522982

Sandia National Laboratories
Operated for the U.S. Department of Energy by
Sandia Corporation

Carlsbad, New Mexico 88220

date: July 25, 2002

to: Laurence H. (Larry) Brush
Geochemistry Team Lead
Org. 6822, MS-1395
Carlsbad Programs Group

from: Emily R. Giambalvo
Performance Assessment
Org. 6821, MS-1395
Carlsbad Programs Group

Emily Giambalvo

subject: Recommended Parameter Values for Modeling An(III) Solubility in WIPP Brines

1. Introduction

Parameter values for modeling speciation and solubility of actinides in the +III oxidation state (An(III)) from the recent literature (Morss and Williams, 1994; Könnecke et al., 1997; Fanghänel and Kim, 1998; Fanghänel et al., 1999) offer the opportunity to improve the An(III) speciation and solubility model implemented in the Fracture Matrix Transport (FMT) geochemical database. Newly available parameters include hydrolysis constants, constants for An(III) complexation with carbonate, chloride, and sulfate, Pitzer parameters describing the interaction of inorganic An(III) species with Na^+, Cl^-, and Ca^{2+}, and solubility product constants (K_{sp}s) for $An(OH)_{3(s)}$ and $AnOHCO_{3(c)}$.

This memo describes the implementation of the An(III) speciation and solubility model in the version of the FMT database used for the Performance Assessment Verification Test (PAVT)*, and recommends a new implementation based on the recent literature. Recommended new values for complexation constants and Pitzer parameters were obtained from spectroscopic analysis of inorganic Cm(III) species in NaCl and $CaCl_2$ brines over a range of ionic strengths appropriate for determining the Pitzer ion interaction parameters necessary to simulate An(III) speciation in Waste Isolation Pilot Plant (WIPP) brines. Recommended new values for solubility product constants were obtained or verified by application of the recommended Pitzer models to independently collected Am solubility data.

2. An(III) Model in the PAVT Database

The An(III) speciation and solubility model in the PAVT version of the FMT geochemical database is that described by Novak (1995; 1996) with some modifications. Parameters are derived from Nd, Pu, Am, and Cm speciation and solubility data and applied to Am, the element that represents the +III oxidation state in the WIPP actinide solubility model (DOE, 1996). Because these four elements behave similarly when in the +III oxidation state (e.g. Rai et al., 1995; Felmy and Rai, 1999), complexation constants, solubility product constants, and Pitzer parameters for analogous species are assumed to be equal. The parameters used in the PAVT An(III) model are derived as follows:

---

* The PAVT database is the most recent documented version of the database (Novak, 1997). It is stored in the CHEMDAT file FMT_970407.CHEMDAT in Sandia National Laboratories' Configuration Management System on the WIPP Alpha Cluster in the library WP$NONPA_CMSROOT:[FMT].

WIPP: 1.3.5.1, 2.1: PA: QA: FMT Database for the CRA

1. Am-chloride complexes are neglected. Am^{3+}-Cl^- Pitzer ion interaction parameters are set equal to Nd^{3+}-Cl^- interaction parameters calculated from the osmotic coefficient for $NdCl_3$ (Felmy et al., 1989; Pitzer and Mayorga, 1973).

2. First and second hydrolysis constants and related Pitzer parameters are derived from Cm speciation as a function of NaCl concentration (0.01-6.15 mol/kg) (Fanghänel et al., 1994). Fanghänel et al. (1994) successfully simulated $Am(OH)_{3(s)}$ solubility in 5 mol/L NaCl (Runde and Kim, 1995) using log $K_{sp} = -28.2$. In the PAVT database log $K_{sp} = -26.2$. Novak (1996) therefore adjusted the hydrolysis constants downward two orders of magnitude before entering them in the database.

3. Novak (1996) estimated the third hydrolysis constant from the K_{sp} for $Am(OH)_{3(s)}$ and experimentally determined $Am(OH)_{3(s)}$ solubility in 5 mol/L NaCl at high pH (>11) (Runde and Kim, 1995). Novak (1996) assumed that the Pitzer parameters describing interaction of $Am(OH)_{3(aq)}$ with Na^+ and Cl^- are zero (i.e., the activity of $Am(OH)_{3(aq)}$ is independent of ionic strength).

