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Sandia National Laboratories
Waste Isolation Pilot Plant

Predictions of Actinide Solubilities as a Function of the Volume of Standard WIPP Brines

Work Carried Out under Task 2 of the Analysis Plan for WIPP Near-Field Geochemical Process Modeling, AP 153, Rev. 0.
To be included in the AP-153 records package

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1 INTRODUCTION

This analysis report provides the results of our predictions of the solubilities of Th(IV), Np(V), and Am(III) in the standard Waste Isolation Pilot Plant (WIPP) brines Generic Weep Brine (GWB) and Energy Research and Development Administration (WIPP Well) 6 (ERDA-6) as a function of the volume of these brines in WIPP disposal rooms. GWB is a synthetic brine representative of intergranular Salado Formation (Fm.) brines at or near the stratigraphic horizon of the repository (Krumhansl et al., 1991; Snider, 2003). ERDA-6 (Popielak et al., 1983) is a synthetic brine representative of fluids in brine reservoirs in the Castile Fm., which underlies the Salado Fm.

This analysis report also provides the predicted compositions of these brines and the values of parameters such as f_{CO_2}, pcH, TIC, etc., as a function of brine volume.

We will provide these actinide solubilities to Sandia National Laboratories' (SNL's) WIPP performance assessment (PA) personnel so that they can use them in an upcoming PA calculation.

We used EQ3/6, Version 8.0a (Wolery and Jarek, 2003; Wolery, 2008; Wolery et al., 2010; Xiong, 2011), for this analysis. Wolery (2008), Wolery et al. (2010) and Xiong (2011) completed the qualification of Version 8.0a of EQ3/6 according to Sandia National Laboratories' (SNL's) WIPP quality assurance (QA) procedures for WIPP compliance-related actinide solubility calculations. We also used FMT, Version 2.4 (Babb and Novak, 1997 and addenda; Wang, 1998), so that we could compare the results obtained with these codes.

This analysis was carried out under Task 2 of AP-153 (Brush and Xiong, 2011).

Table 1 (see next page) defines the abbreviations, acronyms, and initialisms used in this report.

Table 1. Abbreviations, Acronyms, and Initialisms.

Abbreviation, Acronym, or Initialism	Definition
acetate	CH_3COO^- or $CH_3CO_2^-$
Am, Am(III)	americium, americium in the +III oxidation state
am	amorphous
anhydrite	$CaSO_4$
AP	analysis plan
aq	aqueous
aragonite	$CaCO_3$,a polymorph of $CaCO_3$ that is metastable with respect to calcite
atm	atmosphere(s)
B, B(II)	boron, boron in the +II oxidation state
Br, Br(−I)	bromine, bromine in the −I oxidation state
brucite	$Mg(OH)_2$
C	carbon
Ca, Ca(II), Ca^{2+}	calcium, calcium in the +II oxidation state, calcium ion
calcite	$CaCO_3$, the thermodynamically stable polymorph of $CaCO_3$
citrate	$(CH_2COO)_2C(OH)(COO)^{3-}$ or $(CH_2CO_2)_2C(OH)(CO_2)^{3-}$
Cl, Cl(−I), Cl^-	chlorine, chlorine in the −I oxidation state, chloride ion
CMS	(Sandia/WIPP software) Configuration Management System
CO_2	carbon dioxide
CO_3^{2-}	carbonate
CRA-2009	the second WIPP Compliance Recertification Application, submitted to the EPA in March 2009
DB	(thermodynamic) database
DOE	(U.S.) Department of Energy
dolomite	$CaMg(CO_3)_2$, a carbonate mineral that is nucleates and grows slowly under low-temperature conditions and is often suppressed (prevented from forming) in geochemical modeling calculations
DRZ	disturbed rock zone
EDTA	ethylenediaminetetraacetate, $(CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)^{4-}$ or $(CH_2CO_2)_2N(CH_2)_2N(CH_2CO_2)^{4-}$
EPA	(U.S.) Environmental Protection Agency
EQ3/6	a geochemical software package for speciation and solubility calculations (EQ3NR) and reaction-path calculations (EQ6)

