

**Attachment C-5**

**Enchemica.R1.dat Database and Documentation**

**PHREEQC DATABASE  
Enchemica.R1.dat**

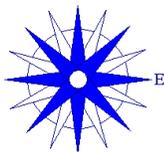
**September 2017**

**Prepared for:**



**Cameco Resources  
PO Box 1210  
Glenrock, Wyoming 82637**

**Prepared by:**



**Enchemica LLC  
2335 Buckingham Circle  
Loveland, Colorado 80538  
(970) 203-0179**

## Table of Contents

Table of Contents .....	i
Tables.....	ii
Figures.....	ii
Appendices .....	ii
1.0 Introduction .....	1
2.0 Data Sources .....	1
3.0 Uranium.....	2
3.1 Aqueous Uranium Data .....	2
3.2 Uranium Solid Phase Data .....	4
3.2.1 Uranium(IV) Solid Phases .....	4
3.2.2 Uranium(VI) Oxide, Hydrated Oxide, Carbonate and Sulfate Solids.....	6
3.2.3 Uranium(VI) Silicate Solids.....	7
3.2.4 Uranium(VI) Phosphate Solids.....	10
4.0 Phosphate .....	11
5.0 Vanadium .....	13
6.0 Sulfide.....	14
7.0 Arsenic .....	15
8.0 Copper.....	17
9.0 Nickel.....	17
10.0 Selenium.....	18
11.0 Iron .....	18
11.1 Aqueous Iron Data .....	19
11.1.1 Ferrous Iron Aqueous Speciation.....	19
11.1.2 Ferric Iron Aqueous Speciation.....	20
11.2 Iron Solid Phase Data .....	22
12.0 Manganese.....	24
13.0 Radium .....	26
14.0 Molybdenum.....	26
15.0 Aluminum and Zinc Solid Phases.....	26
16.0 Surface Complexation Data.....	27
17.0 References .....	27

## Tables

Table 1	Elements/Species Included in the Enchemica Database
Table 2	Uranium(IV) Solid Phases Included in the Enchemica Database
Table 3	Uranium(VI) Solid Phases Included in the Enchemica Database
Table 4	Metaschoepite Solubility Data
Table 5	Phosphate Solid Phases from Vieillard and Tardy (1984)
Table 6	Hydroxyapatite Solubility Data [ $\text{Ca}_5(\text{PO}_4)_3\text{OH}(\text{s}) + \text{H}^+ = 5\text{Ca}^{2+} + 3\text{PO}_4^{3-} + \text{H}_2\text{O}$ ]
Table 7	Equilibrium constants, redox potentials and enthalpies of reaction for $\text{Fe}^{2+} = \text{Fe}^{3+} + \text{e}^-$
Table 8	Ferric iron hydrolysis data
Table 9	Mackinawite solubility data accepted by Lemire et al. (2013, Table IX-3) [ $\text{FeS}(\text{s}) + 2 \text{H}^+ = \text{Fe}^{2+} + \text{H}_2\text{S}(\text{aq})$ ]
Table 10	Manganese(II) aqueous carbonate and bicarbonate speciation data

## Figures

Figure 1	Synthetic metaschoepite solubility calculated with and without polymeric uranium species compared to metaschoepite solubility data from Jang et al. (2006) and from Gorman-Lewis et al. (2008b)
Figure 2	Calculated and measured uranium concentrations in equilibrium with boltwoodite using the $\log_{10}K_{\text{sp}}^0$ value reported by Shvareva et al. (2011)
Figure 3	Calculated solubility of $\text{As}_2\text{S}_3(\text{am})$ as a function of sulfide concentration

## Appendices

Appendix A	Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data
Appendix B	Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data
Appendix C	Revised Surface Complexation Data for Uranyl, Carbonate, Bicarbonate and Radium Adsorption on Hydrous Ferric Oxide

## 1.0 Introduction

A thermodynamic database (Enchemica.R1.dat) was developed for geochemical modeling of aqueous solutions. The database is formatted for use with the PHREEQC geochemical code (Parkhurst and Appelo 2013).<sup>1</sup> It includes major-element aqueous speciation and solid-phase solubility data from the wateq4f.dat database distributed with PHREEQC, combined with selected uranium, phosphate, vanadium, arsenic, copper and nickel aqueous speciation and solid-phase solubility data.

## 2.0 Data Sources

The wateq4f.dat database distributed with PHREEQC by the U.S. Geological Survey was used as the foundation for the database. The wateq4f.dat database was used as the starting point because the its major-element data are consistent with the Nordstrom et al. (1990) data compilation (Table 1), which is a reliable and internally consistent data set. Ball and Nordstrom (1991) provide documentation for the sources of other unmodified aqueous speciation and solid solubility data in the wateq4f.dat database. Exchange reactions from the wateq4f.dat database were included in the Enchemica.R1.dat database. The sources of the exchange reaction constants are provided in the internal documentation for the llnl.dat database distributed with the PHREEQC code.

Table 1. Elements/Species Included in the Database

Data from Nordstrom et al. (1990)		Data from wateq4f.dat		Reviewed Data	
Aluminum	Magnesium	Alkalinity	Lead	Uranium	Iron
Barium	Potassium	Boron	Nitrogen-ammonia	Vanadium	Phosphate
Calcium	Silica	Bromide	Nitrogen-nitrate	Sulfide <sup>c</sup>	Arsenic
Carbonate	Sodium	Cesium	Nitrogen-nitrite	Copper <sup>d</sup>	Nickel <sup>e</sup>
Chloride	Strontium	Copper <sup>a</sup>	Sulfide <sup>b</sup>	Selenium	Manganese
Fluoride	Sulfate	Iodide	Zinc	Radium	Molybdenum
Lithium					

- a – Copper(I) aqueous and solid phases, copper metal, copper(II) fluoride and sulfide aqueous species and copper(II) fluoride, sulfide and sulfate solid phases  
b – Sulfide solid phases other than arsenic sulfides  
c – Hydrogen sulfide dissociation and arsenic-sulfide aqueous and solid phases  
d – Copper(II) hydroxyl, chloride, carbonate, sulfate and phosphate aqueous complexes and copper(II) oxide, hydroxide, and carbonate solid phases  
e – Nickel hydroxyl, chloride, carbonate, sulfate and phosphate aqueous complexes

<sup>1</sup> [http://wwwbrr.cr.usgs.gov/projects/GWC\\_coupled/phreeqc/](http://wwwbrr.cr.usgs.gov/projects/GWC_coupled/phreeqc/)

The wateq4f.dat database was modified by replacing aqueous speciation and solid-phase solubility data for uranium, iron, vanadium, phosphate, sulfide, arsenic, copper, nickel, selenium, manganese, radium and molybdenum with reviewed data as described in Sections 3.0 through 9.0. Data for hydrogen sulfide (H<sub>2</sub>S) dissociation and for arsenic-sulfide aqueous species and solid phases in the database were obtained from reviewed sources. Data for other sulfide solid phases (such as iron sulfides) were retained from the wateq4f.dat database.

## 3.0 Uranium

Under environmental conditions, uranium is chiefly present in aqueous solution and solid phases in either the uranium(IV) (uranous) or uranium(VI) (uranyl) oxidation states. Uranium has also been reported in the uranium(V) oxidation state at pH values less than 7 under moderately reducing conditions (Langmuir 1978). Aqueous speciation and solubility data for uranium(IV), uranium(V) and uranium(VI) were therefore reviewed and included in the database.

Compilations prepared by the Nuclear Energy Agency (NEA) were the principal sources of the uranium data in the database. Grenthe et al. (1992) carried out a critical review and selected thermodynamic properties for uranium solid compounds and aqueous species. This review was updated by Guillaumont et al. (2003). Data for uranium aqueous species and solid phases developed after the Guillaumont et al. (2003) compilation were reviewed and used to revise the uranium data as described below.

### 3.1 Aqueous Uranium Data

The aqueous uranium species included in the database are based on data compiled for uranium hydrolysis, fluoride, chloride, bromide, carbonate, arsenate, phosphate, sulfate, nitrate and silicate species by the NEA (Guillaumont et al. 2003). The aqueous uranium species stability constants included in the database are summarized in Appendix A.

The polymeric uranyl hydroxycarbonate species  $(\text{UO}_2)_2\text{CO}_3(\text{OH})_3^-$ ,  $(\text{UO}_2)_3\text{O}(\text{OH})_2(\text{HCO}_3)^+$  and  $(\text{UO}_2)_{11}(\text{CO}_3)_6(\text{OH})_{12}^{2-}$  reported by Guillaumont et al. (2003) were not included in the database. Jang et al. (2006) investigated the solubility of metaschoepite  $[\text{UO}_3 \cdot 2\text{H}_2\text{O}(\text{cr})]$  and found that the solubility data were best described by omitting these polymeric aqueous species. In addition, Gustafsson et al. (2009) found that the association constants for these species are likely overestimated. The metaschoepite solubility data of Jang et al. (2006) and Gorman-Lewis et al. (2008b) were compared to predicted total uranium solution concentrations as a function of pH using the database with and without the polymeric uranyl hydroxycarbonate species (Figure 1). This comparison confirmed that inclusion of the polymeric uranyl hydroxycarbonate species results in a significant overestimation of uranium concentrations measured at moderate pH values by Jang et al. (2006).

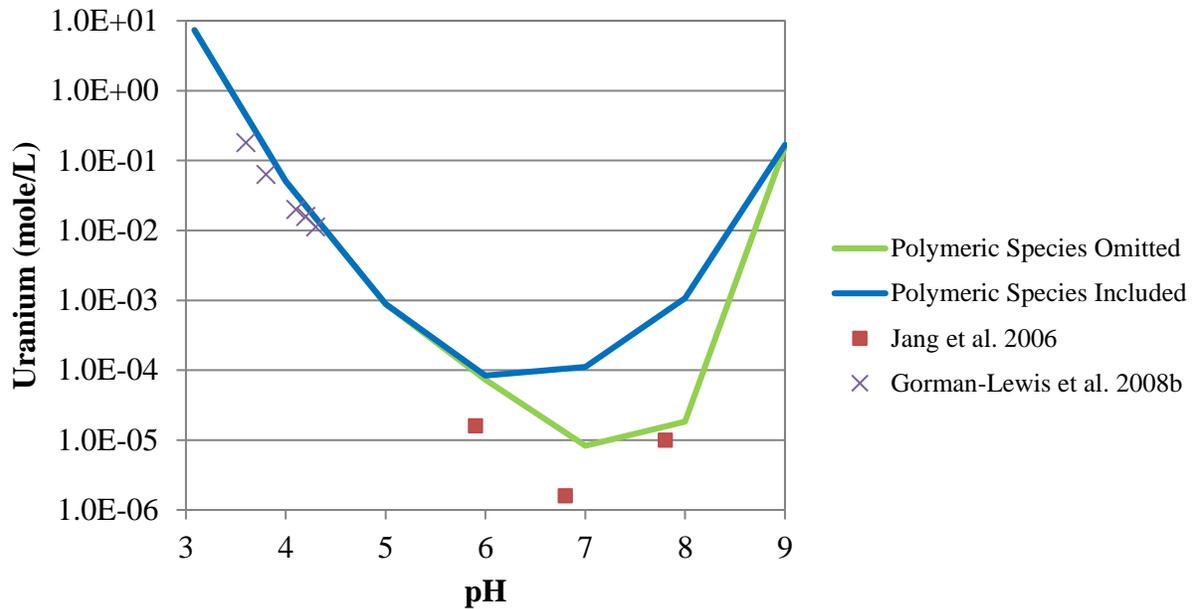


Figure 1. Synthetic metaschoepite solubility calculated with and without polymeric uranium species compared to metaschoepite solubility data from Jang et al. (2006) and from Gorman-Lewis et al. (2008b)

Thermodynamic data for ternary uranium(VI) complexes formed with carbonate and the alkaline earth elements calcium, magnesium, strontium and barium were reported by Dong and Brooks (2006, 2008) after completion of the Guillaumont et al. (2003) review. The existence of the  $\text{CaUO}_2(\text{CO}_3)_3^{2-}$  and  $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$  aqueous species under environmentally relevant conditions and the stability constants of Dong and Brooks (2006) are supported by an investigation using extended X-ray absorption fine structure (Kelly et al. 2007). Consequently, the uranium(VI) complexation data of Dong and Brooks (2006, 2008) for calcium, magnesium, barium and strontium with uranyl and carbonate ions were included in the database.

The uranium(VI)-phosphate aqueous speciation data selected by Guillaumont et al. (2003) were incorporated in the database, with the exception of the data for the  $\text{UO}_2\text{PO}_4^-$  aqueous species. The experimentally determined limiting value of the stability constant ( $\log_{10}K^0$ ) for the association reaction from Rai et al. (2005) was instead selected for this species.

Guillaumont et al. (2003) selected stability constants for the uranium(VI) species  $\text{UO}_2\text{SO}_4^0$ ,  $\text{UO}_2(\text{SO}_4)_2^{2-}$  and  $\text{UO}_2(\text{SO}_4)_3^{2-}$  based on the review by Grenthe et al. (1992). Vercouter et al. (2008) determined the stoichiometries and stabilities of the aqueous uranium(VI)-sulfate complexes at low and high ionic strength at temperatures ranging from 10°C to 75°C. The stability constants for  $\text{UO}_2\text{SO}_4^0$ , and  $\text{UO}_2(\text{SO}_4)_2^{2-}$  extrapolated to zero ionic strength determined by Vercouter et al. (2008) are similar to the Guillaumont et al. (2003) values

and were included in the database. Vercouter et al. (2008) observed the formation of the  $\text{UO}_2(\text{SO}_4)_3^{2-}$  species only at high ionic strength. Because Vercouter et al. (2008) did not extrapolate the stability constant to zero ionic strength, the  $\text{UO}_2(\text{SO}_4)_3^{2-}$  stability constant reported by Guillaumont et al. (2003) was used for this species in the database.

### 3.2 Uranium Solid Phase Data

Thermodynamic data have been reported for a number of uranium-bearing solid phases (e.g., Langmuir 1978, Grenthe et al. 1992, Guillaumont et al. 2003, Gorman-Lewis et al. 2008a), but many of these solid phases are unlikely to form under low-temperature conditions. Only the uranium(IV) and uranium(VI) solid phases that are potentially significant under conditions anticipated in near-surface groundwater were included in the database (Tables 2 and 3).

