

Thermodynamic Data for Predicting Concentrations of Pu(III), Am(III), and Cm(III) in Geologic Environments

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Thermodynamic Data for Predicting Concentrations of Pu(III), Am(III), and Cm(III) in Geologic Environments

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A b s t r a c t

This report provides thermodynamic data for predicting concentrations of Pu(III), Am(III), and Cm(III) in geologic environments, and contributes to an integration of the JNC chemical thermodynamic database, JNC-TDB (previously PNC-TDB), for the performance analysis of geological isolation system for high-level radioactive wastes. Thermodynamic data for the formation of complexes or compounds with hydroxide, chloride, fluoride, carbonate, nitrate, sulfate and phosphate are discussed in this report. Where data for specific actinide(III) species are lacking, the data were selected based on chemical analogy to other trivalent actinides. In this study, the Pitzer ion-interaction model is mainly used to extrapolate thermodynamic constants to zero ionic strength at 25°C.

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地質環境下におけるPu(III), Am(III) およびCm(III)の
濃度予測に関する熱力学データの整備
(研究報告)

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要旨

本研究では、地層処分システム性能評価のための熱力学データベースJNC-TDB（旧PNC-TDB）整備の一環で、III価のアクチニドPu(III), Am(III)およびCm(III)に関する熱力学データ整備を行った。本研究では、これらの元素に対して、水酸化物、塩化物、フッ化物、炭酸、硝酸、硫酸およびリン酸を含む錯体もしくは化合物に関する熱力学データ整備を行った。また、個別の元素に対して信頼できるデータがない場合、アクチニドIII価間での化学的類似性を考慮したデータ選定を行っている。

本研究では主にPitzerイオン相互作用モデルを用いて、25°C, イオン強度0におけるこれらの錯体および化合物の熱力学定数を整備した。

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Introduction

The objective of this study is to provide Japan Nuclear Cycle Development Institute (JNC) with reliable values of thermodynamic constants for environmentally important reactions. These values will be used to predict upper concentration limits of trivalent actinides and selected rare earths that can leach out of high-level waste repositories. The Pitzer ion-interaction model is selected for these applications because 1) it is applicable to low as well as high ionic strengths, 2) specific media effects can be reliably ascertained, 3) it provides a mechanism for cross comparison and reinterpretation of thermodynamic data developed under a wide range of ionic strengths and a variety of aqueous media, and 4) over the last few years we have developed a considerable amount of thermodynamic data in collaboration with JNC for trivalent rare earths (primarily Nd, an analog of trivalent actinides) and Am(III) using this approach and have been instrumental in putting together the trivalent actinide model for applications to the low-level waste repository (Waste Isolation Pilot Project (WIPP), New Mexico); thus, we can take advantage of this expertise and studies conducted for JNC and WIPP.

Compilation of Trivalent Actinide Thermodynamic Data

To develop a comprehensive data base for selecting constants, available pertinent literature was collected. The literature references pertaining to trivalent actinides were collected from several sources including: journal articles and books (e.g., Smith and Martell 1976; Fuger et al. 1992; Silva et al. 1995), subject indices of several pertinent recent journals, listings of articles on Pu compiled by the Nuclear Energy Agency and provided to us by courtesy of Dr. Hans Wanner, and a computer search of different data bases. Thermodynamic constants reported in these publications were summarized in different tables for each element and ligand (Table A1-A6, Appendix). In addition, these tables provide important information regarding experimental techniques and ligand concentration ranges reported in the publications.

The thermodynamic equilibrium constants of OH⁻, Cl⁻, F⁻, NO₃⁻, CO₃²⁻, SO₄²⁻, and PO₄³⁻ complexes with Sm(III) and trivalent actinides (Ac(III), Np(III), Pu(III), Am(III), and Cm(III)) are reported and compiled in Appendix (Tables A1-A6). Several general conclusions can be made from these data: 1) The least amount of experimental data exists for Ac(III), 2) Complexes with Cl⁻ and NO₃⁻ are very weak, 3) Most studies are conducted at relatively high but constant ionic strength and invariably in different ionic media (for an example of Am sulfate complexes, see Rai et al. 1995), thus making it difficult to extrapolate the results to zero ionic strength because of the lack of information on the specific ionic media dependence of the apparent equilibrium constants, 4) Experimental data for many species, expected to be important for trivalent actinides, are not available. For example, no experimental data are available for Pu(OH)₂⁺, Pu(OH)₃⁰, or carbonate complexes of Pu(III), and 5) Other than our work for carbonate (Felmy et al. 1990; Rao et al. 1996a) and sulfate systems (Rai et al. 1995), very few attempts have been made to

reinterpret the literature data for cross comparisons of values obtained using different experimental techniques, at different ionic strengths, and in different ionic media.

As shown in the compilation of literature data (Tables A1-A6, Appendix), a number of authors have used various approaches (e.g., extrapolation, S.I.T., or Pitzer's approach) to estimate values for stability constants at standard state conditions ($I = 0$, $T = 298.15$ K). However, the values for an individual system may vary significantly. To provide a basis for choosing among these values, a screening approach has been used which is based on the strongly ionic nature of the actinide-ligand bonding. A detailed description of this approach is provided in Section A7 of the Appendix. Based on this approach, stability constants of the first complexes of several individual trivalent elements (Sm^{3+} , Am^{3+} , and Pu^{3+}) with relevant ligands were selected and listed in Table A7. This list, including only the constants of the first aqueous complex, is not meant to be used as a complete set of parameters for prediction of the concentrations of $\text{Sm}(\text{III})$, $\text{Am}(\text{III})$, and $\text{Pu}(\text{III})$. Instead, these values can be used for cross-comparison of the stability constants of individual trivalent elements. The values for complexes of Am^{3+} with strongly complexing ligands are essentially the same as those recommended for the Am model described in subsequent sections. Complexation constants of $\text{Sm}(\text{III})$ and $\text{Am}(\text{III})$ with weakly complexing ligands (Cl^- and NO_3^-) are reported in the literature (Tables A1.2-A1.3, A5.2-A5.3, Appendix) and values selected by the screening approach are compared in Table A7 (Appendix). Although no specific ion-interaction parameters for trivalent actinides with Cl^- and NO_3^- are available, values of these parameters for rare earths are available and they are applicable to very dilute to concentrated electrolyte solutions. We have shown that parameters for $\text{Nd}^{3+}-\text{Cl}^-$ are directly applicable to $\text{Pu}^{3+}-\text{Cl}^-$ (Felmy et al. 1989) and $\text{Am}^{3+}-\text{Cl}^-$ (Rai et al. 1992a). For these reasons, and because of the similarity among the values of different complexes of $\text{Am}(\text{III})$, $\text{Pu}(\text{III})$, and $\text{Sm}(\text{III})$ (Table A7, Appendix), an $\text{Am}(\text{III})$ model developed and described in this document is applicable to other trivalent actinides and $\text{Sm}(\text{III})$.

Based on the reported data for solid phases of trivalent actinides or on the available data for solid phases of rare earths (which show similar ionic radii and form isostructural compounds with actinides and that are excellent analogs for trivalent actinides), solid compounds of trivalent actinides which may be of significant importance to waste repository include $\text{M(OH)}_3(\text{am})$, $\text{MPO}_4(\text{c})$, $\text{MF}_3(\text{c})$, $\text{M(OH)CO}_3(\text{c})$, $\text{NaM(CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$, and $\text{M}_2(\text{CO}_3)_3(\text{c})$. These compounds are expected to form readily in geologic environments and they may, under different conditions, help set upper limits on the aqueous concentrations of actinides. Unfortunately, experimental thermodynamic data are not available for all of these solids for all elements. As shown in Table 1 which is based on literature compilation (Tables A1-A6), data are available for all $\text{Am}(\text{III})$ compounds. However, only limited data are available for $\text{Pu}(\text{III})$ compounds ($\text{Pu(OH)}_3(\text{s})$ and $\text{PuPO}_4(\text{s})$) and the only data available for $\text{Cm}(\text{III})$ compounds is for $\text{CmF}_3(\text{s})$.

Because of the lack of experimental data for several important aqueous complexes and solid phases of all trivalent actinides, completely independent thermodynamic models for specific elements cannot be developed. Because trivalent rare earths are excellent analogs for trivalent actinides, as we will show in the next section, and because Pitzer ion-interaction parameters are available for common systems (e.g., Cl^- , ClO_4^- , and NO_3^-) involving trivalent rare earths and for all environmentally important major electrolyte ions, it is currently possible to develop only a generic trivalent model that can be used to

Table 1. Availability of Experimental Data for Solubility of Different Trivalent Actinide Compounds^(a)

Compound	Pu(III)	Am(III)	Cm(III)
M(OH) ₃ (s)	✓	✓	
MPO ₄ (c)	✓	✓	
MF ₃ (c)		✓	✓
M(OH)CO ₃ (c)		✓	
NaM(CO ₃) ₂ •H ₂ O		✓	
M ₂ (CO ₃) ₃ (c)		✓	

(a) Check mark indicates some data are available.

estimate upper concentration limits of all trivalent actinides and important rare earths. In the next section, we discuss such a trivalent model based on the Pitzer approach, and we will provide supporting data on the reliability of this model.

Selected Thermodynamic Data for a Trivalent Actinide Model

Based on extensive experimental studies on Am(III) and Nd(III) and limited studies on Pu(III), a Pitzer model was developed to predict aqueous concentrations in a wide variety of groundwater compositions. While the data specifically pertinent to each actinide element are insufficient for us to develop element-specific models, we do have sufficient data to develop a model for Am. Therefore, modeling parameters specifically for Am(III) are reported in this report. We believe, however, that until more data for other trivalent actinide elements become available, this model can be used to provide reasonable estimates of concentrations for Pu(III), Cm(III), and Sm(III).

The required thermodynamic data for the Am model are reported in Tables 2-8. Tables 2 and 3 provide Pitzer ion-interaction parameters for binary and ternary interactions for the major electrolyte ions (i.e., Na, K, Ca, Mg, F, Cl, ClO₄, NO₃, CO₃, SO₄, PO₄, MoO₄). Table 4 provides the binary and ternary parameters for Am species. Table 5 provides $\Delta_f G^0/RT$ values for major electrolyte ions and Table 6 for Am species. Thermodynamic equilibrium constants for complexation and solid phase dissolution reactions consistent with the reported $\Delta_f G^0/RT$ values appear in Tables 7 and 8.

This model has been tested for a large number of systems involving ion-interactions/ion-pairs of OH, Cl, F, CO₃, SO₄, and PO₄ with Nd(III), Gd(III), Am(III), and Pu(III) and has been shown to reliably predict the concentrations in dilute to concentrated solutions and in simple as well as mixed electrolyte systems. The basis for selection of the model parameters for Am interactions with different ligands is discussed in detail in the following text.

Table 2. Binary Ion-Interaction Parameters for Major Electrolyte Ions

Species	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^ϕ	References
Na ⁺ -Cl ⁻	0.0765	0.2644	0.00	0.00127	Harvie et al. (1984)
Na ⁺ -SO ₄ ²⁻	0.01958	1.113	0.00	0.00497	Harvie et al. (1984)
Na ⁺ -HSO ₄ ⁻	0.0454	0.398	0.00	0.00	Harvie et al. (1984)
Na ⁺ -OH ⁻	0.0864	0.253	0.00	0.0044	Harvie et al. (1984)
Na ⁺ -HCO ₃ ⁻	0.0277	0.0411	0.00	0.00	Harvie et al. (1984)
Na ⁺ -CO ₃ ²⁻	0.0399	1.389	0.00	0.0044	Harvie et al. (1984)
K ⁺ -Cl ⁻	0.04835	0.2122	0.00	-0.00084	Harvie et al. (1984)
K ⁺ -SO ₄ ²⁻	0.04995	0.7793	0.00	0.00	Harvie et al. (1984)
K ⁺ -HSO ₄ ⁻	-0.0003	0.1735	0.00	0.00	Harvie et al. (1984)
K ⁺ -OH ⁻	0.1298	0.320	0.00	0.0041	Harvie et al. (1984)
K ⁺ -HCO ₃ ⁻	0.0296	-0.013	0.00	-0.008	Harvie et al. (1984)
K ⁺ -CO ₃ ²⁻	0.1488	1.43	0.00	-0.0015	Harvie et al. (1984)
Ca ²⁺ -Cl ⁻	0.3159	1.614	0.00	-0.00034	Harvie et al. (1984)
Ca ²⁺ -SO ₄ ²⁻	0.20	3.1973	-54.24	0.00	Harvie et al. (1984)
Ca ²⁺ -HSO ₄ ⁻	0.2145	2.53	0.00	0.00	Harvie et al. (1984)
Ca ²⁺ -OH ⁻	-0.1747	-0.2303	-5.72	0.00	Harvie et al. (1984)
Ca ²⁺ -HCO ₃ ⁻	0.4	2.977	0.00	0.00	Harvie et al. (1984)
Mg ²⁺ -Cl ⁻	0.35235	1.6815	0.00	0.00519	Harvie et al. (1984)
Mg ²⁺ -SO ₄ ²⁻	0.2210	3.343	-37.23	0.025	Harvie et al. (1984)
Mg ²⁺ -HSO ₄ ⁻	0.4746	1.729	0.00	0.00	Harvie et al. (1984)
Mg ²⁺ -HCO ₃ ⁻	0.329	0.6072	0.00	0.00	Harvie et al. (1984)
MgOH ⁻ -Cl ⁻	-0.10	1.658	0.00	0.00	Harvie et al. (1984)
H ⁺ -Cl ⁻	0.1775	0.2945	0.00	0.0008	Harvie et al. (1984)
H ⁺ -SO ₄ ²⁻	0.0298	0.00	0.00	0.0438	Harvie et al. (1984)
H ⁺ -HSO ₄ ⁻	0.2065	0.5556	0.00	0.00	Harvie et al. (1984)
Na ⁺ -H ₂ PO ₄ ⁻	-0.0533	0.0396	0.00	0.00795	Pitzer and Mayorga (1973)
Na ⁺ -HPO ₄ ²⁻	-0.0583	1.4655	0.00	0.02938	Pitzer and Mayorga (1973)
Na ⁺ -PO ₄ ³⁻	0.17813	3.8513	0.00	-0.05154	Pitzer and Mayorga (1973)
Na ⁺ -ClO ₄ ⁻	0.0554	0.2755	0.00	-0.00118	Pitzer (1991)
H ⁺ -ClO ₄ ⁻	0.1747	0.2931	0.00	0.00819	Pitzer (1991)
H ⁺ -NO ₃ ⁻	0.1168	0.3546	0.00	-0.00539	Pitzer (1991)
Na ⁺ -NO ₃ ⁻	0.0068	0.1783	0.00	-0.00072	Pitzer (1991)
K ⁺ -NO ₃ ⁻	-0.0816	0.0494	0.00	0.0066	Pitzer (1991)
Ca ²⁺ -NO ₃ ⁻	0.211	1.409	0.00	-0.0202	Pitzer (1991)
Mg ²⁺ -NO ₃ ⁻	0.3671	1.585	0.00	-0.0207	Pitzer (1991)

Table 3. Ternary Ion-Interaction Parameters for Major Electrolyte Ions

Species	Ternary Parameter	References
$\text{Na}^+ \text{-K}^+$	-0.012	Harvie et al. (1984)
$\text{Na}^+ \text{-K}^+ \text{-Cl}^-$	-0.0018	Harvie et al. (1984)
$\text{Na}^+ \text{-K}^+ \text{-SO}_4^{2-}$	-0.010	Harvie et al. (1984)
$\text{Na}^+ \text{-K}^+ \text{-HCO}_3^-$	-0.003	Harvie et al. (1984)
$\text{Na}^+ \text{-K}^+ \text{-CO}_3^{2-}$	0.003	Harvie et al. (1984)
$\text{Na}^+ \text{-K}^+ \text{-NO}_3^-$	-0.001	Pitzer (1991)
$\text{Na}^+ \text{-Ca}^{2+}$	0.07	Harvie et al. (1984)
$\text{Na}^+ \text{-Ca}^{2+} \text{-Cl}^-$	-0.007	Harvie et al. (1984)
$\text{Na}^+ \text{-Ca}^{2+} \text{-SO}_4^{2-}$	-0.055	Harvie et al. (1984)
$\text{Na}^+ \text{-Mg}^{2+}$	0.07	Harvie et al. (1984)
$\text{Na}^+ \text{-Mg}^{2+} \text{-Cl}^-$	-0.012	Harvie et al. (1984)
$\text{Na}^+ \text{-Mg}^{2+} \text{-SO}_4^{2-}$	-0.015	Harvie et al. (1984)
$\text{Na}^+ \text{-H}^+$	0.036	Harvie et al. (1984)
$\text{Na}^+ \text{-H}^+ \text{-Cl}^-$	-0.004	Harvie et al. (1984)
$\text{Na}^+ \text{-H}^+ \text{-HSO}_4^-$	-0.0129	Harvie et al. (1984)
$\text{K}^+ \text{-Ca}^{2+}$	0.032	Harvie et al. (1984)
$\text{K}^+ \text{-Ca}^{2+} \text{-Cl}^-$	-0.025	Harvie et al. (1984)
$\text{K}^+ \text{-Mg}^{2+} \text{-Cl}^-$	-0.022	Harvie et al. (1984)
$\text{K}^+ \text{-Mg}^{2+} \text{-SO}_4^{2-}$	-0.048	Harvie et al. (1984)
$\text{K}^+ \text{-H}^+$	0.005	Harvie et al. (1984)
$\text{K}^+ \text{-H}^+ \text{-Cl}^-$	-0.011	Harvie et al. (1984)
$\text{K}^+ \text{-H}^+ \text{-SO}_4^{2-}$	0.197	Harvie et al. (1984)
$\text{K}^+ \text{-H}^+ \text{-HSO}_4^-$	-0.0265	Harvie et al. (1984)
$\text{Ca}^{2+} \text{-Mg}^{2+}$	0.007	Harvie et al. (1984)
$\text{Ca}^{2+} \text{-Mg}^{2+} \text{-Cl}^-$	-0.012	Harvie et al. (1984)
$\text{Ca}^{2+} \text{-Mg}^{2+} \text{-SO}_4^{2-}$	0.024	Harvie et al. (1984)
$\text{Ca}^{2+} \text{-H}^+$	0.092	Harvie et al. (1984)
$\text{Ca}^{2+} \text{-H}^+ \text{-Cl}^-$	-0.015	Harvie et al. (1984)
$\text{Mg}^{2+} \text{-MgOH} \text{-Cl}^-$	0.028	Harvie et al. (1984)
$\text{Mg}^{2+} \text{-H}^+$	0.10	Harvie et al. (1984)
$\text{Mg}^{2+} \text{-H}^+ \text{-Cl}^-$	-0.011	Harvie et al. (1984)
$\text{Mg}^{2+} \text{-H}^+ \text{-HSO}_4^-$	-0.0178	Harvie et al. (1984)
$\text{Cl}^- \text{-SO}_4^{2-}$	0.02	Harvie et al. (1984)

Table 3. (contd)