Table 1 continued on next page

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

Abbreviation, Acronym, or Initialism	Definition
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
GWB	Generic Weep Brine, a synthetic brine representative of intergranular Salado brines at or near the stratigraphic horizon of the repository
gypsum	$CaSO_4 \cdot 2H_2O$
H or H_2, H^+	hydrogen or hydrogen ion
halite	NaCl
H_2O	water (aq, g, or contained in solid phases)
hydromagnesite	$Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$
I	ionic strength
K, K(I)	potassium, potassium in the +I oxidation state
kg	kilogram(s)
M	molar
m	meter(s) or molal
magnesite	$MgCO_3$
Mg, Mg(II)	magnesium, magnesium in the +II oxidation state
MgO	magnesium oxide, used to refer to the WIPP engineered barrier, which includes periclase as the primary constituent and various impurities
mM	millimolar
Na, Na(I), Na^+	sodium, sodium in the +I oxidation state, sodium ion
nesquehonite	$MgCO_3 \cdot 3H_2O$
Np, Np(V)	neptunium, neptunium in the +V oxidation state
O or O_2	oxygen
OH, OH^-	hydroxide or hydroxide ion
oxalate	$(COO)^{2-}$ or $C_2O_4^{2-}$
PA	performance assessment
PABC	Performance Assessment Baseline Calculations

Table 1 continued on next page

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

Abbreviation, Acronym, or Initialism	Definition
periclase	pure, crystalline MgO, the primary constituent of the WIPP engineered barrier
pH	the negative, common logarithm of the activity of H^+
pcH	the negative, common logarithm of the molar concentration of H^+
phase 3	$Mg_2Cl(OH)_3 \cdot 4H_2O$
phase 5	$Mg_3(OH)_5Cl \cdot 4H_2O$
polyhalite	$K_2MgCa_2(SO_4)_4 \cdot 2H_2O$
QA	quality assurance
Rev.	revision
RH	relative humidity
S, S(VI), SO_4^{2-}	sulfur, sulfur in the +VI oxidation state, sulfate ion
s	solid
SCA	S. Cohen and Associates
SNL	Sandia National Laboratories
Th, Th(IV)	thorium, thorium in the +IV oxidation state
TIC	total inorganic C
WIPP	(U.S. DOE) Waste Isolation Pilot Plant
wt %	weight percent
μ^0/RT	dimensionless standard chemical potential

2 METHODS

The objective of this analysis was to predict the solubilities of the actinide elements Th(IV), Np(V), and Am(III) in the standard WIPP brines GWB and ERDA-6 as a function of the volume of these brines in WIPP disposal rooms. This report also provides the predicted compositions of GWB and ERDA-6 after equilibration with the important solids in the repository. SNL's WIPP PA personnel will use these actinide solubilities in an upcoming PA calculation. Brush and Xiong (2011) provided a detailed description of the methods used to calculate the solubilities of Th(IV), Np(V), and Am(III) for use in WIPP PA, why the brines GWB and ERDA-6 are used, how these solubilities are applied to other actinides included in WIPP PA, etc.; and the task-specific methods used for this analysis (see their description of Task 2 in Subsection 4.2).

We calculated the dissolved concentrations of the organic ligands acetate (CH_3COO^-), citrate ($(CH_2COO)_2C(OH)(COO)^{3-}$), EDTA (ethylenediaminetetraacetate, $(CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)^{4-}$), and oxalate ($(COO)^{2-}$) in volumes of GWB and ERDA-6 that are 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3, the minimum volume of brine in the repository required for a direct brine release (DBR) from the repository (Clayton, 2008), by dividing the concentrations of these ligands calculated by Brush and Xiong (2009) by 2, 3, 4, and 5, respectively. We used factors of 2, 3, 4, or 5 at the request of WIPP PA personnel, who had determined that all of the DBRs in the Performance Assessment Baseline Calculation (PABC) for the second WIPP Compliance Recertification Application (CRA-2009) had volumes that varied between the minimum brine volume and 5 × the minimum volume. A DBR is defined as a release of brine that occurs directly from the repository to the surface above the repository (i.e., without lateral transport through an offsite transport pathway such as the Culebra Member of the Rustler Fm.). Brush and Xiong (2009) calculated the concentrations of acetate, citrate, EDTA, and oxalate in 17,400 m^3 of brine for the CRA-2009 PABC by assuming that the total masses of these organic ligands in the waste would dissolve completely in this volume of brine.

