Yes
FMT_PABC09_E6_NES_ORGS_024	FMT Run 24	ERDA-6	Brucite-nesquehonite[D]	Yes

[A] f_{CO_2} buffered by the brucite-calcite carbonation reaction (Brush and Xiong, 2009a, Reaction 4).
[B] f_{CO_2} buffered by the brucite-magnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 3).
[C] f_{CO_2} buffered by the brucite-hydromagnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 1).
[D] f_{CO_2} buffered by the brucite-nesquehonite carbonation reaction (U.S. DOE, 2009, Appendix MgO, Reaction MgO.11).

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Table 7. Run-Control Information for the FMT Calculations Carried Out for This Analysis.

	File Names[A, B, C, D, E]	CMS Library	CMS Class
Script:			
Script	EVAL_FMT.COM	LIBPABC09_EVAL	PABC09-0
Script Input	EVAL_FMT_PABC09_w_b_o_n.INP	LIBPABC09_EVAL	PABC09-0
Script Log	EVAL_FMT_PABC09_w_b_o_ n.LOG	LIBPABC09_FMT	PABC09-0
FMT:			
Input	FMT_*.CHEMDAT	LIBPABC09_FMT	PABC09-0
Input	FMT_GENERIC.RHOMIN	LIBPABC09_FMT	PABC09-0
Input	FMT_PABC09_w_b_o_ n.IN	LIBPABC09_FMT	PABC09-0
Input	FMT_PABC09_w_b_o_n.INGUESS	LIBPABC09_FMT	PABC09-0
Output	FMT_PABC09_w_b_o_n.OUT	LIBPABC09_FMT	PABC09-0
Output	FMT_PABC09_w_b_o_n.FOR088	Not kept	Not kept

[A] $w \in \{\text{GWB, E6}\}$.

[B] $b \in \{\text{CAL, HMAG, MAG, NES}\}$ for each W.

[C] $o \in \{\text{ORGS, NOORGS each w}\}$.

[D] $n \in \{001 \text{ through } 024\}$

[E] $* \in \{050405 \text{ for Runs 1 through 16, } 090720 \text{ for Runs 17 through 24}\}$

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3 RESULTS

Tables 8 and 9 provide the chemical compositions and other parameters for GWB and ERDA-6, respectively, before and after equilibration with several of the important solids in WIPP disposal rooms (see Subsection 2.1 above).

Table 8 compares the brine compositions calculated for the CRA-2009 PABC after equilibration with several important solids to those calculated for the CRA-2004 PABC.[1] Most of the results predicted for the CRA-2009 PABC differ somewhat from those predicted for the CRA-2004 PABC. Because we used the same initial brine composition for GWB (Krumhansl et al., 1991; Snider, 2003) and the same thermodynamic database for both of these calculations (FMT_050405.CHEMDAT), these differences were caused by the differences in the organic-ligand concentrations between the CRA-2004 PABC and the CRA-2009 PABC (see Table 2 above). Brush and Xiong (2009b) concluded that "The dissolved concentration of acetate increased by a factor of 1.83; the concentration of citrate increased by a factor of 2.95; the concentration of EDTA increased by a factor of 7.95; and the concentration of oxalate decreased by a factor of 2.63 [between] the CRA-2004 PABC and the CRA-2009 PA [and the CRA-2009 PABC]."

Table 9 shows that there are similar differences for ERDA-6.[2] Again, we used the same initial brine composition for ERDA-6 (Popielak et al., 1983) and the same database for both of these calculations (FMT_050405.CHEMDAT), so these differences were caused by the differences in the organic-ligand concentrations between the CRA-2004 PABC and the CRA-2009 PABC

Tables 8 and 9 also show that when we used the thermodynamic database FMT_050405.CHEMDAT (Xiong, 2005), which includes only phase 3, FMT predicted that phase 3 will precipitate from GWB, but not from ERDA-6.[3] Tables 8 and 9 also show that when we used the database FMT_090720.CHEMDAT (Xiong et al, 2009; Xiong, 2009), which includes both phase 3 and phase 5, FMT predicted that phase 5 will precipitate from GWB instead of phase 3, but that neither of these phases will precipitate from ERDA-6.[4] The results obtained with the database FMT_090720.CHEMDAT agree with the results of laboratory studies of MgO at SNL in Carlsbad (Wang & Bryan, 2000; Wang et al., 2001; Snider and Xiong, 2002; Snider et al., 2004; Deng et al., 2007), which have shown that phase 5 precipitates from GWB instead of phase 3, but that neither of these phases precipitates from ERDA-6.

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[1]See columns labeled "GWB after Eq., w Phase 3 & Organics, CRA-2004 PABC Run 7" and "GWB after Eq., w Phase 3 & Organics, CRA-2009 PABC Run 5."

[2]See columns labeled "ERDA-6 after Eq., w/o Phase 3, w Organics, CRA-2004 PABC Run 11" and "ERDA-6 after Eq., w/o Phase 3, w Organics, CRA-2009 PABC Run 13."

[3]Compare columns labeled "GWB after Eq., w Phase 3 & Organics, CRA-2004 PABC Run 7" and "GWB after Eq., w Phase 3 & Organics, CRA-2009 PABC Run 5" in Table 8 with columns labeled "ERDA-6 after Eq., w/o Phase 3, w Organics, CRA-2004 PABC Run 11" and "ERDA-6 after Eq., w/o Phase 3, w Organics, CRA-2009 PABC Run 13" in Table 9.

[4]Compare column labeled "GWB after Eq., w Phase 5 & Organics, CRA-2009 PABC Run 19" in Table 8 with column labeled "ERDA-6 after Eq., w/o Phase 3, 5, w Organics, CRA-2009 PABC Run 23" in Table 9.