Table 2. Uranium(IV) Solid Phases Included in the Database

Name	Chemical Formula
Uraninite	$\text{UO}_2(\text{cr})^2$
Coffinite	$\text{USiO}_4(\text{cr})$
Synthetic coffinite	$\text{USiO}_4(\text{syn})$
$\text{U}_4\text{O}_9(\text{cr})$	$\text{U}_4\text{O}_9(\text{cr})$
$\alpha\text{-U}_3\text{O}_8(\text{cr})$	$\alpha\text{-U}_3\text{O}_8(\text{cr})$
Amorphous hydrous $\text{UO}_2$	$\text{UO}_2(\text{am,hyd})$
Hydrous uranous phosphate	$\text{U}(\text{HPO}_4)_2 \cdot 4\text{H}_2\text{O}(\text{cr})$
Uranous sulfate	$\text{U}(\text{OH})_2\text{SO}_4(\text{cr})$
Ningyoite	$\text{CaU}(\text{PO}_4)_2 \cdot 2\text{H}_2\text{O}(\text{cr})$

#### 3.2.1 Uranium(IV) Solid Phases

Uraninite and coffinite solubility data from Guillaumont et al. (2003) were included in the database because these phases are common ore minerals in uranium roll-front deposits (Granger and Warren 1969, Ludwig and Grauch 1980). The coffinite solubility from Guillaumont et al. (2003) is based on Langmuir (1978). Szenknect et al. (2013) reported a higher coffinite solubility based on an extrapolation from solubility experiments with uranothorite [ $\text{Th}_x\text{U}_{1-x}\text{SiO}_4(\text{s})$ ] solid solutions. This solubility was not included in the database because different  $\text{U}^{4+}$  aqueous speciation was used in the evaluation. Szenknect et al. (2016) experimentally measured the aqueous solubility of synthetic coffinite, and

---

<sup>2</sup> Notation for solid phase crystallinity follows that of Guillaumont et al. (2003): (am) indicates an amorphous solid phase, (cr) indicates a crystalline solid phase, and (s) indicates that the crystallinity was not specified. The notation (hyd) indicates a hydrous phase with an unspecified amount of water in its crystal structure.

determined a higher solubility than the value based on Langmuir (1978). This solubility for synthetic coffinite was included in the database, with free energy of reaction data based on measurements by Guo et al. (2015) using the same synthetic coffinite.

Table 3. Uranium(VI) Solid Phases Included in the Database

Name	Chemical Formula
$\gamma$ -UO <sub>3</sub> (cr)	$\gamma$ -UO <sub>3</sub> (cr)
Schoepite	$\beta$ -UO <sub>2</sub> (OH) <sub>2</sub> (cr)
Metaschoepite(cr)	UO <sub>3</sub> •2H <sub>2</sub> O(cr)
Metaschoepite(syn)	UO <sub>3</sub> •2H <sub>2</sub> O(syn)
Metaschoepite(am)	UO <sub>3</sub> •2H <sub>2</sub> O(am)
Rutherfordine	UO <sub>2</sub> CO <sub>3</sub> (cr)
UO <sub>2</sub> SO <sub>4</sub> •2.5H <sub>2</sub> O(cr)	UO <sub>2</sub> SO <sub>4</sub> •2.5H <sub>2</sub> O(cr)
UO <sub>2</sub> SO <sub>4</sub> •3H <sub>2</sub> O(cr)	UO <sub>2</sub> SO <sub>4</sub> •3H <sub>2</sub> O(cr)
UO <sub>2</sub> SO <sub>4</sub> •3.5H <sub>2</sub> O(cr)	UO <sub>2</sub> SO <sub>4</sub> •3.5H <sub>2</sub> O(cr)
Clarkeite	Na(UO <sub>2</sub> )O(OH)(s)
Compreignacite	K <sub>2</sub> (UO <sub>2</sub> ) <sub>6</sub> O <sub>4</sub> (OH) <sub>6</sub> •7H <sub>2</sub> O(s)
Sodium-compreignacite	Na <sub>2</sub> (UO <sub>2</sub> ) <sub>6</sub> O <sub>4</sub> (OH) <sub>6</sub> •7H <sub>2</sub> O(s)
Becquerelite	CaU <sub>6</sub> O <sub>19</sub> •11H <sub>2</sub> O(s)
Soddyite	(UO <sub>2</sub> ) <sub>2</sub> SiO <sub>4</sub> •2H <sub>2</sub> O(s)
Boltwoodite	K(UO <sub>2</sub> )(SiO <sub>3</sub> OH)•2H <sub>2</sub> O(s)
Sodium-boltwoodite	Na(UO <sub>2</sub> )(SiO <sub>3</sub> OH)•2H <sub>2</sub> O(s)
Uranophane	Ca(UO <sub>2</sub> ) <sub>2</sub> (SiO <sub>3</sub> OH) <sub>2</sub> •5H <sub>2</sub> O(s)
Sodium-weeksite	Na <sub>2</sub> (UO <sub>2</sub> ) <sub>2</sub> (Si <sub>2</sub> O <sub>5</sub> ) <sub>3</sub> (H <sub>2</sub> O) <sub>4</sub> (s)
Uranyl orthophosphate, anhydrous	(UO <sub>2</sub> ) <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Uranyl orthophosphate	(UO <sub>2</sub> ) <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> •4H <sub>2</sub> O(cr)
Uranyl hydrogen phosphate	UO <sub>2</sub> HPO <sub>4</sub> •3H <sub>2</sub> O(cr)
Hydrogen autunite	UO <sub>2</sub> HPO <sub>4</sub> •4H <sub>2</sub> O(cr)
Autunite	Ca(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> •3H <sub>2</sub> O(cr)
Saleeite	Mg(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Sodium autunite	Na <sub>2</sub> (UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Potassium autunite	K <sub>2</sub> (UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Strontium autunite	Sr(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Uranocircite	Ba(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Bassetite	Fe(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Uramphite	(NH <sub>4</sub> ) <sub>2</sub> (UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Przhevalskite	Pb(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub> (cr)
Metatorbernite	Cu[(UO <sub>2</sub> )(PO <sub>4</sub> )] <sub>2</sub> •8H <sub>2</sub> O(cr)
Torbernite	Cu(UO <sub>2</sub> ) <sub>2</sub> (PO <sub>4</sub> ) <sub>2</sub>
Cesium autunite	CsUO <sub>2</sub> PO <sub>4</sub> •xH <sub>2</sub> O
Carnotite	K <sub>2</sub> (UO <sub>2</sub> ) <sub>2</sub> (VO <sub>4</sub> ) <sub>2</sub> (cr)
Tyuyamunite	Ca(UO <sub>2</sub> ) <sub>2</sub> (VO <sub>4</sub> ) <sub>2</sub> (cr)

Langmuir (1978) reported that natural uraninite ranges in composition from  $\text{UO}_2$  through  $\text{U}_4\text{O}_9$  to  $\text{U}_3\text{O}_8$  as a consequence of partial oxidation of  $\text{UO}_2(\text{cr})$  after deposition. Solubility data selected by Guillaumont et al (2003) for  $\text{U}_4\text{O}_9(\text{cr})$  and  $\text{U}_3\text{O}_8(\text{cr})$  were included in the database to address the possible solubility range of uranium oxide solid phases.

Thermodynamic data reported by Guillaumont et al. (2003) for amorphous hydrous uranium dioxide [ $\text{UO}_2(\text{am,hyd})$ ], hydrous uranous phosphate [ $\text{U}(\text{HPO}_4)_2 \cdot 4\text{H}_2\text{O}(\text{cr})$ ] and uranous sulfate [ $\text{U}(\text{OH})_2\text{SO}_4(\text{cr})$ ] were included in the database. Solubility data for ningyoite [ $\text{CaU}(\text{PO}_4)_2 \cdot 2\text{H}_2\text{O}(\text{cr})$ ] reported by Langmuir (1978) were also included in the database.

### 3.2.2 Uranium(VI) Oxide, Hydrous Oxide, Carbonate and Sulfate Solids

The majority of the uranyl solids listed in Table 3 have been reported to form from solution at low temperatures. Thermodynamic data selected for the solid phases  $\gamma\text{-UO}_3(\text{cr})$ , schoepite [ $\beta\text{-UO}_2(\text{OH})_2(\text{cr})$ ], crystalline metaschoepite [ $\text{UO}_3 \cdot 2\text{H}_2\text{O}(\text{cr})$ ], amorphous metaschoepite, rutherfordine [ $\text{UO}_2\text{CO}_3(\text{cr})$ ], and the hydrous uranyl-sulfate solids were obtained from the NEA compilations (Grenthe et al. 1992, Guillaumont et al. 2003).

A range of solubilities has been reported for crystalline metaschoepite (Table 4). The lowest solubility, reported by Guillaumont et al (2003), was calculated from selected enthalpy and heat capacity data for the crystalline solid. This solubility product ( $\log_{10}K_{\text{sp}}^0$ ) of 4.808 was selected as a lower limit for the solubility of crystalline metaschoepite. Higher metaschoepite solubility product values, ranging from 5.37 to 6.23, were determined on the basis of solubility experiments and may reflect varying degrees of crystallinity of the solid phases in the experiments, different assumptions regarding aqueous speciation, different methods of extrapolating the data to zero ionic strength and possibly, the effects of sodium concentrations on metaschoepite transformation to clarkeite (Giammar and Hering 2004). The results reported by Giammar and Hering (2004) and Gorman-Lewis et al. (2008b) are virtually identical and were obtained using metaschoepite synthesized for the experiments. The metaschoepite solubility from Gorman-Lewis et al. (2008b) was included in the database as metaschoepite(syn). The highest solubility product of 6.33 was reported by Guillaumont et al. (2003) for amorphous metaschoepite, which was also included in the database. The solubility product values selected for crystalline, synthetic and amorphous metaschoepite should represent the expected solubility range as a function of metaschoepite crystallinity.

Table 4. Crystalline Metaschoepite Solubility Data

Log <sub>10</sub> K <sub>sp</sub> <sup>0</sup>	Reference	Source
4.808	Grenthe et al. (1992), Guillaumont et al. (2003)	Calculated from selected enthalpy and heat capacity data
5.23	Langmuir (1978)	Calculated from reported thermodynamic data
5.37	Diaz-Arocas and Grambow (1998)	Solubility experiments
5.39	Jang et al. (2006)	Solubility experiments
5.52	Giammar and Hering (2004)	Solubility experiments
5.60	Gorman-Lewis et al. (2008b)	Solubility experiments
5.96	Sandino (1991)	Solubility experiments reported by Grenthe et al. (1992)
6.23	Sandino and Bruno (1992)	Solubility experiments reported by Gorman-Lewis et al. (2008a)

Gorman-Lewis et al. (2008b) experimentally determined the solubilities of clarkeite, compreignacite, and sodium-compreignacite. Gorman-Lewis et al. (2008b) used the aqueous speciation data summarized by Guillaumont et al. (2003) to calculate solubility products for these solids at 25°C and zero ionic strength (I) from their solubility experiments. The polymeric uranyl-hydroxycarbonate aqueous species excluded from the database (Section 3.1) were included in the Gorman-Lewis (2008b) calculations, which could affect the results under higher pH conditions. However, inclusion of these species did not affect the calculated solubility products because the experiments were conducted at relatively low pH values where the calculated concentrations of the polymeric species would be insignificant.

Gorman-Lewis et al. (2008b) performed solubility experiments from both under- and oversaturation using synthetic becquerelite from pH 3.2 to 5.2 and calculated a solubility product of 40.5. This experimentally determined solubility product equals the value selected by Guillaumont et al. (2003) for becquerelite. Rai et al. (2002) carried out becquerelite solubility experiments only from undersaturation and calculated a solubility product of 41.2. Because Gorman-Lewis et al. (2008b) established their solubility product from both under- and oversaturation, their solubility product value of 40.5 was included in the database.

### 3.2.3 Uranium(VI) Silicate Solids

Guillaumont et al. (2003) selected a solubility product for soddyite of 6.2 based on an average of the results of two experimental investigations – a value of 5.74 reported by Nguyen et al. (1992) from experiments carried out from undersaturation only and a value of 6.7 reported by Moll et al. (1996). Gorman-Lewis et al. (2007) reported a solubility

product of 6.43 for soddyite, which falls within the range reported by the previous investigations. The solubility product reported by Gorman-Lewis et al. (2007) was based on an assumption of different aqueous uranyl species and association constants than those used for the Enchemica.R1 database. However, all experiments were conducted at low pH (less than 3.9), where the uranyl ion ( $\text{UO}_2^{2+}$ ) is the dominant species, minimizing effects of aqueous complexation of the uranyl ion. Because the Gorman-Lewis et al. (2007) data were obtained from both undersaturation and oversaturation and the solid phases were characterized before and after the experiments, the reported solubility product of 6.43 from this study was selected for inclusion in the database.

Guillaumont et al. (2003) identified a solubility product for sodium boltwoodite of 5.82 based on experiments from undersaturation of Nguyen et al. (1992). Ilton et al. (2006) determined a solubility product for sodium boltwoodite of  $5.85 \pm 0.26$  from solubility experiments conducted from undersaturation in sodium carbonate solutions. Shvareva et al. (2011) investigated the solubility of synthetic sodium boltwoodite from both under- and oversaturation in solutions in equilibrium with atmospheric  $\text{CO}_2$  and reported a solubility product of 6.07. As noted by Shvareva et al. (2011), the three reported solubility product values are the same, within the uncertainties of the experimental measurements. Consequently, the intermediate solubility product value of 5.85 from Ilton et al. (2006) was included in the database.