We used EQ3/6, Version 8.0a (Wolery and Jarek, 2003; Wolery, 2008; Wolery et al., 2010) to simulate the reaction of GWB and ERDA-6 with the important solids in the WIPP. In the first step of this reaction (referred to herein as "step 1"), we added the following compounds to GWB and ERDA-6: (1) acetate, citrate, EDTA, and oxalate; and (2) ThO_2(am), $KNpO_2CO_3$, and $Am(OH)_3$(s)), the solids most likely to control the solubilities of Th(IV), Am(III), and Np(V) in the repository (Brush and Xiong, 2011). In step 2, we reacted these brines with the important solids in the repository (see below) in a manner consistent with the conceptual models for WIPP near-field chemistry (SCA, 2008; Brush and Xiong, 2011) and predicted the solubilities of Th(IV), Np(V), and Am(III) and the compositions of GWB and ERDA-6 after equilibration with the important solids. After equilibration, the compositions of these brines define so-called invariant points (one each for GWB and ERDA-6), because the solids specified in the conceptual models — especially brucite ($Mg(OH)_2$) and hydromagnesite ($Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$) — control the new compositions of the brines and

parameters such as the fugacity (similar to the partial pressure) of CO_2 (f_{CO_2}), pH, and total inorganic carbon (TIC).

For step 1, we used the speciation and solubility code EQ3NR to add the organic ligands and the actinide-bearing solids to GWB and ERDA-6. We set the initial concentrations of acetate, citrate, EDTA, and oxalate equal to those in volumes of GWB and ERDA-6 that are 1 ×, 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3. We set the initial value of the total inorganic carbon (TIC) concentrations of both brines at 16 mM for this step because: (1) Popielak et al, (1983) reported that the average TIC content of ERDA-6 was 16 mM, (2) the initial TIC of GWB was not determined, so (3) we assumed that the initial TIC content of GWB was equal to that of ERDA-6. (The initial value of the TIC did not affect the values of the TIC predicted during the rest of the calculations). The code charge balanced on H^+; speciated all of the dissolved elements; and calculated the values of parameters such as f_{CO_2}, pH, and TIC. The code also wrote a "pickup" file (*.3p file) for step 2. The *.3p file is called a pickup file because it is copied and pasted into an EQ6 input file, and provides all of the information on the solution and solids required for the next EQ6 run (see below).

For step 2, we used the reaction-path code EQ6 to titrate the solids halite (NaCl), anhydrite ($CaSO_4$), brucite, and hydromagnesite into GWB. (We used a slightly different procedure, described below, for ERDA-6.) We used halite and anhydrite to simulate the most important minerals in the Salado Fm. at or near the stratigraphic horizon of the repository; and brucite and hydromagnesite to simulate the expected hydration and carbonation products, respectively, of MgO (the WIPP engineered barrier). As EQ6 titrated in these solids, halite and anhydrite dissolved until the brine became saturated with these solids (i.e., until the concentrations of Na^+, Cl^-, Ca^{2+}, and SO_4^{2-} reached their solubility limits). The reaction then continued until brucite and hydromagnesite equilibrated with GWB (i.e., until GWB reached its invariant point). EQ6 then calculated the moles of solids that dissolved and/or precipitated, speciated all of the dissolved elements; and recalculated the values of parameters such as f_{CO_2}, pcH, TIC, etc.

For ERDA-6, we first used EQ6 to titrate just halite and anhydrite into the brine (step 2a). During this step, EQ6 titrated halite and anhydrite into ERDA-6 until this brine became saturated with these solids (i.e., until Na^+, Cl^-, Ca^{2+}, and SO_4^{2-} reached their solubility limits). At the end of step 2a, EQ6 wrote a pickup file (*.6p file), which provided all of the information on the solution and solids required for step 2b. During step 2b, EQ6 titrated in brucite and hydromagnesite as ERDA-6 remained saturated with halite and anhydrite. The reaction continued until brucite and hydromagnesite equilibrated with ERDA-6 (i.e., until this brine reached its invariant point). EQ6 then calculated the moles of solids that dissolved and/or precipitated, speciated all of the dissolved elements; and recalculated the values of parameters such as f_{CO_2}, pcH, TIC, etc.

Table 2 (see next page) summarizes these EQ3/6 calculations for GWB and ERDA-6.