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Finally, Table 8 shows that FMT predicted a somewhat different composition for GWB after precipitation of phase 5 than it predicted after precipitation of phase 3.[5] Because we used the same initial brine composition for GWB (Krumhansl et al., 1991; Snider, 2003) and the same organic-ligand concentrations (Table 2, column labeled "CRA-2009 PABC"), these differences were caused by the differences in the databases used for these calculations (FMT_050405.CHEMDAT, which includes only phase 3; and FMT_090720.CHEMDAT, which includes both phase 3 and phase 5). On the other hand, Table 9 shows that the use of different databases did not result in any predicted differences for ERDA-6.[6] This is because neither phase 3 nor phase 5 precipitated from ERDA-6.

Tables 10, 11, and 12 provide the solubilities of Th(IV), Np(V), and Am(III) from our FMT calculations for the CRA-2009 PABC. These tables also provide the values of f_{CO_2} and pH calculated by FMT. Runs 5 and 13 provide the solubilities that were actually used in the CRA-2009 PABC.

We obtained the Th(IV), Np(V), and Am(III) solubilities in Tables 10, 11, and 12 from the table entitled "Elemental Abundances for Flash Problem" near the end of each FMT output file. Note that each of these solubilities is equal to the sum of all of the aqueous Th(IV), Np(V), and Am(III) species listed in the table entitled "Table of Concentrations for Batch Systems," which follows the table entitled "Elemental Abundances for Flash Problem." We obtained the f_{CO_2} and pH from the data listed below the table entitled "Table of Concentrations for Batch Systems."

Tables 13 and 14 provide the dissolved species distributions for Th(IV) in GWB and ERDA-6, respectively; Tables 15 and 16 give the species distributions for Np(IV) in these brines; Tables 17 and 18 show these results for Am(III). These tables show that FMT predicted somewhat different species distributions for GWB and ERDA-6 for the CRA-2009 PABC for the same actinide oxidation state with the same database.[7] These differences are similar to those obtained for the CRA-2004 PABC (Brush, 2005). These tables also show the effects of using different databases for the same brine. In the case of GWB, the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5, produced results somewhat different from those obtained using FMT_050405.CHEMDAT, which includes only phase 3.[8] For ERDA-6, however, these databases produce identical results.[9] This is because neither phase 3 nor phase 5 precipitated from ERDA-6.

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[5]Compare columns labeled "GWB after Eq., w Phase 3 & Organics, CRA-2009 PABC Run 5" and "GWB after Eq., w Phase 5 & Organics, CRA-2009 PABC Run 19."

[6]Compare columns labeled "ERDA-6 after Eq., w/o Phase 3, w Organics, CRA-2009 PABC Run 13" and "ERDA-6 after Eq., w/o Phase 3, 5, w Organics, CRA-2009 PABC Run 23."

[7]Compare, for example, the columns labeled "GWB, Hydromagnesite, w Phase 3, w Organics (FMT Run 5)" in Table 13 with those labeled "ERDA-6, Hydromagnesite, w/o Phase 3, w Organics (FMT Run 13)" in Table 14).

[8]Compare, for example, columns labeled "GWB, Hydromagnesite, w Phase 3, w Organics (FMT Run 5)" and "GWB, Hydromagnesite, w Phase 5, w Organics (FMT Run 19)" in Table 13.

[9]Compare columns labeled "ERDA-6 Hydromagnesite, w/o Phase 3, w Organics (FMT Run 13)" and "ERDA-6, Hydromagnesite, w/o Phase 3 or 5, w Organics (FMT \ Run 23)" in Table 14.

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Table 8. Composition of GWB (M Unless Otherwise Noted) and Other Parameters (Units as Noted) before and after Equilibration with Solids (see text).

Element or Property	GWB before Eq.,[A]	GWB w Organics after Eq., w Phase 3, CRA-2004 PABC Run 7[B]	GWB w Organics after Eq., w Phase 3, CRA-2009 PABC Run 5 [C]	GWB w Organics after Eq., w Phase 5, CRA-2009 PABC Run 19[D]
B(III)(aq)	0.158	0.166	0.176	0.180
Na(I)(aq)	3.53	4.35	4.31	4.52
Mg(II)(aq)	1.02	0.578	0.584	0.463
K(I)(aq)	0.467	0.490	0.521	0.532
Ca(II)(aq)	0.014	0.00895	0.0098	0.010
S(VI)(aq)	0.177	0.228	0.210	0.214
Cl(-I)(aq)	5.86	5.38	5.40	5.37
Br(-I)(aq)	0.0266	0.0278	0.0297	0.0283
I (m)	-	7.66	7.64	7.52
f_{CO_2} (atm)	-	3.16×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
TIC (mM)	-	0.350	0.350	0.358
pH (Pitzer scale[E])	-	8.69	8.69	8.74
pcH[F]	-	9.39	9.40	9.45
pmH[G]	-	9.33	9.34	9.39
RH (%)	-	73.2	73.2	73.4

Table 8 continued on next page

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Table 8. Composition of GWB (M Unless Otherwise Noted) and Other Parameters (Units as Noted) before and after Equilibration with Solids (continued).

Element or Property	GWB before Eq.[A]	GWB w Organics after Eq., w Phase 3, CRA-2004 PABC Run 7[B]	GWB w Organics after Eq., w Phase 3, CRA-2009 PABC Run 5[C]	GWB w Organics after Eq., w Phase 5, CRA-2009 PABC Run 19[D]
ρ (kg/m^3)	-	1230	1233	1234

[A]Krumhansl et al. (1991) and Snider (2003).
[B]Values calculated by Brush and Xiong (2005a, 2005b) and Brush (2005, FMT Run 7) for the CRA-2004 PABC. Run 7 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[C]This report. Run 5 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[D]This report. Run 19 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.
[E]The Pitzer scale is an unofficial pH scale consistent with pH values calculated using single-ion activity coefficients based on the Pitzer activity-coefficient model for brines and evaporite minerals of Harvie et al. (1984), extended to include Nd(III), Am(III), and Cm(III); Th(IV); and Np(V). The term "Pitzer scale" was proposed unofficially by T. J. Wolery of Lawrence Livermore National Laboratory (LLNL) in Livermore, CA.
[F]The negative, common logarithm of the molar concentration of H^+.
[G]The negative, common logarithm of the molal concentration of H^+.