Shvareva et al. (2011) experimentally determined a solubility product of 4.12 for boltwoodite. The solubility of boltwoodite was calculated as a function of pH with and without the polymeric uranyl hydroxycarbonate species, and the calculated solubility indicates that omitting the polymeric species results in an acceptable level of agreement between the measured and calculated uranium concentrations up to about pH 8.5 (Figure 2). The experimental and calculated results diverge at higher pH. The solubility calculations were performed assuming that the experimental solutions were in equilibrium with atmospheric  $\text{CO}_2$ . Shvareva et al. (2011) did not measure carbonate concentrations in their experiments, and assumed equilibrium with the atmosphere was achieved as a result of reaction vessels being opened periodically for sampling. It is possible that the high-pH experiments did not sufficiently equilibrate with atmospheric  $\text{CO}_2$  and had  $\text{CO}_2$  partial pressures below the value of  $10^{-3.5}$  atm that was assumed for the calculations. This lower  $\text{CO}_2$  partial pressure would result in the lower uranium concentrations observed in Figure 2.

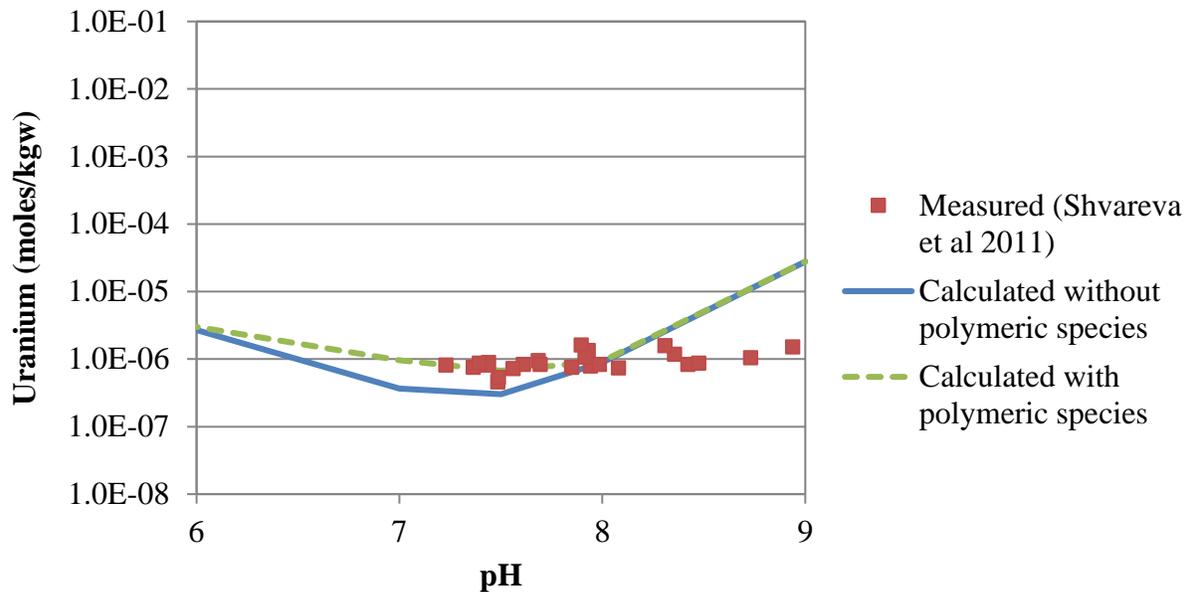


Figure 2. Calculated and measured uranium concentrations in equilibrium with boltwoodite using the solubility product reported by Shvareva et al. (2011)

Nguyen et al (1992) reported a solubility product of 9.42 for uranophane. Three more recent studies have been carried out to determine the solubility of uranophane. Perez et al. (2000) evaluated the dissolution of synthetic uranophane and calculated a solubility product of 11.7. Prikryl (2008) conducted solubility experiments from both under- and oversaturation at approximately pH 6 with synthetic uranophane. The results of the experiments were used with the aqueous speciation of Grenthe et al. (1992) to calculate a solubility product of 11.01 for uranophane. Under the conditions of the Prikryl (2008) experiments, the aqueous speciation of uranium was dominated by  $\text{UO}_2(\text{OH})_2^0$ ,  $\text{UO}_2\text{OH}^+$ ,  $\text{UO}_2^{2+}$  and  $\text{UO}_2\text{CO}_3^0$ . The stability constants for these species changed only slightly from the Grenthe et al. (1992) review to Guillaumont et al. (2003) and these changes are unlikely to have significantly affected the calculations. Shvareva et al. (2011) calculated a solubility product of 10.82 from solubility experiments carried out between pH 3 and 4 using the uranium aqueous speciation data from Guillaumont et al (2003). Although Shvareva et al. (2011) included the uranyl hydroxycarbonate polymeric species in their calculations, these species are not significant at the low pH of the experiments. The reported solubility product values are essentially the same, considering the uncertainties associated with the experimental investigations. Because the experimental study by Prikryl (2008) used well-characterized solid starting materials and was carried out from over- and undersaturation, this solubility product value was included in the database.

Nguyen et al (1992) reported a solubility product of 1.50 for sodium weeksite. Guillaumont et al. (2003) did not select the sodium weeksite data because of uncertainty regarding the

purity of the solid used in the experiments and probable supersaturation of the solutions with respect to silica. However, Guillaumont et al. (2003) suggested this value could be used in scoping calculations with appropriate consideration of the uncertainties in the data. Accordingly, this solubility product value was included in the database for sodium weeksite

#### 3.2.4 Uranium(VI) Phosphate Solids

Guillaumont et al. (2003) selected thermodynamic data for anhydrous uranyl orthophosphate  $[(\text{UO}_2)_3(\text{PO}_4)_2(\text{cr})]$  and hydrogen autunite  $[\text{UO}_2\text{HPO}_4 \cdot 4\text{H}_2\text{O}(\text{cr})]$  that were included in the database. Gorman-Lewis et al. (2009) carried out solubility experiments with autunite  $[\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 3\text{H}_2\text{O}(\text{cr})]$ , uranyl hydrogen phosphate  $[\text{UO}_2\text{HPO}_4 \cdot 3\text{H}_2\text{O}(\text{cr})]$  and uranyl orthophosphate  $[(\text{UO}_2)_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}(\text{cr})]$ . Solubility product values for autunite and uranyl hydrogen phosphate and the  $\Delta H^0_f$  for uranyl hydrogen phosphate were included in the database based on these data.

The uranyl orthophosphate solubility product of -49.36 reported by Gorman-Lewis et al. (2009) was the same as the value selected by Guillaumont et al. (2003). Rai et al. (2005) experimentally determined a solubility product of -49.08 for uranyl orthophosphate, which agrees with the value reported by Gorman-Lewis et al. (2009) within the experimental uncertainties. Armstrong et al. (2010) performed uranyl orthophosphate solubility experiments from undersaturation and determined a solubility product of -49.7, which is in good agreement with the previously reported values. The Rai et al. (2005) solubility product of -49.08 was included in the database because it is consistent with the selected aqueous uranyl phosphate data and because it was determined using data obtained over a range of phosphate concentrations.

Langmuir (1978) reported thermodynamic data for the autunite-group minerals autunite  $[\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$ , hydrogen autunite  $[\text{UO}_2\text{HPO}_4(\text{hyd,cr})]$ , sodium autunite  $[\text{NaUO}_2\text{PO}_4(\text{hyd,cr})]$ , potassium autunite  $[\text{KUO}_2\text{PO}_4(\text{hyd,cr})]$ , uramphite  $[\text{NH}_4\text{UO}_2\text{PO}_4(\text{hyd,cr})]$ , saleeite  $[\text{Mg}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$ , strontium autunite  $[\text{Sr}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$ , uranocircite  $[\text{Ba}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$ , bassetite  $[\text{Fe}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$ , przhhevskite  $[\text{Pb}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$  and torbernite  $[\text{Cu}(\text{UO}_2)_2(\text{PO}_4)_2(\text{hyd,cr})]$ . Langmuir (1978) did not include the moles of water in these mineral formulas because the water contents were poorly established. Langmuir estimated the  $\Delta G^0_f$  data for these uranyl-phosphate solids from solubility, synthesis and exchange data reported by Muto et al. (1968). Although the solubility data were estimated, the Langmuir (1978) data for saleeite, strontium autunite, uranocircite, bassetite, przhhevskite and torbernite were included in the database as the best available for these solids.

Grenthe et al. (1992) corrected the cesium autunite  $[\text{CsUO}_2\text{PO}_4 \cdot x\text{H}_2\text{O}(\text{cr})]$  solubility product from Veselý et al. (1965) for uranyl-phosphate aqueous complexation and ionic strength; this corrected solubility product value is included in the database. Ilton et al.

(2010) experimentally determined the solubility of metatorbernite  $[\text{Cu}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}(\text{cr})]$  using natural mineral samples. The solubility product for this phase was included in the database.

Grenthe et al. (1992) reported a solubility product of -47.3 for sodium autunite based on corrected data from Veselý et al. (1965). Felmy et al. (2005) carried out experiments to determine the solubility of sodium autunite in phosphate and carbonate solutions and confirmed the solubility product value reported by Grenthe et al. (1992), which was included in the database. Gorman-Lewis et al. (2008a) reported a solubility product for potassium autunite based on solubility experiments performed by Pavkovic et al. (1982). Gorman-Lewis et al. (2008a) also reported a solubility product for uramphite based on solubility experiments carried out by Markovic et al. (1988). Both of these solubility product values were included in the database.

The thermodynamic data for uranium solid phases included in the database are summarized in Appendix B.

## 4.0 Phosphate

The wateq4f.dat database included the association constants for the orthophosphoric acid species  $\text{H}_2\text{PO}_4^-$  and  $\text{HPO}_4^{2-}$ . The orthophosphoric acid species association constants in the wateq4f.dat database and the values selected by Guillaumont et al. (2003, Table 8-2) were essentially the same for  $\text{HPO}_4^{2-}$  and  $\text{H}_2\text{PO}_4^-$ . However, the wateq4f.dat database did not include the association constant for  $\text{H}_3\text{PO}_4^0$ . The association constants for  $\text{H}_3\text{PO}_4^0$ ,  $\text{H}_2\text{PO}_4^-$  and  $\text{HPO}_4^{2-}$  from Guillaumont et al. (2003) were included in the database to be consistent with the uranium phosphate aqueous species selected by Guillaumont et al. (2003), which were also included in the database. The orthophosphate and uranium phosphate aqueous species data included in the database are summarized in Appendix A.

The association constants for the major-element phosphate aqueous complexes  $\text{CaPO}_4^-$ ,  $\text{CaHPO}_4^0$ ,  $\text{CaH}_2\text{PO}_4^+$ ,  $\text{FeHPO}_4^0$ ,  $\text{FeH}_2\text{PO}_4^+$ ,  $\text{FeHPO}_4^+$ ,  $\text{FeH}_2\text{PO}_4^{2+}$ ,  $\text{KHPO}_4^-$ ,  $\text{MgPO}_4^-$ ,  $\text{MgHPO}_4^0$ ,  $\text{MgH}_2\text{PO}_4^+$ ,  $\text{NaHPO}_4^-$ ,  $\text{ZnH}_2\text{PO}_4^+$  and  $\text{ZnHPO}_4^0$  (Appendix A) were obtained from the compilation of Vieillard and Tardy (1984).

The solubility product values for the solid phosphates listed in Table 5 were obtained from Vieillard and Tardy (1984). Solubility products for  $\text{MnPO}_4 \cdot 1.5\text{H}_2\text{O}$ ,  $\text{MnHPO}_4 \cdot 3\text{H}_2\text{O}(\text{cr})$ ,  $\text{Mn}_3(\text{PO}_4)_2 \cdot 3\text{H}_2\text{O}(\text{cr})$  and Mn-hureaulite  $[\text{Mn}_5\text{H}_2(\text{PO}_4)_4 \cdot 4\text{H}_2\text{O}(\text{cr})]$  were obtained from Boyle and Lindsay (1985a, 1985b). Solubility data for manganese phosphates  $\text{Mn}_3(\text{PO}_4)_2(\text{s})$  and  $\text{MnHPO}_4(\text{s})$  and for  $\text{Ni}_3(\text{PO}_4)_2(\text{s})$  were retained from the wateq4f.dat database.

Hydroxyapatite  $[\text{Ca}_5(\text{PO}_4)_3\text{OH}(\text{s})]$  solubility data are summarized in Table 6. The range in reported solubility products likely results from differences in crystallinity and methods used to determine solubility. Three values were selected for inclusion in the database,

hydroxyapatite1 based on the results reported by Vieillard and Tardy (1984), hydroxyapatite2 based on the results reported by McDowell et al. (1997) and hydroxyapatite3 based on the estimated solubility provided by Eighmy et al. (1997). Solubility product values for chlorapatite  $[\text{Ca}_5(\text{PO}_4)_3\text{Cl}(\text{cr})]$ , and fluorapatite  $[\text{Ca}_5(\text{PO}_4)_3\text{F}(\text{cr})]$  were obtained from Vieillard and Tardy (1984). The solubility product for fluoride-carbonate-apatite  $[\text{Ca}_{9.31}\text{Na}_{0.36}\text{Mg}_{0.144}(\text{PO}_4)_{4.8}(\text{CO}_3)_{1.2}\text{F}_{2.48}(\text{cr})]$  in the wateq4f.dat database was derived from a study by Chien and Black (1976) and was retained in the database. The phosphate solid solubility data included in the Enchemica.R1.dat database are summarized in Appendix B.

