

Errata & Corrigenda for the 2003 *Update* on the Chemical Thermodynamics of U, Am, Np, Pu and Tc

Guillaumont, R., Fanghänel, T., Fuger, J., Grenthe, I., Neck, V., Palmer, D. A., Rand, M. H., *Update on the chemical thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium*, Nuclear Energy Agency Data Bank, Organisation for Economic Co-operation and Development, Ed., vol. 5, *Chemical Thermodynamics*, North Holland Elsevier Science Publishers B. V., Amsterdam, The Netherlands, (2003).

Authors and readers have pointed out several errata in the 2003 *Update*. These errata are corrected below. Similarly, the *Update* Reviewers contributed new calculations for some systems that could not be incorporated into the final version of the manuscript, as they arrived when the manuscript had already been sent to the publisher for printing. These corrections are also included in the table below.

Errata and corrections are primarily listed by page number but several entries corresponding to the same system have been grouped together for the sake of clarity.

Special thanks go to Prof. Robert Guillaumont (Orsay), Dr. Volker Neck (Karlsruhe), Dr. Ignasi Puigdomenech (Stockholm) and Dr. Lian Wang (Mol) for their efforts to ensure the consistency of text, tables and data files. We are also indebted to Prof. William M. Murphy (Chico, California) who informed us of a misinterpretation of the data reported in one of his papers on the solubility of schoepite.

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Page and position in text	Reads	Should read / corrective action	Affects selected values?	Notes
x		Add in Participating Organisations NAGRA, Switzerland	No	Typographical error.
62, entry for Na ₄ UO ₂ (CO ₃) ₃ (cr)	$\Delta_f G_m^\circ = -(3737.836 \pm 2.342) \text{ kJ}\cdot\text{mol}^{-1}$	$\Delta_f G_m^\circ = -(3739.206 \pm 2.336) \text{ kJ}\cdot\text{mol}^{-1}$	Yes (formation tables)	The <i>Update</i> changed the selection made in [92GRE/FUG] for UO ₂ (CO ₃) ₃ ⁴⁻ but this change was not propagated to the formation value entry for Na ₄ UO ₂ (CO ₃) ₃ (cr) in the NEA database.
93, entry for Np(CO ₃) ₄ ⁴⁻	$4 \text{ CO}_3^{2-} + 2\text{H}_2\text{O}(\text{l}) + \text{NpO}_2(\text{am,hyd}) \rightleftharpoons \text{Np}(\text{CO}_3)_4^{4-} + 4\text{OH}^-$ $\log_{10} K^\circ = -(17.990 \pm 0.220)$ $\Delta_f G_m^\circ = (101.550 \pm 1.256) \text{ kJ}\cdot\text{mol}^{-1}$	Delete entry. See accompanying notes.	Yes (reaction tables)	<p>The <i>Update</i> did not make any new selections for Np(IV) carbonate complexes. The $\Delta_f G_m^\circ$ values for Np(CO₃)₄⁴⁻ and Np(CO₃)₅⁶⁻ selected in [2001LEM/FUG] are adopted in the <i>Update</i>. However, the equilibrium constant for the reaction:</p> $4 \text{ CO}_3^{2-} + 2\text{H}_2\text{O}(\text{l}) + \text{NpO}_2(\text{am,hyd}) \rightleftharpoons \text{Np}(\text{CO}_3)_4^{4-} + 4\text{OH}^-,$ <p>$\log_{10} K^\circ = \log_{10} {}^*K_{s,0}^\circ(\text{NpO}_2, (\text{am,hyd})) + \log_{10} \beta_4^\circ$, depends on the selection for $\log_{10} {}^*K_{s,0}^\circ(\text{NpO}_2(\text{am,hyd}))$, which has been updated from (1.5 ± 1.0) in [2001LEM/FUG] to $-(0.7 \pm 0.5)$ in the <i>Update</i>. As a result, the value shown in the book, $\log_{10} K^\circ = -17.79$, is not consistent with the $\Delta_f G_m^\circ$ selections in the <i>Update</i>. The reaction above should have been replaced by the reaction:</p> $\text{Np}^{4+} + 4 \text{ CO}_3^{2-} \rightleftharpoons \text{Np}(\text{CO}_3)_4^{4-}$ <p>together with reaction</p> $\text{Np}^{4+} + 5 \text{ CO}_3^{2-} \rightleftharpoons \text{Np}(\text{CO}_3)_5^{6-}$ <p>whose equilibrium constants can be calculated from $\Delta_f G_m^\circ$ selections in [2001LEM/FUG] (<i>cf. Update</i>, p. 327).</p>