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Table 9. Composition of ERDA-6 (M Unless Otherwise Noted) and Other Parameters (Units as Noted) before and after Equilibration with Solids (see text).

Element or Property	ERDA-6 before Eq.[A]	ERDA-6 w Organics after Eq., w/o Phase 3, CRA-2004 PABC Run 11[B]	ERDA--6 w Organics after Eq., w/o Phase 3, CRA-2009 PABC Run 13[C]	ERDA-6 w Organics after Eq., w/o Phase 3, 5, CRA-2009 PABC Run 23[D]
B(III)(aq)	0.063	0.0624	0.0624	0.0624
Na(I)(aq)	4.87	5.24	5.28	5.28
Mg(II)(aq)	0.019	0.157	0.136	0.136
K(I)(aq)	0.097	0.0961	0.0961	0.0961
Ca(II)(aq)	0.012	0.0107	0.0112	0.0112
S(VI)(aq)	0.170	0.179	0.176	0.176
Cl(-I)(aq)	4.8	5.24	5.23	5.23
Br(-I)(aq)	0.011	0.0109	0.0109	0.0109
I (m)	-	6.80	6.77	6.77
f_{CO_2} (atm)	-	3.16×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
TIC (mM)	16	0.428	0.448	0.448
pH (Pitzer scale[E])	6.17	8.94	8.98	8.98
pcH[F]	-	9.64	9.68	9.68
pmH[G]	-	9.59	9.63	9.63
RH (%)	-	74.8	74.8	74.8

Table 9 continued on next page

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Table 9. Composition of ERDA-6 (M Unless Otherwise Noted) and Other Parameters (Units as Noted) before and after Equilibration with Solids (continued).

Element or Property	ERDA-6 before Eq.[A]	ERDA-6 w Organics after Eq., w/o Phase 3, CRA-2004 PABC Run 11[B]	ERDA-6 w Organics after Eq., w/o Phase 3, CRA-2009 PABC Run 13[C]	ERDA-6 w Organics after Eq., w/o Phase 3, 5, CRA-2009 PABC Run 23[D]
ρ (kg/m^3)	-	1220	1220	1220

[A]Popielak et al. (1983).
[B]Values calculated by Brush and Xiong (2005a, 2005b) and Brush (2005, FMT Run 11) for the CRA-2004 PABC. Run 11 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[C]This report. Run 13 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[D]This report. Run 23 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.
[E]The Pitzer scale is an unofficial pH scale consistent with pH values calculated using single-ion activity coefficients based on the Pitzer activity-coefficient model for brines and evaporite minerals of Harvie et al. (1984), extended to include Nd(III), Am(III), and Cm(III); Th(IV); and Np(V). The term "Pitzer scale" was proposed unofficially by T. J. Wolery of Lawrence Livermore National Laboratory (LLNL) in Livermore, CA.
[F]The negative, common logarithm of the molar concentration of H^+.
[G]The negative, common logarithm of the molal concentration of H^+.

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Table 10. Actinide Solubilities (M), f_{CO_2} (atm), and pH (Pitzer Scale[A]) from the FMT Runs for the CRA-2009 PABC with GWB and FMT_050405.CHEMDAT. (This database contains only phase 3.) All results rounded to three significant figures.

Property or Actinide Oxidation State	FMT Run 1 (GWB, Calcite,[B] w Phase 3, w Organics	FMT Run 2 (GWB, Calcite,[B] w Phase 3, w/o Organics	FMT Run 3 (GWB, Magnesite,[C] w Phase 3, w Organics)	FMT Run 4 (GWB, Magnesite,[C] w Phase 3, w/o Organics)	FMT Run 5 (GWB, Hydro-magnesite,[D] w Phase 3, w Organics)	FMT Run 6 (GWB, Hydro-magnesite,[D] w Phase 3, w/o Organics)	FMT Run 7 (GWB, Nesque-honite,[E] w Phase 3, w Organics)	FMT Run 8 (GWB, Nesque-honite,[E] Phase 3, w/o Organics)
Th(IV)	5.86×10^{-8}	5.76×10^{-8}	4.56×10^{-8}	4.57×10^{-8}	5.63×10^{-8}	5.64×10^{-8}	6.31×10^{-7}	6.06×10^{-7}
Np(V)	3.40×10^{-7}	2.10×10^{-7}	7.68×10^{-6}	3.22×10^{-6}	3.90×10^{-7}	2.21×10^{-7}	1.84×10^{-7}	1.95×10^{-7}
Am(III)	1.66×10^{-6}	2.25×10^{-7}	1.66×10^{-6}	2.25×10^{-7}	1.66×10^{-6}	2.25×10^{-7}	2.42×10^{-6}	2.05×10^{-6}
f_{CO_2}	3.80×10^{-6}	3.46×10^{-6}	1.20×10^{-7}	1.20×10^{-7}	3.14×10^{-6}	3.14×10^{-6}	1.43×10^{-4}	1.42×10^{-4}
pH[F]	8.69	8.69	8.69	8.69	8.69	8.69	8.69	8.69

Footnotes for Table 10 provided on the next page.

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Footnotes for Table 10:

[A]The Pitzer scale is an unofficial pH scale consistent with pH values calculated using single-ion activity coefficients based on the Pitzer activity-coefficient model for brines and evaporite minerals of Harvie et al. (1984), extended to include Nd(III), Am(III), and Cm(III); Th(IV); and Np(V). The term "Pitzer scale" was proposed unofficially by T. J. Wolery of Lawrence Livermore National Laboratory (LLNL) in Livermore, CA.

[B]f_{CO_2} buffered by the brucite-calcite carbonation reaction (Brush and Xiong, 2009a, Reaction 4).

[C]f_{CO_2} buffered by the brucite-magnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 3).

[D]f_{CO_2} buffered by the brucite-hydromagnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 1).

[E]f_{CO_2} buffered by the brucite-nesquehonite carbonation reaction (U.S. DOE, 2009, Appendix MgO, Reaction MgO.11).