Table 2. Summary of EQ3/6 Calculations Carried Out with GWB and ERDA-6 for This Analysis

Description of Step	GWB	ERDA-6
1	Used EQ3NR to add organic ligands and actinide-bearing solids	Used EQ3NR to add organic ligands and actinide-bearing solids
2	Used EQ6 to titrate in halite, anhydrite, brucite, and hydromagnesite	-
2a	-	Used EQ6 to titrate in halite and anhydrite
2b	-	Used EQ6 to titrate in brucite and hydromagnesite

For step 2 (GWB) or steps 2a and 2b (ERDA-6), we used quantities of brine, halite, anhydrite, brucite, and hydromagnesite similar to those that will be present in the repository after it is filled and sealed, but scaled down by the same factor used to scale down the quantity of water contained in 17,400 m^3 of brine to 1 kg of water. EQ3/6 allows the user to specify the composition and specific gravity of the aqueous phase present at the start of a run. However, the code assumes that exactly 1 kg of H_2O is present in the solution and uses the specific gravity entered by the user to calculate the volume of solution. We used spreadsheet calculations to scale down (1) the quantities of halite and anhydrite present in the disturbed rock zone (DRZ) surrounding the repository, and (2) the quantity of MgO that will be emplaced in the repository. The spreadsheet, entitled "AP-153_Tasks 1 and 2, Scaling of Solids.xls" is in the zip file AP153Task1Data.zip in library LIBEQ36, class AP153, in the Sandia/WIPP software Configuration Management System (CMS.) Table 3 (see next page) provides the locations of this and the other files used for our EQ3/6 calculations. To calculate the quantities of halite and anhydrite, we used the conservatively large DRZ currently implemented in WIPP PA and the assumption that the DRZ comprises 90 wt % halite and 10 wt % anhydrite. This mineralogical composition is similar to Brush's (1990) interpretation of the results of Stein's (1985) mineralogical analysis of the Salado Fm. at or near the stratigraphic horizon of the repository: Brush (1990) concluded that, for use in geochemical modeling, the Salado consists of 93.2 wt % halite and 1.7 wt % each of anhydrite, gypsum ($CaSO_4 \cdot 2H_2O$), magnesite ($MgCO_3$), and polyhalite ($K_2MgCa_2(SO_4)_4 \cdot 2H_2O$). However, we assumed for this analysis that the Salado contains 90 wt % halite and 10 wt % anhydrite because the conceptual models for WIPP near-field chemistry include only halite and anhydrite (SCA, 2008). For run 1, we also assumed that the MgO that will be emplaced in the repository will be present half as brucite and

half as hydromagnesite; this assumption ensured that ample CO_2 was present without having to use a microbial reaction to titrate in CO_2.

Table 3. Locations of the Excel Spreadsheets, I/O Files, etc., Used in the EQ3/6 Calculations for This Analysis. Table 4 provides comparable information for FMT.

Description or Title of File(s)	Location of File(s)
Spreadsheet AP-153_Tasks 1 and 2, Scaling of Solids.xls	In zip file AP153Task1Data.zip, library LIBEQ36, class AP153
EQ3/6 DB DATA0.FMT.R0.16	In zip file DATA0_FMT.ZIP, library LIBEQ36, class DATABASES
Excel macro GetEQData.xls	In zip file AP153Task1Data.zip, library LIBEQ36, class AP153
EQ3/6 I/O files and Excel spreadsheets with extracted data	In zip file AP153Task2Data.zip, library LIBEQ36, class AP153

We used EQ6 in closed-system mode (model variable IOPT1 = 0) for step 2 (GWB) or steps 2a and 2b ERDA-6. Closed-system mode consists of the simulated titration (addition) of the reactants described above to GWB or ERDA-6. "Closed-system" means that no reactants or products can leave the system after the reactants are titrated in, which simulates the WIPP under undisturbed conditions. We suppressed (prevented from precipitating) the solids aragonite ($CaCO_3$), calcite ($CaCO_3$), dolomite ($CaMg(CO_3)_2$), hydromagnesite with the composition $Mg_4(CO_3)_3(OH)_2 \cdot 3H_2O$, and nesquehonite ($MgCO_3 \cdot 3H_2O$) throughout step 2 or steps 2 and 2b. We suppressed these phases to ensure that this analysis was consistent with the near-field chemical conceptual models (SCA, 2008; Brush and Xiong, 2011).