Species	Ternary Parameter	References
$\text{Cl}^- \cdot \text{SO}_4^{2-} \cdot \text{Na}^+$	0.0014	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{SO}_4^{2-} \cdot \text{Ca}^{2+}$	-0.018	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{SO}_4^{2-} \cdot \text{Mg}^{2+}$	-0.004	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{HSO}_4^-$	-0.006	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{HSO}_4^- \cdot \text{Na}^+$	-0.006	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{HSO}_4^- \cdot \text{H}^+$	0.013	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{OH}^-$	-0.050	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{OH}^- \cdot \text{Na}^+$	-0.006	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{OH}^- \cdot \text{K}^+$	-0.006	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{OH}^- \cdot \text{Ca}^{2+}$	-0.025	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{HCO}_3^-$	0.03	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{HCO}_3^- \cdot \text{Na}^+$	-0.015	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{HCO}_3^- \cdot \text{Mg}^{2+}$	-0.096	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{CO}_3^{2-}$	-0.02	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{CO}_3^{2-} \cdot \text{Na}^+$	0.0085	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{CO}_3^{2-} \cdot \text{K}^+$	0.004	Harvie et al. (1984)
$\text{Cl}^- \cdot \text{NO}_3^-$	0.016	Pitzer (1991)
$\text{Cl}^- \cdot \text{NO}_3^- \cdot \text{Na}^+$	-0.006	Pitzer (1991)
$\text{Cl}^- \cdot \text{NO}_3^- \cdot \text{K}^+$	-0.006	Pitzer (1991)
$\text{Cl}^- \cdot \text{NO}_3^- \cdot \text{Mg}^{2+}$	0.00	Pitzer (1991)
$\text{Cl}^- \cdot \text{NO}_3^- \cdot \text{Ca}^{2+}$	-0.017	Pitzer (1991)
$\text{SO}_4^{2-} \cdot \text{HSO}_4^- \cdot \text{Na}^+$	-0.0094	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{HSO}_4^- \cdot \text{K}^+$	-0.0677	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{HSO}_4^- \cdot \text{Mg}^{2+}$	-0.0425	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{OH}^-$	-0.013	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{OH}^- \cdot \text{Na}^+$	-0.009	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{OH}^- \cdot \text{K}^+$	-0.050	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{HCO}_3^-$	0.01	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{HCO}_3^- \cdot \text{Na}^+$	-0.005	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{HCO}_3^- \cdot \text{Mg}^{2+}$	-0.161	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{CO}_3^{2-}$	0.02	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{CO}_3^{2-} \cdot \text{Na}^+$	-0.005	Harvie et al. (1984)
$\text{SO}_4^{2-} \cdot \text{CO}_3^{2-} \cdot \text{K}^+$	-0.009	Harvie et al. (1984)

Table 3. (contd)

Species	Ternary Parameter	References
$\text{OH}^- \text{-CO}_3^{2-}$	0.10	Harvie et al. (1984)
$\text{OH}^- \text{-CO}_3^{2-} \text{-Na}^+$	-0.017	Harvie et al. (1984)
$\text{OH}^- \text{-CO}_3^{2-} \text{-K}^+$	-0.01	Harvie et al. (1984)
$\text{HCO}_3^- \text{-CO}_3^{2-}$	-0.04	Harvie et al. (1984)
$\text{HCO}_3^- \text{-CO}_3^{2-} \text{-Na}^+$	0.002	Harvie et al. (1984)
$\text{HCO}_3^- \text{-CO}_3^{2-} \text{-K}^+$	0.012	Harvie et al. (1984)
$\text{CO}_2^0 \text{-Na}^+$	0.100	Harvie et al. (1984)
$\text{CO}_2^0 \text{-K}^+$	0.051	Harvie et al. (1984)
$\text{CO}_2^0 \text{-Ca}^{2+}$	0.181	Harvie et al. (1984)
$\text{CO}_2^0 \text{-Mg}^{2+}$	0.183	Harvie et al. (1984)
$\text{CO}_2^0 \text{-Cl}^-$	-0.005	Harvie et al. (1984)
$\text{CO}_2^0 \text{-SO}_4^{2-}$	0.097	Harvie et al. (1984)
$\text{CO}_2^0 \text{-HSO}_4^-$	-0.003	Harvie et al. (1984)
$\text{H}_3\text{PO}_4^0 \text{-H}_3\text{PO}_4^0$	0.0503	Pitzer and Silvester (1976)
$\text{H}_3\text{PO}_4^0 \text{-H}_3\text{PO}_4^0 \text{-H}_3\text{PO}_4^0$	0.0109	Pitzer and Silvester (1976)
$\text{H}_3\text{PO}_4^0 \text{-H}_2\text{PO}_4^-$	-0.04	Pitzer and Silvester (1976)
$\text{H}_3\text{PO}_4^0 \text{-H}^+$	0.29	Pitzer and Silvester (1976)
$\text{SO}_4^{2-} \text{-ClO}_4^-$	0.020	Rai et al. (1995) ^(a)
$\text{SO}_4^{2-} \text{-ClO}_4^- \text{-Na}^+$	0.0014	Rai et al. (1995) ^(a)

(a) Assuming Cl^- parameters reported by Harvie et al. (1984) apply to the ClO_4^- system; Rai et al. (1995) showed these values are reasonable.

Table 4. Binary and Ternary Ion-Interaction Parameters for Am Species

Species	Binary Interaction Parameters				References
	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{\ddagger}	
Am ³⁺ -ClO ₄ ⁻	0.754	6.53	0	0.0075	Rai et al. (1995) ^(a)
Am ³⁺ -Cl ⁻	0.6117	5.403	0	-0.019	Rai et al. (1992a) ^(a)
Am ³⁺ -NO ₃ ⁻	0.468	5.13	0	-0.0823	This compilation ^(b)
Am ³⁺ -SO ₄ ²⁻	3.0398	0	-2500	0	Rai et al. (1995)
Am ³⁺ -H ₂ PO ₄ ⁻	0	0	-92.9	0	This compilation ^(c)
Na ⁺ -Am(CO ₃) ₃ ³⁻	-0.256	5.0	0	0.0443	This compilation ^(d)

Ternary Interaction Parameters		
Cl ⁻ -Am(CO ₃) ₃ ³⁻	0.168	This compilation ^(e)
Na ⁺ -Cl ⁻ -Am(CO ₃) ₃ ³⁻	0.0273	This compilation ^(e)

(a) Assumed to be analogous to corresponding Nd(III) parameters.
 (b) Assumed to be analogous to the parameter for Nd³⁺-NO₃⁻ (Pitzer 1991).
 (c) Assumed to be analogous to the parameter for Nd³⁺-H₂PO₄⁻ (Rai and Felmy 1991). This ion-interaction parameter and the complexation reaction for AmH₂PO₄²⁺ reported in Table 8 are redundant. Thus, only one of these values should be used in calculations.
 (d) Assumed to be analogous to the parameter for Na⁺-Nd(CO₃)₃³⁻ (Rao et al. 1996a).
 (e) Assumed to be analogous to the parameter for Na⁺-Cl⁻-Nd(CO₃)₃³⁻ (Rao et al. 1996c).

Table 5. Dimensionless Standard Molar Gibbs Energies of Formation of Major Electrolyte Species

Species	$\Delta_f G^\circ / RT$	References
H ₂ O(1)	-95.6635	Harvie et al. (1984)
Na ⁺	-105.651	Harvie et al. (1984)
K ⁺	-113.957	Harvie et al. (1984)
Ca ²⁺	-223.30	Harvie et al. (1984)
Mg ²⁺	-183.468	Harvie et al. (1984)
MgOH ⁺	-251.94	Harvie et al. (1984)
H ⁺	0.00	Harvie et al. (1984)
Cl ⁻	-52.955	Harvie et al. (1984)
SO ₄ ²⁻	-300.386	Harvie et al. (1984)
HSO ₄ ⁻	-304.942	Harvie et al. (1984)
OH ⁻	-63.435	Harvie et al. (1984)
HCO ₃ ⁻	-236.751	Harvie et al. (1984)
CO ₃ ²⁻	-212.944	Harvie et al. (1984)
CaCO ₃ ⁰	-443.5	Harvie et al. (1984)
MgCO ₃ ⁰	-403.155	Harvie et al. (1984)
CO ₂ (aq) ⁰	-155.68	Harvie et al. (1984)
CO ₂ (g)	-159.092	Harvie et al. (1984)
Mg(OH) ₂ (c) Brucite	-335.40	Harvie et al. (1984)
CaCO ₃ (c) Calcite	-455.6	Harvie et al. (1984)
MgCO ₃ (c) Magnesite	-414.45	Harvie et al. (1984)
Ca(OH) ₂ (c) Portlandite	-362.12	Harvie et al. (1984)
H ₃ PO ₄ ⁰	-460.90	Pitzer and Silvester (1976)
H ₂ PO ₄ ⁻	-455.96	Pitzer and Silvester (1976)
HPO ₄ ²⁻	-439.367	Wagman et al. (1982)
PO ₄ ³⁻	-410.947	Wagman et al. (1982)
MoO ₄ ²⁻	-337.366	Wagman et al. (1982)
HMnO ₄ ⁻	-348.62	Felmy et al. (1995)
H ₂ MoO ₄ ⁰	-358.86	Felmy et al. (1995)
Mo ₇ O ₂₀ (OH) ₄ ²⁻	-2152.05	Felmy et al. (1995)
F ⁻	-112.465	Wagman et al. (1982)
HF(aq)	-119.738	Wagman et al. (1982)
NO ₃ ⁻	-43.868	Wagman et al. (1982)

Table 6. Dimensionless Standard Molar Gibbs Energies of Formation of Am Species

Species	$\Delta_f G^\circ / RT$	References
Am ³⁺	-241.694	Fuger and Oetting (1976)
AmOHCO ₃ (c)	-569.98	Felmy et al. (1990)
AmCO ₃ ⁺	-472.06	Felmy et al. (1990)
Am(CO ₃) ₂ ⁻	-695.88	Felmy et al. (1990)
Am(CO ₃) ₃ ³⁻	-915.46	Felmy et al. (1990)
AmPO ₄ ·xH ₂ O(am)	-709.75	Rai et al. (1992a)
Am(MoO ₄) ₂ ⁻	-942.22	Felmy et al. (1995)
Am ₂ (Mo ₂ O ₂₄)(aq)	-2644.30	Felmy et al. (1995)
NaAm(MoO ₄) ₂ (c)	-1069.29	Felmy et al. (1995)
Am ₂ (MoO ₄) ₃ ·xH ₂ O	-1555.59	Felmy et al. (1995)
AmF ²⁺	-361.988	Silva et al. (1995)
AmF ₂ ⁺	-479.979	Silva et al. (1995)
AmF ₃ ⁰	>-604.878	This compilation ^(b)
AmF ₃ (c)	-624.00	This compilation ^(b)
AmOH ²⁺	-322.621	Silva et al. (1995)
Am(OH) ₂ ⁺	-400.554	Silva et al. (1995)
Am(OH) ₃ ⁰	>-462.95	Felmy et al. (1990)
Am(OH) ₃ (am)	-489.564	Silva et al. (1995)
Am(OH) ₃ (c)	-493.709	Silva et al. (1995)
NaAm(CO ₃) ₂ ·6H ₂ O(c)	-1396.48	This compilation ^(c)
Am ₂ (CO ₃) ₃ (c)	-1199.126	Silva et al. (1995) ^(d)

(a) Assumed to be analogous to the values for corresponding Nd(III) species (Felmy et al. 1995).
 (b) Assumed to be analogous to the parameters for NdF₃(c) (Rai et al. 1992b).
 (c) Assumed to be analogous to the parameters for NaNd(CO₃)₂·6H₂O(c) (Rao et al. 1996a).
 (d) Value based on solubility product reported by Silva et al. (1995) and consistent with the $\Delta_f G^\circ / RT$ values for Am³⁺ and CO₃²⁻ used in this report.

Table 7. Log K_{sp}° Values (25°C) of Am(III) Solid Compounds

Reaction	$\log K_{sp}^{\circ}$	Reference
$\text{Am(OH)}_3(\text{s}) \rightleftharpoons \text{Am}^{3+} + 3\text{OH}^-$	-25.0	Silva et al. (1995)
$\text{Am(OH)}_3(\text{c}) \rightleftharpoons \text{Am}^{3+} + 3\text{OH}^-$	-26.8	Silva et al. (1995)
$\text{AmOHCO}_3(\text{c}) \rightleftharpoons \text{Am}^{3+} + \text{OH}^- + \text{CO}_3^{2-}$	-22.5	Felmy et al. (1990)
$\text{Am}_2(\text{CO}_3)_3(\text{c}) \rightleftharpoons 2\text{Am}^{3+} + 3\text{CO}_3^{2-}$	-33.4	Silva et al. (1995)
$\text{NaAm}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c}) \rightleftharpoons \text{Na}^+ + \text{Am}^{3+} + 2\text{CO}_3^{2-} + 6\text{H}_2\text{O}$	-21.40	This compilation ^(a)
$\text{AmPO}_4 \cdot x\text{H}_2\text{O}(\text{am}) \rightleftharpoons \text{Am}^{3+} + \text{PO}_4^{3-} + x\text{H}_2\text{O}$	-24.79	Rai et al. (1992a)
$\text{AmF}_3(\text{c}) \rightleftharpoons \text{Am}^{3+} + 3\text{F}^-$	-19.5	This compilation ^(b)
$\text{NaAm}(\text{MoO}_4)_2(\text{c}) \rightleftharpoons \text{Na}^+ + \text{Am}^{3+} + 2\text{MoO}_4^{2-}$	-20.5	This compilation ^(c)
$\text{Am}_2(\text{MoO}_4)_3 \cdot x\text{H}_2\text{O} \rightleftharpoons 2\text{Am}^{3+} + 3\text{MoO}_4^{2-} + x\text{H}_2\text{O}$	-26.1	This compilation ^(c)

(a) Assumed to be analogous to the value for $\text{NaNd}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$ (Rao et al. 1996a).(b) Assumed to be analogous to the value for $\text{NdF}_3(\text{c})$ (Rai et al. 1992b).(c) Assumed to be analogous to the values for $\text{NaNd}(\text{MoO}_4)_2(\text{c})$ and $\text{Nd}_2(\text{MoO}_4)_3 \cdot x\text{H}_2\text{O}$ (Felmy et al. 1995).**Table 8.** Log K° Values (25°C) for the Formation of Aqueous Species

Reaction	$\log K^{\circ}$	Reference
$\text{Am}^{3+} + \text{H}_2\text{O} \rightleftharpoons \text{Am}(\text{OH})^{2+} + \text{H}^+$	-6.4	Silva et al. (1995)
$\text{Am}^{3+} + 2\text{H}_2\text{O} \rightleftharpoons \text{Am}(\text{OH})^{+}_2 + 2\text{H}^+$	-14.1	Silva et al. (1995)
$\text{Am}^{3+} + 3\text{H}_2\text{O} \rightleftharpoons \text{Am}(\text{OH})^0 + 3\text{H}^+$	<-28.6	Felmy et al. (1990)
$\text{Am}^{3+} + \text{CO}_3^{2-} \rightleftharpoons \text{AmCO}_3^{+}$	7.6	Felmy et al. (1990)
$\text{Am}^{3+} + 2\text{CO}_3^{2-} \rightleftharpoons \text{Am}(\text{CO}_3)_2^{-}$	12.3	Felmy et al. (1990)
$\text{Am}^{3+} + 3\text{CO}_3^{2-} \rightleftharpoons \text{Am}(\text{CO}_3)_3^{3-}$	15.2	Felmy et al. (1990)
$\text{Am}^{3+} + \text{H}_2\text{PO}_4^{-} \rightleftharpoons \text{AmH}_2\text{PO}_4^{2+}$	2.74	This compilation ^(a)
$\text{Am}^{3+} + 2\text{MoO}_4^{2-} \rightleftharpoons \text{Am}(\text{MoO}_4)_2^{-}$	11.2	This compilation ^(b)
$\text{Am}^{3+} + \text{Mo}_{20}\text{O}_{20}(\text{OH})_4^{2-} \rightleftharpoons \text{Am}_2\text{Mo}_{20}\text{O}_{24}(\text{aq}) + 4\text{H}^+$	3.85	This compilation ^(b)
$\text{Am}^{3+} + \text{F}^- \rightleftharpoons \text{AmF}^{2+}$	3.4	Silva et al. (1995)
$\text{Am}^{3+} + 2\text{F}^- \rightleftharpoons \text{AmF}_2^{+}$	5.8	Silva et al. (1995)
$\text{Am}^{3+} + 3\text{F}^- \rightleftharpoons \text{AmF}_3(\text{aq})$	< 11.2	This compilation ^(c)

(a) Assumed to be analogous to the values for corresponding Nd/phosphate species (Rai and Felmy 1991). This complexation reaction and the ion-interaction parameters reported in Table 4 for $\text{Am}^{3+} - \text{H}_2\text{PO}_4^{-}$ are redundant. Thus, only one of these values should be used in calculations.

(b) Assumed to be analogous to the values for corresponding Nd(III)/molybdate species (Felmy et al. 1995).

(c) Estimated from the results for solubility of $\text{NdF}_3(\text{c})$ (Rai et al. 1992b).

Chloride and Nitrate

Felmy et al. (1989) studied the solubility of $\text{Pu}(\text{OH})_3(\text{am})$ in dilute solutions and in chloride and sulfate brines (Figure 1). The observed solubility of $\text{Pu}(\text{OH})_3(\text{am})$ in brine at a given pC_{H}^+ was about 3.5 orders of magnitude higher than in dilute solutions. Using the solubility product, calculated from the results of dilute solutions, and assuming that the binary ion-interaction parameters for $\text{Nd}^{3+}\text{-Cl}^-$ (Pitzer and Mayorga 1973) apply to $\text{Pu}^{3+}\text{-Cl}^-$, they were able to very closely predict the solubility of $\text{Pu}(\text{OH})_3(\text{am})$ in brines in the entire range of measured pC_{H}^+ values. These data, as expected, indicated that parameters for $\text{Nd}^{3+}\text{-Cl}^-$ equally apply to the corresponding Pu(III) system. Since Pu(III) and Am(III) show similar behavior in solubility, ion-interaction parameters for $\text{Nd}^{3+}\text{-Cl}^-$ are assumed to be applicable to the Am³⁺-Cl⁻ system. In fact, these parameters were also found to provide consistent predictions for the solubility of $\text{AmPO}_4(\text{am})$ in chloride media (Rai et al. 1992a).

Because of the analogous behavior of trivalent rare earths and actinides, binary parameters for $\text{Nd}^{3+}\text{-NO}_3^-$ reported by Pitzer (1991) are assumed to apply to the corresponding Am system.

Hydroxide

The solubility of Nd, Pu, and Am hydroxides are reported in Figure 2. In general, solubilities of amorphous hydroxides are slightly higher than crystalline hydroxides. There is remarkable similarity in the pH dependence of the solubility behavior. However, different combinations of hydrolysis constants and solubility products may equally explain the observed behavior. For example, the solubility of $\text{Nd}(\text{OH})_3(\text{c})$ can be reasonably explained by using the values of hydrolysis constants reported by Silva et al. (1995) or by Rai et al. (1983) for Am in combination with the solubility product reported by Rao et al. (1996b) (Figure 3). These values of constants and solubility product also provide reasonable predictions for the solubility of $\text{Pu}(\text{OH})_3(\text{am})$ (Felmy et al. 1989) and $\text{Am}(\text{OH})_3(\text{am})$ (Rai et al. 1983). We do notice that the solubilities of $\text{Nd}(\text{OH})_3(\text{c})$ predicted using hydrolysis constant data of Rai et al. (1983) are different from those by Silva et al. (1995) in the pH range above 7 (Figure 3). However, the experimental data in this pH range are quite scattered (close to the detection limit), making it difficult to exclusively justify the constants either by Rai et al. (1983) or by Silva et al. (1995). Furthermore, data in this pH range are not critical in evaluating the solubility product of $\text{Nd}(\text{OH})_3(\text{c})$. To be consistent with the published data base, values for first and second hydrolysis constants reported by Silva et al. (1995) are selected for this compilation. Reliable values for the formation of $\text{Am}(\text{OH})_3^0$ are not available, but are generally set at the detection limits obtained by solubility measurement in the OH system. Based on the data for Am in the carbonate system (Felmy et al. 1990) and on recent data for Gd in the carbonate system (Weger et al. 1998), the limiting value for this complex is expected to be several orders of magnitude lower than selected by Silva et al. (1995). Hence, the equilibrium constant value for the formation of $\text{Am}(\text{OH})_3^0$ is selected based on data by Felmy et al. (1990).