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101, entry for PuCl ²⁺	$\Delta_f G_m^\circ = - (717.051 \pm 2.923) \text{ kJ}\cdot\text{mol}^{-1}$	Delete entry. See accompanying notes.	Yes (formation tables)	The <i>Update</i> did not retain the selection made by [2001LEM/FUG] for $\log_{10} \beta^\circ$ for the reaction $\text{Pu}^{3+} + \text{Cl}^- \rightleftharpoons \text{PuCl}^{2+}$ for the reasons given in section 11.3.1.1, page 322. Therefore it is currently not possible to select a value for $\Delta_f G_m^\circ$ for PuCl ²⁺ .
726, entry for PuCl ²⁺	$\varepsilon(\text{PuCl}^{2+}, \text{ClO}_4^-) = (0.39 \pm 0.16)$	Delete entry. See accompanying notes.	Yes (SIT interaction coefficients)	For modeling An(III) in chloride solutions, a suggested procedure is to use the SIT coefficients based on $\varepsilon(\text{Nd}^{3+}, \text{Cl}^-) = \varepsilon(\text{Am}^{3+}, \text{Cl}^-) = (0.23 \pm 0.02)$.
121, entry for Am ₂ (CO ₃) ₃ (am)	$\log_{10} K^\circ = (16.7 \pm 1.1)$ $\Delta_r G_m^\circ = - (95.324 \pm 6.279) \text{ kJ}\cdot\text{mol}^{-1}$	$\log_{10} K^\circ = (33.4 \pm 2.2)$ $\Delta_r G_m^\circ = - (190.649 \pm 12.558) \text{ kJ}\cdot\text{mol}^{-1}$	Yes (reaction tables)	The value given in the printed tables corresponds to the same reaction but referred to half a mol of the Am ₂ (CO ₃) ₃ (am) product. See pages 380 and 381 of the <i>Update</i> .
117	Missing entry for AmO ₂ (CO ₃) ₃ ⁴⁻	$\Delta_f G_m^\circ = - (2277.831 \pm 9.177) \text{ kJ}\cdot\text{mol}^{-1}$	Yes (formation tables)	It is actually possible to select a value for $\Delta_f G_m^\circ$ (AmO ₂ (CO ₃) ₃ ⁴⁻ , 298.15 K) since the <i>Update</i> retains the selection made in [95SIL/BID] for the redox reaction $\text{AmO}_2(\text{CO}_3)_3^{4-} + \text{e}^- \rightleftharpoons \text{AmO}_2(\text{CO}_3)_3^{5-}$, based on the re-evaluation of [83BOU/GUI].
122	Missing entry for AmO ₂ (CO ₃) ₃ ⁴⁻	$\text{AmO}_2(\text{CO}_3)_3^{5-} \rightleftharpoons \text{AmO}_2(\text{CO}_3)_3^{4-} + \text{e}^-$ $\log_{10} K^\circ = - (13.100 \pm 0.600)$ $\Delta_r G_m^\circ = (74.775 \pm 3.425) \text{ kJ}\cdot\text{mol}^{-1}$	Yes (reaction tables)	The other value obtained from the re-evaluation of data from this latter source, [83BOU/GUI] is $\log_{10} K^\circ = (20.1 \pm 0.9)$ for $\text{Am}(\text{CO}_3)_5^{6-} + \text{e}^- \rightleftharpoons \text{Am}(\text{CO}_3)_3^{3-} + 2 \text{CO}_3^{2-}.$ This value agrees well with the result that may be calculated using the selections for Am in the <i>Update</i> .