Table 5. Phosphate Solid Phases from Vieillard and Tardy (1984)

Name	Chemical Formula
Variscite	$\text{AlPO}_4 \cdot 2\text{H}_2\text{O}$
Barium hydrogen phosphate	$\text{BaHPO}_4$
Brusheite	$\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$
Monetite	$\text{CaHPO}_4$
$\beta$ -Whitlockite	$\text{Ca}_3(\text{PO}_4)_2$
Octacalcium phosphate	$\text{Ca}_8\text{H}_2(\text{PO}_4)_6 \cdot 5\text{H}_2\text{O}$
$\text{CaAlH}(\text{PO}_4)_2 \cdot 6\text{H}_2\text{O}(\text{cr})$	$\text{CaAlH}(\text{PO}_4)_2 \cdot 6\text{H}_2\text{O}$
$\text{Cu}_3(\text{PO}_4)_2(\text{cr})$	$\text{Cu}_3(\text{PO}_4)_2$
$\text{Cu}_3(\text{PO}_4)_2 \cdot 3\text{H}_2\text{O}(\text{cr})$	$\text{Cu}_3(\text{PO}_4)_2 \cdot 3\text{H}_2\text{O}$
Vivianite	$\text{Fe}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$
Strengite	$\text{FePO}_4 \cdot 2\text{H}_2\text{O}$
$\text{Fe}_3(\text{PO}_4)_2(\text{cr})$	$\text{Fe}_3(\text{PO}_4)_2$
$\text{FePO}_4(\text{cr})$ ,	$\text{FePO}_4$
$\text{KMgPO}_4 \cdot 6\text{H}_2\text{O}(\text{cr})$	$\text{KMgPO}_4 \cdot 6\text{H}_2\text{O}$
Potassium taranakite	$\text{K}_3\text{Al}_5(\text{PO}_4)_8\text{H}_6 \cdot 18\text{H}_2\text{O}$
Minyulite	$\text{KAl}_2(\text{PO}_4)_2\text{OH} \cdot 2\text{H}_2\text{O}$
Farringtonite	$\text{Mg}_3(\text{PO}_4)_2$
Newberryite	$\text{MgHPO}_4 \cdot 3\text{H}_2\text{O}$
Magnesium hydrogen phosphate	$\text{MgHPO}_4$
Bobbierrite	$\text{Mg}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$
$\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O}$	$\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O}$
$\text{Ni}_3(\text{PO}_4)_2(\text{cr})$	$\text{Ni}_3(\text{PO}_4)_2$
$\text{SrHPO}_4(\text{cr})$	$\text{SrHPO}_4$
$\alpha$ -Hopeite	$\text{Zn}_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}$
$\text{Zn}_3(\text{PO}_4)_2(\text{cr})$	$\text{Zn}_3(\text{PO}_4)_2$

Table 6. Hydroxyapatite Solubility Data [ $\text{Ca}_5(\text{PO}_4)_3\text{OH}(\text{s}) + \text{H}^+ = 5\text{Ca}^{2+} + 3\text{PO}_4^{3-} + \text{H}_2\text{O}$ ]

Log <sub>10</sub> K <sub>sp</sub> <sup>0</sup>	Reference	Source
-42.886	Vieillard and Tardy (1984)	Lindsay and Moreno (1960), calculated from thermodynamic data
-44.201	Avnimelech et al. (1973)	Dissolution experiments, synthetic material
-45.531	McDowell et al. (1977)	Dissolution experiments, synthetic material
-38.15	Taylor Eighmy et al. (1997)	Estimated value

## 5.0 Vanadium

Vanadium in natural waters may occur in the +III, +IV, and +V oxidation states. Wanty and Goldhaber (1992) critically reviewed and summarized the thermodynamic data for vanadium aqueous species and solid phases, and these data were used to calculate stability constants and solubility products for inorganic aqueous species and solid phases for the database. Wanty and Goldhaber (1992) also provided thermodynamic data for vanadium(IV) complexes with oxalate ( $\text{C}_2\text{O}_4^{2-}$ ) and acetate ( $\text{CH}_3\text{COO}^-$ ) and for vanadium(V) complexes with oxalate, but these organic complexes were not included in the database.

The vanadium(III) aqueous species  $\text{V}^{3+}$ , the hydrolysis species  $\text{VOH}^{2+}$  and  $\text{V}(\text{OH})_2^+$ , the dimeric species  $\text{V}_2(\text{OH})_2^{4+}$  and the vanadium sulfate complex  $\text{VSO}_4^+$  were included in the database. Wanty and Goldhaber (1992) noted that strong vanadium complexes would be expected to form with chloride by analogy with iron(III). However, no thermodynamic data are available for the vanadium-chloride aqueous species. Solubility data for  $\text{V}(\text{OH})_3(\text{ppt})$  and  $\text{V}_2\text{O}_3(\text{cr})$  were included in the database. Wanty and Goldhaber (1992) noted that  $\text{V}_2\text{O}_3(\text{cr})$  is unlikely to form in natural systems at low temperatures and that  $\text{V}(\text{OH})_3(\text{s})$  and montroseite [ $\text{VOOH}(\text{s})$ ] are more likely solubility controls for vanadium(III).

Thermodynamic data for  $\text{V}(\text{OH})_3(\text{ppt})$  were provided by Wanty and Goldhaber (1992), but they noted that experimental results obtained during vanadium(IV) reduction by  $\text{H}_2\text{S}$  suggested the formation of an amorphous vanadium(III) oxide or hydroxide that is three orders of magnitude less soluble than predicted by the thermodynamic data. The  $\Delta G^\circ_f$  of montroseite has been estimated by analogy with iron and manganese (Wanty and Goldhaber 1992, Langmuir 1979). This value was used to calculate the solubility product for montroseite that was included in the database. However, it is likely that montroseite formed in groundwater systems would contain appreciable amounts of iron and aluminum, which would affect its solubility (Wanty and Goldhaber 1992). The vanadium(III) aqueous speciation and solid phase solubility data included in the database are summarized in Appendix A and Appendix B, respectively.

Vanadium(IV) aqueous species contain the vanadyl ion ( $\text{VO}^{2+}$ ). Thermodynamic data for aqueous vanadyl complexes summarized by Wanty and Goldhaber (1992) and incorporated in the database include the hydrolysis, fluoride, chloride, and sulfate species (Appendix A). Wanty and Goldhaber (1992) estimated the thermodynamic data for the vanadyl-carbonate species  $\text{VOCO}_3^0$  and  $\text{VO}(\text{OH})\text{CO}_3^-$ . Because these data were estimated and their stability is uncertain, they were not included in the database. Wanty and Goldhaber (1992) provided thermodynamic data for a single vanadium(IV) solid phase,  $\text{VO}(\text{OH})_2(\text{s})$ , which was included in the database (Appendix B).

Wanty and Goldhaber (1992) provided thermodynamic data for the vanadium(V) vanadate ion ( $\text{VO}_4^{3-}$ ) and its hydrolysis species, which were included in the database (Appendix A). Vanadium(V) can polymerize, but Wanty and Goldhaber (1992) reported that vanadate ion is likely to predominate over polymeric species at total vanadium concentrations encountered in most natural waters. Thermodynamic data reported by Larson (1995) indicate that polymeric vanadium species such as  $\text{V}_4\text{O}_{12}^{4-}$  become significant at moderate pH values only at vanadium concentrations exceeding 50 mg/L. Langmuir (1978) noted that aqueous uranyl vanadate complexes may be significant by analogy with phosphate. However, thermodynamic data have not been reported for uranyl vanadate complexes.

Because of the relatively high solubility of most vanadium(V) solid phases, few of these solids form in natural systems. However, the minerals carnotite [ $\text{K}_2(\text{UO}_2)_2(\text{VO}_4)_2(\text{cr})$ ] and tyuyamunite [ $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2(\text{cr})$ ] have been observed to form in some systems containing uranium and vanadium (Langmuir 1978). Langmuir (1978) calculated the solubility product for carnotite from a solubility measurement reported by Hostetler and Garrels (1962) for carnotite in carbonate-free water at pH 7. Because the uranium and vanadium aqueous speciation data used by Langmuir (1978) differed from the data in the database, the carnotite solubility product was recalculated using the aqueous speciation data in the database. Langmuir (1978) estimated the  $\Delta G^0_f$  of tyuyamunite by assuming that the  $\Delta G^0_r$  for the reaction of tyuyamunite to carnotite equals the  $\Delta G^0_r$  for the reaction of autunite to potassium autunite. These calculations were repeated to derive a revised  $\Delta G^0_f$  for tyuyamunite based on the thermodynamic data in the database, which was then used to calculate a revised solubility product for tyuyamunite. The solubility data for carnotite and tyuyamunite are provided in Appendix B.

## 6.0 Sulfide

The dissociation constant for hydrogen sulfide ( $\text{H}_2\text{S}$ ) in the wateq4f.dat database (6.99) was modified slightly (to 6.994) to be consistent with the value used by Guillaumont et al. (2003). The dissociation constant for bisulfide ( $\text{HS}^-$ ) and the  $\Delta H^0_f$  values of the hydrogen sulfide and bisulfide dissociation reactions were retained from the wateq4f.dat database. Sulfide mineral solubility data were retained from the wateq4f.dat database, with the exception of mackinawite (Section 11.3).

## 7.0 Arsenic

Under redox conditions typically encountered in groundwater, arsenic may be present in the arsenite (+III) or arsenate (+V) oxidation states. Nordstrom and Archer (2003) provided an evaluation of thermodynamic data for arsenic-oxide and arsenic-sulfide aqueous species and solid phases. The  $\Delta G^0_f$  data provided by Nordstrom and Archer (2003) were used to calculate the stability constants in the database for the arsenic(III) and arsenic(V) hydrolysis species (Appendix A) and solubility products for the solid phases arsenolite [ $\text{As}_2\text{O}_3(\text{cr})$ , cubic], claudetite [ $\text{As}_2\text{O}_3(\text{cr})$ , monoclinic], orpiment [ $\alpha\text{-As}_2\text{S}_3(\text{cr})$ ], realgar [ $\alpha\text{-AsS}(\text{cr})$ ] and native arsenic [ $\text{As}(\text{s})$ ] (Appendix B). Thermodynamic data for metal-arsenate complexes and solubility data for amorphous ferric arsenate [ $\text{FeAsO}_4(\text{am})$ ] and scorodite [ $\text{FeAsO}_4(\text{cr})$ ] were provided by Langmuir et al. (2006); these thermodynamic data are consistent with the thermodynamic data of Nordstrom and Archer (2003) and were included in the database (Appendix A and Appendix B).

Paktunc and Bruggeman (2010) experimentally evaluated the solubility of nanocrystalline scorodite and amorphous ferric arsenate. Their results for nanocrystalline scorodite were virtually identical to the results reported by Langmuir et al. (2006), indicating that crystal size did not affect the solubility of this phase. For amorphous ferric arsenate, the solubility reported by Paktunc and Bruggeman (2010) was between the solubilities reported by Langmuir et al. (2006) for amorphous ferric arsenate and scorodite. The Langmuir et al. (2006) solubilities for amorphous ferric arsenate and crystalline scorodite were included in the database.

There has been considerable disagreement regarding the aqueous arsenic species that form in sulfide solutions. Nordstrom and Archer (2003) selected thermodynamic data for the thioarsenite species  $\text{H}_2\text{As}_3\text{S}_6^-$  and  $\text{AsO}(\text{SH})_2^-$  based on results reported by Helz et al. (1995). Wilkin et al. (2003) used ion chromatography-inductively coupled plasma-mass spectrometry (IC-ICP-MS) to identify the principal aqueous arsenic species in anoxic sulfide systems as arsenite [ $\text{H}_3\text{AsO}_3^0$ ] and a mononuclear thioarsenite with a sulfur/arsenic ratio of 3:1. Wilkin et al. (2003) determined formation constants for the thioarsenite species  $\text{H}_2\text{AsO}_2\text{S}^-$ ,  $\text{H}_2\text{AsOS}_2^-$ ,  $\text{HASOS}_2^{2-}$ ,  $\text{HAS}_3^{2-}$  and  $\text{H}_4\text{AsS}_4^-$ .

Stauder et al. (2005) used IC-ICP-MS to identify the thioarsenate species  $\text{HASO}_3\text{S}^{2-}$ ,  $\text{HASO}_2\text{S}_2^{2-}$ ,  $\text{AsOS}_3^{3-}$  and  $\text{AsS}_4^{3-}$ . The presence of thioarsenate species instead of thioarsenites under reducing, sulfidic conditions was attributed to the high affinity of arsenic for sulfur, and the disproportionation of arsenic(III) to thioarsenate and native arsenic [ $\text{As}(0)$ ]. Stauder et al. (2005) did not provide thermodynamic data for the thioarsenate species identified in their investigation.

One source of uncertainty regarding the presence of either thioarsenites or thioarsenates under reducing, sulfidic conditions is that IC-ICP-MS does not measure the arsenic

oxidation state. Beak et al. (2008) used spectroscopic methods, speciation modeling and chromatography to determine arsenic speciation in sulfidic solutions. X-ray absorption spectroscopy (XAS) indicated that arsenite and monomeric thioarsenites were present in the experimental solutions, with increasing substitution of sulfur for oxygen in the thioarsenites observed as sulfide concentrations increased. The arsenic speciation was similar to the speciation modeled using the thermodynamic data of Wilkin et al. (2003). Helz and Tossell (2008) performed quantum-chemical calculations indicating that complex mixtures of thioarsenite and thioarsenate species might occur simultaneously in sulfidic solutions.

Planer-Friedrich et al. (2010) evaluated arsenite-sulfide solutions by both XAS and IC-ICP-MS. The results of their investigation indicated that thioarsenites are the primary reaction products in the complete absence of oxygen. The presence of thioarsenates in some previous investigations was attributed to oxidation of the thioarsenites by trace amounts of oxygen during IC-ICP-MS analysis and also to reaction during dilution of the samples prior to analysis.

The thermodynamic data provided by Wilkin et al. (2003) are consistent with the spectroscopic measurements performed by Beak et al. (2008) and provide a reasonable description of the solubility data for amorphous  $\text{As}_2\text{S}_3(\text{s})$  measured by Eary (1992) (Figure 3). Consequently, the aqueous stability data for the thioarsenite species provided by Wilkin et al. (2003) were included in the database (Appendix A).