We used the EQ3/6 thermodynamic database (DB) DATA0.FMT.R0.16 (Wolery et al., 2010) for this analysis. (DATA0.FMT.R0.16 is in DATA0_FMT.ZIP, LIBEQ36, class DATABASES, in the CMS.) This DB is identical to FMT_050405.CHEMDAT (Nowak, 2005; Xiong, 2005; LIBFMT, class CHEMDAT_2005), the DB used by Brush et al. (2009) to predict the solubilities of Th(IV), Np(V), and Am(III) for the CRA-2009 PABC. Both of these DBs contain values of the dimensionless standard chemical potential (μ^0/RT) for phase 3 ($Mg_2Cl(OH)_3 \cdot 4H_2O$), but not phase 5 ($Mg_3Cl(OH)_5 \cdot 4H_2O$). Brush and Xiong (2011) provided a detailed history of the DBs used for WIPP compliance-related actinide-solubility calculations.

We extracted the output from these EQ6 *.6o files by running the Excel macro "GetEQData.xls." This macro extracts all of the EQ6 output into an Excel spreadsheet.

All of our EQ3/6 input and output (I/O) files, the Excel macro GetEQData.xls, and the Excel spreadsheets that contain the output extracted with GetEQData.xls are in zip file AP153Task2Data.zip in library LIBEQ36, class AP153, in the CMS.

We also used FMT, Version 2.4 (Babb and Nowak, 1997 and addenda; Wang, 1998), supported by the thermodynamic DB FMT_050405.CHEMDAT (Xiong, 2005), to predict the solubilities of Th(IV), Np(V), and Am(III) and the compositions of GWB and ERDA-6 in volumes of GWB and ERDA-6 that are 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3. We used procedures identical to those used by Brush et al. (2009) for the CRA-2009 PABC. For the minimum brine volume of 17,400 m^3, we used the results of Brush et al. (2009, runs 5 and 13).

Table 4 provides run-control information for the FMT calculations carried out for this analysis.

Table 4. Run-Control Information for the FMT Calculations Carried Out for This Analysis.

	File Names[A, B]	CMS Library	CMS Class
Script:			
Script	EVAL_FMT.COM	LIBAP153_EVAL	AP153-0
Script Input	EVAL_FMT_AP153_w.INP	LIBAP153_EVAL	AP153-0
Script Log	EVAL_FMT_AP153_w.LOG	LIBAP153_FMT	AP153-0
FMT:			
Input	FMT_050405.CHEMDAT	LIBAP153_FMT	AP153-0
Input	FMT_GENERIC.RHOMIN	LIBAP153_FMT	AP153-0
Input	FMT_AP153_w_n.IN	LIBAP153_FMT	AP153-0
Input	FMT_AP153_w_n.INGUESS	LIBAP153_FMT	AP153-0
Output	FMT_AP153_w_n.OUT	LIBAP153_FMT	AP153-0
Output	FMT_ AP153_w_n.FOR088	Not kept	Not kept

[A] $w \in \{\text{GWB, ERDA6}\}$.

[B] $n \in \{002 \text{ through } 005\}$

The methods described above did not deviate from those described in AP-153, Task 2 Brush and Xiong (2011, Subsection 4.2).

3 RESULTS

Table 5 provides the dissolved concentrations of the organic ligands acetate, citrate EDTA, and oxalate in volumes of GWB and ERDA-6 that are 1 ×, 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3, the minimum volume required for a DBR from the repository (Clayton, 2008). The concentrations of these ligands for a volume of 17,400 m^3 are from Brush and Xiong (2009); the concentrations for brine volumes greater than 17,400 m^3 are from this analysis.

Table 6 provides the solubilities of Th(IV), Np(V), and Am(III); the compositions; and parameters such as f_{CO_2}, pcH, and TIC for volumes of GWB that are 1 ×, 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3. Table 7 provides comparable information for ERDA-6. All of the results predicted with EQ3/6 are from this analysis. The results predicted with FMT for 17,400 m^3 of GWB or ERDA-6 are from Brush and Xiong (2009, runs 5 and 13, respectively); the results from FMT for brine volumes greater than 17,400 m^3 are from this analysis.

It is worth noting that the actinide solubilities, brine compositions, and other parameters predicted with EQ3/6 and FMT are essentially identical, given the uncertainties inherent in geochemical process modeling and total system PA.