[F]pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

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Table 11. Actinide Solubilities (M), f_{CO_2} (atm), and pH (Pitzer Scale[A]) from the FMT Runs for the CRA-2009 PABC with ERDA-6 and FMT_050405.CHEMDAT. (This database contains only phase 3.) All results rounded to three significant figures.

Property or Actinide Oxidation State	FMT Run 9 (EDRA-6, Calcite,[B] w/o Phase 3, w Organics	FMT Run 10 (EDRA-6, Calcite,[B] w/o Phase 3, w/o Organics	FMT Run 11 (EDRA-6, Magnesite,[C] w/o Phase 3, w Organics)	FMT Run 12 (EDRA-6, Magnesite,[C] w/o Phase 3, w/o Organics)	FMT Run 3 (EDRA-6, Hydro-magnesite,[D] w/o Phase 3, w Organics)	FMT Run 14 (EDRA-6, Hydro-magnesite,[D] w/o Phase 3, w/o Organics)	FMT Run 15 (EDRA6, Nesque-honite,[E] w/o Phase 3, w Organics)	FMT Run 16 (EDRA-6, Nesque-honite,[E] w/o Phase 3, w/o Organics)
Th(IV)	5.31×10^{-8}	5.27×10^{-8}	4.85×10^{-8}	4.87×10^{-8}	6.98×10^{-8}	7.20×10^{-8}	1.00×10^{-6}	1.06×10^{-6}
Np(V)	2.35×10^{-6}	1.07×10^{-6}	1.23×10^{-5}	3.65×10^{-6}	8.75×10^{-7}	5.38×10^{-7}	9.70×10^{-7}	9.89×10^{-7}
Am(III)	1.55×10^{-6}	9.14×10^{-8}	1.51×10^{-6}	8.60×10^{-8}	1.51×10^{-6}	8.67×10^{-8}	5.62×10^{-7}	5.42×10^{-7}
f_{CO_2}	8.21×10^{-7}	6.94×10^{-7}	1.23×10^{-7}	1.23×10^{-7}	3.14×10^{-6}	3.14×10^{-6}	1.37×10^{-4}	1.36×10^{-4}
pH[F]	8.96	8.99	8.98	9.02	8.98	9.02	8.98	9.00

Footnotes for Table 11 provided on the next page.

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Footnotes for Table 11:

[A]The Pitzer scale is an unofficial pH scale consistent with pH values calculated using single-ion activity coefficients based on the Pitzer activity-coefficient model for brines and evaporite minerals of Harvie et al. (1984), extended to include Nd(III), Am(III), and Cm(III); Th(IV); and Np(V). The term "Pitzer scale" was proposed unofficially by T. J. Wolery of Lawrence Livermore National Laboratory (LLNL) in Livermore, CA.

[B]f_{CO_2} buffered by the brucite-calcite carbonation reaction (Brush and Xiong, 2009a, Reaction 4).

[C]f_{CO_2} buffered by the brucite-magnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 3).

[D]f_{CO_2} buffered by the brucite-hydromagnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 1).

[E]f_{CO_2} buffered by the brucite-nesquehonite carbonation reaction (U.S. DOE, 2009, Appendix MgO, Reaction MgO.11).

[F]pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

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Table 12. Actinide Solubilities (M), f_{CO_2} (atm), and pH (Pitzer Scale[A]) from the FMT Runs for the CRA-2009 PABC with GWB or ERDA-6 and FMT_090720.CHEMDAT. (This database contains both phase 3 and phase 5.) All results rounded to three significant figures.

Property or Actinide Oxidation State	FMT Run 17 (GWB, Calcite,[B] w Phase 5, w Organics	FMT Run 18 (GWB, Magnesite,[C] w Phase 5, w Organics	FMT Run 19 (GWB, Hydro-magnesite,[D] w Phase 5, w Organics)	FMT Run 20 (GWB, Nesque-honite,[E] w Phase 5, w Organics)	FMT Run 21 (EDRA-6, Calcite,[B] w/o Phase 3 or 5, w Organics)	FMT Run 22 (EDRA-6, Magnesite,[C] w/o Phase 3 or 5, w Organics)	FMT Run 23 (EDRA-6, Hydro-magnesite,[D] w/o Phase 3 or 5, w Organics)	FMT Run 24 (EDRA-6, Nesque-honite,[E] w/o Phase 3 or 5, w Organics)
Th(IV)	5.71×10^{-8}	4.59×10^{-8}	5.78×10^{-8}	6.24×10^{-7}	5.31×10^{-8}	4.85×10^{-8}	6.98×10^{-8}	1.00×10^{-6}
Np(V)	3.45×10^{-7}	6.33×10^{-6}	3.30×10^{-7}	1.60×10^{-7}	2.35×10^{-6}	1.23×10^{-5}	8.75×10^{-7}	9.70×10^{-7}
Am(III)	1.97×10^{-6}	1.97×10^{-6}	1.97×10^{-6}	9.88×10^{-7}	1.55×10^{-6}	1.51×10^{-6}	1.51×10^{-6}	5.62×10^{-7}
f_{CO_2}	2.95×10^{-6}	1.21×10^{-7}	3.14×10^{-6}	1.42×10^{-4}	8.21×10^{-7}	1.23×10^{-7}	3.14×10^{-6}	1.37×10^{-4}
pH[F]	8.74	8.74	8.74	8.74	8.96	8.98	8.98	8.98

Footnotes for Table 12 provided on the next page.

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Footnotes for Table 12:

[A]The Pitzer scale is an unofficial pH scale consistent with pH values calculated using single-ion activity coefficients based on the Pitzer activity-coefficient model for brines and evaporite minerals of Harvie et al. (1984), extended to include Nd(III), Am(III), and Cm(III); Th(IV); and Np(V). The term "Pitzer scale" was proposed unofficially by T. J. Wolery of Lawrence Livermore National Laboratory (LLNL) in Livermore, CA.