4. Carbonate complexation constants are derived from $AmOHCO_{3(c)}$ solubility data in dilute (<0.1 mol/L) Na_2CO_3 solution (Felmy et al., 1990). Related Pitzer parameters used in the database were calculated by Rao et al. (1996), who used the Felmy et al. (1990) Am-carbonate complexation constants to simulate $NaNd(CO_3)_2{\cdot}6H_2O_{(c)}$ solubility in concentrated (0.1-2 mol/L) Na_2CO_3 solutions. Rao et al. (1996) verified the Pitzer parameters by successfully simulating the Felmy et al. (1990) $AmOHCO_{3(c)}$ solubility data.

5. Am-sulfate complexes are neglected. Am^{3+}-SO_4^{2-} Pitzer ion interaction parameters were calculated from Am solvent-extraction data in Na_2SO_4-H_2SO_4 solutions up to 0.5 mol/kg (Rai et al., 1995). The Pitzer parameters were verified by successfully simulating two independent data sets for Am-sulfate complexation in 1 mol/kg $NaClO_4$ and the solubility of $NdPO_{4(s)}$ in the presence of SO_4^{2-} in dilute solution (Rai et al., 1995).

6. Log K_{sp} for $Am(OH)_{3(s)}$ is set equal to that for $Pu(OH)_{3(s)}$ as determined from supersaturation and undersaturation experiments (-26.2, Felmy et al., 1989[*]). Because these experiments were conducted at room temperature, the resulting precipitate can be assumed to be amorphous $Pu(OH)_3$ (e.g., Morss and Williams, 1994).

7. Log K_{sp} for $AmOHCO_{3(c)}$ (-22.5) was determined by monitoring the transformation of an amorphous Am-carbonate phase to a crystalline phase (essentially a supersaturation experiment) and by resuspending the crystalline phase in various solutions (undersaturation experiments) (Felmy et al., 1990).

The An(III) model also includes Pitzer ion interaction parameters describing the interaction of Am^{3+} with $H_2PO_4^-$ (Rai et al., 1995) and Am^{3+} with ClO_4^- (Felmy et al., 1990), and solubility product constants for the solid phases $NaAm(CO_3)_2{\cdot}6H_2O$ (Rao et al., 1996) and $AmPO_{4(s)}$ (Rai et al., 1992). In addition, the Pitzer parameters describing interaction of SO_4^{2-} with ClO_4^- and Na^+ were derived from Rai et al.'s (1995) study of Am-sulfate interaction. At this time I do not recommend any changes to these parameters except for setting the SO_4^{2-} - ClO_4^- - Na^+ Pitzer parameters to zero for consistency with the recommended changes to the Am-sulfate model (see Section 3). Parameters describing Am interactions

---

[*] Novak (1996) incorrectly references this value to Rai et al. (1983).

with perchlorate and phosphate should possibly be re-evaluated if the FMT database is to be used to simulate solutions in which either perchlorate or phosphate is a dominant component. Neither is a component in WIPP geochemical models.

Tables 1-4 list the parameter values used in the PAVT An(III) model.

3. Shortcomings of the PAVT An(III) Model

The primary shortcomings of the PAVT An(III) model are related to (1) the way in which the Fanghänel et al. (1994) hydrolysis model was incorporated into the database, and (2) the limited range of ionic strength over which certain data were collected.

3.1. Hydrolysis model

The Fanghänel et al. (1994) parameters are appropriate for use in simulating WIPP brines, but their inclusion in the PAVT database was awkwardly implemented. Fanghänel et al. (1994) did not use the PAVT Am^{3+}-Cl^- Pitzer parameters to develop the Pitzer parameters for Am-hydroxide species. Thus use of the Fanghänel et al. (1994) Pitzer parameters for Am-hydroxide species creates inconsistencies among Pitzer parameters in the database.