We recommend that PA personnel use the Th(IV), Np(V), and Am(III) solubilities calculated using EQ3/6 in their upcoming PA calculations. However, use of the results obtained with FMT would make essentially no difference, because the results from these codes are essentially identical.

Table 5. Concentrations of Organic Ligands (M) in Brine Volumes That Are 1 ×, 2 ×, 3 ×, 4 ×, and 5 × the Minimum Volume Required for a Release from the Repository.

Organic Ligand	1 × Minimum[A, B]	2 × Minimum	3 × Minimum	4 × Minimum	5 × Minimum
Acetate	1.94×10^{-2}	9.70×10^{-3}	6.47×10^{-3}	4.85×10^{-3}	3.88×10^{-3}
Citrate	2.38×10^{-3}	1.19×10^{-3}	7.93×10^{-4}	5.95×10^{-4}	4.76×10^{-4}
EDTA	6.47×10^{-5}	3.24×10^{-5}	2.16×10^{-5}	1.62×10^{-5}	1.29×10^{-5}
Oxalate	1.73×10^{-2}	8.65×10^{-3}	5.77×10^{-3}	4.32×10^{-3}	3.46×10^{-3}

A. Based on a minimum brine volume of 17,400 m^3 (Clayton, 2008)
B. Organic ligand concentrations calculated by Brush and Xiong (2009) for the WIPP CRA-2009 PABC.

Table 6. Comparisons of the Compositions of Five Different Volumes of GWB (M Unless Otherwise Noted) Predicted by EQ3/6 and FMT.

Element or Property	1 × Minimum,[A] EQ3/6[B]	1 × Minimum,[A] FMT[C]	2 × Minimum,[A] EQ3/6[B]	2 × Minimum,[A] FMT[B]
B(III)(aq)	0.178	0.176	0.178	0.176
Na(I)(aq)	4.30	4.31	4.31	4.32
Mg(II)(aq)	0.587	0.584	0.575	0.572
K(I)(aq)	0.528	0.521	0.527	0.521
Ca(II)(aq)	0.00979	0.00980	0.0101	0.0101
S(VI)(aq)	0.212	0.210	0.203	0.200
Cl(-I)(aq)	5.40	5.40	5.42	5.42
Br(-I)(aq)	0.0300	0.0297	0.0300	0.0297
Th(IV)(aq)	5.62×10^{-8}	5.63×10^{-8}	5.63×10^{-8}	5.63×10^{-8}
Np(V)(aq)	3.88×10^{-7}	3.90×10^{-7}	3.21×10^{-7}	3.23×10^{-7}
Am(III)(aq)	1.67×10^{-6}	1.66×10^{-6}	9.54×10^{-7}	9.49×10^{-7}
f_{CO_2} (atm)	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
I (m)	7.66	7.64	7.63	7.62
pH[D]	8.69	8.69	8.69	8.69
pcH	9.40	9.40	9.40	9.40
RH (%)[E]	73.1	73.2	73.2	73.2
TIC (mM)	0.350	0.350	0.350	0.350

Table 6 continued on next page. Footnotes provided on last page of Table 6.

Table 6. Comparisons of Compositions of Different Volumes of GWB (M Unless Otherwise Noted) Predicted by EQ3/6 and FMT (continued).

Element or Property	3 × Minimum.[A] EQ3/6[B]	3 × Minimum,[A] FMT[B]	4 × Minimum.[A] EQ3/6[B]	4 × Minimum,[A] FMT[B]
B(III)(aq)	0.178	0.176	0.178	0.176
Na(I)(aq)	4.32	4.32	4.32	4.32
Mg(II)(aq)	0.571	0.568	0.568	0.566
K(I)(aq)	0.527	0.521	0.526	0.521
Ca(II)(aq)	0.0102	0.0102	0.0103	0.0103
S(VI)(aq)	0.199	0.197	0.198	0.196
Cl(-I)(aq)	5.43	5.43	5.43	5.43
Br(-I)(aq)	0.0300	0.0297	0.0300	0.0297
Th(IV)(aq)	5.63×10^{-8}	5.64×10^{-8}	5.64×10^{-8}	5.64×10^{-8}
Np(V)(aq)	2.98×10^{-7}	3.01×10^{-7}	2.87×10^{-7}	2.89×10^{-7}
Am(III)(aq)	7.12×10^{-7}	7.09×10^{-7}	5.91×10^{-7}	5.89×10^{-7}
f_{CO_2} (atm)	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
I (m)	7.62	7.61	7.62	7.60
pH[D]	8.69	8.69	8.69	8.69
pcH	9.40	9.40	9.40	9.40
RH (%)[E]	73.2	73.2	73.2	73.2
TIC (mM)	0.350	0.350	0.350	0.350

Table 6 continued on next page. Footnotes provided on last page of Table 6.