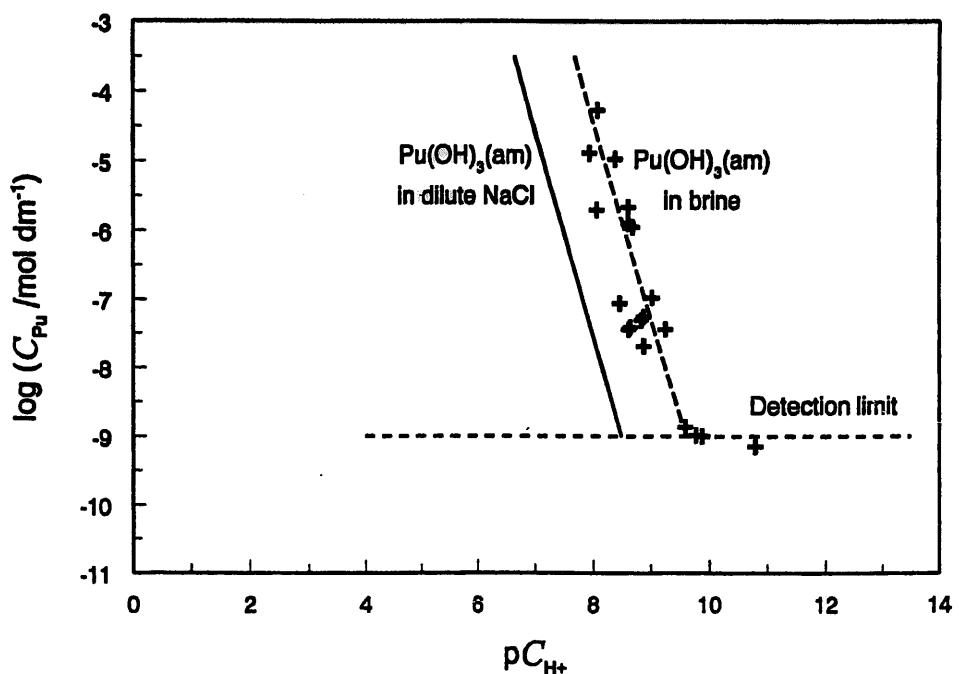


Figure 1. The Effect of PBB1 Brine ($[Na^+] = 5.2\text{ M}$, $[Ca^{2+}] = 0.034\text{ M}$, $[Cl^-] = 4.6\text{ M}$, $[SO_4^{2-}] = 0.044\text{ M}$, Ionic Strength $\sim 5.5\text{ M}$) on the Solubility of $Pu(OH)_3(\text{am})$ (Felmy et al. 1989)

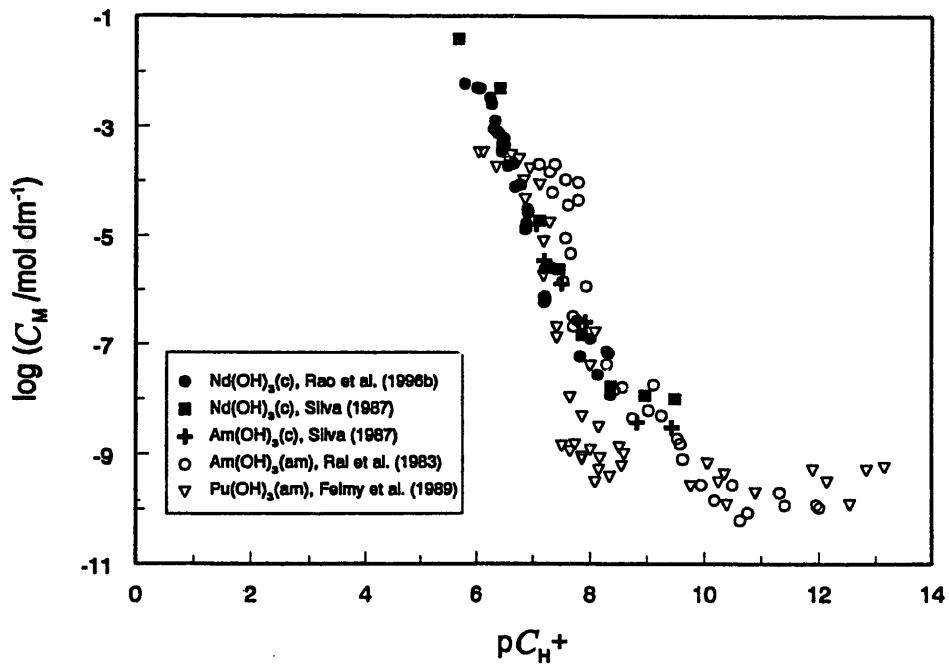


Figure 2. Solubility of Trivalent Actinide and Lanthanide Hydroxides

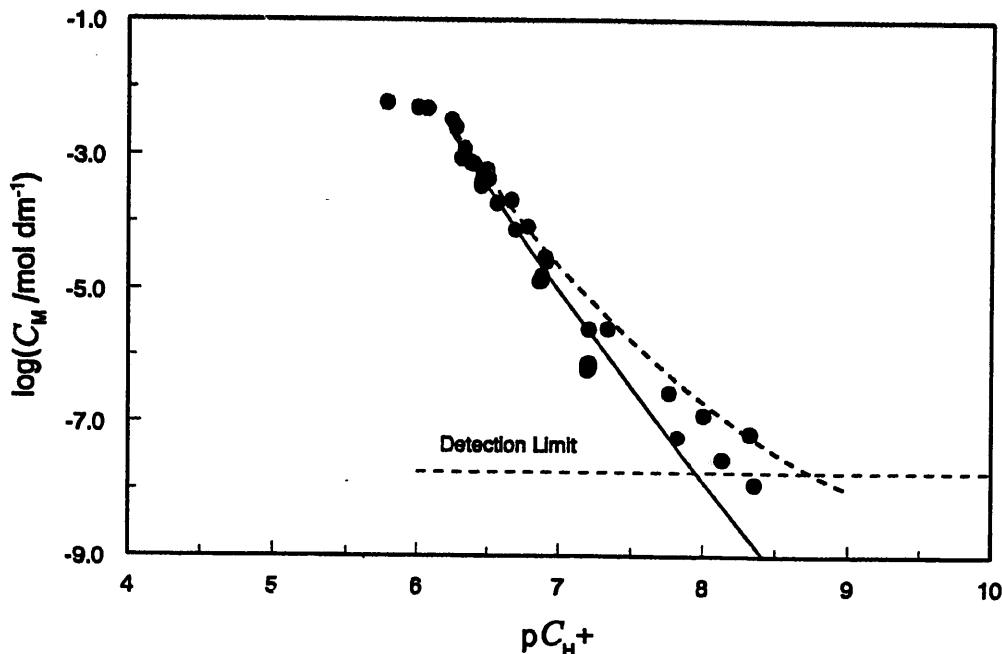


Figure 3. Comparison of Measured (points, Rao et al. 1996b) and Predicted Solubility of $\text{Nd}(\text{OH})_3(\text{c})$. Predictions are made with the hydrolysis constants from Rai et al. 1983 (solid line) and from Silva et al. 1995 (dashed line).

Fluoride

Fluoride forms strong aqueous complexes and fairly insoluble fluorides with trivalent actinides and rare earths. The values of formation constants of AmF^{2+} and AmF_2^+ from Silva et al. (1995) are selected and, in general, appear to be in close agreement with the values reported for other trivalent actinides (Tables A4.1, A5.1, and A6.1 in Appendix). The solubilities for $\text{AmF}_3(\text{c})$ (Phillips et al. 1988) and $\text{PuF}_3(\text{c})$ (Wanner 1986) predicted from estimated thermodynamic data are over 4 to 9 orders of magnitude higher than those experimentally determined for $\text{NdF}_3(\text{c})$ by Rai et al. (1992b) (Figure 4).

These predicted solubilities cannot possibly be correct, because they are all expected to be similar. The solubility product selected for the compilation is based on the solubility data reported by Rai et al. (1992b) for $\text{NdF}_3(\text{c})$ in NaF solutions ranging in concentrations from 10^{-5} to 1 M. Based on the same set of solubility data of $\text{NdF}_3(\text{c})$, an upper limit for the formation constant of NdF_3^0 is estimated and assumed applicable to AmF_3^0 .

Phosphate

Aqueous complexes of phosphate are not very strong, but phosphate forms very insoluble solids. Nd and Am phosphates show similar solubility and pH dependence (Figure 5). Based on the solubility of

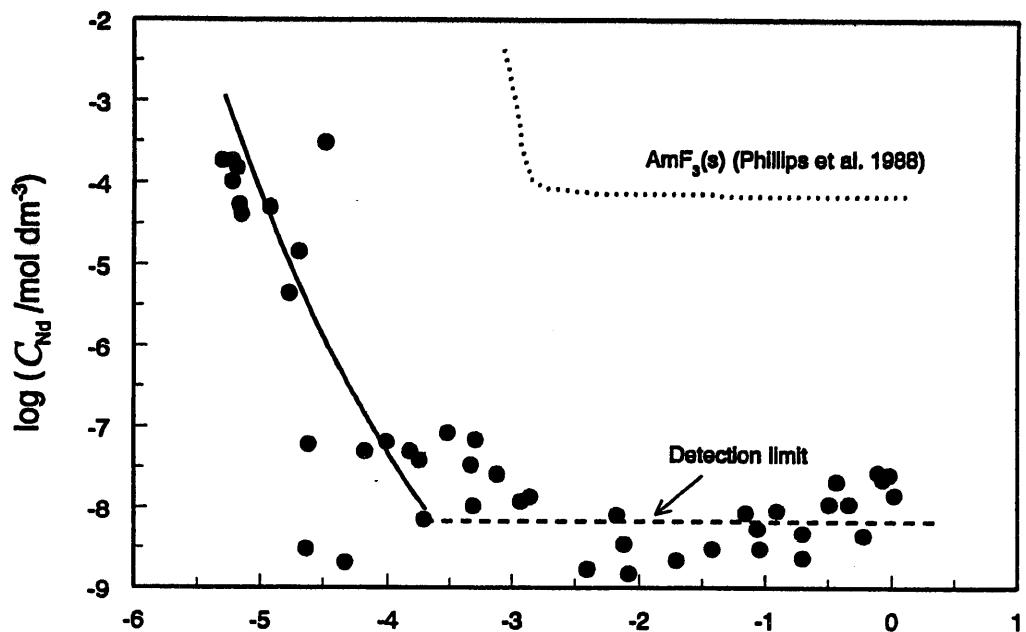


Figure 4. Comparison of Experimental Solubility of $\text{NdF}_3(\text{c})$ (Rai et al. 1992b) with the Predicted Solubilities of $\text{AmF}_3(\text{s})$ (Phillips et al. 1988)

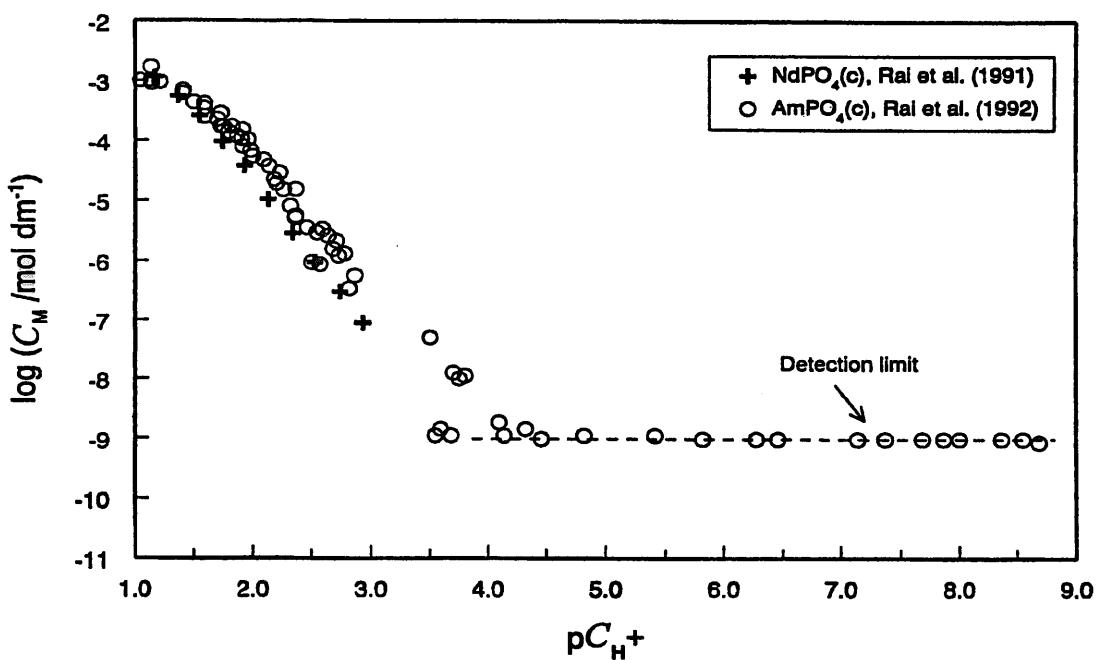


Figure 5. Solubility of Nd(III) and Am(III) Phosphates at Fixed $[\text{phosphate}]_{\text{total}} \approx 10^{-3.5} \text{ M}$

$\text{NdPO}_4(\text{c})$ as a function of $[\text{NaH}_2\text{PO}_4]$ ranging from 10^{-4} to 1.0 M (Figure 6) and as a function of pH in $10^{-3.5} \text{ M}$ total phosphate, Rai et al. (1991) developed reliable values of the solubility product of $\text{NdPO}_4(\text{c})$ and the ion-interaction parameter $\beta^{(2)}$ for $\text{Nd}^{3+}\text{-H}_2\text{PO}_4^-$. The solubility product of $\text{NdPO}_4(\text{c})$ was later found, as expected, to be similar to the solubility product of $\text{AmPO}_4 \cdot x\text{H}_2\text{O}$ (Rai et al. 1992a). This value is quoted by Silva et al. (1995) and selected for this review along with the $\beta^{(2)}$ value for $\text{Am}^{3+}\text{-H}_2\text{PO}_4^-$ (identical to the experimentally determined $\beta^{(2)}$ for $\text{Nd}^{3+}\text{-H}_2\text{PO}_4^-$). These values will provide reliable predictions of the solubility of Am phosphate in a large range of pH and phosphate concentrations.

Alternatively, the solubility of Am phosphate as a function of phosphate concentrations can be interpreted by assuming the formation of Am phosphate complexes (e.g., $\text{AmH}_2\text{PO}_4^{2+}$), instead of using the ion-interaction parameters $\beta^{(2)}$ for $\text{Am}^{3+}\text{-H}_2\text{PO}_4^-$. Rai and Felmy (1991) calculated the value of $\Delta_f G^\circ/\text{RT}$ for $\text{NdH}_2\text{PO}_4^{2+}$ from the best fit of their solubility data, which corresponds to $\log \beta = 2.74$ for $\text{Nd}^{3+} + \text{H}_2\text{PO}_4^- \rightleftharpoons \text{NdH}_2\text{PO}_4^{2+}$. Using either this value to account for the formation of $\text{NdH}_2\text{PO}_4^{2+}$ or the $\beta^{(2)}$ to account for the ion-interaction of $\text{Nd}^{3+}\text{-H}_2\text{PO}_4^-$, but not both, provides satisfactory predictions of the solubility of $\text{NdPO}_4(\text{c})$ in the presence of phosphate.

Sulfate

A large quantity of data are available on the complexation constants of sulfate with Pu(III), Am(III), and Cm(III) (Tables A4.13, A4.14, A5.11, A5.12, and A6.9 in Appendix). Most of these data were collected at high but constant ionic strengths to avoid variations in activity coefficients. Reliable

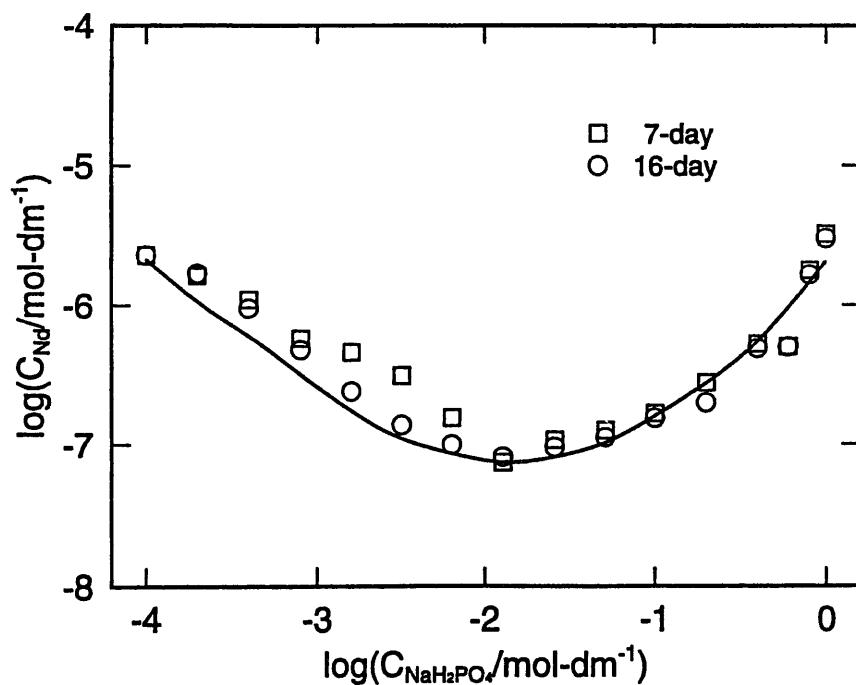


Figure 6. Solubility of $\text{NdPO}_4(\text{c})$ as a Function of the Concentration of NaH_2PO_4 at a Constant pH of 2.5 (Rai and Felmy 1991)

extrapolations of these data to low or zero ionic strengths is difficult. However, the values obtained by different authors under a constant set of conditions for different trivalent actinides agree closely. For example, values of $\log \beta_1$ for the formation of MSO_4^+ at $I = 2 \text{ M}$ reported by Rao et al. (1978) for Pu, Am, and Cm are 1.89, 1.71, and 1.59, respectively, and those reported by Carvalho and Choppin (1967) for Am and Cm are 1.65 and 1.61, respectively. However, there are several difficulties in unambiguously interpreting these data. The major difficulty results from the lack of ion-interaction parameters for bulk electrolyte ions to extrapolate these values to zero ionic strength or to have a capability to predict equilibrium constants at different ionic strengths. To overcome this difficulty, we chose data obtained by McDowell and Coleman (1972) in the absence of bulk electrolyte to determine binary ion-interaction parameters for $\text{Am}^{3+}-\text{SO}_4^{2-}$ (Rai et al. 1995). Values of $\beta^{(0)} = 3.0398$ and $\beta^{(2)} = -2500$ for $\text{Am}^{3+}-\text{SO}_4^{2-}$ provided the best representation of McDowell and Coleman's data (Figure 7). To check whether these parameters obtained from a simple system are applicable to mixed systems such as $\text{ClO}_4^--\text{SO}_4^{2-}$ and $\text{H}_2\text{PO}_4^--\text{SO}_4^{2-}$, we examined other solvent extraction and ion exchange Am data obtained in 1 M NaClO_4 solutions (Sekine 1965; Nair 1968) and data on the solubility of $\text{NdPO}_4(\text{c})$ in sulfate solutions (Rai et al. 1995). For these calculations we used $\beta^{(0)} = 3.0398$ and $\beta^{(2)} = -2500$ for $\text{Am}^{3+}-\text{SO}_4^{2-}$. We also used ion-interaction parameters for $\text{Nd}^{3+}-\text{ClO}_4^-$ reported by Pitzer (1991) to represent $\text{Am}^{3+}-\text{ClO}_4^-$ interactions for perchlorate systems or $\text{Nd}^{3+}-\text{H}_2\text{PO}_4^-$ parameters discussed above for the $\text{NdPO}_4(\text{c})$ solubility in sulfate solutions. The calculated concentrations in all cases are in excellent agreement with the solvent extraction and ion exchange data for Am perchlorate systems and with the solubility data of $\text{NdPO}_4(\text{c})$ in sulfate solutions (Figures 8 through 10). Several important conclusions can be drawn from these studies:

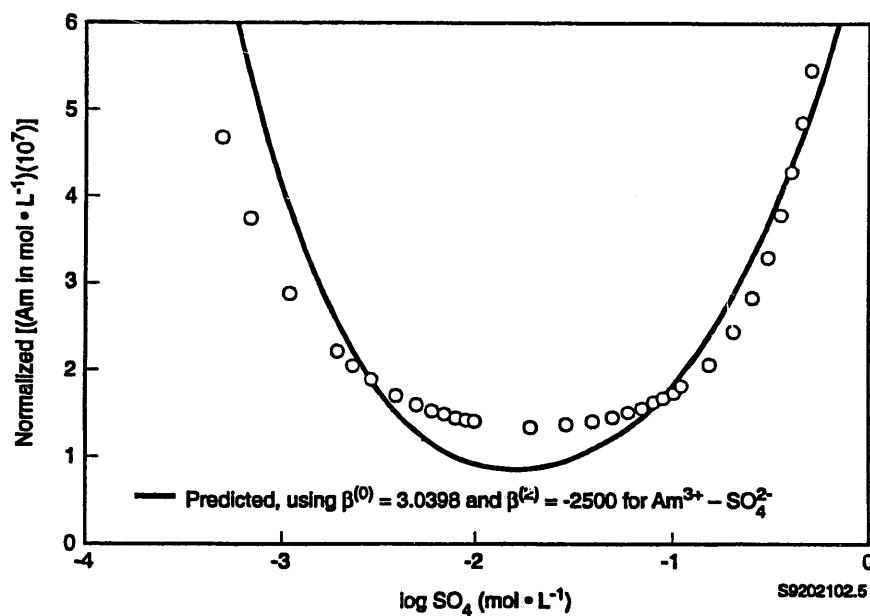


Figure 7. Normalized Equilibrium Aqueous Am Concentrations from Solvent Extraction Data of McDowell and Coleman (1972) Obtained at Constant Acid Activity and Extractant-Phase Composition. The solid line represents the predicted concentrations (see text for details). (Rai et al. 1995)