The combination of the Fanghänel et al. (1994) first and second hydrolysis constants with the Felmy et al. (1989) K_{sp} for $Am(OH)_{3(s)}$ and Novak's (1996) estimated third hydrolysis constant imposes inconsistent constraints on $Am(OH)_{3(s)}$ solubility. Novak (1996) adjusted the first and second hydrolysis constants for use in the PAVT database using the constraint that

$$\beta^0_{1n}K^0_{sp}(\text{Fanghänel}) = \beta^0_{1n}K^0_{sp}(\text{database}),$$

where β^0_{1n} is the appropriate hydrolysis constant at infinite dilution and the product $\beta^0_{1n}K^0_{sp}$ is the equilibrium constant at infinite dilution for the dissolution reaction

$$Am(OH)_{3(s)} = Am(OH)_n^{3-n} + (3-n)OH^-.$$

Use of the adjusted hydrolysis constants and the Felmy et al. (1989) log K_{sp} produces the same amount of $AmOH^{2+}$ and $Am(OH)_2^+$ in equilibrium with $Am(OH)_{3(s)}$ as would using the complete Fanghänel et al. (1994) model. In effect, if $AmOH^{2+}$ or $Am(OH)_2^+$ is the dominant Am species in solution (as predicted for WIPP brines; Novak et al., 1997), the log K_{sp} for $Am(OH)_{3(s)}$ is -28.2. If $Am(OH)_{3(aq)}$, Am^{3+}, or other species is the dominant Am species in solution, the log K_{sp} for $Am(OH)_{3(s)}$ is -26.2.

3.2. Ionic strength

Pitzer parameters for the PAVT Am-carbonate and Am-sulfate models were derived and/or verified using data collected in solutions containing ≤ 4 mol/L Na^+ and ≤1 mol/kg Na^+, respectively. Neither set of Pitzer parameters was tested on a solution containing Cl^-. The PAVT database includes parameters describing the ternary interaction of $Am(CO_3)_3^{3-}$, Cl^-, and Na^+, but the origin of these parameters is unknown. Models derived from data collected in solutions more similar to WIPP brines (i.e. solutions containing more Na^+ and Cl^-) would be preferable.

4. Recommended An(III) Model

The recommended parameter values discussed in this section are derived from some of the same data that resulted in the PAVT version of the An(III) model plus more recently collected experimental data. The recommended model includes revisions to the third hydrolysis constant and Pitzer parameters for hydrolysis species, revisions to carbonate complexation constants and corresponding Pitzer parameters, addition of chloride and sulfate complexes, and revisions to $Am(OH)_{3(s)}$ and $AmOHCO_{3(c)}$ solubility product constants. The recommended model closely follows that proposed by Fanghänel and Kim (1998). Specifically, parameters are derived as follows:

1. Am-chloride complexes ($AmCl_2^+$ and $AmCl^{2+}$) are included on the basis of spectroscopic evidence for the existence of $CmCl_2^+$ and $CmCl^{2+}$ in 2-6 mol/kg $CaCl_2$ (4-12 mol/kg Cl^-) solution (Fanghänel et al., 1995). Complexation constants for these chloride complexes, Pitzer parameters describing interaction of Am^{3+}, $AmCl_2^+$, and $AmCl^{2+}$ with Cl^- and Ca^{2+} are set equal to those calculated by Könnecke et al. (1997) from the Cm speciation data of Fanghänel et al. (1995).

2. First and second hydrolysis constants remain equal to those calculated by Fanghänel et al. (1994) from Cm speciation data. Related Pitzer parameters are set equal to those calculated by Könnecke et al. (1997) from the Fanghänel et al. (1994) data set. The Könnecke et al. (1997) recalculation of these Pitzer parameters accounts for the presence of Cm-chloride complexes and includes a term describing the interaction of Cm^{3+} with Na^+ (θ).Könnecke et al.'s (1997) successful use of their model to simulate Cm speciation in NaCl solution validates the absence of Pitzer parameters describing interaction between Am-chloride complexes and Na^+.

3. The third hydrolysis constant is set equal to that recommended by the Nuclear Energy Agency (Silva et al., 1995). Könnecke et al. (1997) used this value and the K_{sp} for $Am(OH)_{3(s)}$ recommended by Morss and Williams (1994) to successfully simulate the $Am(OH)_{3(s)}$ solubility data of Runde and Kim (1995). Könnecke et al.'s (1997) model included Pitzer parameters describing the interaction of $Am(OH)_{3(aq)}$ with Na^+ and Cl^-.