Table 6. Comparisons of Compositions of Different Volumes of GWB (M Unless Otherwise Noted) Predicted by EQ3/6 and FMT (continued).

Element or Property	5 × Minimum,[A] EQ3/6[B]	5 × Minimum,[A] FMT[B]
B(III)(aq)	0.178	0.176
Na(I)(aq)	4.32	4.33
Mg(II)(aq)	0.567	0.565
K(I)(aq)	0.526	0.521
Ca(II)(aq)	0.0103	0.0103
S(VI)(aq)	0.197	0.195
Cl(-I)(aq)	5.43	5.43
Br(-I)(aq)	0.0300	0.0297
Th(IV)(aq)	5.64×10^{-8}	5.64×10^{-8}
Np(V)(aq)	2.80×10^{-7}	2.83×10^{-7}
Am(III)(aq)	5.17×10^{-7}	5.15×10^{-7}
f_{CO_2} (atm)	3.14×10^{-6}	3.14×10^{-6}
I (m)	7.61	7.60
pH[D]	8.69	8.69
pcH	9.40	9.40
RH (%)[E]	73.2	73.2
TIC (mM)	0.350	0.350

Footnotes for Table 6 provided on next page

Footnotes for Table 6:

A. Based on a minimum brine volume of 17,400 m^3 (Clayton, 2008)
B. Calculated for this analysis (AP-153, Task 2)
C. FMT run 5 for the CRA-2009 PABC (Brush et al., 2009)
D. 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.
E. The value of the RH predicted by EQ3/6 divided by 100 yields the value of the activity of H_2O in GWB.

Table 7. Comparisons of the Compositions of Five Different Volumes of ERDA-6 (M Unless Otherwise Noted) Predicted by EQ3/6 and FMT.

Element or Property	1 × Minimum,[A] EQ3/6[B]	1 × Minimum,[A] FMT[C]	2 × Minimum. EQ3/6[B]	2 × Minimum, FMT[B]
B(III)(aq)	0.0624	0.0624	0.0624	0.0624
Na(I)(aq)	5.29	5.28	5.31	5.31
Mg(II)(aq)	0.136	0.136	0.121	0.121
K(I)(aq)	0.0960	0.0961	0.0960	0.0961
Ca(II)(aq)	0.0113	0.0112	0.0112	0.0111
S(VI)(aq)	0.177	0.176	0.176	0.175
Cl(-I)(aq)	5.23	5.23	5.24	5.24
Br(-I)(aq)	0.0109	0.0109	0.0109	0.0109
Th(IV)(aq)	6.99×10^{-8}	6.98×10^{-8}	7.09×10^{-8}	7.08×10^{-8}
Np(V)(aq)	8.72×10^{-7}	8.75×10^{-7}	7.19×10^{-7}	7.21×10^{-7}
Am(III)(aq)	1.50×10^{-6}	1.51×10^{-6}	7.86×10^{-7}	7.92×10^{-7}
f_{CO_2} (atm)	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
I (m)	6.77	6.77	6.77	6.75
pH[D]	8.99	8.98	9.00	9.00
pcH	9.69	9.68	9.71	9.70
RH (%)[E]	74.8	74.8	74.8	74.8
TIC (mM)	0.449	0.448	0.460	0.458

Table 7 continued on next page. Footnotes provided on last page of Table 7.

Table 7. Comparisons of Compositions of Five Different Volumes of ERDA-6 (M Unless Otherwise Noted) Predicted by EQ3/6 and FMT (continued).