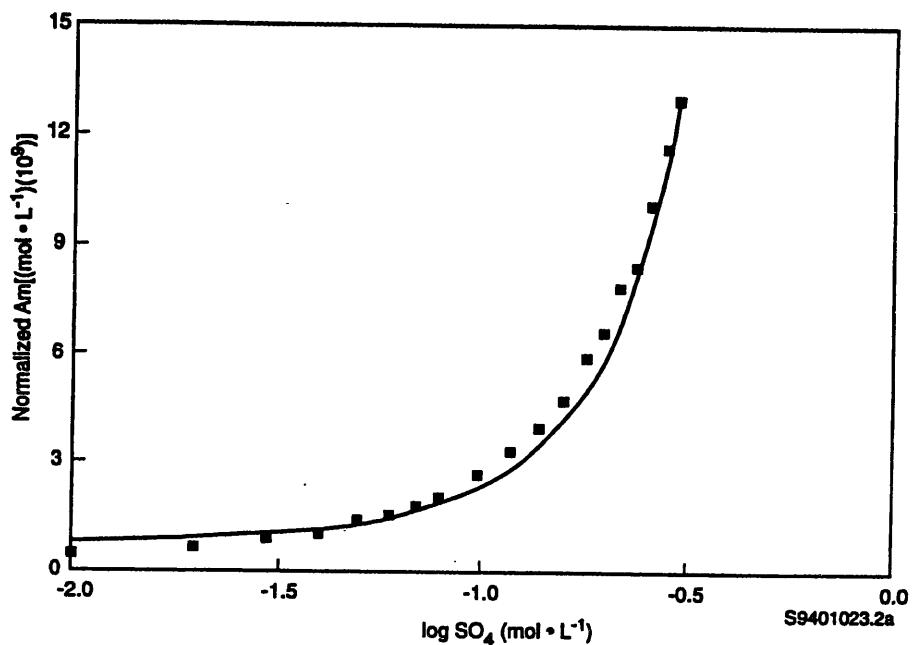


Figure 8. Normalized Equilibrium Aqueous Am Concentrations from Ion Exchange Experimental Data of Nair (1968) Obtained at Constant pH and 1 M NaClO₄. The solid line represents the predicted concentrations (see text for details). (Rai et al. 1995)

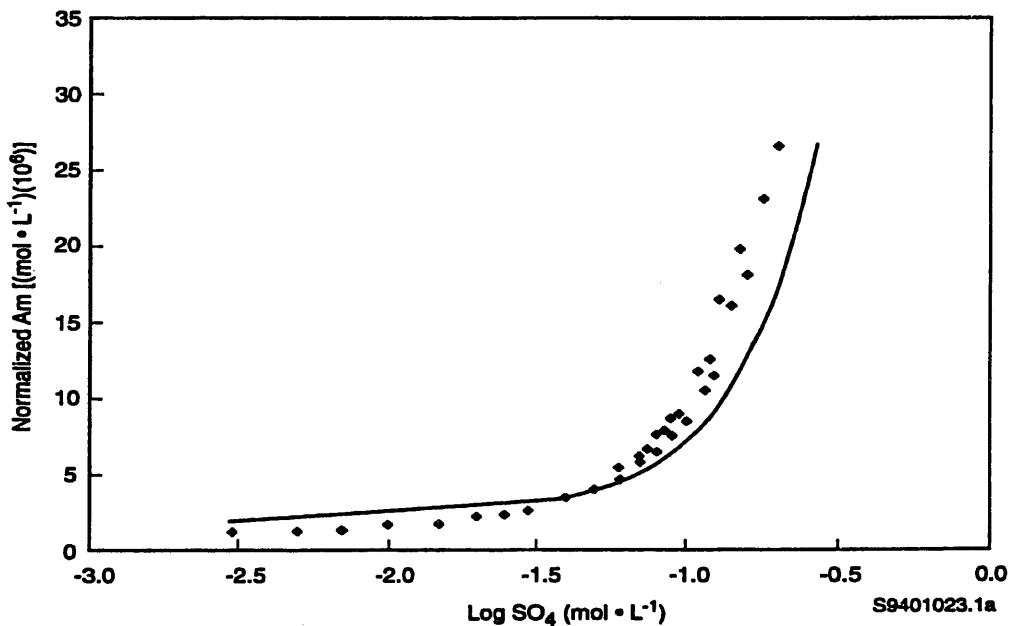


Figure 9. Normalized Equilibrium Aqueous Am Concentrations from Solvent Extraction Data of Sekine (1965) Obtained at Constant Acid Activity and Extractant-Phase Composition. The solid line represents the predicted concentrations (see text for details). (Rai et al. 1995)

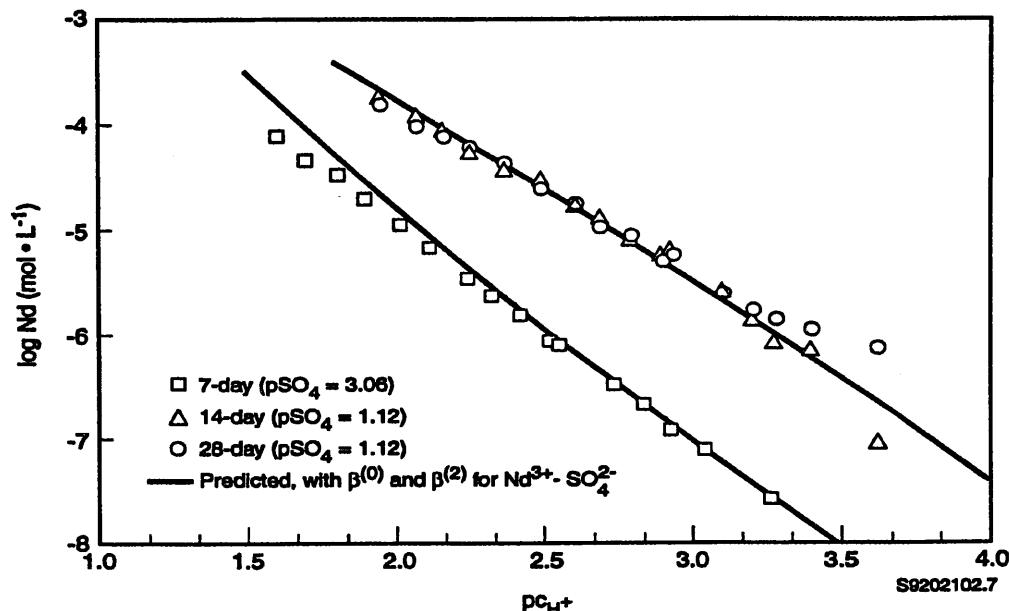


Figure 10. Solubility of $\text{NdPO}_4(\text{c})$ at $[\text{NaH}_2\text{PO}_4] = 10^{-3.5} \text{ M}$ and as a Function of Sulfate Concentration. Predicted concentrations based on Pitzer parameters obtained for Am-SO₄ data (see text for details). (Rai et al. 1995)

1) the values of Am³⁺-SO₄²⁻ interaction parameters are consistent with literature data and are applicable to sulfate concentrations as high as 0.5 M, 2) the values of Am³⁺-ClO₄⁻ interactions estimated in this study are reasonable, 3) the interaction parameters obtained for Am³⁺-SO₄²⁻ apply equally well to Nd³⁺-SO₄²⁻, and 4) the Pitzer parameters obtained in this study from simple systems can be used for predictions in concentrated mixed systems containing ClO₄⁻-SO₄²⁻ or H₂PO₄⁻-SO₄²⁻.

Carbonate

Carbonate forms strong complexes with trivalent rare earths and actinides. A large quantity of experimental data are available on carbonate complexes of Am (Tables A5.6-A5.7 in Appendix). Very limited experimental data are available for Cm(III) or Pu(III) (Tables A4.7 and A6.6 in Appendix). Felmy et al. (1990) determined the solubility of Am(OH)CO₃(c) in a wide pH range, fixed CO₂(g) atmosphere, and CO₃²⁻ concentrations extending to 0.1 M. Based on these results, Pitzer modeling parameters were developed, including the solubility product of Am(OH)CO₃(c) and values of formation constants of mono-, di-, and tricarbonate complexes of Am³⁺, that closely depicted the solubility of Am(OH)CO₃(c) (Figure 11). Felmy et al. (1990) also showed that their model was consistent with the solvent extraction data of Bidoglio (1982) (Figure 12) and the Am(OH)CO₃(c) solubility data of Silva and Nitsche (1984) (Figure 11). A recent review of Am thermodynamic data (Silva et al. 1995) based their values on Felmy et al. (1990).

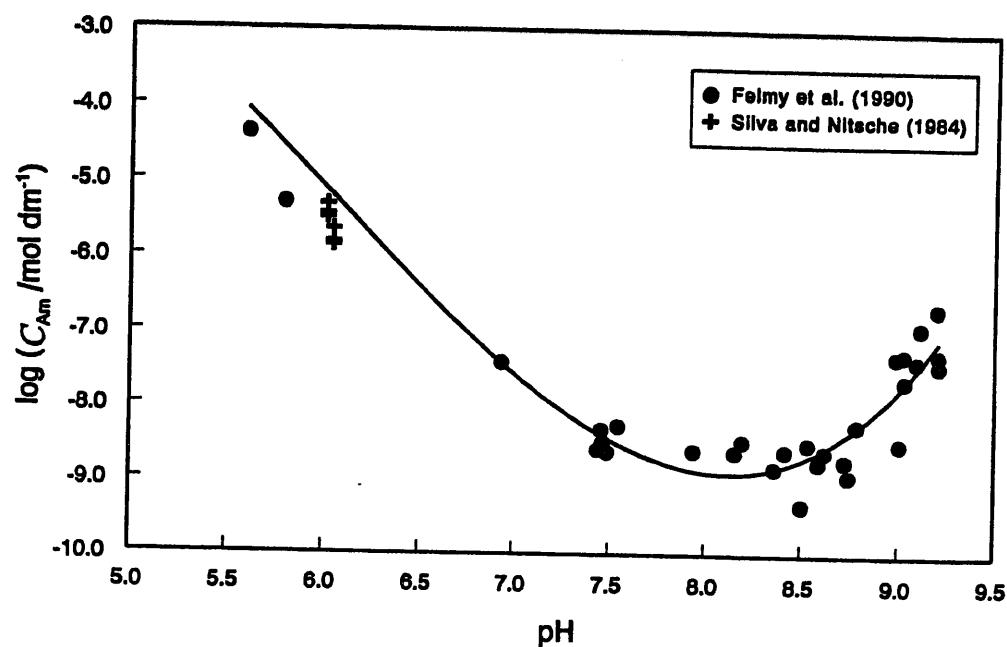


Figure 11. Solubility of $\text{AmOHCO}_3(\text{c})$ at a Fixed P_{CO_2} of 0.001 atm

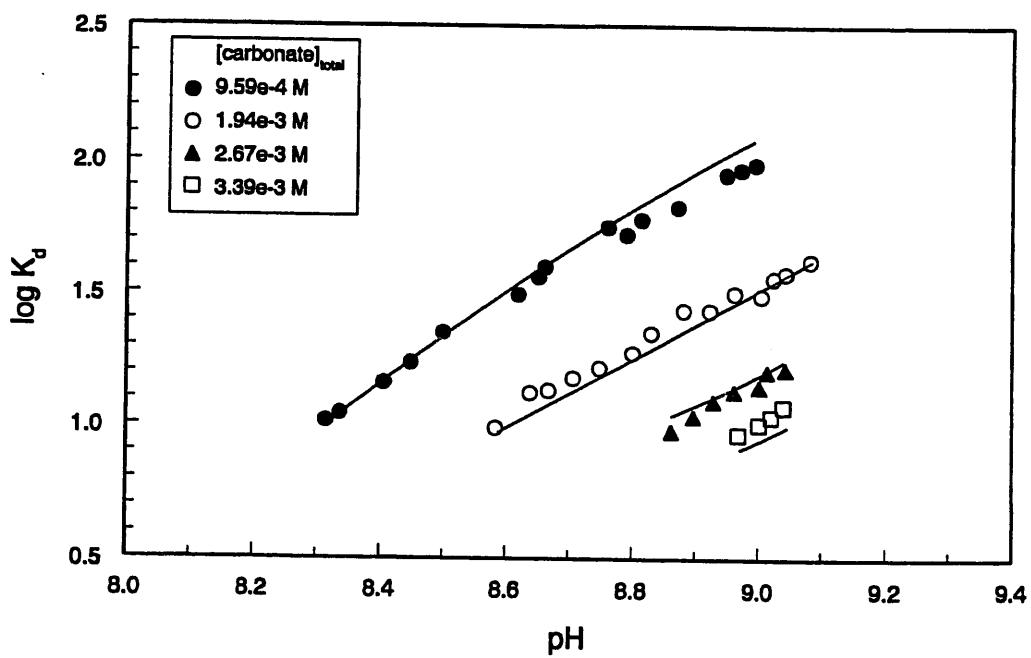


Figure 12. Comparison Between the Measured K_d by Solvent Extraction (points, Bidoglio 1982) and the Calculated K_d (solid lines, Felmy et al. 1990) at Different Total Carbonate Concentrations

To extend this model to high carbonate concentrations, determine the solubility controlling carbonate solid in the high carbonate region, and obtain reliable values of binary and ternary parameters applicable to concentrated mixed systems ($\text{Na}_2\text{CO}_3/\text{NaCl}$), Rao et al. (1996a, 1996c) conducted solubility studies in solutions of high $[\text{Na}_2\text{CO}_3]$ and $[\text{NaCl}]$ using $\text{Nd}_2(\text{CO}_3)_3(\text{c})$ and $\text{NaNd}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$ as the initial solids (Figures 13 and 14). Several conclusions were drawn from these data: 1) important carbonato complexes and their complexation constants with Nd^{3+} were identical to those for the analogous Am^{3+} system, 2) the solubility controlling solid in solutions of high carbonate concentrations was $\text{NaNd}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$, whereas $\text{Nd}_2(\text{CO}_3)_3(\text{c})$ is not stable under these conditions, 3) solubility of $\text{NaNd}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$ increased with the increase in $[\text{NaCl}]$ up to 4 M, and 4) revised values for the binary and ternary parameters of Nd^{3+} complexes were required to interpret these data. The final model selected in this report is based on these studies and is consistent with all of the previous data for $\text{Nd}(\text{III})/\text{Am}(\text{III})$ -carbonate systems (Figures 13 and 14).

As a final test of the robustness of the carbonate model and its ability to reliably predict solubility behavior in mixed and concentrated solutions, the model-predicted concentrations are compared with the experimental data of Runde and Kim (1995) for the solubility of $\text{NaAm}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$ in 5 M NaCl (Figure 15). Close agreement between the predicted and the experimental data showed that the model is applicable to the Am system and that the recommended values of the parameters are reliable.

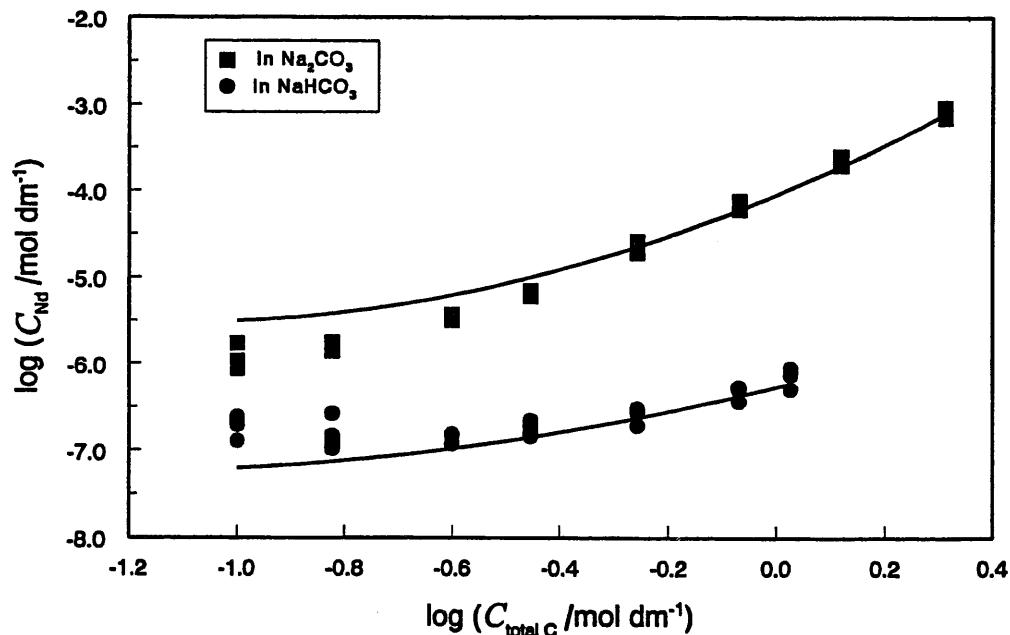


Figure 13. Observed (data points) and Model Predicted (solid lines, see text for model parameters) Solubility of $\text{NaNd}(\text{CO}_3)_2(\text{s})$ (Rao et al. 1996a)

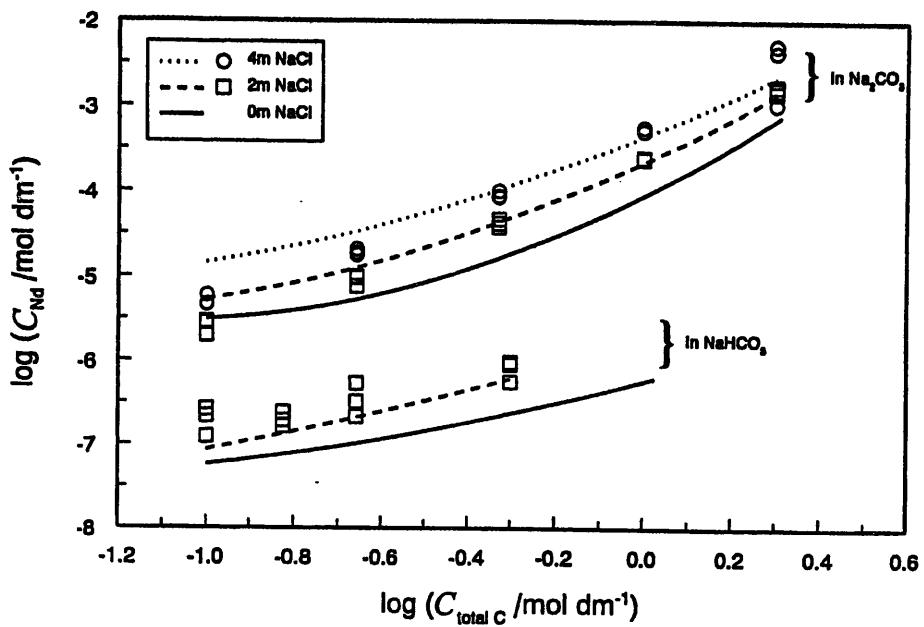


Figure 14. Solubility of $\text{NaNd}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}$ (c) in NaHCO_3 and Na_2CO_3 Solutions, in the Absence (solid lines) and Presence of NaCl (Rao et al. 1996c)

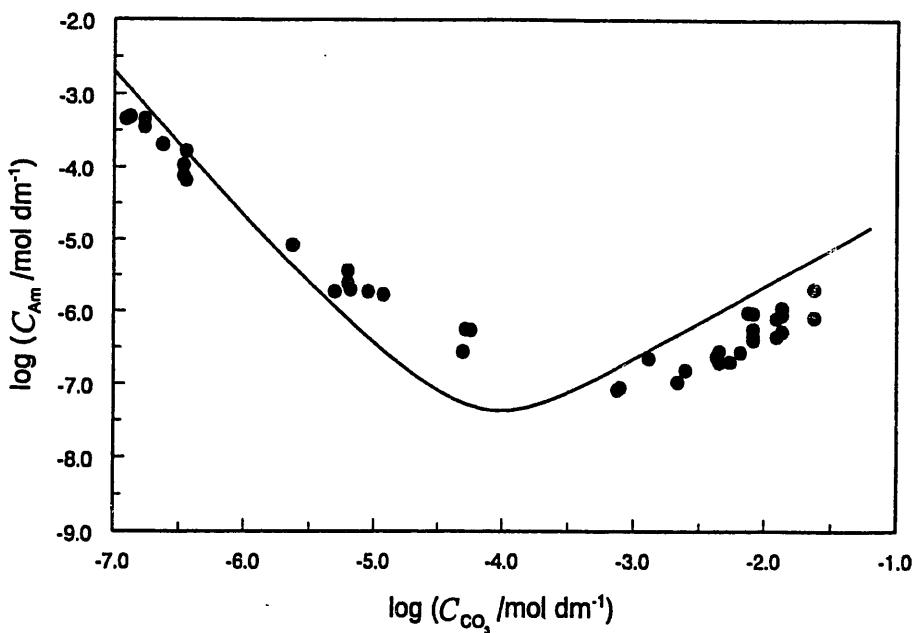


Figure 15. Solubility of $\text{NaAm}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}$ (s) as a Function of the Concentration of Carbonate (data points from Runde and Kim 1995, the solid line based on predictions with modeling parameters discussed in the text)

This model has recently been shown to be applicable to another trivalent rare earth system (Weger et al. 1998). The predictions by this model were completely consistent with the measured solubility of $\text{Gd}_2(\text{CO}_3)_3(\text{c})$ in K_2CO_3 and of $\text{NaGd}(\text{CO}_3)_2 \cdot 6\text{H}_2\text{O}(\text{c})$ in Na_2CO_3 , (Figure 16). Since the shape of the solubility curve is dependent on the aqueous phase model, the close agreement of the predicted solubility curve to the solubility data suggests that the aqueous phase model is applicable to the Gd-carbonate system. As further proof of the reliability of the aqueous phase model, the EXAFS analyses of the Gd solutions in equilibrium with $\text{Gd}_2(\text{CO}_3)_3(\text{c})$ in relatively concentrated carbonate solutions showed the presence of $\text{Gd}(\text{CO}_3)_3^{3-}$, in agreement with the model predictions (Weger et al. 1998).