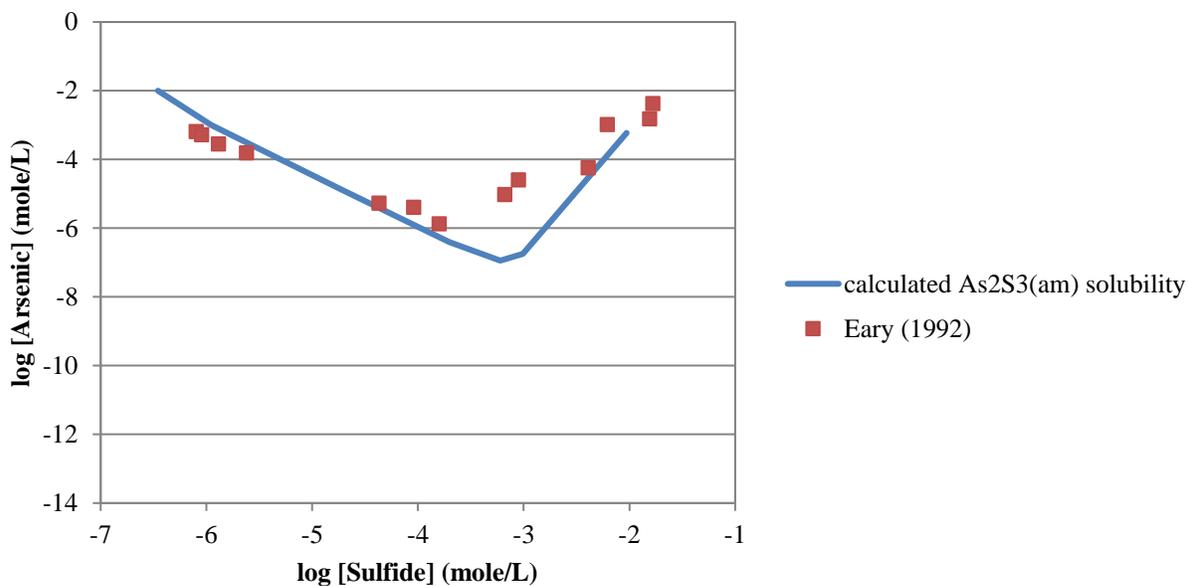


Figure 3. Calculated and measured solubility of  $\text{As}_2\text{S}_3(\text{am})$  as a function of sulfide concentration

## 8.0 Copper

Copper(II) is typically the predominant oxidation state for copper in most groundwater systems. Copper may also occur in the copper(I) oxidation state or as copper metal. Powell et al. (2007) critically reviewed copper(II) aqueous speciation and solid phase data for systems that included a number of common inorganic ligands. Ilton et al. (2010) used the copper(II) hydrolysis and phosphate aqueous speciation data from Powell et al. (2007) in their evaluation of metatorbernite solubility. The Powell et al. (2007) data for copper(II) hydrolysis species and aqueous complexes of copper(II) with chloride, carbonate, bicarbonate, sulfate and phosphate were included in the database. Powell et al. (2007) did not evaluate data for copper(II)-sulfide or copper(II)-fluoride aqueous species, so the aqueous speciation data for these aqueous species were retained from the wateq4f.dat database. The copper(II) aqueous speciation data included in the database are summarized in Appendix A.

Powell et al. (2007) provided critically reviewed solubility product values for tenorite [CuO(s)], Cu(OH)<sub>2</sub>(s), malachite [Cu<sub>2</sub>CO<sub>3</sub>(OH)<sub>2</sub>(s)] and azurite [Cu<sub>3</sub>(CO<sub>3</sub>)<sub>2</sub>(OH)<sub>2</sub>(s)], which were included in the database. The data for the solid phases covellite [CuS(s)], CuF<sub>2</sub>(s), CuF<sub>2</sub>•2H<sub>2</sub>O(s), antlerite [Cu<sub>3</sub>(OH)<sub>4</sub>SO<sub>4</sub>(s)], brochantite [Cu<sub>4</sub>(OH)<sub>6</sub>SO<sub>4</sub>(s)] and langite [Cu<sub>4</sub>(OH)<sub>6</sub>SO<sub>4</sub>•H<sub>2</sub>O(s)] were retained from the wateq4f.dat database. The copper(II) solid phase data included in the database are summarized in Appendix B.

Data for copper(I) aqueous species (Cu<sup>+</sup>, CuCl<sub>2</sub><sup>-</sup> and CuCl<sub>3</sub><sup>2-</sup>) were retained from the wateq4f.dat database (Appendix A). Solubility product values for copper metal and the copper(I) solid phases nantokite [CuCl(s)], CuF(s), cuprite [Cu<sub>2</sub>O(s)], chalcocite [Cu<sub>2</sub>S(s)] and Cu<sub>2</sub>SO<sub>4</sub>(s) were also retained from the wateq4f.dat database (Appendix B).

## 9.0 Nickel

Nickel(II) is the predominant oxidation state in aqueous solutions. Gamsjäger et al. (2005) provided a compilation and review of thermodynamic data for nickel and these data were included in the database for the hydroxyl, fluoride, chloride, bisulfide, sulfate, nitrate, phosphate, arsenate and carbonate aqueous species (Appendix A). Solubility data from Gamsjäger et al. (2005) for nickel oxide, hydroxide, chloride, sulfate and carbonate solid phases were included in the database. Solubility product values for Ni<sub>4</sub>(OH)<sub>6</sub>SO<sub>4</sub>, millerite [NiS(s)], retgersite [NiSO<sub>4</sub>•6H<sub>2</sub>O], nickel silicate, nickel arsenate and nickel phosphate were retained from the wateq4f.dat database (Appendix B).

## 10.0 Selenium

Available selenium aqueous speciation and solids solubility data have been critically reviewed and summarized by Séby et al. (2001) and Olin et al. (2005). Torres et al. (2011) evaluated the speciation of selenium(IV) and selenium(VI) using potentiometric measurements and Berner (2014) updated the Olin et al. (2005) compilation using more recent data. Comparison of the aqueous speciation data from these sources indicates relatively good agreement, so the selenium aqueous speciation data from Berner (2014) was selected for inclusion in the Enchemica.R1.dat database. Because these data are internally consistent with previous OECD thermodynamic data (e.g., Guillaumont et al. 2003), they are also internally consistent with other data in the Enchemica.R1.dat database. The selenium aqueous speciation data are summarized in Appendix A.

Barium-, calcium-, copper-, magnesium-, nickel- and zinc-selenate solid solubilities in the compilations by Berner (2014) or Olin et al. (2005) were included in the database. Ammonium-, sodium-, potassium- and lithium-selenate solids included in the Olin et al. (2005) compilation were omitted because of the high solubilities of these solids. Selenite solids selected by Berner (2014) were incorporated in the database, including nickel-, manganese-, magnesium-, calcium-, strontium-, barium- and iron-selenite solids. The FeSe(s) solubility reported by Elrashidi et al. (1987) was included in the database as the best available, because Olin et al. (2005) and Berner (2014) did not select a solubility for this solid. However, because this value was calculated from thermodynamic data reported by Smith and Martell (1976), it should be considered only an approximate value.

Crystalline elemental selenium can exist in two forms: 1) trigonal, black elemental selenium that is the most stable form of elemental selenium at low temperatures and 2) monoclinic, red elemental selenium that may form as a metastable phase. Solubility data selected for these crystalline forms of elemental selenium by Olin et al. (2005) were included in the database. The solubility of amorphous elemental selenium reported by Iida et al. (2010) was also included in the database. This solubility is consistent with the thermodynamic data of Olin et al. (2005), although Iida et al. (2010) determined slightly different association constants for the polymeric selenium aqueous species  $\text{Se}_3^{-2}$  and  $\text{Se}_4^{-2}$ .

## 11.0 Iron

Nordstrom et al. (1990) reviewed chemical equilibrium data for iron. Lemire et al. (2013) provided a more recent data review and compilation for iron aqueous species and solid phases.

## 11.1 Aqueous Iron Data

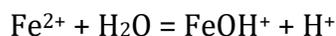
Thermodynamic data for the equilibrium between aqueous ferrous and ferric iron from Nordstrom et al. (1990) are compared to data from Parker and Khodakovskii (1995) and Lemire et al. 2013 (Table 7). These data are in very good agreement, so the data reported by Nordstrom et al. (1990) were included in the Enchemica.R1.dat database.

Table 7. Equilibrium constants, redox potentials and enthalpies of reaction for  
 $\text{Fe}^{2+} = \text{Fe}^{3+} + \text{e}^-$

$\log_{10}K^0$	$E^0$ (volts)	$\Delta H^0_{\text{R}}$ (kjoules/mol)	Source
-13.02	-0.770	40.5	Nordstrom et al. (1990)
-13.01	-0.770	41.0	Parker and Khodakovskii (1995)
-13.05	-0.772	40.2	Lemire et al. 2013

### 11.1.1 Ferrous Iron Aqueous Speciation

Nordstrom et al. (1990) selected an aqueous stability constant of -9.5 for the first hydrolysis reaction of ferrous iron based on the value recommended by Baes and Mesmer (1976):



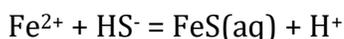
The selected value has an estimated uncertainty of  $\pm 0.16$ . Lemire et al. (2013) recommended a value of  $-9.1 \pm 0.4$  for this reaction, which is equivalent within the stated uncertainties. The Baes and Mesmer (1976) log K of -9.5 was included in the database.

The  $\text{FeF}^+$  aqueous stability constant selected by Lemire et al. (2013) was included in the database because the value selected by Nordstrom et al. (1990) was an estimated value. Lemire et al. (2013) did not select an aqueous stability constant value for  $\text{FeCl}^+$ , so the value recommended by Nordstrom et al. (1990) was included in the database.

The ferrous-carbonate speciation assumed by Nordstrom et al. (1990) was based on an evaluation of aqueous iron carbonate speciation and estimated stability constants by Fouillac and Criaud (1984) that included the species  $\text{FeHCO}_3^+$  and  $\text{FeCO}_3(\text{aq})$ . However, Bruno et al. (1992b) and Silva et al. (2002) found no evidence of significant  $\text{FeHCO}_3^+$  formation in siderite [ $\text{FeCO}_3(\text{s})$ ] solubility experiments. Bruno et al. (1992b) developed stability constants for  $\text{FeCO}_3(\text{aq})$  and  $\text{Fe}(\text{CO}_3)_2^{2-}$  based on the results of their solubility experiments. Lemire et al. (2013) recalculated the  $\Delta G^0_{\text{f}}$  data for these species using the data reported by Bruno et al. (1992b) that yielded stability constants values for  $\text{FeCO}_3(\text{aq})$  and  $\text{Fe}(\text{CO}_3)_2^{2-}$  of 5.27 and 7.03, respectively. These stability constants were included in the database.

Lemire et al. (2013) reviewed the available aqueous speciation data for ferrous iron-sulfate complexes and determined an aqueous stability constant for  $\text{FeSO}_4(\text{aq})$  of  $2.44 \pm 0.03$ , which is similar to the value selected by Nordstrom et al. (1990). Nordstrom et al. (1990) also included a stability constant for  $\text{FeHSO}_4^+$ . Lemire et al. (2013) did not select a value for this stability constant because the ionic strength corrections and cumulative uncertainties for the available data are greater than the stability constant. Consequently, only the stability constant for  $\text{FeSO}_4(\text{aq})$  from Lemire et al. (2013) was included in the database.

Lemire et al. (2013) reviewed the available data for ferrous iron-sulfide complexes and determined that the data were inadequate for selection of recommended values for the aqueous species. However, solubility studies have demonstrated that mackinawite dissolution is pH-independent in aqueous solutions above pH 6, evidence that a neutral ferrous iron-sulfide species such as  $\text{Fe}(\text{HS})_2(\text{aq})$  (Davison et al. 1999) or  $\text{FeS}(\text{aq})$  is formed in solution (Rickard 2006). Rickard (2006) determined that in solutions equilibrated with mackinawite [ $\text{FeS}(\text{s})$ ] only  $\text{Fe}^{2+}$  is present in acid solutions and  $\text{FeS}(\text{aq})$  with minor amounts of  $\text{Fe}^{2+}$  is present in alkaline solutions. Consequently, the  $\text{FeS}(\text{aq})$  species was included in the database, with a stability constant of -2.2 (Rickard 2006) for the reaction:



The sources of the ferrous iron-phosphate and ferrous iron-arsenate stability constant data are summarized in Sections 4.0 and 7.0, respectively.

### 11.1.2 Ferric Iron Aqueous Speciation

Nordstrom et al. (1990) and Lemire et al. (2013) identify the same ferric iron hydrolysis species:  $\text{FeOH}^{2+}$ ,  $\text{Fe}(\text{OH})_2^+$ ,  $\text{Fe}(\text{OH})_3(\text{aq})$ ,  $\text{Fe}(\text{OH})_4^-$ ,  $\text{Fe}_2(\text{OH})_2^{4+}$  and  $\text{Fe}_3(\text{OH})_4^{5+}$  (Table 8). However, Lemire et al. (2013) did not recommend stability constants for  $\text{Fe}(\text{OH})_3(\text{aq})$  and  $\text{Fe}(\text{OH})_4^-$ . Lemire et al. (2013) also did not recommend a log K value for  $\text{Fe}_3(\text{OH})_4^{5+}$  because they found the existence of this species to be questionable.

Stefánsson (2007) performed UV-vis spectrophotometric measurements, potentiometric titrations and solubility measurements to establish hydrolysis constants for aqueous ferric iron and the solubility of 2-line ferrihydrite (Table 8). The hydrolysis results determined by Stefánsson (2007) are consistent with the aqueous stability constants summarized by Nordstrom et al. (1990) for  $\text{FeOH}^{2+}$ ,  $\text{Fe}(\text{OH})_2^+$ ,  $\text{Fe}(\text{OH})_4^-$  and  $\text{Fe}_2(\text{OH})_2^{4+}$ . Consequently, these values were selected for inclusion in the Enchemica.R1.dat database. The maximum aqueous stability constant value established by Stefánsson (2007) for  $\text{Fe}(\text{OH})_3(\text{aq})$  is less than the stability constant selected by Nordstrom et al. (1990) and the smaller limiting value from Stefánsson (2007) was included in the database.

Lemire et al. (2013) did not address complexation of ferric iron by fluoride, so the stability constants selected by Nordstrom et al. (1990) were included in the database. Nordstrom et

al. (1990) and Lemire et al. (2013) determined stability constants for  $\text{FeCl}_2^+$ ,  $\text{FeCl}_2^+$  and  $\text{FeCl}_3(\text{aq})$  that were essentially identical, so the values summarized by Nordstrom et al. (1990) were included in the database. Lemire et al. (2013) also selected a log K value for  $\text{FeCl}_4^-$ . Because this species is only significant in very concentrated chloride solutions, this species was not included in the database.