[B]f_{CO_2} buffered by the brucite-calcite carbonation reaction (Brush and Xiong, 2009a, Reaction 4); pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

[C]f_{CO_2} buffered by the brucite-magnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 3); pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

[D]f_{CO_2} buffered by the brucite-hydromagnesite carbonation reaction (Brush and Xiong, 2009a, Reaction 1); pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

[E]f_{CO_2} buffered by the brucite-nesquehonite carbonation reaction (U.S. DOE, 2009, Appendix MgO, Reaction MgO.11); pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

[F]pH controlled by the brucite dissolution-precipitation reaction (Brush and Xiong, 2009a, Reaction 2).

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Table 13. Comparisons of Distributions of Dissolved Th(IV) Species in GWB Predicted for the CRA-2009 PABC with Phase 3 or Phase 5. Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was hydrous, amorphous ThO_2.

GWB, Hydromagnesite, w Phase 3, w Organics (FMT Run 5[A])			GWB, Hydromagnesite, w Phase 5, w Organics (FMT Run 19[B])		
Th(IV) Species	Concentration (M)	Percent of Solubility	Th(IV) Species	Concentration (M)	Percent of Solubility
$Th(OH)_4(aq)$	4.52×10^{-8}	80.3	$Th(OH)_4(aq)$	4.54×10^{-8}	78.5
$Th(OH)_3CO_3^-$	1.11×10^{-8}	19.7	$Th(OH)_3CO_3^-$	1.25×10^{-8}	21.5
$Th(CO_3)_5^{6-}$	4.02×10^{-16}	-	$Th(CO_3)_5^{6-}$	8.30×10^{-17}	-
ThEDTA(aq)	6.17×10^{-17}	-	ThEDTA(aq)	6.26×10^{-17}	-
$Th(SO_4)_3^{2-}$	1.15×10^{-17}	-	$Th(SO_4)_3^{2-}$	6.08×10^{-18}	-
$Th(SO_4)_2(aq)$	2.44×10^{-19}	-	$ThCit^+$	1.34×10^{-19}	-
$ThCit^+$	1.68×10^{-19}	-	$Th(SO_4)_2(aq)$	1.49×10^{-19}	-
$ThAc_2^{2+}$	2.63×10^{-20}	-	$ThAc_2^{2+}$	2.48×10^{-20}	-
$ThAc^{3+}$	1.67×10^{-21}	-	$ThAc^{3+}$	1.41×10^{-21}	-
$ThOx^{2+}$	1.16×10^{-22}	-	$ThOx^{2+}$	7.36×10^{-23}	-
Total dissolved Th(IV) species (solubility)	5.63×10^{-8}	100	Total dissolved Th (IV) species (solubility)	5.78×10^{-8}	100

[A]Run 5 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[B]Run 19 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.

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Table 14. Comparisons of Distributions of Dissolved Th(IV) Species in ERDA-6 Predicted for the CRA-2009 PABC w/o Phase 3 or Phase 5. Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was hydrous, amorphous ThO_2.

ERDA-6, Hydromagnesite, w/o Phase 3, w Organics (FMT Run 13[A])			ERDA-6, Hydromagnesite, w/o Phase 3 or 5, w Organics (FMT Run 23[B])		
Th(IV) Species	Concentration (M)	Percent of Solubility	Th(IV) Species	Concentration (M)	Percent of Solubility
$Th(OH)_4(aq)$	4.76×10^{-8}	68.2	$Th(OH)_4(aq)$	4.76×10^{-8}	68.2
$Th(OH)_3CO_3^-$	2.22×10^{-8}	31.8	$Th(OH)_3CO_3^-$	2.22×10^{-8}	31.8
ThEDTA(aq)	2.83×10^{-17}	-	ThEDTA(aq)	2.83×10^{-17}	-
$Th(CO_3)_5^{6-}$	1.92×10^{-17}	-	$Th(CO_3)_5^{6-}$	1.92×10^{-17}	-
$Th(SO_4)_3^{2-}$	5.18×10^{-19}	-	$Th(SO_4)_3^{2-}$	5.18×10^{-19}	-
$ThCit^+$	9.83×10^{-20}	-	$ThCit^+$	9.83×10^{-20}	-
$Th(SO_4)_2(aq)$	1.03×10^{-20}	-	$Th(SO_4)_2(aq)$	1.03×10^{-20}	-
$ThAc_2^{2+}$	7.38×10^{-21}	-	$ThAc_2^{2+}$	7.38×10^{-21}	-
$ThAc^{3+}$	3.12×10^{-22}	-	$ThAc^{3+}$	3.12×10^{-22}	-
Total dissolved Th(IV) species (solubility)	6.98×10^{-8}	100	Total dissolved Th(IV) species (solubility)	6.98×10^{-8}	100

[A]Run 13 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[B]Run 23 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.

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Table 15. Comparisons of Distributions of Dissolved Np(V) Species in GWB Predicted for the CRA-2009 PABC with Phase 3 or Phase 5. Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was $KNpO_2CO_3$.

GWB, Hydromagnesite, w Phase 3, w Organics (FMT Run 5[A])			GWB, Hydromagnesite, w Phase 5, w Organics (FMT Run 19[B])		
Np(V) Species	Concentration (M)	Percent of Solubility	Np(V) Species	Concentration (M)	Percent of Solubility
$NpO_2Ac(aq)$	1.32×10^{-7}	33.9	$NpO_2Ac(aq)$	1.20×10^{-7}	36.3
NpO_2^+	1.17×10^{-7}	29.9	$NpO_2CO_3^-$	8.86×10^{-8}	26.9
$NpO_2CO_3^-$	9.84×10^{-8}	25.2	NpO_2^+	8.66×10^{-8}	26.3
NpO_2Ox^-	3.72×10^{-8}	9.53	NpO_2Ox^-	3.01×10^{-8}	9.12
$NpO_2(OH)(aq)$	4.08×10^{-9}	1.05	$NpO_2(OH)(aq)$	3.42×10^{-9}	1.04
$NpO_2(CO_3)_2^{3-}$	1.13×10^{-9}	0.288	$NpO_2(CO_3)_2^{3-}$	1.05×10^{-9}	0.319
NpO_2Cit^{2-}	4.30×10^{-10}	0.0110	NpO_2Cit^{2-}	3.90×10^{-10}	0.0118
$NpO_2(CO_3)_3^{5-}$	1.34×10^{-11}	-	$NpO_2(CO_3)_3^{5-}$	9.08×10^{-12}	-
$NpO_2(OH)_2^-$	5.97×10^{-12}	-	$NpO_2(OH)_2^-$	5.59×10^{-12}	-
NpO_2EDTA^{3-}	3.47×10^{-12}	-	NpO_2EDTA^{3-}	2.17×10^{-12}	-
NpO_2HEDTA^{2-}	6.64×10^{-15}	-	NpO_2HEDTA^{2-}	5.83×10^{-15}	-
$NpO_2H_2EDTA^-$	1.01×10^{-18}	-	$NpO_2H_2EDTA^-$	9.32×10^{-19}	-
Total dissolved Np(V) species (solubility)	3.90×10^{-7}	100	Total dissolved Np(V) species (solubility)	3.30×10^{-7}	100