Molybdate

Based on thermodynamic analysis of solubility measurements of $\text{NdPO}_4(\text{c})$ as a function of pH and molybdate, and of $\text{NaNd}(\text{MoO}_4)_2(\text{c})$ and $\text{Nd}_2(\text{MoO}_4)_3 \cdot x\text{H}_2\text{O}$ as a function of NdCl_3 , Felmy et al. (1995) determined the solubility products of $\text{NaNd}(\text{MoO}_4)_2(\text{c})$ and $\text{Nd}_2(\text{MoO}_4)_3 \cdot x\text{H}_2\text{O}$ and the equilibrium constants of Nd-molybdate complexes. The predictions based on these constants agreed closely with the extensive experimental data not only for the solubility of molybdate solids but also with the solubility of $\text{NdPO}_4(\text{c})$ in molybdate solutions (Figure 17). No other reliable data for these reactions are available. The thermodynamic data selected for the trivalent model are based on this study.

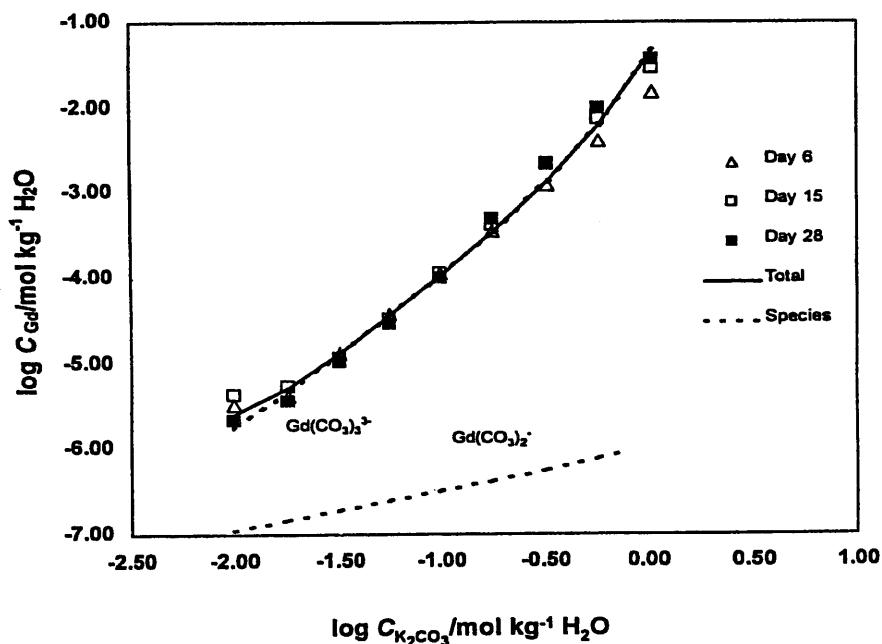


Figure 16. Measured Aqueous Gd Concentrations in Equilibrium with $\text{Gd}_2(\text{CO}_3)_3(\text{c})$. Predicted concentrations of major Gd-carbonate aqueous species are reported. EXAFS analysis confirmed that $\text{Gd}(\text{CO}_3)_3^{3-}$ was the dominant aqueous species.

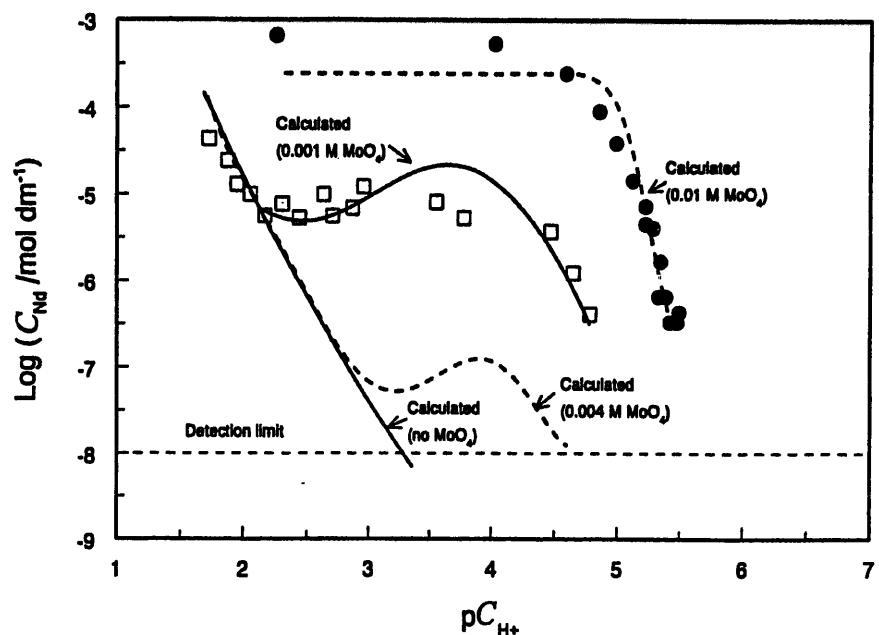


Figure 17. Solubility of NdPO₄(c) as Functions of pC_{H+} and Molybdate Concentrations at Fixed [phosphate]_{total} of 10^{-3.5} M (Felmy et al. 1995)

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Appendix

Compilation of Thermodynamic Data for Trivalent Actinides

(In selecting the data for this compilation, primary emphasis was placed on the literature sources where the original experimental data are reported.)

Contents

<p>List of Abbreviations Used for Methods</p> <p>A1 Samarium(III)</p> <p>Table A1.1 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with F^- ($\text{Sm}^{3+} + n\text{F}^- \rightleftharpoons \text{SmF}_n^{(3-n)+}$)</p> <p>Table A1.2 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with Cl^- ($\text{Sm}^{3+} + n\text{Cl}^- \rightleftharpoons \text{SmCl}_n^{(3-n)+}$)</p> <p>Table A1.3 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with NO_3^- ($\text{Sm}^{3+} + n\text{NO}_3^- \rightleftharpoons \text{SmNO}_3^{(3-n)+}$)</p> <p>Table A1.4 Equilibrium Constants (β_{mn}) for Formation of Aqueous Sm^{3+} Complexes with OH^- ($m\text{Sm}^{3+} + n\text{H}_2\text{O} \rightleftharpoons \text{Sm}_m\text{OH}_n^{(3m-n)+} + n\text{H}^+$)</p> <p>Table A1.5 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with CO_3^{2-} ($\text{Sm}^{3+} + n\text{CO}_3^{2-} \rightleftharpoons \text{Sm}(\text{CO}_3)_n^{(3-2n)+}$)</p> <p>Table A1.6 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with HCO_3^- ($m\text{Sm}^{3+} + n\text{HCO}_3^- \rightleftharpoons \text{Sm}_m(\text{HCO}_3)_n^{(3m-n)+}$)</p> <p>Table A1.7 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with HPO_4^{2-} ($m\text{Sm}^{3+} + n\text{HPO}_4^{2-} \rightleftharpoons \text{Sm}_m(\text{HPO}_4)_n^{(3m-2n)+}$)</p> <p>Table A1.8 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with H_2PO_4^- ($m\text{Sm}^{3+} + n\text{H}_2\text{PO}_4^- \rightleftharpoons \text{Sm}_m(\text{H}_2\text{PO}_4)_n^{(3m-n)+}$)</p> <p>Table A1.9 Equilibrium Constants (β_n) for Formation of Aqueous Sm^{3+} Complexes with SO_4^{2-} ($\text{Sm}^{3+} + n\text{SO}_4^{2-} \rightleftharpoons \text{Sm}(\text{SO}_4)_n^{(3-2n)+}$)</p> <p>Table A1.10 Solubility Products (K_{sp}) of the Different Sm^{3+} Solid Phases</p> <p>Table A1.11 Binary Interaction Parameters</p> <p>A2 Actinium(III)</p> <p>Table A2.1 Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with F^- ($\text{Ac}^{3+} + n\text{F}^- \rightleftharpoons \text{AcF}_n^{(3-n)+}$)</p> <p>Table A2.2 Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with Cl^- ($\text{Ac}^{3+} + n\text{Cl}^- \rightleftharpoons \text{AcCl}_n^{(3-n)+}$)</p> <p>Table A2.3 Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with NO_3^- ($\text{Ac}^{3+} + n\text{NO}_3^- \rightleftharpoons \text{AcNO}_3^{(3-n)+}$)</p> <p>Table A2.4 Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with OH^- ($m\text{Ac}^{3+} + n\text{H}_2\text{O} \rightleftharpoons \text{Ac}_m\text{OH}_n^{(3m-n)+} + n\text{H}^+$)</p> <p>Table A2.5 Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with H_2PO_4^- ($\text{Ac}^{3+} + n\text{H}_2\text{PO}_4^- \rightleftharpoons \text{Ac}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)</p>	<p>A.5</p> <p>A.6</p> <p>A.6</p> <p>A.7</p> <p>A.7</p> <p>A.7</p> <p>A.8</p> <p>A.8</p> <p>A.8</p> <p>A.9</p> <p>A.9</p> <p>A.9</p> <p>A.9</p> <p>A.9</p> <p>A.10</p> <p>A.11</p> <p>A.12</p> <p>A.13</p> <p>A.13</p> <p>A.13</p> <p>A.14</p> <p>A.14</p> <p>A.14</p>
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Table A2.6	Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with SO_4^{2-} ($\text{Ac}^{3+} + n\text{SO}_4^{2-} \rightleftharpoons \text{Ac}(\text{SO}_4)_n^{(3-2n)+}$)	A.15
Table A2.7	Solubility Products (K_{sp}) of the Different Ac^{3+} Solid Phases	A.15
A3	Neptunium(III)	A.16
Table A3.1	Equilibrium Constants (B_n) for Formation of Aqueous Np^{3+} Complexes with Cl^- ($\text{Np}^{3+} + n\text{Cl}^- \rightleftharpoons \text{NpCl}_n^{(3-n)+}$)	A.16
Table A3.2	Equilibrium Constants (β_{mn}) for Formation of Aqueous Np^{3+} Complexes with OH^- ($m\text{Np}^{3+} + n\text{OH}^- \rightleftharpoons \text{Np}_m\text{OH}_n^{(3m-n)+}$)	A.16
Table A3.3	Equilibrium Constants (β_{mn}) for Formation of Aqueous Np^{3+} Complexes with OH^- ($m\text{Np}^{3+} + n\text{H}_2\text{O} \rightleftharpoons \text{Np}_m\text{OH}_n^{(3m-n)+} + n\text{H}^+$)	A.17
Table A3.4	Equilibrium Constants (B_n) for Formation of Aqueous Np^{3+} Complexes with CO_3^{2-} ($m\text{Np}^{3+} + n\text{CO}_3^{2-} \rightleftharpoons \text{Np}(\text{CO}_3)_n^{(3-2n)+}$)	A.17
Table A3.5	Equilibrium Constants (B_n) for Formation of Aqueous Np^{3+} Complexes with H_2PO_4^- ($\text{Np}^{3+} + n\text{H}_2\text{PO}_4^- \rightleftharpoons \text{Np}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)	A.17
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List of Abbreviations Used for Methods

Abbreviation	Method
cal	calorimetry
calc	calculated
chr	chromatography
cix	cation exchange
col	colorimetry
dis	distribution between two phases
emf	electromotive force, not specified
est	estimated
gl	glass electrode
ise-X	ion selective electrode with ion X stated
ix	ion exchange
nmr	nuclear magnetic resonance
pot	potentiometry
recal	recalculation using data from another paper
sol	solubility
sp	spectroscopy
tp	electrical migration or transference number

A1. Samarium(III)

Table A1.1. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with F⁻ ($\text{Sm}^{3+} + n\text{F}^- \rightleftharpoons \text{SmF}_n^{(3-n)+}$)

log β_1	log β_2	log β_3	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
4.2			H ₂ O	n.a.	est	[78ALL/BEA]
4.17			H ₂ O	n.a.	est	[90WOO]
3.58			H ₂ O	n.a.	est	[92MIL]
3.15	6.95		0.5 M NH ₄ NO ₃	M:1.5mM, H: ^(a) , L:1mM	pot,ise- F ⁻	[89MEN/JAM]
3.12			1 M NaClO ₄	M:0.01M, H:0.1M, L:2mM	pot	[67WAL/CHO]
3.12			I = 1 M	n.a.	est	[80BON/HEF]
3.09	4.56		1 M NaCl	M:0.4mM, H:0.25mM, L:1mM	pot,ise- F ⁻	[85BEC/BIL]
3.23	5.81	10.54	1 M (H,Na)ClO ₄	M:1mM, H:1mM, L:0.4mM	pot,ise- F ⁻	[93SAW/MAH]

(a) pH not given in the paper.

Table A1.2. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with Cl⁻ ($\text{Sm}^{3+} + n\text{Cl}^- \rightleftharpoons \text{SmCl}_n^{(3-n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
0.36		H ₂ O	n.a.	est	[90WOO]
0.30		H ₂ O	n.a.	est	[92MIL]
0.09 ^(a)		1 M LiCl	M:tr, pH=1.5, L:1M	dis	[75MOU/HUS]
0.13		1 M LiCl	M:tr, pH=1.5, L:1M	dis	[75MOU/HUS]
0.29 ^(b)		1 M LiCl	M:tr, pH=1.5, L:1M	dis	[75MOU/HUS]
-0.02		3 M Li(Cl,ClO ₄)	M:0.4M, H: ^(c) , L:2.5M	sp	[71KOZ/BAT]
-0.39 -0.60		3 M Li(Cl,ClO ₄)	M:tr, H:0.15M, L:3M	dis	[82FUK/KAW]

(a) T=323 K.

(b) T=273 K.

(c) Limited experimental details given.

Table A1.3. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with NO₃⁻ ($\text{Sm}^{3+} + n\text{NO}_3^- \rightleftharpoons \text{SmNO}_3^{(3-n)+}$)

log β_1	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
1.2	H ₂ O	n.a.	est	[78ALL/BEA]
0.78	H ₂ O	n.a.	est	[92MIL]
1.06 ^(a)	1 M LiNO ₃	M:tr, pH=1.5, L:1M	dis	[75MOU/HUS]
1.25	1 M LiNO ₃	M:tr, pH=1.5, L:1M	dis	[75MOU/HUS]
1.50 ^(b)	1 M LiNO ₃	M:tr, pH=1.5, L:1M	dis	[75MOU/HUS]

(a) T=323 K.

(b) T=273 K.

Table A1.4. Equilibrium Constants (β_{mn})^(a) for Formation of Aqueous Sm³⁺ Complexes with OH⁻ ($mSm^{3+} + nH_2O \leftrightarrow Sm_mOH_n^{(3m-n)+} + nH^+$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Other $\log \beta_{mn}$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
-7.9					H ₂ O	n.a.	est	[81BAE/MES]
-7.9					H ₂ O	n.a.	est	[90WOO]
-8.19					H ₂ O	n.a.	est	[92MIL]
-4.4					0.1 M (H,Li)ClO ₄	M:tr, pH=3	dis	[71GUI/DES]
-7.2	-21.7	-24.9			0.1 M NaClO ₄	M:1mM, pH=5	sol	[95SHI/SHI]
-8.84	-15.73				1 M NaClO ₄	n.a. ^(b)	gl	[82NAI/CHA]
-7.5	-15.0	-22.7		$\log \beta_{(4,3)} = -19.5$	1 M NaClO ₄	M:0.1M, pH=7	sol	[79KRA/CHA]
				-36.7	1 M NaClO ₄	M:0.1M, pH=7	sol	[83KRA/CHA]

(a) If $m = 1$, β_{mn} is written β_n .

(b) Limited experimental details given.

Table A1.5. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with CO₃²⁻ ($Sm^{3+} + nCO_3^{2-} \leftrightarrow Sm(CO_3)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
7.3	12.11	H ₂ O	n.a.	est	[92MIL]
8.6	13.4	0.1 M NaClO ₄	M:tr, pH=5, L:1mM	sol	[95SHI/MAK]

Table A1.6. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with HCO₃⁻ ($m\text{Sm}^{3+} + n\text{HCO}_3^- \leftrightarrow \text{Sm}_m(\text{HCO}_3)_n^{(3m-n)^+}$)

log β_1	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
1.75	H ₂ O	n.a.	est	[92MIL]

Table A1.7. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with HPO₄²⁻ ($m\text{Sm}^{3+} + n\text{HPO}_4^{2-} \leftrightarrow \text{Sm}_m(\text{HPO}_4)_n^{(3m-2n)^+}$)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
5.35	8.96	H ₂ O	n.a.	est	[92MIL]

Table A1.8. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with H₂PO₄⁻ ($m\text{Sm}^{3+} + n\text{H}_2\text{PO}_4^- \leftrightarrow \text{Sm}_m(\text{H}_2\text{PO}_4)_n^{(3m-n)^+}$)

log β_1	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
2.23	H ₂ O	n.a.	est	[92MIL]

Table A1.9. Equilibrium Constants (β_n) for Formation of Aqueous Sm³⁺ Complexes with SO₄²⁻ (Sm³⁺ + nSO₄²⁻ ⇌ Sm(SO₄)_n⁽³⁻²ⁿ⁾⁺)

log β ₁	log β ₂	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
3.52	5.19	H ₂ O	n.a. ^(a)	cal	[69IZA/EAT]
3.67	5.20	H ₂ O	n.a.	est	[78ALL/BEA]
3.28		H ₂ O	n.a.	est	[92MIL]
1.30	1.91	2 M NaClO ₄	M:0.1M, pH=3, L:0.7M	pot	[67CAR/CHO]
4.34	var		n.a. ^(a)	sol	[54SPE/JAF]

(a) Limited experimental details given.