Table 8. Ferric iron hydrolysis data

Reaction	$\text{Log}_{10}K^0$	$\Delta H^0_{\text{R}}$ (kjoules/mol)	Source
$\text{Fe}^{3+} + \text{H}_2\text{O} = \text{FeOH}^{2+} + \text{H}^+$	-2.19	43.515	Nordstrom et al. (1990)
	-2.19	n.d.	Stefánsson (2007)
	-2.15	36.000	Lemire et al. (2013)
$\text{Fe}^{3+} + 2 \text{H}_2\text{O} = \text{Fe}(\text{OH})_2^+ + 2 \text{H}^+$	-5.67	71.548	Nordstrom et al. (1990)
	-5.76	n.d.	Stefánsson (2007)
	-4.80	n.d.	Lemire et al. 2013
$\text{Fe}^{3+} + 3 \text{H}_2\text{O} = \text{Fe}(\text{OH})_3(\text{aq}) + 3 \text{H}^+$	-12.56	103.766	Nordstrom et al. (1990)
	$\leq -14.3$	n.d.	Stefánsson (2007)
$\text{Fe}^{3+} + 4 \text{H}_2\text{O} = \text{Fe}(\text{OH})_4^- + 4 \text{H}^+$	-21.6	133.473	Nordstrom et al. (1990)
	-21.71	n.d.	Stefánsson (2007)
$2 \text{Fe}^{3+} + 2 \text{H}_2\text{O} = \text{Fe}_2(\text{OH})_2^{4+} + 2 \text{H}^+$	-2.95	56.485	Nordstrom et al. (1990)
	-2.92	n.d.	Stefánsson (2007)
	-2.82	44.000	Lemire et al. 2013
$3 \text{Fe}^{3+} + 4 \text{H}_2\text{O} = \text{Fe}_3(\text{OH})_4^{5+} + 4 \text{H}^+$	-6.3	59.833	Nordstrom et al. (1990)

n.d. = not determined

Nordstrom et al. (1990) did not include aqueous ferric-carbonate aqueous species in their review. However, Lemire et al. (2013) summarized the results of solubility studies by Bruno et al. (1992a) and Grivé (2005). Based on a review of these data, Lemire et al. (2013) recommended stability constants of  $10.7 \pm 2.0$  for  $\text{FeCO}_3\text{OH}(\text{aq})$  and  $24.0 \pm 2.0$  for  $\text{Fe}(\text{CO}_3)_3^{3-}$ . However, inclusion of these ferric-carbonate aqueous species in the database results in the formation of  $\text{FeCO}_3\text{OH}(\text{aq})$  and stabilization of ferric iron solution species even at relatively low Eh values. Consequently, these ferric-carbonate aqueous species were omitted from the database.

Nordstrom et al. (1990) provided aqueous stability constants for the ferric iron-sulfate species  $\text{FeSO}_4^+$ ,  $\text{FeHSO}_4^{2+}$  and  $\text{Fe}(\text{SO}_4)_2^-$ . Lemire et al. (2013) reviewed the available data and determined that sufficient data exist for development of a recommended stability constant for  $\text{FeSO}_4^+$ . Lemire et al. (2013) noted that although the existence of  $\text{Fe}(\text{SO}_4)_2^-$  and  $\text{FeHSO}_4^{2+}$  has been established, these species will be present in solution only at low concentrations and insufficient data are available for selecting stability constants.

Consequently, only the stability constant for  $\text{FeSO}_4^+$  from Lemire et al. (2013) was included in the database.

## 11.2 Iron Solid Phase Data

Lemire et al. (2013) reviewed available iron oxide and hydroxide data, but did not recommend solubilities for these solids. Available data for ferric iron hydroxide minerals were evaluated, and the  $\text{Fe}(\text{OH})_3(\text{am})$  solubility of  $\log_{10}K_{\text{sp}}^0 = 4.81$  in the wateq4f.dat database (Ball and Nordstrom 1991) was retained because it is consistent with the upper limit of the solubility range summarized by Nordstrom et al. (1990). The variation of ferric hydroxide solubility with increasing crystallinity was addressed by incorporating the solubility data reported for 2-line and 6-line ferrihydrite. The two-line ferrihydrite solubility product of 3.5 used in the database is the arithmetic mean of the solubility products from Majzlan et al. (2004a) and Stefánsson (2007), whereas the six-line ferrihydrite solubility product of 3.2 is the value determined by Majzlan et al. (2004a).

Lemire et al. (2013) did not recommend solubilities for hematite, maghemite or magnetite, so the solubility data included in the wateq4f.dat database (Ball and Nordstrom 1991) were retained in the database. Goethite solubility values recommended by Lemire et al. (2013) and by Stefánsson (2007) were the same. This goethite solubility was included in the database, with the  $\Delta H^0_{\text{r}}$  from the wateq4f.dat database (Ball and Nordstrom 1991).

Data selection for iron-arsenate, iron-phosphate and iron-selenide/selenite solids is described in Sections 4.0, 7.0 and 10.0, respectively. Solubility data for bassetite  $[\text{Fe}(\text{UO}_2)_2(\text{PO}_4)_2(\text{s})]$  is addressed in Section 3.2.4.

Data for schwertmannite  $[\text{Fe}_8\text{O}_8(\text{OH})_{4.8}(\text{SO}_4)_{1.6}(\text{s})]$  solubility has been reported in a number of studies (Bigham et al. 1996, Yu et al. 2002, Majzlan et al. 2004a, Sánchez-España et al. 2011). The solubility of naturally occurring schwertmannite reported by Bigham et al. (1996) was confirmed by Sánchez-España et al. (2011), and this solubility data was included in the database.

Nordstrom et al. (1990) included a solubility product for melanterite  $[\text{FeSO}_4 \cdot 7\text{H}_2\text{O}(\text{s})]$  of -2.209. Lemire et al. (2013) took into account more recent data and derived a melanterite solubility product of  $-2.279 \pm 0.23$ , which is the same value within the uncertainty limits. Consequently, the solubility value for melanterite from Nordstrom et al. (1990) was included in the database.

Alpers et al. (1989) cited literature solubility product values for Na-jarosite  $[\text{NaFe}_3(\text{SO}_4)_2(\text{OH})_6(\text{s})]$ , K-jarosite  $[\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6(\text{s})]$  and H-jarosite  $[(\text{H}_3\text{O})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6(\text{s})]$  and experimentally determined the solubility of jarosite solid solution  $[(\text{K}_{0.77}\text{Na}_{0.03}\text{H}_{0.2})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6(\text{s})]$ . The literature solubility product value for Na-jarosite and the experimentally determined solubility product for jarosite solid solution

were included in the database. Baron and Palmer (1996) experimentally determined a slightly lower solubility for K-jarosite and this solubility product was included in the database. Majzlan et al. (2004b) determined a  $\Delta G^0_f$  of -3226.4 kJ/mole for end-member H-jarosite. This  $\Delta G^0_f$  value was used to calculate a solubility product of -5.21, which is in excellent agreement with the literature value of -5.39 from Alpers et al. (1989). The solubility product calculated from the Majzlan et al (2004b)  $\Delta G^0_f$  was included in the database.

Lemire et al. (2013) reviewed the available solubility data for mackinawite and accepted the results of the investigations listed in Table 9. Based on the available data, Lemire et al. (2013) recommended a solubility product of  $3.8 \pm 0.4$ . Two values were included in the database to capture the possible solubility range for FeS(s). FeS(ppt) with solubility product of 4.1 was selected to represent rapidly precipitated, amorphous iron sulfide. This value is similar to the best-estimate value of 4.04 determined by Davison (1991) for amorphous material. Mackinawite with a solubility product of 3.5 was selected to represent the solubility of a more-crystalline FeS(s) solid phase, identified as synthetic nanoparticulate mackinawite (Rickard 2006). Solubility data for pyrite [FeS<sub>2</sub>(s)] and greigite [Fe<sub>3</sub>S<sub>4</sub>(s)] were retained from the wateq4f.dat database (Ball and Nordstrom 1991).

Table 9. Mackinawite solubility data accepted by Lemire et al. (2013, Table IX-3) [FeS(s) + 2 H<sup>+</sup> = Fe<sup>2+</sup> + H<sub>2</sub>S(aq)]

Log <sub>10</sub> K <sub>sp</sub> <sup>0</sup>	Source
3.7 ± 0.5	Berner (1967)
4.1 ± 0.3	Theberge and Luther (1997)
4.0 ± 0.3	Davison et al. (1999)
3.2 ± 0.5	Benning et al. (2000)
3.5 ± 0.3	Rickard (2006)

Lemire et al. (2013) evaluated the available solubility and thermodynamic data for siderite and determined a solubility product of -10.68 for dissolution of crystalline siderite. Lemire et al. (2013) found that the low-temperature results of Bénézech et al (2009) are consistent with their selected data. Bénézech et al (2009) determined a solubility product of -10.9 for crystalline siderite, and this value was included in the database. This solubility product for crystalline siderite is essentially equal to the solubility product of -11.0 determined by Jensen et al. (2002) for dried, crystalline siderite. Because less-crystalline siderite can have a solubility product approximately 0.5 higher than crystalline siderite (Lemire et al. 2013), the solubility product value of -10.4 reported for wet crystals by Jensen et al. (2002) was selected for siderite(ppt).

## 12.0 Manganese

The manganese aqueous speciation and solubility data in the wateq4f.dat database were reviewed. In most groundwater or surface water samples, manganese speciation is dominated by manganese(II) because of the low solubilities of manganese(III) and manganese(IV) solids. The manganese(II) aqueous species most likely to be important in groundwater samples include the hydrolysis species, bicarbonate and carbonate species, sulfate species, chloride and fluoride species.

The manganese hydroxide speciation data for  $\text{MnOH}^+$  and  $\text{Mn(OH)}_3^-$  included in the wateq4f.dat database were obtained from Baes and Mesmer (1976), who also provided stability constants for  $\text{Mn(OH)}_2(\text{aq})$  and  $\text{Mn(OH)}_4^{2-}$ . Wolfram and Krupp (1996) calculated a stability constant for  $\text{Mn(OH)}_2(\text{aq})$  as the only hydrolysis species in rhodochrosite solubility experiments. However, de Abreu et al. (2008) used computational chemistry methods to confirm the speciation established by Baes and Mesmer (1976). Accordingly, data for the  $\text{Mn(OH)}_2(\text{aq})$  and  $\text{Mn(OH)}_4^{2-}$  species in the wateq4f.dat database were retained.

The stability constants for  $\text{MnHCO}_3^+$  and  $\text{MnCO}_3(\text{aq})$  from various studies are summarized in Table 10. Both Morgan (1967) and Lesht and Bauman (1978) experimentally determined stability constants for  $\text{MnHCO}_3^+$ , but the results are questionable because the effects of  $\text{MnCO}_3(\text{aq})$  were not considered in the evaluation of their titration experiments. Palmer and Van Eldik (1983) and Fouillac and Criaud (1984) performed critical reviews of the available data and selected the stability constant for  $\text{MnHCO}_3^+$  from Morgan (1967). Palmer and Van Eldik (1983) selected a  $\text{MnCO}_3(\text{aq})$  stability constant of 4.9 based on a correlation with electronegativity developed by Zirino and Yamamoto (1972), whereas Fouillac and Criaud (1984) selected a stability constant of 4.1 based on a correlation between oxalate and carbonate complexes. Nordstrom et al. (1990) and Ball and Nordstrom (1991) selected the stability constants from Morgan (1967) and Palmer and Eldik (1983) for  $\text{MnHCO}_3^+$  and  $\text{MnCO}_3(\text{aq})$ , respectively.

Wolfram and Krupp (1996) used rhodochrosite solubility experiments to determine stability constants for  $\text{MnHCO}_3^+$  and  $\text{MnCO}_3(\text{aq})$  (Table 10). However, these results were determined assuming the presence of  $\text{Mn(OH)}_2(\text{aq})$  as the only hydrolysis species, which is inconsistent with the hydrolysis species in the database [ $\text{MnOH}^+$  and  $\text{Mn(OH)}_3^-$ ]. Sternbeck (1997) used the  $\text{MnHCO}_3^+$  stability constant from Lesht and Bauman (1978) and estimated a  $\text{MnCO}_3(\text{aq})$  stability constant of 3.8 from rhodochrosite crystal growth experiments. Jensen et al. (2002) used the stability constants from Fouillac and Criaud (1984) in solubility and precipitation rate experiments with rhodochrosite.

Table 10. Manganese(II) aqueous carbonate and bicarbonate speciation data

Reaction	Log <sub>10</sub> K <sup>0</sup>	Source
Mn <sup>2+</sup> + HCO <sub>3</sub> <sup>-</sup> = MnHCO <sub>3</sub> <sup>+</sup>	1.95	Morgan (1967)
	1.27	Lesht and Bauman (1978)
	1.95	Palmer and Van Eldik (1983)
	1.95	Fouillac and Criaud (1984)
	1.95	Nordstrom et al. (1990), Ball and Nordstrom (1991)
	2.2	Wolfram and Krupp (1996)
	1.27	Sternbeck (1997)
	1.95	Jensen et al. (2002)
Mn <sup>2+</sup> + CO <sub>3</sub> <sup>2-</sup> = MnCO <sub>3</sub>	4.9	Palmer and Van Eldik (1983)
	4.1	Fouillac and Criaud (1984)
	4.9	Nordstrom et al. (1990), Ball and Nordstrom (1991)
	4.97	Wolfram and Krupp (1996)
	3.8	Sternbeck (1997)
	4.1	Jensen et al. (2002)

The stability constant of 1.95 determined by Morgan (1967) appears to be the most reliable value for MnHCO<sub>3</sub><sup>+</sup>. There is uncertainty related to the stability constant for MnCO<sub>3</sub>(aq), with either of the values of 4.1 established by Fouillac and Criaud (1984) or 4.9 established by Palmer and Eldick (1983) appearing to be reasonably consistent with the electronegativity or oxalate correlations. In the absence of conclusive justification for revision, the data in the wateq4f.dat database were retained. Stability constants for MnSO<sub>4</sub>(aq), MnCl<sup>+</sup>, MnCl<sub>2</sub>(aq), MnCl<sub>3</sub><sup>-</sup>, and MnF<sup>+</sup> were retained from the wateq4f.dat database. Manganese-arsenate aqueous speciation data (Langmuir et al. 2006) were included as described in Section 7.0.