Footnotes for Table 15 provided on next page.

Information Only

Footnotes for Table 15.

[A]Run 5 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[B]Run 19 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.

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Table 16. Comparisons of Distributions of Dissolved Np(V) Species in ERDA-6 Predicted for the CRA-2009 PABC w/o Phase 3 or Phase 5. Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was $KNpO_2CO_3$.

ERDA-6, Hydromagnesite, w/o Phase 3, w Organics (FMT Run 13[A])			ERDA-6, Hydromagnesite, w/o Phase 3 or 5, w Organics (FMT Run 23[B])		
Np(V) Species	Concentration (M)	Percent of Solubility	Np(V) Species	Concentration (M)	Percent of Solubility
$NpO_2CO_3^-$	3.99×10^{-7}	45.6	$NpO_2CO_3^-$	3.99×10^{-7}	45.7
$NpO_2Ac(aq)$	2.59×10^{-7}	29.6	$NpO_2Ac(aq)$	2.59×10^{-7}	29.6
NpO_2^+	1.44×10^{-7}	16.4	NpO_2^+	1.44×10^{-7}	16.4
NpO_2Ox^-	5.09×10^{-8}	5.82	NpO_2Ox^-	5.09×10^{-8}	5.82
$NpO_2(CO_3)_2^{3-}$	9.81×10^{-9}	1.12	$NpO_2(CO_3)_2^{3-}$	9.81×10^{-9}	1.12
$NpO_2(OH)(aq)$	9.41×10^{-9}	1.08	$NpO_2(OH)(aq)$	9.41×10^{-9}	1.08
NpO_2Cit^{2-}	2.01×10^{-9}	0.230	NpO_2Cit^{2-}	2.01×10^{-9}	0.229
$NpO_2(CO_3)_3^{5-}$	7.79×10^{-11}	-	$NpO_2(CO_3)_3^{5-}$	7.79×10^{-11}	-
$NpO_2(OH)_2^-$	2.60×10^{-11}	-	$NpO_2(OH)_2^-$	2.60×10^{-11}	-
NpO_2EDTA^{3-}	3.81×10^{-12}	-	NpO_2EDTA^{3-}	3.81×10^{-12}	-
NpO_2HEDTA^{2-}	1.87×10^{-14}	-	NpO_2HEDTA^{2-}	1.87×10^{-14}	-
$NpO_2H_2EDTA^-$	3.54×10^{-18}	-	$NpO_2H_2EDTA^-$	3.54×10^{-18}	-
Total dissolved Np(V) species (solubility)	8.75×10^{-7}	100	Total dissolved Np(V) species (solubility)	8.75×10^{-7}	100

Footnotes for Table 16 provided on next page.

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Footnotes for Table 16.

[A]Run 13 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[B]Run 23 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.

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Table 17. Comparisons of Distributions of Dissolved Am(III) Species in GWB Predicted for the CRA-2009 PABC with Phase 3 or Phase 5. Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was $Am(OH)_3$.

GWB, Hydromagnesite, w Phase 3, w Organics (FMT Run 5[A])			GWB, Hydromagnesite, w Phase 5, w Organics (FMT Run 19[B])		
Am(III) Species	Concentration (M)	Percent of Solubility	Am(III) Species	Concentration (M)	Percent of Solubility
$AmEDTA^-$	1.44×10^{-6}	86.8	$AmEDTA^-$	1.79×10^{-6}	90.5
$Am(OH)_2^+$	2.11×10^{-7}	12.8	$Am(OH)_2^+$	1.81×10^{-7}	9.18
$Am(OH)^{2+}$	2.41×10^{-9}	0.146	$Am(OH)^{2+}$	1.90×10^{-9}	0.0962
$AmAc^{2+}$	2.12×10^{-9}	0.128	$AmAc^{2+}$	1.77×10^{-9}	0.0899
AmCit(aq)	1.40×10^{-9}	0.0847	AmCit(aq)	1.23×10^{-9}	0.0625
$Am(OH)_3(aq)$	6.03×10^{-10}	0.0364	$Am(OH)_3(aq)$	6.64×10^{-10}	0.0336
$AmCO_3^+$	3.73×10^{-10}	0.0226	$AmCO_3^+$	3.24×10^{-10}	0.0164
$Am(CO_3)_2^-$	1.33×10^{-10}	-	$Am(CO_3)_2^-$	1.58×10^{-10}	-
$AmSO_4^+$	1.13×10^{-10}	-	$AmSO_4^+$	7.70×10^{-11}	-
$Am(CO_3)_3^{3-}$	3.39×10^{-11}	-	$Am(CO_3)_3^{3-}$	3.49×10^{-11}	-
$AmOx^+$	2.33×10^{-11}	-	$AmOx^+$	1.56×10^{-11}	-
$Am(SO_4)_2^-$	1.60×10^{-11}	-	$Am(SO_4)_2^-$	1.16×10^{-11}	-
Am^{3+}	1.24×10^{-11}	-	Am^{3+}	8.75×10^{-12}	-
$Am(CO_3)_4^{5-}$	1.05×10^{-11}	-	$Am(CO_3)_4^{5-}$	3.54×10^{-12}	-

Table 17 continued on next page.