Table A1.10. Solubility Products (K_{sp}) of the Different Sm^{3+} Solid Phases

Reaction	Log K_{sp}	Medium	Concentration (Molarity) of metal, ligand, H	Method	References
$\text{Sm(OH)}_3(\text{s}) \rightleftharpoons \text{Sm}^{3+} + 3\text{OH}^-$	-25.2	0	m:4.3e-5, pH:7.1	col	[62KOV/BAG]
	-25.9	0 (0.02-4 M NaCl) ^(e)	M:0.3, pH:7.8	sol, gl	[60AKS/SPI]
	-25.20	0 (0.1 M LiCl) ^(e)	M:6E-4, pH:7	pol, gl	[66KOV/AZH]
	-23.7	SmCl_3 in H_2O	M:0.01, pH:6.9	pH	[55KOR]
	-22.08	0.1 M HClO_4	M:n.a., pH:7.2	gl	[51MOE/FOG]
	-21.2	0.1 M $\text{Sm(NO}_3)_3$, SmCl_3 or $\text{Sm(C}_2\text{H}_3\text{O}_2)_3$	M:0.1, OH:0.3	gl	[44MOE/KRE]
$\text{Sm(OH)}_3(\text{s}) + 3\text{H}^+ \rightleftharpoons \text{Sm}^{3+} + 3\text{H}_2\text{O}$	16.5	0	n.a.	est	[76BAE/MES]
	22.12	HClO_4 + NaOH ^(a)	M:0.09, pH:n.a.	gl	[59MEL/VRÁ]
	21.3	HClO_4 + NaOH ^(b)	M:0.09, pH:n.a.	gl	[59MEL/VRÁ]
	20.6	HClO_4 + NaOH ^(c)	M:0.09, pH:n.a.	gl	[59MEL/VRÁ]
	19.9	HClO_4 + NaOH ^(d)	M:0.09, pH:n.a.	gl	[59MEL/VRÁ]
$\text{Sm(OH)}_2\text{Cl}(\text{s}) \rightleftharpoons \text{Sm}^{3+} + 2\text{OH}^- + \text{Cl}^-$	-17.3	0 (0.02-4 M NaCl) ^(e)	M:0.3, pH:7.8	sol, gl	[60AKS/SPI]
$\text{SmOHCO}_3 \cdot 0.5\text{H}_2\text{O} \rightleftharpoons \text{Sm}^{3+} + \text{OH}^- + \text{CO}_3^{2-} + 0.5\text{H}_2\text{O}$	-21.0	0 (1 M HCl) ^(e)	M:0.012,L:0.012,H:1	cal	[96MER/FUG]
$\text{Sm}_2(\text{CO}_3)_3(\text{s}) \rightleftharpoons 2\text{Sm}^{3+} + 3\text{CO}_3^{2-}$	-32.5	0 (0.1 M NaClO_4) ^(e)	M:0.001, pH:8	sol, gl	[66JOR/HAV]
	-34.41	0.01 M HClO_4	M:n.a.,L:n.a.,pH:5.7	pot, sp	[86FIR/MOH]

(a) T = 283 K.

(b) T = 293 K.

(c) T = 303 K.

(d) T = 313 K.

(e) Thermodynamic value calculated using data from medium listed in parenthesis.

Table A1.11. Binary Interaction Parameters

Cation	Anion	β^0	β^1	β^2	C^ϕ	Reference
Sm^{3+}	Cl^-	0.6	5.6		-0.0204	[91PIT]
Sm^{3+}	ClO_4^-	0.76	6.53		0.0058	[91PIT]
Sm^{3+}	NO_3^-	0.467	5.13		-0.0504	[91PIT]

A2. Actinium(III)

Table A2.1. Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with f ($\text{Ac}^{3+} + n \text{F}^- \rightleftharpoons \text{AcF}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
3.6	6.6	9.8	0	n.a.	est	[78ALL/BEA]
2.95			0.1 M NaClO ₄	M:tr, H:0.016M, L:0.01M	tp	[73MAK/STE]
2.72	5.22	7.90	0.5 M NaClO ₄	M:tr, pH=3.6, L:4e-3M	dis	[70AZI/LYL]
2.69	5.18	7.78	0.5	n.a.	est	[80BON/HEF]
2.7	5.2	7.9	0.5	n.a.	est	[92FUG/KHO]

Table A2.2. Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with Cl⁻ ($\text{Ac}^{3+} + n\text{Cl}^- \rightleftharpoons \text{AcCl}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
1.1	0	n.a.		est	[78ALL/BEA]
1.3	0	n.a.		est	[92FUG/KHO]
-0.10	-0.82	1 M H(ClO ₄ ,Cl)	M:tr, H:1M, L:1M	dis	[68SHA/MAT]
-0.1	-0.8	1	n.a.	est	[92FUG/KOH]
0.27		1 M H(ClO ₄ ,Cl)	M:tr, H:1M, L:0.5M	dis	[91SZE/KUB]
-0.36	-0.51	3 M Li(Cl,ClO ₄)	M:tr, H:0.15M, L:3M	dis	[82FUK/KAW]
-0.05	-1.0	4 M NaClO ₄	M:tr, H:0.01M, L: ^(a)	dis	[69SEK/SAK]
-0.05	-1	4	n.a.	est	[92FUG/KHO]

(a) No or limited experimental details.

Table A2.3. Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with NO_3^- ($\text{Ac}^{3+} + n\text{NO}_3^- \rightleftharpoons \text{AcNO}_3^{(3-n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
0.12	0.01	1 M H(ClO ₄ ,NO ₃)	M:tr, H:1M, L:1M	dis	[68SHA/MAT]
0.57		1 M H(ClO ₄ ,NO ₃)	M:tr, H:1M, L:0.5M	dis	[91SZE/KUB]
0.1	1	n.a.		est	[92FUG/KHO]

Table A2.4. Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with OH⁻ ($m\text{Ac}^{3+} + n\text{H}_2\text{O} \rightleftharpoons \text{Ac}_m\text{OH}_n^{(3m-n)+} + n\text{H}^+$)

log β_1	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
<-10.4	H ₂ O	n/a	est	[76BAE/MES]

Table A2.5. Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with H₂PO₄⁻ ($\text{Ac}^{3+} + n\text{H}_2\text{PO}_4^- \rightleftharpoons \text{Ac}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)

log β_1	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
2.5	0	n.a.	est	[92FUG/KHO]
1.59	0.5 M NH ₄ ClO ₄	M:tr, pH=2, L:0.14M	dis	[70RAO/SHA]

Table A2.6. Equilibrium Constants (B_n) for Formation of Aqueous Ac^{3+} Complexes with SO_4^{2-} ($\text{Ac}^{3+} + n\text{SO}_4^{2-} \rightleftharpoons \text{Ac}(\text{SO}_4)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
3.6	5.5	0	n.a.	est	[78ALL/BEA]
2.47		0.1 M NH_4Cl	M:tr, H^{2+} , L: $9\text{e-}4$	tp	[73STE]
1.75	2.64	0.5 M NaClO_4	M:tr, pH=3.6, H:0.12M	dis	[70AZI/LYL]
1.36	2.68	1 M NaClO_4	M:tr, H: $1\text{e-}3$ M, L: ^(a)	dis	[69SEK/SAK]
1.20	1.85	1 M $\text{H}(\text{ClO}_4, \text{HSO}_4)$	M:tr, H:1M, L:0.16M	dis	[68SHA/MAT]

(a) Limited experimental details given.

Table A2.7. Solubility Products (K_{sp}) of the Different Ac^{3+} Solid Phases

Reaction	Log K_{sp}	Medium	Maximum Metal/ Ligand/ H Concentration	Method	References
$\text{Ac(OH)}_3(\text{s,freash}) \rightleftharpoons \text{Ac}^{3+} + 3\text{OH}^-$	-18.68	0 (0-0.01 M NH_4NO_3) ^(a)	M:0.32, pH:10.4	sol	[65ZIV/SHE]
$\text{Ac(OH)}_3(\text{s,aged}) \rightleftharpoons \text{Ac}^{3+} + 3\text{OH}^-$	-20.89	0 (0-0.01 M NH_4NO_3) ^(a)	M:0.32, pH:10.4	sol	[65ZIV/SHE]

(a) Thermodynamic value calculated using data from medium listed in parenthesis.

A3. Neptunium(III)

Table A3.1. Equilibrium Constants (B_n) for Formation of Aqueous Np^{3+} Complexes with Cl^- ($\text{Np}^{3+} + n\text{Cl}^- \rightleftharpoons \text{NpCl}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
-2.39	-2.89	>7 M LiCl	M: ^(a) , H:<7M, L:<7M	sp	[64SHI/MAR]

(a) Limited experimental details given.

Table A3.2. Equilibrium Constants (β_{mn})^(a) for Formation of Aqueous Np^{3+} Complexes with OH^- ($m\text{Np}^{3+} + n\text{OH}^- \rightleftharpoons \text{Np}_m\text{OH}_n^{(3m-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Other $\log \beta_{mn}$	Medium	Maximum Metal/ H^+ / Ligand Concentration	Method	Reference
7	13	19	23	2,2=15	H_2O	n/a	est	[78ALL/BEA]
6.6	11	15	18	2,2=15	H_2O	n/a	est	[80ALL/KIP]

(a) If $m = 1$, $\beta_{1,x}$ is written β_x .

Table A3.3. Equilibrium Constants (β_{mn})^(a) for Formation of Aqueous Np³⁺ Complexes with OH⁻ ($m\text{Np}^{3+} + n\text{H}_2\text{O} \rightleftharpoons \text{Np}_m\text{OH}_{n-(3m-n)}^{(3m-n)+} + n\text{H}^+$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Other $\log \beta_{mn}$	Medium	Maximum Metal/H ⁺ / Ligand Concentration	Method	Reference
-7	-15	-25	-35	2,2=-14	H ₂ O	n/a	est	[86WAN]
-7.43					I=0.3	n/a	est	[80FUG/KHO]
-7.43					n/a	n/a	est	[74MED/KRO]

(a) If $m = 1$, $\beta_{1,x}$ is written β_x .

Table A3.4. Equilibrium Constants (B_n) for Formation of Aqueous Np³⁺ Complexes with CO₃²⁻ ($m\text{Np}^{3+} + n\text{CO}_3^{2-} \rightleftharpoons \text{Np}(\text{CO}_3)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
9.6	12.9	16.2	H ₂ O	n.a.	est	[86WAN]
7.4	12.3		H ₂ O	n.a.	calc	[88CAN]

Table A3.5. Equilibrium Constants (B_n) for Formation of Aqueous Np³⁺ Complexes with H₂PO₄⁻ ($\text{Np}^{3+} + n\text{H}_2\text{PO}_4^- \rightleftharpoons \text{Np}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
2.40	3.73	5.64	H ₂ O	n.a. ^(a)	ix	[69MOS]
2.5			H ₂ O	n.a.	est	[92FUG/KHO]

(a) Limited experimental details given.

A4. Plutonium(III)

Table A4.1. Equilibrium Constants (B_n) for Formation of Aqueous Pu^{3+} Complexes with F^- ($\text{Pu}^{3+} + n\text{F}^- \rightleftharpoons \text{PuF}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
3.3			H_2O	n.a.	est	[83FUG2]
4.3	7.6	10.8	H_2O	n.a.	est	[86WAN]
3.58	6.40	12.61	1 M $(\text{H},\text{Na})\text{ClO}_4$	M:4e-3M:, H:6e-2M, L:5e-3M	pot, ise- F^-	[93SAW/MAH]

Table A4.2. Equilibrium Constants (B_n) for Formation of Aqueous Pu^{3+} Complexes with Cl^- ($\text{Pu}^{3+} + n\text{Cl}^- \rightleftharpoons \text{PuCl}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
1.1		H_2O	n.a.	est	[78ALL/BEA]
-0.1		H_2O	n.a.	est	[82JEN]
1.2		H_2O	n.a.	est	[83FUG2]
-1.9		H_2O	n.a.	est	[85COW/JEN]
0.35		0.2 M HCl	M:tr, H:0.2M, L:0.2M	cix	[56WAR/WEL]
0.24		0.5 M HCl	M:tr, H:0.5M, L:0.5M	cix	[56WAR/WEL]
0.25		0.5 M HCl/HClO_4	M:tr, H:0.5M, L:0.5M	cix	[56WAR/WEL]
0.1		I = 0.5 M	n.a.	est	[92FUG/KHO]
-0.15		1 M HClO_4	n.a.	est	[53CON/MCV]
-0.04		1 M HCl	M:tr, H:1M, L:1M	cix	[56WAR/WEL]
-0.1		I = 1 M	n.a.	est	[76SMI/MAR]
0.5		I = 1 M	n.a.	est	[92FUG/KHO]
-2.4	-5	3-13 M LiCl	M:tr, H:0.5M, L:13M	sp	[66SHI/MAR]

Table A4.3. Equilibrium Constants (B_n) for Formation of Aqueous Pu^{3+} Complexes with NO_3^- ($\text{Pu}^{3+} + n\text{NO}_3^- \rightleftharpoons \text{PuNO}_3^{(3-n)+}$)

log β_1	log β_2	log β_3	Medium	Maximum Metal/H^+/Ligand Concentration	Method	Reference
1.41	2.22	2.52	H_2O	n.a.	est	[85COW/JEN]
0.77			1 M HClO_4	n.a. ^(a)	dis	[59SHE/TIM]
1.18	0.07	-0.72	8 M HClO_4	M:tr, H:8M, L:3M	dis	[70LAH/KNO]

(a) Limited experimental details given.

Table A4.4. Equilibrium Constants (β_{mn})^(a) for Formation of Aqueous Pu³⁺ Complexes with OH⁻ ($mPu^{3+} + nOH^- \leftrightarrow Pu_mOH_n^{(3m-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Other $\log \beta_{mn}$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
7.0					H ₂ O	n/a	est	[76SMI/MAR]
7.2	14	19	24	2,2=16	H ₂ O	n/a	est	[78ALL/BEA]
7.2	12.1	16.7	20.2	2,2=16	H ₂ O	n/a	est	[80ALL/KIP]
6.7					H ₂ O	n/a	est	[82JEN]
6.5	11.5	15.5	19	2,2=14	H ₂ O	n/a	est	[83ALL]

(a) If $m = 1$, $\beta_{1,x}$ is written β_x .

Table A4.5. Equilibrium Constants (B_{mn})^(a) for Formation of Aqueous Pu³⁺ Complexes with OH⁻ ($Pu^{3+} + nH_2O \leftrightarrow PuOH_n^{(3-n)+} + nH^+$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Other $\log \beta_{mn}$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
-5.54					H ₂ O	n.a. ^(b)	gl	[49KRA/DAM]
-8					H ₂ O	n.a.	est	[80LEM/TRE]
-7.5	-16.5	-26.5	-37	2,2=-14	H ₂ O	n.a.	est	[86WAN]
-7.37					I = 0.024 M	n.a.	est	[80FUG/KHO]
-7.22					I = 0.069	n.a.	est	[80FUG/KHO]
-3.8					0.1 M (H,Li)ClO ₄	M:tr, pH=2.5	dis	[76HUB/HUS]
-3.8					0.2 M LiClO ₄	M:1e-3M, pH=2.5	dis	[75HUB/HUS]
-3.8					I = 0.2 M	n.a.	est	[80FUG/KHO]

(a) If $m = 1$, $\beta_{1,x}$ is written β_x .

(b) Limited experimental details given.

Table A4.6. Equilibrium Constants (B_n) for Formation of Aqueous Pu^{3+} Complexes with CO_3^{2-} ($Pu^{3+} + nCO_3^{2-} \leftrightarrow Pu(CO_3)_n^{(3-2n)+}$)

log β_1	log β_2	log β_3	Medium	Maximum Metal/H^+/Ligand Concentration	Method	Reference
9.6	12.9	16.2	H_2O	n.a.	est	[82JEN]
6	10	13	H_2O	n.a.	est	[83ALL]
6	10	13	H_2O	n.a.	est	[86WAN]
7.5	12.4		H_2O	n.a.	calc	[88CAN]

Table A4.7. Equilibrium Constants (B_n) for Formation of Aqueous Pu^{3+} Complexes with PO_4^{3-} ($Pu^{3+} + nPO_4^{3-} \leftrightarrow Pu(PO_4)_n^{(3-3n)+}$)

log β_1	Medium	Maximum Metal/H^+/Ligand Concentration	Method	Reference
22	H_2O	n.a. ^(a)	ix	[69MOS]
19.3	1 M NH_4Cl	M:0.015M, H:>0.5M, L:0.25M	sol	[71MOS2]

(a) Limited experimental details given.

Table A4.8. Equilibrium Constants (β_n) for Formation of Aqueous Pu^{3+} Complexes with HPO_4^{2-} ($mPu^{3+} + nHPO_4^{2-} \leftrightarrow Pu_m(HPO_4)_n^{(3m-2n)+}$)

log β_1	Medium	Maximum Metal/H^+/Ligand Concentration	Method	Reference
9.7	H_2O	n/a	est	[80LEM/TRE]

Table A4.9. Equilibrium Constants (B_n) for Formation of Aqueous Pu^{3+} Complexes with H_2PO_4^- ($\text{Pu}^{3+} + n\text{H}_2\text{PO}_4^- \rightleftharpoons \text{Pu}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
2.39	3.7	5.63	6.2	H_2O	n.a. ^(a)	ix	[69MOS]
1.48	2.2	2.9	3.5	1 M NH_4Cl	M:0.015M, H:>0.5M, L:0.25M	sol	[71MOS2]

(a) Limited experimental details given.