Solubility data for manganese oxide solids (e.g., manganite and birnessite), manganese sulfides and manganese arsenate were retained from the wateq4f.dat database. Manganese phosphate solid solubilities were reviewed and included in the database as described in Section 4.0. Rhodochrosite solubility data reported by various investigations are summarized in Table 11. Because different aqueous speciation was assumed in the studies by Wolfram and Krupp (1996), Sternbeck (1997) and Jensen et al. (2002), the rhodochrosite solubilities selected by Nordstrom et al. (1990) were retained in the database.

Table 11. Rhodochrosite solubility data [ $\text{MnCO}_3(\text{s}) = \text{Mn}^{2+} + \text{CO}_3^{2-}$ ]

$\text{Log}_{10}K_{\text{sp}}^0$	$\Delta H^0_{\text{R}}$ (kjoules/mol)	Source
-11.13	-5.98	Crystalline, Nordstrom et al. (1990)
-10.39	n.d.	Synthetic, Nordstrom et al. (1990)
-12.19	n.d.	Crystalline, Wolfram and Krupp (1996)
-9.27 to -9.82	n.d.	Disordered, Sternbeck (1997)
-12.51	n.d.	Wet crystals, Jensen et al. (2002)
-11.39	n.d.	Dry crystals, Jensen et al. (2002)

### 13.0 Radium

Radium aqueous speciation and solubility data were summarized by Langmuir and Riese (1985). Their data for the aqueous species  $\text{RaOH}^+$ ,  $\text{RaCl}^+$ ,  $\text{RaSO}_4(\text{aq})$  and  $\text{RaCO}_3(\text{aq})$  and solid phases  $\text{RaCO}_3(\text{s})$  and  $\text{RaSO}_4(\text{s})$  were included in the database (Appendices A and B). Sajih et al. (2014) used the aqueous speciation data from Langmuir and Riese (1985) along with data for the  $\text{Ra}(\text{OH})_2(\text{aq})$  species to develop surface complexation constants for radium. Because the source of the  $\text{Ra}(\text{OH})_2(\text{aq})$  species data is uncertain and this species was shown by Sajih et al (2014) to be unimportant up to pH 10,  $\text{Ra}(\text{OH})_2(\text{aq})$  was not included in the database.

### 14.0 Molybdenum

Low molybdenum concentrations are sometimes observed in groundwater associated with uranium deposits. Aqueous molybdenum is typically present in neutral to alkaline groundwater in the +VI oxidation state as molybdate [ $\text{MoO}_4^{2-}$ ]. The aqueous molybdate species  $\text{H}_2\text{MoO}_4(\text{aq})$ ,  $\text{HMoO}_4^-$  and  $\text{MoO}_4^{2-}$  were included in the database using the aqueous speciation data from EPA (1999). Solid phase data from EPA (1999) were also included for molybdite [ $\text{MoO}_3(\text{s})$ ] and molybdenite [ $\text{MoS}_2(\text{s})$ ].

### 15.0 Aluminum and Zinc Solid Phases

Aluminum solid phases in the wateq4f.dat database were retained in the database. Solubility data reported for microcrystalline gibbsite by Nordstrom et al. (1990) and for hydrobasaluminite by Sánchez-España et al. (2011) were added to the solid phase data (Appendix B).

Zinc phases in the wateq4f.dat database were also retained in the database. The solubility for synthetic hydrozincite reported by Zachara et al. (1989) was added to the database.

## 16.0 Surface Complexation Data

PHREEQC can be used to model constituent sorption on aquifer solids using the surface complexation modeling (SCM) approach (Dzombak and Morel 1990). In this approach, sorbing ions are assumed to react with surface hydroxyl groups in a manner analogous to aqueous complex formation.

This version of the database contains data from the wateq4f.dat database for the weak and strong binding sites of hydrous ferric hydroxide summarized by Dzombak and Morel (1990). Surface complexation data from Dzombak and Morel (1990) were included in the database for the cations calcium, strontium, barium, nickel, copper, zinc, magnesium and manganese. Anion data were included from Dzombak and Morel (1990) for phosphate, arsenate, arsenite, borate ( $H_3BO_3$ ), sulfate, selenate (+VI), selenite (+IV), fluoride and molybdate. Strong-site data for ferrous iron were included from Appelo et al. (2002) and weak-site ferrous iron data were based on data from Liger et al (1999), re-optimized for the Dzombak and Morel model by Appelo et al. (2002).

The Dzombak and Morel (1990) surface complexation constants for uranyl ion were replaced with the constants determined by Mahoney et al. (2009) (Appendix C). Carbonate and bicarbonate surface complexation constants from the wateq4f.dat database were also replaced by values reported by Appelo et al. (2002) (Appendix C). Sajih et al. (2014) performed experiments determining surface complexation constants for radium adsorption onto ferrihydrite, and these data were included in the database (Appendix C). The Dzombak and Morel (1990) surface complexation constants for molybdate were also included in the database.

## 17.0 References

- Alpers, C.N. and D.K. Nordstrom. 1989. Solubility of jarosite solid solutions precipitated from acid mine waters, Iron Mountain, California, U.S.A. *Sci. Géol. Bull.* 42:281-298.
- Appelo, C.A.J., M.J.J. van der Weiden, C. Tournassat, and L. Charlet. 2002. Surface complexation of ferrous iron and carbonate on ferrihydrite and the mobilization of arsenic. *Environmental Science and Technology* 36:3096-3103.
- Armstrong, C.R., A.R. Felmy and S.B. Clark. 2010. Solubility of triuranyl diphosphate tetrahydrate (TDT) and Na autunite at 23 and 50°C. *Radiochimica Acta* 98:549-554.
- Avnimelech, Y., E.C. Moreno and W.E. Brown. 1973. Solubility and surface properties of finely divided hydroxyapatite. *Journal of Research of the National Bureau of Standards – A. Physics and Chemistry* 77A:149-155.
- Baes, C.F. and R.E. Mesmer. 1976. *The Hydrolysis of Cations*. John Wiley & Sons, New York.

Ball, J.W., and D. K. Nordstrom. 1991. *User's Manual for WATEQ4F, with Revised Thermodynamic Data Base and Test Cases for Calculating Speciation of Major, Trace, and Redox Elements in Natural Waters*. Open-File Report 91-183, United States Geological Survey, Menlo Park, California.

Baron, D. and C.D. Palmer. 1996. Solubility of jarosite at 4-35°C. *Geochimica Cosmochimica Acta* 60:185-195.

Beak, D.G., R.T. Wilkin, R.G. Ford and S.D. Kelly. 2008. Examination of arsenic speciation in sulfidic solutions using X-ray absorption spectroscopy. *Environmental Science and Technology* 42:1643-1650.

Bénézeth, P., J.L. Dandurand and J.C. Harrichoury. 2009. Solubility product of siderite (FeCO<sub>3</sub>) as a function of temperature (25-250 °C). *Chemical Geology* 265:3-12.

Berner, R.A. 1967. Thermodynamic stability of sedimentary iron sulphides. *American Journal of Science* 265:773-785.

Berner, U. 2014. *The PSI/Nagra Chemical Thermodynamic Database 12/07 (update of the Nagra/PSI TDB 01/01) Data Selection of Selenium*. Paul Scherrer Institute, TM-44-14-07.

Bigham, J.M., U. Schwertmann, S.J. Traina, R.L. Winland and M. Wolf. 1996. Schwertmannite and the chemical modeling of iron in acid sulfate waters. *Geochimica et Cosmochimica Acta* 60:2111-2121.

Boyle, F.W. and W.L. Lindsay. 1985a. Preparation, X-ray diffraction pattern, and solubility product of manganese(III) phosphate hydrate. *Soil Science Society of America Journal* 49:758-760.

Boyle, F.W. and W.L. Lindsay. 1985b. Diffraction patterns and solubility products of several divalent manganese phosphate compounds. *Soil Science Society of America Journal* 49:761-766.

Bruno, J., W. Stumm, P. Wersin and F. Brandberg. 1992a. On the influence of carbonate in mineral dissolution: I. The thermodynamics and kinetics of hematite dissolution in bicarbonate at T = 25°C. *Geochimica et Cosmochimica Acta* 56:1139-1147.

Bruno, J., P. Wersin and W. Stumm. 1992b. On the influence of carbonate in mineral dissolution: II. The solubility of FeCO<sub>3</sub>(s) at 25°C and 1 atm total pressure. *Geochimica et Cosmochimica Acta* 56:1149-1155.

Chien, S.H. and C.A. Black. 1976. Free energy of formation of carbonate apatites in some phosphate rocks. *Soil Science Society of America Journal* 40:234-239.

- Davis, J.A. and G.P. Curtis. 2003. *Application of Surface Complexation Modeling to Describe Uranium(VI) Adsorption and Retardation at the Uranium Mill Tailings Site at Naturita, Colorado*. NUREG/CR-6820, Prepared for the U.S. Nuclear Regulatory Commission, Washington DC by the U.S. Geological Survey, Menlo Park, California.
- Davison, W. 1991. The solubility of iron sulphides in synthetic and natural waters at ambient temperature. *Aquatic Sciences* 53/4:309-329.
- Davison, W., N. Phillips and B.J. Tabner. 1999. Soluble iron sulfide species in natural waters: Reappraisal of their stoichiometry and stability constants. *Aquatic Sciences* 61:23-43.
- de Abreu, H.A., L. Guimarães and H.A. Duarte. 2008. DFT/PCM investigation of the Mn(II) chemical speciation in aqueous solution. *International Journal of Quantum Chemistry* 108:2467-2475.
- Diaz Arocas, P. and B. Grambow. 1998. Solid-liquid phase equilibria of U(VI) in NaCl solutions. *Geochimica et Cosmochimica Acta* 62:245-263.
- Dong, W., and S.C. Brooks. 2006. Determination of the formation constants of ternary complexes of uranyl and carbonate with alkaline earth metals ( $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Sr^{2+}$ , and  $Ba^{2+}$ ) using anion exchange method. *Environmental Science and Technology* 40:4689-4695.
- Dong, W., and S.C. Brooks. 2008. Formation of aqueous  $MgUO_2(CO_3)_3^{2-}$  complex and uranium anion exchange mechanism onto an exchange resin. *Environmental Science and Technology* 42:1979-1983.
- Dzombak, D.A., and F.M.M. Morel. 1990. *Surface Complexation Modeling, Hydrous Ferric Oxide*. Wiley-Interscience, New York.
- Eary, L.E. 1992. The solubility of amorphous  $As_2S_3$  from 25 to 90°C. *Geochimica et Cosmochimica Acta* 56:2267-2280.
- Eighmy, T.T., B.S. Crannell, L.G. Butler, F.K. Cartledge, E.F. Emery, D. Oblas, J.E. Krzanowski, J. D. Eusden, E.L. Shaw and C.A. Francis. 1997. Heavy metal stabilization in municipal solid waste combustion dry scrubber residue using soluble phosphate. *Environmental Science and Technology* 31:3330-3338.
- Elrashidi, M.A., D.C. Adriano, S.M. Workman and W.L. Lindsay. 1987. Chemical equilibria of selenium in soils: a theoretical development. *Soil Science* 144:141-152.
- EPA (U.S. Environmental Protection Agency). 1999. *MINTEQA2/PRODEFA2, A Geochemical Assessment Model for Environmental Systems: User Manual Supplement for Version 4.0*. Prepared by HydroGeoLogic Inc. and Allison Geoscience Consultants Inc.

- Felmy, A.R., Y. Xia and Z. Wang. 2005. The solubility product of  $\text{NaUO}_2\text{PO}_4 \cdot x\text{H}_2\text{O}$  determined in phosphate and carbonate solutions. *Radiochimica Acta* 93:401-408.
- Fouillac, C. and A. Criaud. 1984. Carbonate and bicarbonate trace metal complexes: critical reevaluation of stability constants. *Geochemical Journal* 18:297-303.
- Gamsjäger, H., J. Bugajski, T. Gajda, R.J. Lemire and W. Preis. 2005. *Chemical Thermodynamics of Nickel*. Nuclear Energy Agency, Organization for Economic Cooperation and Development.
- Giammar, D.E., and J.G. Hering. 2004. Influence of dissolved sodium and cesium on uranyl oxide hydrate solubility. *Environmental Science and Technology* 38:171-179.
- Gorman-Lewis, D., L. Maxeina, J.B. Fein, J.E.S. Szymanowski, P.C. Burns, and A. Navrotsky. 2007. Thermodynamic properties of soddyite from solubility and calorimetry measurements. *Journal of Chemical Thermodynamics* 39:568-575.
- Gorman-Lewis, D., P.C. Burns, and J.B. Fein. 2008a. Review of uranyl mineral solubility measurements. *Journal of Chemical Thermodynamics* 40:335-352.
- Gorman-Lewis, D., J.B. Fein, P.C. Burns, J.E.S. Szymanowski, and J. Converse. 2008b. Solubility measurements of the uranyl oxide hydrate phases metaschoepite, compreignacite, Na-compreignacite, becquerelite, and clarkeite. *Journal of Chemical Thermodynamics* 40:980-990.
- Gorman-Lewis, D., T. Shvareva, K.A. Kubatko, P.C. Burns, D.M. Wellman, B. McNamara, J.E.S. Szymanowski, A. Navrotsky and J.B. Fein. 2009. Thermodynamic properties of autunite, uranyl hydrogen phosphate, and uranyl orthophosphate from solubility and calorimetric measurements. *Environmental Science and Technology* 43:7416-7422.
- Granger, H.C., and C.G. Warren. 1969. Unstable sulfur compounds and the origin of roll-type uranium deposits. *Economic Geology* 64:160-171.
- Grenthe, I., J. Fuger, R.J.M. Konings, R.J. Lemire, A.B. Muller, C. Nguyen-Trung Cregu and H. Wanner. 1992. *Chemical Thermodynamics of Uranium*. Nuclear Energy Agency, Organization for Economic Cooperation and Development.
- Grivé, M. 2005. *The Linkage Between Uranium, Iron and Carbon Cycles. Processes at Interfaces: Evidences from Solution Chemical and Spectroscopic Studies*. PhD Dissertation, Universidad Politécnic de Cataluña.
- Guillaumont, R., T. Fanghänel, V. Neck, J. Fuger, D.A. Palmer, I. Grenthe, and M.H. Rand. 2003. *Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium*. Nuclear Energy Agency, Organization for Economic Cooperation and Development, Elsevier.