4. An additional Am-carbonate species ($Am(CO_3)_4^{5-}$) is included on the basis of spectroscopic evidence for the existence of $Cm(CO_3)_4^{5-}$ (Fanghänel et al., 1999). Carbonate complexation constants and related Pitzer parameters are derived from Cm-carbonate speciation data collected over a range of NaCl concentrations from 0-6 mol/kg (Fanghänel et al., 1999). Fanghänel et al. (1999) neglected the formation of Am-chloride complexes in their model, a valid decision at chloride concentrations <6 mol/kg (Fanghänel et al., 1995). They verified their model by successfully simulating the solubility of $AmOHCO_{3(c)}$ (including the Felmy et al. (1990) data set) and $NaAm(CO_3)_{2(s)}$.

5. Am-sulfate complexes are included on the basis of spectroscopic evidence for the existence of $CmSO_4^+$ and $Cm(SO_4)_2^-$ (Paviet et al., 1996). Related Pitzer parameters are derived from Cm-sulfate speciation data collected over a range of NaCl concentrations from 0-6 mol/kg (Fanghänel and Kim, 1998).

6. The log K_{sp} for $Am(OH)_{3(s)}$ (-27.5) is equal to that recommended by Morss and Williams (1994) on the basis of a compilation of values from the literature. This is the value Könnecke et al. (1997) successfully used to simulate Runde and Kim's (1995) $Am(OH)_{3(s)}$ solubility data.

7. The log K_{sp} for $AmOHCO_{3(c)}$ (-22.7) is equal to that calculated by Fanghänel et al. (1999) on the basis of Felmy et al.'s (1990) $AmOHCO_{3(s)}$ solubility data.

Tables 1-4 list the recommended parameter values and compare them to the values used in the PAVT version of the database.

5. Benefits of the Recommended Model

The recommended model is superior to the PAVT model in that:

1. The Am-hydrolysis model is self-consistent, consistent with the Am-chloride model, and consistent with the choice of $Am(OH)_{3(s)}$ K_{sp}. The recommended value for log K_{sp} of $Am(OH)_{3(s)}$ (-27.5) in combination with the recommended Pitzer model predicts essentially the same Am solubility (in 5 M NaCl) predicted by the PAVT model in the pcH (-log [H^+]) region relevant to the WIPP (pcH = 9-10). At higher and lower pH, the recommended model provides a better fit to the Am solubility data of Runde and Kim (1995) (Figure 1).

2. The Am-carbonate and Am-sulfate models were derived from data collected over a range of NaCl concentration from 0-6 mol/kg (Fanghänel and Kim, 1998; Fanghänel et al., 1999). Therefore the Pitzer ion interaction parameters associated with these models can be more confidently applied to WIPP brines than the parameters in the PAVT model, which were derived from data collected at Na^+ concentrations $\leq$4 mol/L and in the absence of chloride.

3. The inclusion of Am-chloride species extends the useful range of the An(III) model to chloride concentrations up to 12 mol/kg (the maximum amount of Cl^- present in the experiments from which the Am-chloride model is derived). Extension of the model to higher chloride concentrations will result in a slight improvement in simulations of WIPP brines, which (as represented in the PAVT) have chloride concentrations as high as 6.7 mol/kg (Novak et al., 1997). The inclusion of Pitzer parameters describing the interaction of Am-chloride species with Ca^{2+} will also slightly improve simulations of WIPP brines, and will allow use of the An(III) model to simulate high ionic strength brines dominated by $CaCl_2$. Application of the Ca^{2+} Pitzer parameters to Mg^{2+} interactions with Am-chloride species may also improve the model for use in modeling high Mg brines, including WIPP brines. Whether such application is appropriate remains to be determined.

6. References

DOE, 1996. Appendix SOTERM in *Title 40 CFR Part 191 Compliance Certification Application for the Waste Isolation Pilot Plant*, US Department of Energy, Carlsbad, NM.