Table A4.10. Equilibrium Constants (β_n) for Formation of Aqueous Pu^{3+} Complexes with PO_4^{3-} ($\text{Pu}^{3+} + n\text{PO}_4^{3-} + 2n\text{H}^+ \rightleftharpoons \text{Pu}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
21.64	42.31	H_2O	n/a	est	[85COW/JEN]
22		H_2O	n/a	est	[86WAN]

Table A4.11. Equilibrium Constants (β_n) for Formation of Aqueous Pu^{3+} Complexes with PO_4^{3-} ($\text{Pu}^{3+} + n\text{PO}_4^{3-} + n\text{H}^+ \rightleftharpoons \text{Pu}(\text{HPO}_4)_n^{(3-2n)+}$)

$\log \beta_1$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
18.3	H_2O	n/a	est	[86WAN]

Table A4.12. Equilibrium Constants (β_n) for Formation of Aqueous Pu³⁺ Complexes with SO₄²⁻ ($\text{Pu}^{3+} + n\text{SO}_4^{2-} \rightleftharpoons \text{Pu}(\text{SO}_4)_n^{(3-2n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
3.7		H ₂ O	n.a.	est	[78ALL/BEA]
3.5		H ₂ O	n.a.	est	[80LEM/TRE]0
1.26			n.a.	est	[82JEN]
3.61	5.47	H ₂ O	n.a.	est	[82COW/JEN]
3.5	5.2	H ₂ O	n.a.	est	[86WAN]
4.5	6.7	H ₂ O	n.a.	est	[92FUG/KHO]
1.26		1 M HClO ₄	M:tr, H:1M, L:0.17M	cix	[67NAI/RAO]
1.99	3.04	1 M HClO ₄	M:tr, H:1M, L:0.7M	dis	[78RAO/BAG1]
1.90	3.18	1 M HClO ₄	M:tr, H:1M, L:0.17M	cix	[67NAI/RAO]
1.73	3.39	1 M HClO ₄	M:tr, H:1M, L:0.9M	cix	[76FAR/BUC]
1.26		I = 1 M	n.a.	est	[79CLE]
1.65	3.29	2 M HClO ₄	M:tr, H:2M, L:0.9M	cix	[76FAR/BUC]
1.89	2.90	2 M HClO ₄	M:tr, H:2M, L:0.7M	dis	[78RAO/BAG1]
1	1.18	2 M (Na,H)Cl	M:5e-3M, H:0.3M, L:0.4M	est	[56NEW/BAK]

Table A4.13. Equilibrium Constants (β_n) for Formation of Aqueous Pu³⁺ Complexes with SO₄²⁻ (Pu³⁺ + nHSO₄⁻ ⇌ Pu(SO₄)_n⁽³⁻²ⁿ⁾⁺ + nH⁺)

log β ₁	log β ₂	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
0.34		I = 2 M	n.a.	est	[83NAS/CLE]

Table A4.14. Solubility Products (K_{sp}) of the Different Pu³⁺ Solid Phases

Reaction	Log K _{sp}	Medium	Maximum Metal/ H ⁺ / Ligand Concentration	Method	References
PuF ₃ ⇌ Pu + 3F	-15.6	0.06-2.4 M HNO ₃	M:0.02, L:7.94, H:2.4	sol	[61MAN/FRA]
Pu(OH) ₃ (c) ⇌ Pu ³⁺ + 3OH ⁻	-26.2	H ₂ O	M:0.001, pH:13	sol	[89FEL/RAI]
	-19.7	H ₂ O	n.a.	est	[76BAE/MES]
Pu(OH) _{2.5} Cl _{0.5} (s) ⇌ Pu ³⁺ + 2.5OH ⁻ + 0.5Cl ⁻	-20.0	0.0073 M PuCl ₃ + NaOH	M:7E-3, L:0.02, OH:0.025	pot, gl	[50BUS/COW]
PuPO ₄ •xH ₂ O ⇌ Pu ³⁺ + PO ₄ ³⁻ + xH ₂ O	-24.4	0.5 M NH ₄ ClO ₄	M:0.015, L:n.a., pH:8.9	sol	[71MOS2]

A5. Americium(III)

(For the Americium data, tracer amounts (<1mM) of the metal concentration are assumed unless other notification)

Table A5.1. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with F^- ($\text{Am}^{3+} + n\text{F}^- \leftrightarrow \text{AmF}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
4.3	7.5	10.8	14	H_2O	n.a.	est	[78ALL/BEA]
3.39	6.11	9		H_2O	n.a.	est	[82JEN]
4.3	7.4	10.6		H_2O	n.a.	est	[82PHI]
3.4	5.8			H_2O	n.a.	est	[85SIL/BID]
4.3	7.6	10.8		H_2O	n.a.	est	[86WAN]
3.24				H_2O	n.a.	est	[88PHI/HAL]
3.4	5.8			H_2O	n.a.	est	[82JEN]
3.32				0.1 M NaClO_4	L:0.65mM	emf	[73MAK/STE]
2.59	4.76			0.1 M NaClO_4	L:0.01M	cix	[84NAS/CLE2]
3.39	6.11			0.5 M NaClO_4	L:4mM	dis	[69AZI/LYL]
3.39	6.11	9.0		I = 0.5	n.a.	est	[76SMI/MAR]
2.39				1 M NaClO_4		dis	[69JON/CHO]
2.93				1 M NaClO_4		dis	[75DEG/CHO]
2.39 ^(a)				1 M NaClO_4		dis	[76CHO/UNR]
2.49				1 M NaClO_4		dis	[76CHO/UNR]
2.57 ^(b)				1 M NaClO_4		dis	[76CHO/UNR]
2.71 ^(c)				1 M NaClO_4		dis	[76CHO/UNR]

(a) T=283 K.

(b) T=313 K.

(c) T=328 K.

Table A5.2. Equilibrium Constants (B_n) for Formation of Aqueous Am³⁺ Complexes with Cl⁻ ($\text{Am}^{3+} + n\text{Cl}^- \rightleftharpoons \text{AmCl}_n^{(3-n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
0.013		H ₂ O	M:1.4E-3,H:12.6,L:10.75	sp	[69BAR/MIK]
1	0.5	H ₂ O	n.a.	est	[78ALL/BEA]
-0.1		H ₂ O	n.a.	est	[82JEN]
1.3	1.4	H ₂ O	n.a.	est	[92FUG/KHO]
1.05		H ₂ O	n.a.	est	[95SIL/BID]
0.5		I = 0.1	n.a.	est	[92FUG/KHO]
0.35		0.21 M HCl	H:0.21M, L:0.21M	cix	[56WAR/WEL]
0.24		0.5 M HCl	H:0.5M, L:0.5M	cix	[56WAR/WEL]
0.1		I = 0.5	n.a.	est	[92FUG/KHO]
-0.05		1 M H(ClO ₄ ,Cl)	H:1M,L:1M	dis	[62PEP/MAS]
-0.05		1 M H(ClO ₄ ,Cl)	H:1M,L:1M	cix	[64BAN/PAT]
0.15		1 M Na(ClO ₄ ,Cl)	L:1M	cix	[64BAN/PAT]
-0.6		1 M Na(ClO ₄ /Cl)		dis	[64KHO/SHA]
-0.14	-0.52	1 M H(ClO ₄ ,Cl)	H:1M,L:1M	dis	[71KHO/NAR]
-0.25		1 M Li(ClO ₄ ,Cl)	L:1M	dis	[71KHO/NAR]
0.02	-0.37	1 M Na(ClO ₄ ,Cl)	L:1M	dis	[71KHO/NAR]
0.12	0.03	1 M NH ₄ (ClO ₄ ,Cl)	L:1M	dis	[71KHO/NAR]
	-0.09	1 M NH ₄ (ClO ₄ ,Cl)	L:1M	dis	[80KHO/MAT]
-0.1	-0.5	I = 1 M	n.a.	est	[92FUG/KHO]
-0.66		3 M (Li,H) (ClO ₄ ,Cl)		dis	[82FUK/KAW]

Table A5.2. (contd)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
-0.16	-0.74	4 M H(ClO ₄ ,Cl)	H:4M,L:3.5M	cix	[62GRE]
-0.15	-0.69	4 M Na(ClO ₄ ,Cl)	L:3.3M	dis	[64SEK]
-0.15	-0.7	I = 4 M	n.a.	est	[92FUG/KHO]
0.03	-0.97	var. LiCl	L:4M	nmr	[66VDO/KOL]
1	0.34	var LiCl		tp	[69MAR2]
-2.21	-4.7	var LiCl	L:13.7M	sp	[69MAR/SHI]
-0.26	-0.05	var LiCl	L:4M	cppt	[69VDO/STE]

Table A5.3. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with NO_3^- ($\text{Am}^{3+} + n\text{NO}_3^- \rightleftharpoons \text{AmNO}_3^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
-1	0.5		H_2O	n.a.	est	[78ALL/BEA]
1.5			H_2O	n.a.	est	[92FUG/KHO]
1.33			H_2O	n.a.	est	[95SIL/BID]
0.15	-0.4		1 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:1M,L:1M	cix	[64BAN/PAT]
0.2			1 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:1M,L:1M	cix	[64BAN/PAT]
0.6			1 M NH_4X	L:1M	cix	[60LEB/PIR]
0.26			1 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:1M,L:1M	dis	[62PEP/MAS]
0.25			1 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:1M,L:1M	dis	[65CHO/STR]
0.29			1 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:1M,L:1M	dis	[71KHO/NAR]
0.30			1 M $\text{Li}(\text{ClO}_4, \text{NO}_3)$	L:1M	dis	[71KHO/NAR]
0.26			1 M $\text{Na}(\text{ClO}_4, \text{NO}_3)$	L:1M	dis	[71KHO/NAR]
0.23	1.34		1 MNH ₄ ($\text{ClO}_4, \text{NO}_3$)	L:1M	dis	[71KHO/NAR]
0.25			I = 1 M	n.a.	est	[92FUG/KHO]
0.2			2 MNH ₄ (SCN, NO_3)	L:2M	dis	[73CHI/DAN]
-0.33	-0.77	-1.4	8 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:8M,L:8M	dis	[70LAH/KNO]
-1.3			1-15 M $\text{H}(\text{ClO}_4, \text{NO}_3)$	H:15M,L:15M	sp	[69SHI/GIV]
-1.3			var ($\text{H}, \text{Li}\text{NO}_3$)		sp	[66GIV]

Table A5.4. Equilibrium Constants (B_n) for Formation of Aqueous Am³⁺ Complexes with OH⁻ ($\text{Am}^{3+} + n\text{OH}^- \rightleftharpoons \text{AmOH}_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
8.2	15	21	H ₂ O	n.a.	est	[78ALL/BEA]
7.9			H ₂ O	n.a.	est	[82JEN]
6.5			H ₂ O	n.a.	est	[83ALL]
11.3			0.005 M (H,K)ClO ₄		tp	[69MAR/KIK]
10.7	20.09		0.005 M NH ₄ ClO ₄	M:0.01M,pH5	tp	[72SHA/STE]
7.9			I = 0.1	n.a.	est	[76SMI/MAR]
5.85	12.79	16.63	0.1 M NaClO ₄	M:1E-5,pH:10	sol	[84BER/KIM]
6.44	13.8	17.86	0.1 M NaClO ₄	M:1E-5,pH:10	sp	[84BER/KIM]
6.3	12.2	14.4	0.1 M NaClO ₄	M:5Ci/L,pH13	sol	[88STA/KIM]
5.6	11.6	14.1	0.1 M NaClO ₄	M:5Ci/L,pH13	sol	[88STA/KIM]
7.6	14.6	21.7	3 M NaClO ₄	M:0.15M,pH10	sol	[89PAZ]

Table A5.5. Equilibrium constants (β_n) for Formation of Aqueous Am³⁺ Complexes with OH⁻ ($\text{Am}^{3+} + n\text{H}_2\text{O} \rightleftharpoons \text{AmOH}_n^{(3-n)+} + n\text{H}^+$)

log β_1	log β_2	log β_3	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
-8.2	-17.1	-27	H ₂ O	M:1mM,pH11	sol	[83RAI/STR]
-7.5	-16.5	-26.5	H ₂ O	n.a.	est	[86WAN]
-8	-16.9	-29	H ₂ O	n.a.	est	[92EWA/SMI]
-6.4	-14.1	-25.7	H ₂ O	n.a.	est	[95SIL/BID]
-3.3		0.005 M NH ₄ ClO ₄		n.a.	tp	[72SHA/STE] ^(a)
-5.3		0.1 M (H;Li)ClO ₄	pH:6		dis	[73HUS/HUB]
-2.5	-6.6	0.1 M LiNO ₃			chr, tp	[73KOR2]
-5.3		0.1 M (H;Li)ClO ₄	pH:6		dis	[76HUB/HUS]
-7.7		0.1 M NaClO ₄			sol	[87SIL]
	-16.7	0.1 M NaClO ₄			sol	[84SIL]
-6.9	-23.8	0.1 M XClO ₄			tp	[89ROS/REI]
	-14.76	0.2 M NaClO ₄			dis	[82BID]
-7.5		1 M NaClO ₄	pH:9		dis	[82LUN]
-7.03		1 M NaClO ₄			pot	[82NAI/CHA]
-7.78		1 M NaClO ₄	pcH:7.8		dis	[96CHO/CHE]
-7.83		3 M NaClO ₄	pcH:7.8		dis	[96CHO/CHE]
-8		5 M NaClO ₄	pcH:8.3		dis	[96CHO/CHE]

(a) Information obtained from [80FUG/KHO]

Table A5.6. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with CO_3^{2-} ($\text{Am}^{3+} + n\text{CO}_3^{2-} \rightleftharpoons \text{Am}(\text{CO}_3)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
			11	H_2O	n.a.	est	[78ALL/BEA]
6	10	13		H_2O	n.a.	est	[83ALL]
7.7	12.8			H_2O	n.a.	calc	[88CAN]
8.3				H_2O	n.a.	est	[92FUG/KHO]
6.4	14	13.38		H_2O	n.a.	est	[92EWA/SMI]
7.8	12.3	15.2		H_2O	n.a.	est	[95SIL/BID]
8.16				H_2O	M:0.14mM,L:1.4x10 ⁻⁶ M, pH4.4-6.4	sp	[89NIT/STA]
7.6	15.2	15.2		H_2O	L:0.1M	sol	[90FEL/RAI]
6.69				0.1 M NaClO_4	M:0.14mM,L:1.4x10 ⁻⁶ M, pH4.4-6.4	sp	[89NIT/STA]
6.9				I = 0.1 M	n.a.	est	[92FUG/KHO]
7.7	12.8	12.8		0.1 M NaCl		sol	[93GIF/VIT]
5.08	9.27	12.12		0.1-0.3 M NaClO_4	M:0.01mM,L:0.06M,pH6-10	sol	[84BER/KIM]
5.97	9.58			0.1-0.3 M NaClO_4	M:0.1mM,L:0.05M,pH6-9	sol	[91MEI/KIM]
6.48	9.94			0.1-0.3 M NaClO_4	M0.06mM,0.03atm CO_2 at pH9.7	sp	[91MEI/KIM]
	11.45			0.2 M NaClO_4		dis	[82BID]
5.7	9.4			I = 1 M	n.a.	est	[82ALL]
5.81	9.72			1 M NaClO_4	1atm CO_2 at pH5-7	dis	[82LUN]
5.3	8.5			1 M NaClO_4	L:1M,pH7.8	tp	[82LUN]
5.7				I = 1 M	n.a.	est	[92FUG/KHO]
5.45	8.92	11.44		3 M NaCl		sol	[89ROB]
5.3	11.4	11.4		4 M NaCl		sol	[93GIF/VIT]

Table A5.7. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with HCO_3^- ($\text{Am}^{3+} + n\text{HCO}_3^- \leftrightarrow \text{Am}(\text{HCO}_3)_n^{(3-n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H^+/Ligand Concentration	Method	Reference
4.79	8.15	0.2 M NaClO_4		dis	[82BID]
1.91		0.5 M NaClO_4	L:0.025M,pH7	dis	[88RAO/MAD]
2		0.5 M NaClO_4	L:0.30M,pH7	dis	[88RAO/MAD]
2.13	3.83 ^(a)	0.5 M NaClO_4	L:0.05M,pH7	dis	[88RAO/MAD]
(a) T=308 K.					

Table A5.8. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with HPO_4^{2-} ($\text{Am}^{3+} + n\text{HPO}_4^{2-} \leftrightarrow \text{Am}(\text{HPO}_4)_n^{(3-2n)+}$)

log β_1	Medium	Maximum Metal/H^+/Ligand Concentration	Method	Reference
4.14	H_2O		dis	[86RAO/MAH]
3.63	0.5 M NaClO_4	L:0.025M,pH7	dis	[88RAO/MAH]
3.76	0.5 M NaClO_4	L:0.025M,pH7	dis	[88RAO/MAH]
3.92	0.5 M NaClO_4	L:0.025M,pH7	dis	[86RAO/MAH]

Table A5.9. Equilibrium Constants (β_n) for Formation of Aqueous Am³⁺ Complexes with H₂PO₄⁻ (Am³⁺ + nH₂PO₄⁻ ⇌ Am(H₂PO₄)_n⁽³⁻ⁿ⁾⁺)

log β ₁	log β ₂	log β ₃	log β ₄	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
2.5				H ₂ O	n.a.	est	[78ALL/BEA]
2.39	3.63	5.62	6.3	H ₂ O		cix	[69MOS]
2.5				H ₂ O	n.a.	est	[92FUG/KHO]
3				H ₂ O	n.a.	est	[95SIL/BID]
2.13				H ₂ O	n/a	dis	[86RAO/MAH]
1.97				0.5 M (Na,NH ₄)ClO ₄	L:0.15M,pH2	dis	[86RAO/MAH]
1.64				0.5 M (Na,NH ₄)ClO ₄	L:0.15M,pH2	dis	[88RAO/MAH]
1.69				0.5 M (Na,NH ₄)ClO ₄	L:0.15M,pH2	dis	[88RAO/MAH]
2.36 ^(a)				0.5 M (Na,NH ₄)ClO ₄	L:0.15M,pH2	dis	[88RAO/MAH]
1.69				0.5 M NH ₄ ClO ₄	L:0.1M,pH2	cix	[66BOR/ELE]
1.48	2.1	2.85	3.4	1 M NH ₄ Cl	L:0.72M,pH1.8	cix	[71MOS2]
2.73	3.72			1-13 M H ₃ PO ₄	M:8mM,H:1M,L:13M	sp	[79LEB/FRE2]

(a) T=314.

Table A5.10. Equilibrium Constants (β_n) for Formation of Aqueous Am³⁺ Complexes with PO₄³⁻ (Am³⁺ + nPO₄³⁻ + nH⁺ ⇌ Am(HPO₄)_n⁽³⁻²ⁿ⁾⁺)

log β ₁	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
18.4	H ₂ O	n/a	est	[86WAN]

Table A5.11. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with SO_4^{2-} ($\text{Am}^{3+} + n\text{SO}_4^{2-} \leftrightarrow \text{Am}(\text{SO}_4)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
3.76	5.64	5.29	H_2O	H:0.5mM,L:0.5M	sol	[72MCD/COL]
3.6	5.6		H_2O	n.a.	est	[78ALL/BEA]
1.57	2.66		H_2O	n.a.	est	[82JEN]
3.5	5.2		H_2O	n.a.	est	[86WAN]
4.2	6.1		H_2O	n.a.	est	[92FUG/KHO]
3.85	5.4		H_2O	n.a.	est	[95SIL/BID]
2.5	3.1		0.1 M $\text{Na}(\text{ClO}_4,\text{HSO}_4)$	n.a.	tp	[90ROS/REI]
2.45 ^(b)			0.1 M NH_4Cl	H:n.a.,L:0.04M	tp	[73STE]
2.41			0.1 M NH_4Cl	H:n.a.,L:0.02M	tp	[73STE]
2.48 ^(c)			0.1 M NH_4Cl	H:n.a.,L:0.024M	tp	[73STE]
1.86	2.79		0.5 M $\text{Na}(\text{ClO}_4,\text{SO}_4)$	L:0.1M,pH3.6	cix	[68AZI/LYL]
1.85	2.83		0.5 M $\text{Na}(\text{ClO}_4,\text{SO}_4)$	L:0.1M,pH3.6	dis	[68AZI/LYL]
1.86	2.82		I = 0.5 M	n.a.	est	[76SMI/MAR]
1.78			0.75 M $\text{NH}_4(\text{Cl},\text{SO}_4)$	L:0.25M,pH3.5-4	cix	[60LEB/PIR]
1.2	1.4		1 M $\text{H}(\text{ClO}_4,\text{HSO}_4)$	H:1M,L:0.14M	cix	[64BAN/PAT]
1.49	2.5		1 M $\text{Na}(\text{ClO}_4,\text{SO}_4)$	L:0.23M,pH3	cix	[64BAN/PAT]
1.57	2.66		1 M $\text{Na}(\text{ClO}_4,\text{SO}_4)$	L:0.50M,pH3-4	dis	[64SEK], [65SEK2]

Table A5.11. (contd)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
1.22			1 M H(ClO ₄ ,HSO ₄)	H:1M,L:1M	cix	[67NAI], [68NAI]
1.49	2.36		1 M Na (ClO ₄ ,SO ₄)	n.a.	cix	[67NAI], [68NAI]
1.82	3.12		1 M H(ClO ₄ ,HSO ₄)	H:1M,L:0.7M	dis	[78RAO/BAG]
1.72			1 M NH ₄ (ClO ₄ ,SO ₄)	L:0.05M,pH3	dis	[80KHO/MAT]
1.76	2.11		1.5 M NH ₄ (Cl,SO ₄)	L:0.51M,pH3.5-4	cix	[60LEB/PIR]
1.43	1.85		2 M Na (ClO ₄ ,SO ₄)	L:0.1M,pH3	dis	[67CAR/CHO]
1.11 ^(a)	1.73 ^(a)		2 M Na (ClO ₄ ,SO ₄)	L:0.1M,pH3	dis	[67CAR/CHO2]
1.58 ^(d)	2.03 ^(d)		2 M Na (ClO ₄ ,SO ₄)	L:0.1M,pH3	dis	[67CAR/CHO2]
2.033	1.56	2.5	2 M Na (ClO ₄ ,SO ₄)	L:0.44M,pH3.1	pot	[67CAR/CHO2]
1.65 ^(e)	2.38 ^(e)		2 M Na (ClO ₄ ,SO ₄)	L:0.1M,pH3	dis	[67CAR/CHO2]
1.71			2 M H(ClO ₄ ,HSO ₄)	H:2M,L:0.7M	dis	[78RAO/BAG]

(a) T=273 K.

(b) T=288 K.

(c) T=308 K.

(d) T=313 K.

(e) T=328 K.