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117, entry for $\text{Am}(\text{CO}_3)_5^{6-}$	$\Delta_f G_m^\circ = -(3210.227 \pm 7.919) \text{ kJ}\cdot\text{mol}^{-1}$	$\Delta_f G_m^\circ = -(3209.086 \pm 9.141) \text{ kJ}\cdot\text{mol}^{-1}$	Yes (formation tables)	The <i>Update</i> changed the selection made in [95SIL/BID] for $\log_{10} K^\circ$ for the reaction $\text{Am}^{3+} + 3 \text{CO}_3^{2-} \rightleftharpoons \text{Am}(\text{CO}_3)_3^{3-}$ from (15.2 ± 0.6) to (15.0 ± 1.0) but this change was not propagated to the formation values in the NEA database.
327, line 16	$\log_{10} \beta_5^\circ (\text{Pu}(\text{CO}_3)_5^{6-}, 298.15 \text{ K}) = (36.65 \pm 1.13)$.	$\log_{10} \beta_5^\circ (\text{Pu}(\text{CO}_3)_5^{6-}, 298.15 \text{ K}) = (35.65 \pm 1.13)$.	No	Typographical error with no further implications for other parts of the Review.
168, table 9-5, entry for $\text{Na}_2\text{U}_2\text{O}_7 \cdot x\text{H}_2\text{O}(\text{s})$	$-(29.7 \pm 0.1)$	$-(30.1 \pm 0.1)$	No	<p>Explanations for corrections concerning solubility constant of $\text{Na}_2\text{U}_2\text{O}_7 \cdot x\text{H}_2\text{O}(\text{cr})$ from data of [98MEI/FIS]:</p> <p>Meinrath <i>et al.</i> [98MEI/FIS] determined solubility data in 0.1 M NaClO_4 for the reaction:</p> $\text{Na}_2\text{U}_2\text{O}_7(\text{s}) + 6 \text{H}^+ + 6 \text{CO}_3^{2-} \rightleftharpoons 2 \text{Na}^+ + 2 \text{UO}_2(\text{CO}_3)_3^{4-} + 3 \text{H}_2\text{O}$ <p>The equilibrium constant ($\log_{10} {}^*K_{s,0} + 2\log_{10} \beta_3$) is not explicitly given. However, using $\log_{10} \beta_3 = 21.86$ (similar to 21.84 selected in the <i>Update</i>), it is possible to calculate $\log_{10} {}^*K_{s,0} = (24.2 \pm 0.2)$ in 0.1 M NaClO_4 for the reaction:</p> $\text{Na}_2\text{U}_2\text{O}_7(\text{s}) + 6 \text{H}^+ \rightleftharpoons 2 \text{Na}^+ + 2 \text{UO}_2^{2+} + 3 \text{H}_2\text{O}.$
168, table 9-5, footnote (e)	(24.6 ± 0.2)	(23.8 ± 0.2)	No	
181, line 28	(24.6 ± 0.2)	(23.8 ± 0.2)	No	
608, line 31	$\log_{10} \beta_3^\circ$	$\log_{10} \beta_3$	No	

(continued on next page)