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Table 17. Comparisons of Distributions of Dissolved Am(III) Species in GWB Predicted for the CRA-2009 PABC with Phase 3 or Phase 5 (continued). Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was $Am(OH)_3$.

GWB, Hydromagnesite, w Phase 3, w Organics (FMT Run 5[A])			GWB, Hydromagnesite, w Phase 5, w Organics (FMT Run 19[B])		
Am(III) Species	Concentration (M)	Percent of Solubility	Am(III) Species	Concentration (M)	Percent of Solubility
$AmCl^{2+}$	1.75×10^{-12}	-	$AmCl^{2+}$	1.16×10^{-12}	-
$AmCl_2^+$	9.21×10^{-14}	-	$AmCl_2^+$	5.41×10^{-14}	-
Total dissolved Am(III) species (solubility)	1.66×10^{-6}	100	Total dissolved Am(III) species (solubility)	1.97×10^{-6}	100

[A]Run 5 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[B]Run 19 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.

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Table 18. Comparisons of Distributions of Dissolved Am(III) Species in ERDA-6 Predicted for the CRA-2009 PABC w/o Phase 3 or Phase 5. Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was $Am(OH)_3$.

ERDA-6 Hydromagnesite, w/o Phase 3, w Organics (FMT Run 13[A])			ERDA-6, Hydromagnesite, w/o Phase 3 or 5, w Organics (FMT Run 23[B])		
Am(III) Species	Concentration (M)	Percent of Solubility	Am(III) Species	Concentration (M)	Percent of Solubility
$AmEDTA^-$	1.42×10^{-6}	93.9	$AmEDTA^-$	1.42×10^{-6}	93.9
$Am(OH)_2^+$	9.03×10^{-8}	5.96	$Am(OH)_2^+$	9.03×10^{-8}	5.96
$Am(OH)_3(aq)$	6.93×10^{-10}	0.0458	$Am(OH)_3(aq)$	6.93×10^{-10}	0.0458
$Am(OH)^{2+}$	5.85×10^{-10}	0.0387	$Am(OH)^{2+}$	5.85×10^{-10}	0.0386
AmCit(aq)	5.79×10^{-10}	0.0383	AmCit(aq)	5.79×10^{-10}	0.0382
$Am(CO_3)_2^-$	3.02×10^{-10}	0.0200	$Am(CO_3)_2^-$	3.02×10^{-10}	0.0200
$AmAc^{2+}$	2.98×10^{-10}	0.0197	$AmAc^{2+}$	2.98×10^{-10}	0.0196
$AmCO_3^+$	1.86×10^{-10}	0.0123	$AmCO_3^+$	1.86×10^{-10}	0.0123
$Am(CO_3)_3^{3-}$	7.75×10^{-11}	-	$Am(CO_3)_3^{3-}$	7.75×10^{-11}	-
$AmSO_4^+$	1.16×10^{-11}	-	$AmSO_4^+$	1.16×10^{-11}	-
$AmOx^+$	3.25×10^{-12}	-	$AmOx^+$	3.25×10^{-12}	-
Am^{3+}	2.27×10^{-12}	-	Am^{3+}	2.27×10^{-12}	-
$Am(SO_4)_2^-$	1.41×10^{-12}	-	$Am(SO_4)_2^-$	1.41×10^{-12}	-
$Am(CO_3)_4^{5-}$	7.82×10^{-13}	-	$Am(CO_3)_4^{5-}$	7.82×10^{-13}	-

Table 18 continued on next page.

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Table 18. Comparisons of Distributions of Dissolved Am(III) Species in ERDA-6 Predicted for the CRA-2009 PABC w/o Phase 3 or Phase 5 (continued). Percentages of solubilities < 0.01 were not reported. Percentages may not add up to 100% because concentrations were rounded to three significant figures before calculating percentages. The solubility-controlling solid was $Am(OH)_3$.

ERDA-6 Hydromagnesite, w/o Phase 3, w Organics (FMT Run 13[A])			ERDA-6, Hydromagnesite, w/o Phase 3 or 5, w Organics (FMT Run 23[B])		
Am(III) Species	Concentration (M)	Percent of Solubility	Am(III) Species	Concentration (M)	Percent of Solubility
$AmCl^{2+}$	1.77×10^{-13}	-	$AmCl^{2+}$	1.77×10^{-13}	-
$AmCl_2^{+}$	6.71×10^{-15}	-	$AmCl_2^{+}$	6.71×10^{-15}	-
Total dissolved Am(III) species (solubility)	1.51×10^{-6}	100	Total dissolved Am(III) species (solubility)	1.51×10^{-6}	100

[A]Run 13 carried out with the database FMT_050405.CHEMDAT, which includes only phase 3.
[B]Run 23 carried out with the database FMT_090720.CHEMDAT, which includes both phase 3 and phase 5.

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4 CONCLUSIONS

This analysis report provides the results of our calculations of the speciation and solubilities of actinide elements for the CRA-2009 PABC. Table 19 (see next page) is the parameter data entry table for the Th(IV), Np(V), and Am(III) solubilities predicted by FMT Run 5 (see Table 10 above) and 13 (Table 11), the results that will actually be used for the CRA-2009 PABC. The results of the other runs in Tables 10, 11, and 12 will be used for various sensitivity analyses.

Table 20 (see next page) compares the solubilities of An(III), An(IV), and An(V) calculated for the PABC with the An solubilities calculated for the CCA PA, the PAVT, and the CRA-2004 PA. Table 20 also compares the values of f_{CO_2} and pH calculated by FMT. The CRA-2004 PA results are from Brush and Xiong (2003c).

Comparison of these results is not straightforward because several assumptions or other factors changed from calculation to calculation. These include: (1) assumptions as to which carbonation reaction will buffer f_{CO_2} (i.e., brucite-magnesite of brucite-hydromagnesite; see Brush and Xiong, 2009a, Subsection 2.1.2), (2) changes in the concentrations of organic ligands in the brines used to calculate these solubilities (Table 2 above), and (3) changes in the FMT thermodynamic database (Brush and Xiong, 2009a, Subsection 2.2.4).