Fanghänel, T., J.I. Kim, P. Paviet, R. Klenze and W. Hauser, 1994. Thermodynamics of radioactive trace elements in concentrated electrolyte solutions: Hydrolysis of Cm^{3+} in NaCl-solutions, *Radiochimica Acta*, **66/67**:81-87.

Fanghänel, T., J.I. Kim, R. Klenze and Y. Kato, 1995. Formation of Cm(III) Chloride Complexes in $CaCl_2$ Solutions, *Journal of Alloys and Compounds*, **225**:308-311.

Fanghänel, T. and J.I. Kim, 1998. Spectroscopic evaluation of thermodynamics of trivalent actinides in brines, *Journal of Alloys and Compounds*, **271-273**:728-737.

Fanghänel, T., T. Könnecke, H.T. Weger, P. Paviet-Hartmann, V. Neck and J.I. Kim, 1999. Thermodynamics of Cm(III) in concentrated salt solutions: Carbonate complexation in NaCl solution at 25°C, *Journal of Solution Chemistry*, **28**:447-462.

Felmy, A.R., D. Rai, J.A. Schramke and J.L. Ryan, 1989. The solubility of plutonium hydroxide in dilute solution and in high-ionic strength chloride brines, *Radiochimica Acta*, **48**:29-35. WPO 40690.

Felmy, A.R., D. Rai and R.W. Fulton, 1990. The solubility of $AmOHCO_3(c)$ and the aqueous thermodynamics of the system Na^+-Am^{3+}-HCO_3^--OH^--H_2O, *Radiochimica Acta*, **50**:193-204.

Felmy, A.R. and D. Rai, 1999. Application of Pitzer's equations for modeling the aqueous thermodynamics of actinide species in natural waters: A review, *Journal of Solution Chemistry*, **28**:533-553. SAND99-0340J

Fuger, J. and F.L. Oetting, 1976. *The Chemical Thermodynamics of Actinide Elements and Compounds. Part 2. The Actinide Aqueous Ions.*, Int. Atomic Energy Agency, Vienna, Austria.

Könnecke, T., T. Fanghänel and J.I. Kim, 1997. Thermodynamics of trivalent actinides in concentrated electrolyte solutions: Modelling the chloride complexation of Cm(III), *Radiochimica Acta*, **76**:131-135.

Morss, L.R. and C.W. Williams, 1994. Synthesis of crystalline americium hydroxide, $Am(OH)_3$, and determination of its enthalpy of formation; estimation of the solubility-product constants of actinide(III) hydroxides, *Radiochimica Acta*, **66/67**:89-93.

Novak, C.F., 1995. "A Data Base for Thermodynamic Modeling of +III Actinide Solubility in Concentrated Na-Cl-SO4-CO3-PO4 Electrolytes," 31 July 1995, Sandia National Laboratories, Albuquerque, NM, 17 pp. SAND95-2010C,

Novak, C.F., 1996. "Parameter values for +III actinide hydrolysis species," Memo to R. V. Bynum, June 11, 1996, Albuquerque: Sandia National Laboratories. WPO 40692

Novak, C.F., 1997. "Calculation of actinide solubilities in WIPP SPC and ERDA-6 brines under MgO backfill scenarios containing either nesquehonite or hydromagnesite as the Mg-CO_3 solubility-limiting phase," Memo to R. V. Bynum, April 21, 1997, Albuquerque, NM: Sandia National Laboratories. WPO 46124

Paviet, P., T. Fanghänel, R. Klenze and J.I. Kim, 1996. Thermodynamics of curium(III) in concentrated electrolyte solutions: Formation of sulfate complexes in NaCl/Na_2SO_4 solutions, *Radiochimica Acta*, **74**:99-103.

Pitzer, K.S. and G. Mayorga, 1973. Thermodynamics of electrolytes. II. Activity and osmotic coefficients for strong electrolytes with one or both ions univalent, *The Journal of Physical Chemistry*, **77**:2300-2308.

Rai, D., A.R. Felmy and R.W. Fulton, 1992. Solubility and ion activity product of $AmPO_4 \cdot xH_2O(am)$, *Radiochimica Acta*, **56**:7-14.