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609, lines 3 to 6	This review has recalculated this value using the selected value of $\log_{10} \beta_3^{\circ} = 21.60$ and obtains $\log_{10} {}^*K_{s,0}(\text{A.100}) = (23.7 \pm 0.2)$ which recalculated to zero ionic strength gives $\log_{10} {}^*K_{s,0}^{\circ}(\text{A.100}) = (24.1 \pm 0.2)$.	This review has recalculated this value using the selected value of $\log_{10} \beta_3^{\circ} = 21.60$ [92GRE/FUG] and obtains $\log_{10} {}^*K_{s,0}(\text{A.100}) = (24.7 \pm 0.2)$, which recalculated to zero ionic strength gives $\log_{10} {}^*K_{s,0}^{\circ}(\text{A.100}) = (24.3 \pm 0.2)$.	No	Extrapolation to $I = 0$ with the SIT ($\Delta z^2 = 4$, $\Delta \varepsilon = 0.14$,) yields: $\log_{10} {}^*K_{s,0}^{\circ} = \log_{10} {}^*K_{s,0} - \Delta z^2 D + \Delta \varepsilon I = (23.8 \pm 0.2)$ (instead of 24.6 ± 0.2). Accordingly the solubility product given in Table 9-5 for the reaction $1/2 \text{Na}_2\text{U}_2\text{O}_7(\text{s}) + 1.5 \text{H}_2\text{O} \rightleftharpoons \text{Na}^+ + \text{UO}_2^{2+} + 3 \text{OH}^-$ should be $\log_{10} K_{s,0}^{\circ} = 1/2 \log_{10} {}^*K_{s,0}^{\circ} + 3 \log_{10} K_w^{\circ} = -(30.1 \pm 0.1)$ (instead of $-(29.7 \pm 0.1)$). In <i>Appendix A</i> , the equilibrium constant $\log_{10} {}^*K_{s,0} = (24.2 \pm 0.2)$ [98MEI/FIS] is recalculated by using $\log_{10} \beta_3^{\circ} = 21.60$ from [92GRE/FUG] instead of $\log_{10} \beta_3 = 21.86$ used by Meinrath <i>et al.</i> [98MEI/FIS] (probably for better comparison with the corresponding value from [98YAM/KIT]). Using a value of $\log_{10} \beta_3 (= \log_{10} \beta_3^{\circ})$ which is 0.26 log-units lower, the corresponding $\log_{10} {}^*K_{s,0}$ value must be 0.5 log-units higher to give the experimentally determined sum ($\log_{10} {}^*K_{s,0} + 2 \log_{10} \beta_3$). From that, it follows for the recalculation of the data from [98MEI/FIS]: $\log_{10} {}^*K_{s,0}(\text{A.100}) = (24.7 \pm 0.2)$ and $\log_{10} {}^*K_{s,0}^{\circ}(\text{A.100}) = (24.3 \pm 0.2)$.
618, line 20	$-(5.47 \pm 0.25)$	$-(9.47 \pm 0.25)$	No	Typographical error with no further implications for other parts of the Review.

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555, Appendix A entry for [97MUR]	The calculated solubility of schoepite decreases from $10^{-3.1}$ to $10^{-4.8}$ M when T decreases from 100 to 25°C	The equilibrium solubility constant of schoepite (<i>i.e.</i> ; the equilibrium constant for the schoepite dissolution reaction – not molarity) increases with decreasing temperature from $10^{3.1}$ to $10^{4.8}$ between 100 and 25°C.	No	The statement in the original Appendix A entry is incorrect and implies the opposite of the correct conclusion in [97MUR].
732, entry for SO_4^{2-}		Missing (†) dagger footnote for both $\epsilon(\text{Li}^+, \text{SO}_4^{2-})$ and $\epsilon(\text{Na}^+, \text{SO}_4^{2-})$	No	These SIT coefficients can be described more accurately with an ionic strength dependent function, as listed in Table B-6.
719, figure B-1	$\log_{10} \beta_1^0 = (0.17 \pm 002)$	$\log_{10} \beta_1^0 = (0.17 \pm 0.02)$	No	Typographical error.
751, line 3	(0.09 ± 0.92)	(8.09 ± 0.96)	No	Typographical error.