Table A5.12. Equilibrium Constants (B_n) for Formation of Aqueous Am^{3+} Complexes with HSO_4^- ($\text{Am}^{3+} + n\text{HSO}_4^- \rightleftharpoons \text{Am}(\text{HSO}_4)_n^{(3-n)+}$)

$\log \beta_1$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
0.54	1 M $\text{H}(\text{ClO}_4, \text{HSO}_4)$	n.a.	cix	[67NAI,68NAI]

Table A5.13. Solubility Products (K_{sp}) of the Different Am^{3+} Solid Phases

Reaction	Log K_{sp}	Medium	Maximum Metal/ H^+ /Ligand Concentration (M)	Method	References
$\text{AmF}_3(s) \rightleftharpoons \text{AmF}_3(\text{aq})$	-5.09	0.1 M $\text{HClO}_4^{(a)}$	n.a. ^(c)	sol	[54FEA]
	-4.87	0.1 M HClO_4	n.a. ^(c)	sol	[54FEA]
	-4.68	0.1 M $\text{HClO}_4^{(b)}$	n.a. ^(c)	sol	[54FEA]
$\text{AmF}_3(s) \rightleftharpoons \text{Am}^{3+} + 3\text{F}^-$	-13.9	0	n.a.	est	[88PHI/HAL]
$\text{Am(OH)}_3(\text{am}) + 3\text{H}^+ \rightleftharpoons \text{Am}^{3+} + 3\text{H}_2\text{O}$	17.00	0	n.a.	est	[95SIL/BID]
	17.5	0.1 M NaClO_4	M:1E-6, pH:10	sol	[83EDE/BUC]
$\text{Am(OH)}_3(\text{c}) + 3\text{H}^+ \rightleftharpoons \text{Am}^{3+} + 3\text{H}_2\text{O}$	15.2	0	n.a.	est	[95SIL/BID]
$\text{Am(OH)}_3(\text{s}) + 3\text{H}^+ \rightleftharpoons \text{Am}^{3+} + 3\text{H}_2\text{O}$	15.9	0 (0.1 M NaClO_4) ^(d)	M:1.6e-5, pH:9.4	sol	[87SIL]
	16.6 ^s	0 (0.1 M NaClO_4) ^(d)	M:1.6e-5, pH:9.4	recal	[95SIL/BID] ^(e)
	16.5	0.1 M NaClO_4	M:n.a., pH:9.5	sol	[84SIL]
	17.5	0 (0.0015 CaCl_2) ^(d)	M:1E-3, pH:10	sol	[83RAI/STR]
$\text{Am(OH)}_3(\text{s}) \rightleftharpoons \text{Am}^{3+} + 3\text{OH}^-$	-18.7	0	n.a.	est	[76BAE/MES]
	-28.56	0 (0.1 M NaClO_4) ^(e)	M:0.002, pH:13.5	sol	[84KIM/BER]
	-25.7	0.1 M NaClO_4	M:0.004, pH:13	sol	[88STA/KIM]

Table A5.13. (contd)

Reaction	Log K _{sp}	Medium	Maximum Metal/H ⁺ /Ligand Concentration (M)		
				Method	References
$\text{AmOHCO}_3(\text{s}) + 2\text{H}^+ \rightleftharpoons \text{Am}^{3+} + \text{HCO}_3^- + \text{H}_2\text{O}$	-27.35	0.002 M AmCl ₃	M:0.002, pH:7	sol, est	[90PER/SAP]
	-28.89	0.1 M NaClO ₄	M:2E-5, pH:13	sol	[84BER/KIM]
	-27.16	0.1 M NaClO ₄	M:0.002, pH:13.5	sol	[84KIM/BER]
	-27.4	3 NH ₄ ClO ₄	M:0.02, pH:9.8	sol	[89PAZ/KOC]
$\text{AmOHCO}_3(\text{s}) + 2\text{H}^+ \rightleftharpoons \text{Am}^{3+} + \text{HCO}_3^- + \text{H}_2\text{O}$	2.53	0 (0.1 M NaClO ₄) ^(c)	M:3E-6,L:8E-3 atm,pH:6.1	sol	[85SIL]
	2.77	0.1 M NaClO ₄	M:3E-6,L:8E-3 atm,pH:6.1	sol	[85SIL]
$\text{AmOHCO}_3(\text{s}) + 2\text{H}^+ \rightleftharpoons \text{Am}^{3+} + \text{HCO}_3^- + \text{H}_2\text{O}$	2.74	0.1 M NaClO ₄	M:n.a.,L:2E-4,pH:6.1	sol	[84SIL] [84SIL/NIT]
	$\text{AmOHCO}_3(\text{s}) \rightleftharpoons \text{Am}^{3+} + \text{OH}^- + \text{CO}_3^{2-}$	21.20	0	n.a.	est
		-23.1	0 (1 M HCl) ^(c)	M:2.4E-3,L:2.4E-3,H:1	cal
		-22.5	0 (0-0.1 M NaCO ₃) ^(c)	M:1E-4,L:0.1,pH:13.0	sol
		-18.70	0.1 M NaClO ₄	M:n.a.,L:1E-7,pH:7	sol
		-21.03	0.1 - 0.3 M NaClO ₄	M:2E-5,L:0.1,pH:13	sol
$\text{Am}_2(\text{CO}_3)_3(\text{s}) \rightleftharpoons 2\text{Am}^{3+} + 3\text{CO}_3^{2-}$	-33.40	0	n.a.	est	[95SIL/BID]
	-35.06	0	n.a.	est	[93GIF/VIT]
	-29.89	0.1 M NaClO ₄	M:1E-4,L:0.05,pH:9.7	sol	[91MEI/KIM]
	-29.45	0.1 M NaClO ₄	M:n.a.,L:1E-7,pH:7	sol	[92RUN/MEI]
	-34.2	0.1 M NaCl	M:1E-4,L:0.1,pC _H :9.5	sol	[93GIF/VIT]
	-30.16	3.0 M NaClO ₄	M:1E-4,L:1.0,pH:10.5	sol	[89ROB] ^(d)

Table A5.13. (contd)

Reaction	Log K_{sp}	Medium	Maximum Metal/H⁺/Ligand Concentration (M)	Method	References
	-30.4	4 M NaCl	M:1E-4,L:0.1,pC _H :9.5	sol	[93GIF/VIT]
NaAm(CO ₃) ₂ (s) ⇌ Na ⁺ + Am ³⁺ + 2CO ₃ ²⁻	-17.38	0	n.a.	sol	[86AVO/BIL] ^(b)
	-18.32	0.1 M	n.a.	sol	[85KIM] ^(b)
	-17.4	5 M NaCl	M:5E-3,L:0.02,pH:8.3	sol	[95RUN/KIM]
AmPO ₄ (am) ⇌ Am ³⁺ + PO ₄ ³⁻	-24.79	0	n.a.	est	[95SIL/BID]
	-24.79	0	M:1E-3, L:1E-3, pC _H :10	sol	[92RAI/FEL]

(a) T = 283 K.

(b) T = 320 K.

(c) Information obtained from [64SIL/MAR] or [71SIL/MAR].

(d) Thermodynamic value calculated using data from medium listed in parenthesis.

(e) Recalculation of data from [87SIL].

(f) Information obtained from [95SIL/BID].

Table A5.14. Binary Interaction Parameters

Cation	Anion	β^0	β^1	β^2	C^\ddagger	Reference
Am ³⁺	SO ₄ ²⁻	3.0398	0	-2500	0	[95RAI/FEL]
Am ³⁺	ClO ₄ ⁻	0.80	5.35	0	-0.0048	[90FEL/RAI]
		0.754	6.53	0	0.0075	[95RAI/FEL]
Na ⁺	Am(CO ₃) ₃ ³⁻	0.24	8.1	0	0	[90FEL/RAI]

A6. Curium(III)

Table A6.1. Equilibrium Constants (β_n) for Formation of Aqueous Cm^{3+} Complexes with F^- ($mCm^{3+} + nF^- \leftrightarrow Cm_m F_n^{(3m-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
4.3	7.6	10.9	H ₂ O	n.a.	est	[78ALL/BEA]
3.34	6.18	9.1	H ₂ O	n.a.	est	[82JEN]
3.34	6.17	9.07	0.5 M NaClO ₄	n.a.	est	[80BON/HEF]
6.2	9.1	I=0.5		n.a.	est	[92FUG/KHO]

Table A6.2. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with Cl⁻ ($Cm^{3+} + nCl^- \leftrightarrow CmCl_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
1.3		H ₂ O	n.a.	est	[92FUG/KHO]
0.18		0.195 M HCl	H:0.195M, L:0.195M	cix	[56WAR/WEL]
0.37		0.5 M HCl	H:0.5M, L:0.5M	cix	[82JEN]
0.215	-0.027	1 M NH ₄ ClO ₄	L:n.a.	dis	[80KHO/MAT]

Table A6.3. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with NO_3^- ($Cm^{3+} + nNO_3^- \rightleftharpoons CmNO_3^{(3-n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
0.57		1.0 M NH_4Cl	L:1.0M	dis	[60LEB/PIR]
0.34	0.097	1 M NH_4ClO_4	L:1.0M	dis	[80KHO/MAT]
-0.125		2.01 M NH_4SCN	L:2.01M	dis	[73CHI/DAN]

Table A6.4. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with OH^- ($Cm^{3+} + nOH^- \rightleftharpoons CmOH_n^{(3-n)+}$)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
8.4		H_2O	n.a.	est	[78ALL/BEA]
7.9		H_2O	n.a.	est	[82JEN]
10.6	18.9	0.005 M NH_4ClO_4	M:10 ⁻⁵ M,pH5	tp	[72SHA/STE]
6.67	12.06	0.1 M $NaClO_4$	pH7	sp	[92WIM/KEL]
6.09	11.32	0.011 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
5.76	11.1	0.099 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
5.58	10.98	0.576 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
5.47	11.01	0.985 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
5.48	10.9	0.99 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
5.6	11.15	1.00 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
5.72	11.39	1.022 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
6.06	12.19	2.00 M $NaCl$	pH:8.6	sp	[94FAN/KIM]
6.16	12.24	2.648 M $NaCl$	pH:8.6	sp	[94FAN/KIM]

Table A6.4. (contd)

log β_1	log β_2	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
6.26	12.39	3.476 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.39	13	3.743 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.43	12.7	3.828 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.21	13.25	4.00 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.6	13.23	4.244 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.6	13.22	4.981 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.48	13.96	5.357 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.62	13.24	5.693 M NaCl	pH:8.6	sp	[94FAN/KIM]
6.8	14.1	6.15 M NaCl	pH:8.6	sp	[94FAN/KIM]

Table A6.5. Equilibrium Constants (B_n) for Formation of Aqueous Cm³⁺ Complexes with OH⁻ ($Cm^{3+} + nH_2O \rightleftharpoons CmOH_n^{(3-n)+} + nH^+$)

log β_1	Medium	Maximum Metal/H⁺/Ligand Concentration	Method	Reference
-2.7	0.005M KCl	n.a.	tp	[68MAR/KIK]
-3.30 ^(a)	0.01 NaAc	pH:4.5	dis	[89MOH]
-5.92	0.1 M LiClO ₄	pH:6	dis	[69DES/HUS]
-6.05	0.1 M LiClO ₄	n.a.	dis	[69GUI/FER]
-5.4	0.1 M LiClO ₄	pH:6	dis	[73HUB/HUS]
-7.7	I = 0.1	n.a.	gl	[83EDE/BUC]

(a) T=303 K.

Table A6.6. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with CO_3^{2-} ($\text{Cm}^{3+} + n\text{CO}_3^{2-} \leftrightarrow \text{Cm}(\text{CO}_3)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
7.7	12.8		H_2O	n.a.	calc	[88CAN]
6.65 ^(a)	9.6 ^(b)	10.7 ^(c)	0.1 M NaClO_4	L:100% CO_2	sp	[92WIM/KIM]
6.32		n.a.		n.a.	sol	[91MEI]

(a) at pH 6.
 (b) at pH 9.4.
 (c) in 1M NaCO_3 .

Table A6.7. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with PO_4^{3-} ($\text{Cm}^{3+} + n\text{PO}_4^{3-} \leftrightarrow \text{Cm}(\text{PO}_4)_n^{(3-3n)+}$)

$\log \beta_1$	$\log \beta_2$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
20.2	36.8	H_2O	n.a.	sol, ix	[69MOS]
17.5	34.1	1 M NH_4Cl	M:0.015M, pH:1.8, L:0.57M	sol, ix	[71MOS2]

Table A6.8. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with H_2PO_4^- ($\text{Cm}^{3+} + n\text{H}_2\text{PO}_4^- \leftrightarrow \text{Cm}(\text{H}_2\text{PO}_4)_n^{(3-n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	$\log \beta_4$	Medium	Maximum Metal/ H^+ /Ligand Concentration	Method	Reference
2.4	3.6	5.61	6.2	H_2O	n.a.	sol, ix	[69MOS]
1.48	2.08	2.84	3.1	1 M NH_4Cl	M:0.015M, pH:1.8, L:0.57M	sol, ix	[71MOS2]

Table A6.9. Equilibrium Constants (B_n) for Formation of Aqueous Cm^{3+} Complexes with SO_4^{2-} ($Cm^{3+} + nSO_4^{2-} \rightleftharpoons Cm(SO_4)_n^{(3-2n)+}$)

$\log \beta_1$	$\log \beta_2$	$\log \beta_3$	Medium	Maximum Metal/H ⁺ /Ligand Concentration	Method	Reference
3.88	5.7	5.15	H ₂ O	L:0.5M,H5x10 ⁻⁴ M	sol	[72MCD/COL]
3.6	5.6		H ₂ O	n.a.	est	[78ALL/BEA]
1.86	2.7		H ₂ O	n.a.	est	[82JEN]
4.2	6.1		H ₂ O	n.a.	est	[92FUG/KHO]
2.41 ^(d)		0.1 M NH ₄ Cl		L:5mM,pH1.8	tp	[73STE]
2.45		0.1 M NH ₄ Cl		L:9mM,pH1.8	tp	[73STE]
2.48 ^(e)		0.1 M NH ₄ Cl		L:5mM,pH1.8	tp	[73STE]
1.85	2.7	0.5 M NaClO ₄		L:0.1M,pH3.6	dis	[68AZI/LYL]
1.86	2.37	0.5 M NaClO ₄		L:0.1M,pH3.6	ix	[68AZI/LYL]
1.75		0.75 M NH ₄ Cl		L:0.25M,pH4	dis	[60LEB/PIR]
1.7	3.19	1 M HClO ₄		H:1M,L:0.7M	dis	[78RAO/BAG]
1.509	2.382	1 M NH ₄ ClO ₄		L:0.05M,pH3	dis	[80KHO/MAT]
1.34	1.86	2 M NaClO ₄		L:0.1M,pH3	dis	[67CAR/CHO]
1.08 ^(a)	1.66 ^(a)	2 M NaClO ₄		L:0.1M,pH3	dis	[67CAR/CHO2]
1.49 ^(b)	2.05 ^(b)	2 M NaClO ₄		L:0.1M,pH3	dis	[67CAR/CHO2]
1.61 ^(c)	2.30 ^(c)	2 M NaClO ₄		L:0.1M,pH3	dis	[67CAR/CHO2]
1.59	3.11	2 M HClO ₄		H:1M,L:0.7M	dis	[78RAO/BAG]
0.93	0.61	3 M Na(Cl,SO ₄)		L:0.367m,pH2	sp	[96PAV/FAN]

(a) T=273K.

(b) T=313K.

(c) T=328K.

(d) T=288K.

(e) T=308K.

Table A6.10. Solubility Products (K_{sp}) of the Different Cm^{3+} Solid Phases

Reaction	Log K_{sp}	Medium	Maximum Metal/ H^+/ Ligand Concentration	Method	References
$\text{CmF}_3(s) = \text{CmF}_3(aq)$	-5.22	0.1 M $\text{HClO}_4^{(b)}$	n.a.	sol	[54FEA] ^(a)
	-4.91	0.1 M HClO_4	n.a.	sol	[54FEA] ^(a)
	-4.75	0.1 $\text{HClO}_4^{(c)}$	n.a.	sol	[54FEA] ^(a)

(a) Information obtained from [64SIL/MAR] or [71SIL/MAR].
(b) T = 283K.
(c) T = 320K.

Table A6.11. Binary Interaction Parameters

Cation	Anion	β^0	β^1	β^2	C^\ddagger	Reference
Cm^{3+}	OH^-	-0.6	3.0		0.2	[94FAN/KIM]
$\text{Cm}(\text{OH})_2^+$	Cl^-	-0.58	-0.9		0	[94FAN/KIM]
Cm^{3+}	Cl^-	0.5856	5.6		-0.019	[94FAN/KIM2]

A7. Selection of $\log \beta^{\circ}$ Values with the Screening Approach

A number of authors have used various approaches (e.g., extrapolation, S.I.T. or Pitzer modeling, etc.) to estimate values at classical standard state ($I = 0$, $T = 298K$) conditions for stability constants. These have been cited in Tables A1 - A6. However, the values for an individual system may vary significantly. To provide a basis for choosing among these values, a screening approach has been used which is based on the strongly ionic nature of the actinide-ligand bonding. An extended Born equation is used for estimation of $\log \beta^{\circ}_{101}$ for AnX complexation of various actinide cations with the same ligand X where An includes both simple and dioxo cations [94CHO/RIZ]. The equation takes into account the different "effective" ionic charges of the actinide cations, the different effective dielectric constants for these and the differences in An - X bond distances:

$$\Delta G_{101} = \frac{Ne^2 Z_1 Z_2}{418.7 D_e d_{12}} - vRT \ln 55.51$$

$Z_1 = +3$	$D_e = 57$	$d_{12} = \text{based on Shannon radii [76SHA]}$
$= +4$	$= 40$	
$= +2.2 (\text{NpO}_2^+)$	$= 70$	
$= +3.3 (\text{UO}_2^{2+})$	$= 50$	

The screening method calculated the unknown $\log \beta^{\circ}(\text{An}^{11})$ values from known values of another actinide, $\log \beta(\text{An}^I)$, by the formula:

$$\log \beta^{\circ}(\text{An}^{11}) = \log \beta(\text{An}^I) \cdot \frac{Z_1(\text{An}^{11})}{Z_1(\text{An}^I)} \cdot \frac{D_e(\text{An}^I)}{D_e(\text{An}^{11})} \cdot \frac{d_{12}(\text{An}^I)}{d_{12}(\text{An}^{11})}$$

Sm(III) values of $\log \beta^{\circ}$ were obtained from measurements by a variety of techniques at concentrations above tracer scale. For this reason, they were chosen as a reliable set of more reliable known values and unknown values of $\log \beta^{\circ}$ were calculated for the An^{3+} , An^{4+} , AnO_2^+ and AnO_2^{2+} complexation. UO_2^{2+} values of $\log \beta^{\circ}$ from the careful evaluation of the NEA seem reliable and were used in a second series of calculations of unknown values for the III - VI series of actinide cations. Only values for the 1:1 complexation were calculated as the use of the

Born equation for 1:2, etc., requires more "extension" and results in less reliable estimates. However, it is reasonable to assume that values for 1:2, 1:3, etc. from the authors responsible for the most reliable 1:1 values would be the values to use.

The values from the Sm³⁺ and UO₂²⁺ based estimates were compared and found to be in acceptable agreement. In turn, they were used to select, by the degree of agreement, a set of values from those at I = 0 in the data tables. These selected values which best agreed with these estimates are listed below as our recommended values for I = 0.

Table A7. Values of Log B_I⁰ Selected with the Screening Approach

Trivalent Ions	Cl ⁻	F ⁻	NO ₃ ⁻	OH ⁻	CO ₃ ²⁻	HPO ₄ ²⁻	H ₂ PO ₄ ⁻	SO ₄ ²⁻
Sm ³⁺		3.58 ^(a)	1.2 ^(c)	6.8 ^(a)		5.35 ^(a)	2.23 ^(a)	3.67 ^(e)
Am ³⁺	1.05 ^(a)	3.4 ^(b)	1.33 ^(b)	7.6 ^(b)	7.8 ^(b)	5.35 ^(a)	3 ^(b)	3.85 ^(b)
Pu ³⁺	(1.03)	3.3 ^(c)	1.41 ^(d)	7.6	(7.7)	(5.31)	2.95	(3.80)

() estimate.
 (a) [92MIL].
 (b) [95SIL/BID].
 (c) [86WAN].
 (d) [83FUG2].
 (e) [78ALL/BEA].

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Selection of $\log \beta_1^0$ Values with the Screening Approach

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