Rai, D., A.R. Felmy and R.W. Fulton, 1995. Nd^{3+} and Am^{3+} ion interactions with sulfate ion and their influence on $NdPO_{4(c)}$ solubility, *Journal of Solution Chemistry*, **24**:879-895.

Rai, D., R.G. Strickert, D.A. Moore and J.L. Ryan, 1983. Am(III) hydrolysis constants and solubility of Am(III) hydroxide, *Radiochimica Acta*, **33**:201-206.

Rao, L., D. Rai, A.R. Felmy, R.W. Fulton and C.F. Novak, 1996. Solubility of $NaNd(CO_3)_3 \cdot 6H_2O(c)$ in concentrated Na_2CO_3 and $NaHCO_3$ solutions, *Radiochimica Acta*, **75**:141-147. SAND94-1948J (also in SAND94-2274).

Runde, W. and J.I. Kim, 1995. "Study of the Extrapolability of Laboratory Data to Natural Conditions: Chemical Behavior of Trivalent and Pentavalent Americium in Saline NaCl Solutions," 28 July 1995, Institute for Radiochemistry, Technical University of Munich, Munich, 270 pp. SAND95-1720, WPO 41862.

Silva, R., G. Bidoglio, M.H. Rand, P.B. Robouch, H. Wanner and I. Puigdomenech, 1995. *Chemical Thermodynamics of Americium*, Nuclear Energy Agency OECD, Amsterdam.

ERG:6821:erg/(2002-1006, Rev. A)

Copy to:

MS-1395,	M.K. Knowles	[Dept. 6821]
MS-1395,	D.E. Wall	[Dept. 6822]
MS-1395,	N.A. Wall	[Dept. 6822]
MS-1395,	Y. Xiong	[Dept. 6822]
MS-1395,	Y. Wang	[Dept. 6823]
MS-0779,	R.C. Moore	[Dept. 6849]
MS-1395,	E.R. Giambalvo	[Dept. 6821]
MS-1395,	Day File	[Dept. 6821]