Element or Property	3 × Minimum. EQ3/6[B]	3 × Minimum, FMT[B]	4 × Minimum. EQ3/6[B]	4 × Minimum, FMT[B]
B(III)(aq)	0.0624	0.0624	0.0624	0.0624
Na(I)(aq)	5.32	5.31	5.32	5.32
Mg(II)(aq)	0.115	0.116	0.113	0.114
K(I)(aq)	0.0960	0.0961	0.0961	0.0961
Ca(II)(aq)	0.0112	0.0111	0.0113	0.0112
S(VI)(aq)	0.173	0.173	0.172	0.172
Cl(-I)(aq)	5.24	5.24	5.25	5.25
Br(-I)(aq)	0.0109	0.0109	0.0109	0.0109
Th(IV)(aq)	7.13×10^{-8}	7.12×10^{-8}	7.15×10^{-8}	7.14×10^{-8}
Np(V)(aq)	6.70×10^{-7}	6.72×10^{-7}	6.47×10^{-7}	6.48×10^{-7}
Am(III)(aq)	5.48×10^{-7}	5.53×10^{-7}	4.31×10^{-7}	4.35×10^{-7}
f_{CO_2} (atm)	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}	3.14×10^{-6}
I (m)	6.75	6.75	6.74	6.74
pH[D]	9.01	9.01	9.01	9.01
pcH	9.71	9.71	9.72	9.71
RH (%)[E]	74.8	74.8	74.8	74.8
TIC (mM)	0.464	0.462	0.466	0.464

Table 7 continued on next page. Footnotes provided on last page of Table 7.

Table 7. Comparisons of Compositions of Five Different Volumes of ERDA-6 (M Unless Otherwise Noted) Predicted by EQ3/6 and FMT (continued).

Element or Property	5 × Minimum. EQ3/6[B]	5 × Minimum, FMT[B]
B(III)(aq)	0.0624	0.0624
Na(I)(aq)	5.32	5.32
Mg(II)(aq)	0.111	0.112
K(I)(aq)	0.0961	0.0961
Ca(II)(aq)	0.0113	0.0112
S(VI)(aq)	0.171	0.171
Cl(-I)(aq)	5.25	5.25
Br(-I)(aq)	0.0109	0.0109
Th(IV)(aq)	7.16×10^{-8}	7.15×10^{-8}
Np(V)(aq)	6.32×10^{-7}	6.34×10^{-7}
Am(III)(aq)	3.60×10^{-7}	3.63×10^{-7}
f_{CO_2} (atm)	3.14×10^{-6}	3.14×10^{-6}
I (m)	6.74	6.74
pH[D]	9.02	9.01
pcH	9.72	9.72
RH (%)[E]	74.8	74.8
TIC (mM)	0.467	0.465

Footnotes for Table 7 provided on next page

Footnotes for Table 7

A. Based on a minimum brine volume of 17,400 m^3 (Clayton, 2008)
B. Calculated for this analysis (AP-153, Task 2)
C. FMT run 13 for the CRA-2009 PABC (Brush et al., 2009)
D. 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.
E. The value of the RH predicted by EQ3/6 divided by 100 yields the value of the activity of H_2O in ERDA-6GWB.

4 CONCLUSIONS

Table 6 provides the solubilities of Th(IV), Np(V), and Am(III); the compositions; and parameters, such as f_{CO_2}, pcH, and TIC for GWB and ERDA-6 as a function of brine volume. We will provide these actinide solubilities to SNL's WIPP PA personnel for use in an upcoming PA calculation. We used EQ3/6, Version 8.0a (Wolery and Jarek, 2003; Wolery, 2008; Wolery et al., 2010); and FMT, Version 2.4 (Babb and Nowak, 1997 and addenda; Wang, 1998), for this analysis. We carried out this analysis under Task 2 of AP-153 (Brush and Xiong, 2011).

Table 5 (see Section 3 above) provides the dissolved concentrations of the organic ligands acetate, citrate EDTA, and oxalate in volumes of GWB and ERDA-6 that are 1 ×, 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3, the minimum volume required for a DBR from the repository.

Tables 6 and 7 provide the solubilities of Th(IV), Np(V), and Am(III); the compositions; and parameters, such as f_{CO_2}, pcH, and TIC for volumes of GWB and ERDA-6, respectively, that are 1 ×, 2 ×, 3 ×, 4 ×, and 5 × 17,400 m^3. These tables show that the results obtained with EQ3/6 and FMT are essentially identical.

We recommend that PA personnel use the Th(IV), Np(V), and Am(III) solubilities calculated using EQ3/6 in their upcoming PA calculations. However, use of the results obtained with FMT would make no difference, because the results from these codes are nearly identical.

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