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Table 19. Information Required for Parameter Data Entry for the Actinide Solubilities for the CRA-2009 PABC.

Parameter Description	Material	Property	Value	Units
Oxidation state III model, solubility in Salado brine with organics included controlled by the $Mg(OH)_2$-hydromagnesite(5424) buffer	SOLMOD3	SOLSOH	1.66×10^{-6}	mol/L (M)
Oxidation state III model, solubility in Castile brine with organics included controlled by the $Mg(OH)_2$-hydromagnesite(5424) buffer	SOLMOD3	SOLCOH	1.51×10^{-6}	M
Oxidation state IV model, solubility in Salado brine with organics included controlled by the $Mg(OH)_2$-hydromagnesite(5424) buffer	SOLMOD4	SOLSOH	5.63×10^{-8}	M
Oxidation state IV model, solubility in Castile brine with organics included controlled by $Mg(OH)_2$-hydromagnesite(5424) buffer	SOLMOD4	SOLCOH	6.98×10^{-8}	M
Oxidation state V model, solubility in Salado brine with organics included controlled by the $Mg(OH)_2$-hydromagnesite(5424) buffer	SOLMOD5	SOLSOH	3.90×10^{-7}	M
Oxidation state V model, solubility in Castille brine with organics included controlled by the $Mg(OH)_2$-hydromagnesite(5424) buffer	SOLMOD5	SOLCOH	8.75×10^{-7}	M

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Table 20. Comparison of Actinide Solubilities (M), f_{CO_2} (atm), and pH (Pitzer scale[A]) from These and Previous Compliance-Related FMT Runs. Results rounded to three significant figures if more than three were provided.

Property or Actinide Oxidation State	CCA (SPC, Magnesite, w/o Organics, All Vectors)[B]	CCA (ERDA-6, Magnesite, w/o Organics, All Vectors)[B]	PAVT (SPC, Hydro-magnesite, w/o Organics, All Vectors)[C]	PAVT (ERDA-6, Hydro-magnesite, w/o Organics, All Vectors)[C]	CRA-2004 PA (GWB, Hydro-magnesite, w Organics, Microbial Vectors)[D]	CRA-2004 PA (ERDA-6, Hydro-magnesite, w Organics, Microbial Vectors)[D]	CRA-2004 PABC (GWB, Hydro-magnesite, w Organics, All Vectors)[E]	CRA-2004 PABC (ERDA-6, Hydro-magnesite, w Organics, All Vectors)[E]	CRA-2009 PABC (GWB, Hydro-magnesite, w Organics, All Vectors)[F]	CRA-2009 PABC (ERDA-6, Hydro-magnesite, w Organics, All Vectors)[G]
Th(IV)	4.4×10^{-6}	6.0×10^{-9}	1.3×10^{-8}	4.1×10^{-8}	1.19×10^{-8}	2.47×10^{-8}	5.64×10^{-8}	6.79×10^{-8}	5.63×10^{-8}	6.98×10^{-8}
Np(V)	2.3×10^{-6}	2.2×10^{-6}	2.4×10^{-7}	4.8×10^{-7}	1.02×10^{-6}	5.08×10^{-6}	3.55×10^{-7}	8.24×10^{-7}	3.90×10^{-7}	8.75×10^{-7}
Am(III)	5.82×10^{-7}	6.52×10^{-8}	1.2×10^{-7}	1.3×10^{-8}	3.07×10^{-7}	1.69×10^{-7}	3.87×10^{-7}	2.88×10^{-7}	1.66×10^{-6}	1.51×10^{-6}
f_{CO_2}	1.29×10^{-7}	1.29×10^{-7}	3.16×10^{-6}	3.16×10^{-6}	3.16×10^{-6}	3.16×10^{-6}	3.16×10^{-6}	3.16×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
pH	8.69	9.24	8.69	9.24	8.69	9.02	8.69	8.94	8.69	8.98

[A]The Pitzer scale is an unofficial pH scale consistent with pH values calculated using single-ion activity coefficients based on the Pitzer activity-coefficient model for brines and evaporite minerals of Harvie et al. (1984), extended to include Nd(III), Am(III), and Cm(III); Th(IV); and Np(V). The term "Pitzer scale" was proposed unofficially by T. J. Wolery of Lawrence Livermore National Laboratory (LLNL) in Livermore, CA.

[B]From Novak et al. (1996) and U.S. DOE (1996, Appendix SOTERM), except that Novak et al. (1996) used molal instead of molar units. U.S. EPA (1998) cited Novak and Moore (1996) as the source of the CCA PA solubilities, but the Am(III) solubilities from Novak and Moore (1996) differ from those in Novak et al. (1996) and U.S. DOE (1996).

[C]From Trovato (1997). Novak (1997) also calculated actinide solubilities for the PAVT, but the EPA used the results of its own calculations.

[D]From Brush and Xiong (2003a, 2003b, 2003c, 2003d) and U.S. DOE (2004, Appendix SOTERM).

[E]Brush and Xiong (2005a, 2005b) and Brush (2005). These solubilities were also used for the CRA-2009 PA calculations.

[F]This report, FMT Run 5.

[G]This report, FMT Run 13.

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Yongliang,

I hereby authorize you to sign for me as a review requester on any of the Document Review and Comment Forms, NP 6-1-1, if necessary; or as an author on my approval line on page 1 of our analysis report "Results of the Calculation of Actinide Solubilities for the WIPP CRA-2009 PABC" while I am out of my Carlsbad office in October 2009.

The final version of this analysis report is attached for you to print out, sign for you and me, obtain all of the other signatures required for approval, and have Shannon put into the SNL/WIPP Records Center. Please check the Table of Contents and the List of Tables to make sure that the page numbers are still correct. Sometimes these have gotten scrambled when I have e-mailed this file. If you don't know how to get Word to correct the numbers, please ask Shannon. Also, please check to see that the numerical entries in Table 2 are still centered. Sometimes these have gotten uncentered too.

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Larry Brush

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