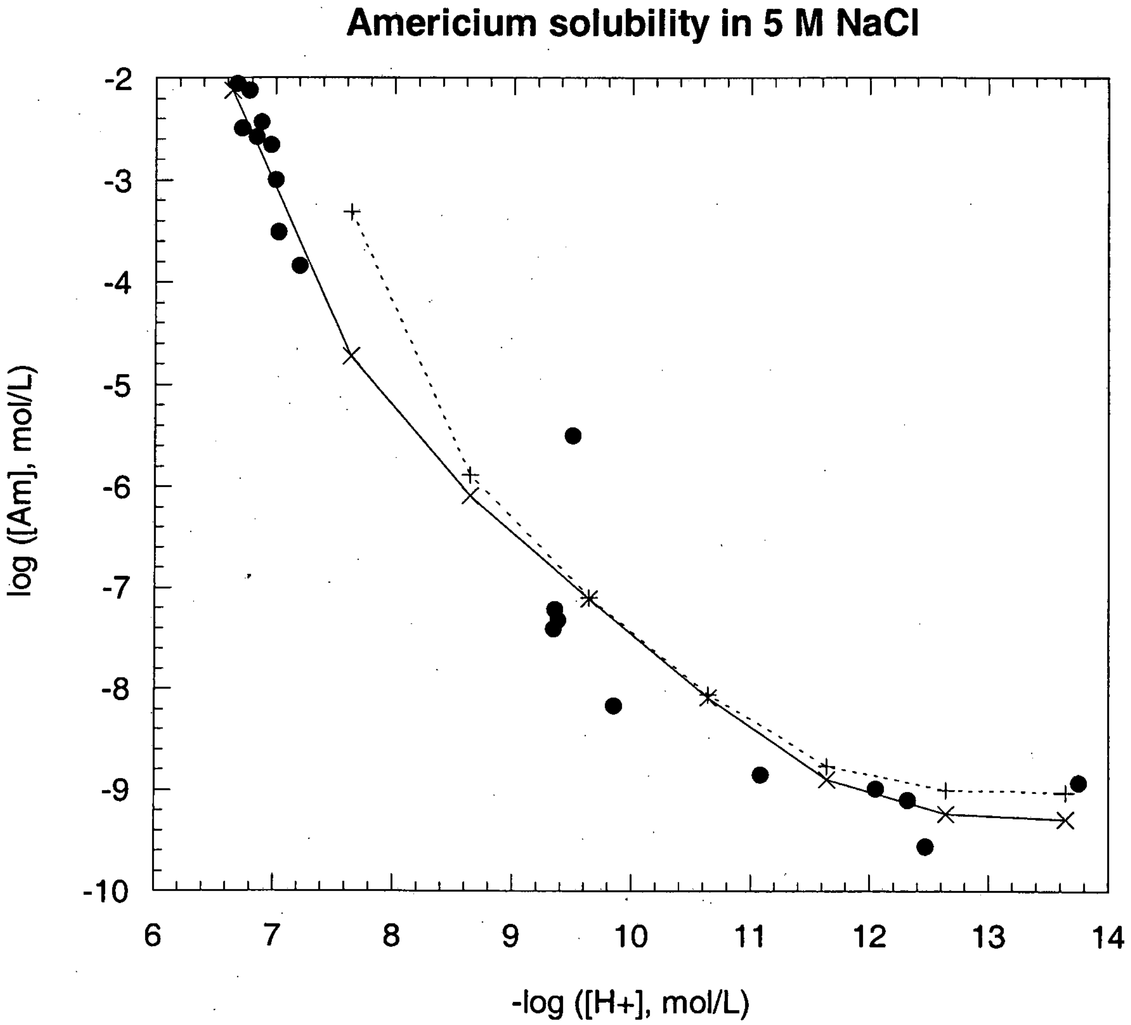

Figure 1. Am(OH)$_{3(s)}$ solubility in 5 mol/L NaCl. Circles are measured values (Runde and Kim, 1995). Solid line is predicted using the recommended model. Dashed line is predicted using the PAVT model. X and + symbols indicate points for which calculations were performed. The PAVT model predicts Am solubility in excess of 1 mol/L at pcH = 6.6.

Table 1. Normalized Chemical Potential (μ^0/RT)

	PAVT		Recommended	
Species	μ^0/RT	Source	μ^0/RT	Source
Am^{3+}	-241.694	Fuger and Oetting (1976)	-241.694	Fuger and Oetting (1976)
$AmCO_3^+$	-472.06	Felmy et al. (1990)	-473.29	Fanghänel et al. (1999)
$Am(CO_3)_2^-$	-695.88	Felmy et al. (1990)	-697.52	Fanghänel et al. (1999)
$Am(CO_3)_3^{3-}$	-915.46	Felmy et al. (1990)	-915.53	Fanghänel et al. (1999)
$Am(CO_3)_4^{5-}$	not included	NA	-1123.40	Fanghänel et al. (1999)
$AmSO_4^+$	not included	NA	-549.56	Fanghänel and Kim (1998)
$Am(SO_4)_2^-$	not included	NA	-850.99	Fanghänel and Kim (1998)
$AmOH^{2+}$	-315.352	Novak (1996), based on Fanghänel et al. (1994)	-319.96	Fanghänel et al. (1994)
$Am(OH)_2^+$	-392.281	Novak (1996), based on Fanghänel et al. (1994)	-396.89	Fanghänel et al. (1994)
$Am(OH)_{3(aq)}$	-471.603	Novak (1996), based on Runde and Kim (1995)	-469.53	Könnecke et al. (1997), from Silva et al. (1995)
$AmCl^{2+}$	not included	NA	-295.20	Könnecke et al. (1997)
$AmCl_2^+$	not included	NA	-345.90	Könnecke et al. (1997)
$AmOHCO_{3(c)}$	-569.98	Felmy et al. (1990)	-570.34	Fanghänel et al. (1999), based on Felmy et al. (1990)
$Am(OH)_{3(s)}$	-492.294	Novak (1996), based on Felmy et al. (1989)	-495.32	Könnecke et al. (1997), from Morse and Williams (1994)
$NaAm(CO_3)_2.6H_2O_{(c)}$	-1396.468	Rao et al. (1996)	-1396.468	Rao et al. (1996)
$AmPO_{4(c)}$	-709.75	Rai et al. (1992)	-709.75	Rai et al. (1992)

Table 2. Binary Pitzer Parameters ($\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(2)}$, C^{ϕ})

		PAVT					Recommended				
Cation	Anion	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	Source	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	Source
Na^+	$Am(CO_3)_2^-$	0	0	0	0	Rao et al. (1996)	-0.24	0.224	0	0.0284	Fanghänel et al. (1999)
Na^+	$Am(CO_3)_3^{3-}$	-0.256	5.0	0	0.0443	Rao et al. (1996)	0.125	4.73	0	0.0007	Fanghänel et al. (1999)
Na^+	$Am(CO_3)_4^{5-}$	NA	NA	NA	NA	NA	2.022	19.22	0	-0.305	Fanghänel et al. (1999)
$AmCO_3^+$	Cl^-	0	0	0	0	NA	-0.072	0.403	0	0.0388	Fanghänel et al. (1999)
Am^{3+}	SO_4^{2-}	3.0398	0	-2500	0	Rai et al. (1995)	1.792	15.04	0	0.600	Fanghänel and Kim (1998)
$AmSO_4^+$	Cl^-	NA	NA	NA	NA	NA	-0.091	-0.39	0	0.048	Fanghänel and Kim (1998)
Na^+	$Am(SO_4)_2^-$	NA	NA	NA	NA	NA	-0.354	0.4	0	0.051	Fanghänel and Kim (1998)
Am^{3+}	$H_2PO_4^-$	0	0	-92.9	0	Rai et al. (1995)	0	0	-92.9	0	Rai et al. (1995)
Am^{3+}	ClO_4^-	0.8	5.35	0	-0.0048	Felmy et al. (1990)	0.8	5.35	0	-0.0048	Felmy et al. (1990)
Am^{3+}	Cl^-	0.6117	5.403	0	-0.0284	Felmy et al. (1989)	0.5856	5.60	0	-0.016	Könnecke et al. (1997)
$AmOH^{2+}$	Cl^-	-0.6	3.0	0	0.2	Fanghänel et al. (1994)	-0.055	1.60	0	0.050	Könnecke et al. (1997)
$Am(OH)_2^+$	Cl^-	-0.58	-0.9	0	0	Fanghänel et al. (1994)	-0.616	-0.45	0	0.050	Könnecke et al. (1997)
$AmCl^{2+}$	Cl^-	NA	NA	NA	NA	NA	0.593	3.15	0	-0.006	Könnecke et al. (1997)
$AmCl_2^+$	Cl^-	NA	NA	NA	NA	NA	0.516	1.75	0	0.010	Könnecke et al. (1997)

Table 3. Neutral-Ion Interaction Parameter (λ)

		PAVT		Recommended	
Neutral	Ion	λ	Source	λ	Source
$Am(OH)_{3(aq)}$	Na^+	0	Novak (1996)	-0.2	Könnecke et al. (1997)
$Am(OH)_{3(aq)}$	Cl^-	0	Novak (1996)	-0.2	Könnecke et al. (1997)

Table 4. Ternary Interaction Parameters (θ, ψ)

			PAVT			Recommended		
ion *i*	ion *j*	ion *k*	θ_{ij}	ψ_{ijk}	Source	θ_{ij}	ψ_{ijk}	Source
Ca^{2+}	Am^{3+}	Cl^-	0	0	NA	0.2	0	Könnecke et al. (1997)
Ca^{2+}	$AmCl^{2+}$	Cl^-	0	0	NA	-0.014	0	Könnecke et al. (1997)
Ca^{2+}	$AmCl_2^+$	Cl^-	0	0	NA	-0.196	0	Könnecke et al. (1997)
Na^+	Am^{3+}	Cl^-	0	0	NA	0.1	0	Könnecke et al. (1997)
Cl^-	$Am(CO_3)_3^{3-}$	Na^+	0.168	0.0273	unknown	0	0	Fanghänel et al. (1999)
SO_4^{2-}	ClO_4^-	Na^+	0.020	0.0014	Rai et al. (1995)	0	0	*

*Eliminate Rai et al. (1995) Am^{3+}-SO_4^{2-} model.