Guo, X., S. Szenknect, A. Mesbah, S. Labs, N. Clavier, C. Poinssot, S.V. Ushakov, H. Curtius, D. Bosbach, R.C. Ewing, P.C. Burns, N. Dacheux, and A. Navrotsky. 2015. Thermodynamics of formation of coffinite,  $USiO_4$ . *Proceedings of the National Academy of Sciences* 112:6551:6555.

Gustafsson, J.P., E. Dässman, and M. Bäckström. 2009. Towards a consistent geochemical model for prediction of uranium(VI) removal from groundwater by ferrihydrite. *Applied Geochemistry* 24:454-462.

Helz, G.R., and J.A. Tossell. 2008. Thermodynamic model for arsenic speciation in sulfidic waters: a novel use of *ab initio* computations. *Geochimica et Cosmochimica Acta* 72:4457-4468.

Helz, G.R., J.A. Tossell, J.M. Charnock, R.A.D. Patrick, D.J. Vaughn, and C.D. Garner. 1995. Oligomerization in As(III) sulfide solution: theoretical constraints and spectroscopic evidence. *Geochimica et Cosmochimica Acta* 59:4591-4604.

Hostetler, P.B., and R.M. Garrels. 1962. Transportation and precipitation of uranium and vanadium at low temperatures, with special reference to sandstone-type uranium deposits. *Economic Geology* 57:137-167.

Iida, Y., T. Yamaguchi, T. Tanaka and S. Hakayama. 2010. Solubility of selenium at high ionic strength under anoxic conditions. *Journal of Nuclear Science and Technology* 47:431-438.

Ilton, E.S., C. Liu, W. Yantasee, Z. Wang, D.A. Moore, A.R. Felmy, and J.M. Zachara. 2006. The dissolution of synthetic Na-boltwoodite in sodium carbonate solutions. *Geochimica et Cosmochimica Acta*. 70:4836-4849.

Ilton, E.S., J.M. Zachara, D.A. Moore, J.P. McKinley, A.D. Eckberg, C.L. Cahill and A.R. Felmy. 2010. Dissolution study of metatorbernite: thermodynamic properties and the effect of pH and phosphate. *Environmental Science and Technology* 44:7521-7526.

Jang, J.H., B.A. Dempsey, and W.D. Burgos. 2006. Solubility of schoepite: comparison and selection of complexation constants for U(VI). *Water Research* 40:2738-2746.

Jensen, D.L., J.K. Boddum, J.C. Tjell and T.H. Christensen. 2002. The solubility of rhodochrosite ( $MnCO_3$ ) and siderite in anaerobic aquatic environments. *Applied Geochemistry* 17:503-511.

Kelly, S.D., K.M. Kemner, and S.C. Brooks. 2007. X-ray absorption spectroscopy identifies calcium-uranyl-carbonate complexes at environmental concentrations. *Geochimica et Cosmochimica Acta* 71:821-834.

Langmuir, D. 1978. Uranium solution-mineral equilibria at low temperatures with applications to sedimentary ore deposits. *Geochimica et Cosmochimica Acta* 42:547-569.

- Langmuir, D. 1979. Techniques of estimating thermodynamic properties for some aqueous complexes of geochemical interest. In *Chemical Modeling in Aqueous Systems* (ed. E.A. Jenne), American Chemical Society Symposium Series 93:353-387.
- Langmuir, D. J.J. Mahoney and J. Rowson. 2006. Solubility products of amorphous ferric arsenate and crystalline scorodite ( $\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$ ) and their application to arsenic behavior in buried mine tailings. *Geochimica et Cosmochimica Acta* 70:2942-2956.
- Langmuir, D. and A.C. Riese. 1985. The thermodynamic properties of radium. *Geochimica et Cosmochimica Acta* 49:1593-1601.
- Larson, J. W. 1995. Thermochemistry of vanadium(5+) in aqueous solutions. *J. Chem. Eng. Data* 40:1276-1280.
- Lesht, D. and J.E. Bauman. 1978. Thermodynamics of the manganese(II) bicarbonate system. *Inorganic Chemistry* 17:3332-3334.
- Lemire, R.J., U. Berner, C. Musikas, D.A. Palmer, P. Taylor and O. Tochiyama. 2013. *Chemical Thermodynamics of Iron Part 1*. Nuclear Energy Agency, Organization for Economic Cooperation and Development.
- Lindsay, W.L. and E.C. Moreno. 1960. Phosphate phase equilibria in soils. *Soil Science Society Proceedings* 24:177-182.
- Liger, E., L. Charlet, and P. Van Cappellen. 1999. Surface catalysis of uranium(VI) reduction by iron(II). *Geochimica et Cosmochimica Acta* 63:2939-2955.
- Ludwig, K.R., and R.I. Grauch. 1980. Coexisting coffinite and uraninite in some sandstone-host uranium ores of Wyoming. *Economic Geology* 75:296-302.
- Mahoney, J.J., S.A. Cadle and R.T. Jakubowski. 2009. Uranyl adsorption onto hydrous ferric oxide – a re-evaluation for the diffuse layer model database. *Environmental Science and Technology* 43:9260-9266.
- Majzlan, J., A. Navrotsky and U. Schwertmann. 2004a. Thermodynamics of iron oxides: Part III. Enthalpies of formation and stability of ferrihydrite ( $\sim\text{Fe}(\text{OH})_3$ ), schwertmannite ( $(\sim\text{FeO}(\text{OH})_{3/4}(\text{SO}_4)_{1/8})$ ), and  $\epsilon\text{-Fe}_2\text{O}_3$ . *Geochimica et Cosmochimica Acta* 68:1049-1059.
- Majzlan, J., R. Stevens, J. Boerio-Goates, B.F. Woodfield, A. Navrotsky, P.C. Burns, M.K. Crawford and T.G. Amos. 2004b. Thermodynamic properties, low-temperature heat-capacity anomalies, and single-crystal X-ray refinement of hydronium jarosite,  $(\text{H}_3\text{O})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6$ . *Physics and Chemistry of Minerals* 31:518-531.

Marković, M., N. Pavković, and N.D. Pavković. 1988. Precipitation of  $\text{NH}_4\text{UO}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}$  – solubility and structural comparison with alkali uranyl(2+) phosphates. *Journal of Research of the National Bureau of Standards* 93:557–563.

McDowell, H., T.M. Gregory and W.E. Brown. 1977. Solubility of  $\text{Ca}_5(\text{PO}_4)_3\text{OH}$  in the system  $\text{Ca}(\text{OH})_2\text{-H}_3\text{PO}_4\text{-H}_2\text{O}$  at 5, 15, 25, and 37°C. *Journal of Research of the National Bureau of Standards – A. Physics and Chemistry* 81A:273-281.

Moll, H., G. Geipel, W. Matz, G. Bernhard, and H. Nitsche. 1996. Solubility and speciation of  $(\text{UO}_2)_2\text{SiO}_4 \cdot 2\text{H}_2\text{O}$  in aqueous systems. *Radiochimica Acta* 74: 3-7.

Morgan, J.J. 1967. In *Principles and Applications of Water Chemistry*, S.D. Faust and J.V. Hunter, eds., John Wiley & Sons, pp 561-624.

Muto, T., S. Hirono, and H. Kurata. 1968. *Some Aspects of Fixation of Uranium from Natural Waters*. Japan Atomic Energy Research Inst. Report NSJ Transl. No. 91: From: *Mining Geol.* (Japan) (1965) 15, 287-298 (cited by Langmuir 1978).

Nguyen, S.N., R.J. Silva, H.C. Weed, and J.E. Andrews. 1992. Standard Gibbs free energies of formation at the temperature 303.15 K of four uranyl silicates: soddyite, uranophane, sodium boltwoodite, and sodium weeksite. *Journal of Chemical Thermodynamics* 24:359-376.

Nordstrom, D.K. and D.G. Archer. 2003. Arsenic thermodynamic data and environmental geochemistry. In *Arsenic in Ground Water, Geochemistry and Occurrence* (ed. A.H. Welch and K.G. Stollenwerk), Kluwer Academic Publishers.

Nordstrom, D.K., L.N. Plummer, D. Langmuir, E. Busenberg, H.M. May, B.F. Jones and D.L. Parkhurst, D.L. 1990. Revised chemical equilibrium data for major water-mineral reactions and their limitations. In *Chemical Modeling in Aqueous Systems II* (eds. Bassett, R.L. and D. Melchior), American Chemical Society Symposium Series 416, pp. 398–413.

Olin, Å., B. Noläng, E.G. Osadchii, L.-O. Öhman and E. Rosén. 2005. *Chemical Thermodynamics of Selenium*. Nuclear Energy Agency, Organization for Economic Cooperation and Development.

Paktunc, D. and K. Bruggeman. 2010. Solubility of nanocrystalline scorodite and amorphous ferric arsenate: implications for stabilization of arsenic in mine wastes. *Applied Geochemistry* 25:674-683.

Palmer, D.A. and R. van Eldik. 1983. The chemistry of metal carbonato and carbon dioxide complexes. *Chemical Reviews* 83:651-731.

- Parker, V.B, and I.L. Khodakovskii. 1995. Thermodynamic properties of the aqueous ions (2+ and 3+ of iron and the key compounds of iron. *Journal of Physical Chemistry Reference Data* 24:1699-1745.
- Parkhurst, D.L. and C.A.J. Appelo. 2013. Description of input and examples for PHREEQC version 3 – A computer program for speciation, batch-reaction, one-dimensional transport and inverse geochemical calculations. *U.S. Geological Survey Techniques and Methods, Book 6*. Chapter A43. <http://pubs.usgs.gov/tm/06/a43>.
- Pavkovic, N., M. Markovic, and B. Kojicprodic. 1982. *Croat. Chem. Acta* 55:393–403 (cited by Gorman-Lewis et al. 2008a).
- Perez, I., I. Casas, M. Martin and J. Bruno. 2000. Thermodynamics and kinetics of uranophane dissolution in bicarbonate test solutions. *Geochimica et Cosmochimica Acta* 64:603-608.
- Planer-Friedrich, B., E. Suess, A.C. Scheinost and D. Wallschläger. 2010. Arsenic speciation in sulfidic waters: reconciling contradictory spectroscopic and chromatographic evidence. *Analytical Chemistry* 82:10228-10235.
- Powell, K.J., P.L. Brown, R.H. Byrne, T. Gajda, G. Hefer, S. Sjöberg, and H. Wanner. 2007. Chemical speciation of environmentally significant metals with inorganic ligands. Part 2: the  $\text{Cu}^{2+}$  -  $\text{OH}^-$ ,  $\text{Cl}^-$ ,  $\text{CO}_3^{2-}$ ,  $\text{SO}_4^{2-}$ , and  $\text{PO}_4^{3-}$  systems. *Pure and Applied Chemistry* 79:895-950.
- Prikryl, J.D. 2008. Uranophane dissolution and growth in  $\text{CaCl}_2$ - $\text{SiO}_2(\text{aq})$  test solutions. *Geochimica et Cosmochimica Acta* 72:4508-4520.
- Rai, D., A.R. Felmy, N.J. Hess, V.L. LeGore and D.E. McCready. 2002. Thermodynamics of the  $\text{U(VI)}\text{-Ca}^{2+}\text{-Cl}^-\text{-OH}^-\text{-H}_2\text{O}$  system: Solubility product of becquerelite. *Radiochimica Acta* 90:495-503.
- Rai, D., Y. Xia, L. Rao, N.J. Hess, A.R. Felmy, D.A. Moore and D.E. McCready. 2005. Solubility of  $(\text{UO}_2)_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}$  in  $\text{H}^+\text{-Na}^+\text{-OH}^-\text{-H}_2\text{PO}_4^-\text{-HPO}_4^{2-}\text{-PO}_4^{3-}\text{-H}_2\text{O}$  and its comparison to the analogous  $\text{PuO}_2^{2+}$  system. *Journal of Solution Chemistry* 34:469-498.
- Rickard, D. 2006. The solubility of  $\text{FeS}$ . *Geochimica et Cosmochimica Acta* 70:5779-5789.
- Sajih, M., N.D. Bryan, F.R. Livens, D.J. Vaughan, M. Descostes, V. Phrommavanh, J. Nos and K. Morris. 2014. Adsorption of radium and barium on goethite and ferrihydrite: A kinetic and surface complexation modeling study. *Geochimica et Cosmochimica Acta* 146:150-163.
- Sánchez-España, J. I. Yusta and M. Diez-Ercilla. 2011. Schwertmannite and hydrobasaluminite: A re-evaluation of their solubility and control on the iron and aluminum concentration in acidic pit lakes. *Applied Geochemistry* 26:1752-1774.

Sandino, A. 1991. *Processes Affecting the Mobility of Uranium in Natural Waters*. Ph.D. thesis, The Royal Institute of Technology, Stockholm, Sweden, as cited by Grenthe et al. (1992).

Sandino, A. and J. Bruno. 1992. Solubility of  $(\text{UO}_2)_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}(\text{s})$  and the formation of U(VI) phosphate complexes. Their influence in uranium speciation in natural waters. *Geochimica et Cosmochimica Acta* 56:4135-4145.

Schindler, P., M. Reinert and H. Gämsjäger. 1969. Löslichkeitskonstanten und free bildun senthal pien von  $\text{ZnCO}_3$  and  $\text{Zn}_5\text{OH}_6(\text{CO}_3)_2$  bei 25°C. *Helvetic Chimica Acta* 52:2327-2332 (as cited by Zachara et al. 1989).

Séby, F., M. Potin-Gautier, E. Giffaut, G. Borge, and O.F.X. Donard. 2001. A critical review of thermodynamic data for selenium species at 25°C. *Chemical Geology* 171:173-194.

Shvareva, T.Y., L. Mazeina, D. Gorman-Lewis, P.C. Burns, J.E.S. Szymanowski, J.F. Fein. and A. Navrotsky. 2011. Thermodynamic characterization of boltwoodite and uranophane: Enthalpy of formation and aqueous solubility. *Geochimica et Cosmochimica Acta* 75:5269 - 5282.

Silva, C.A.R., X. Liu and F.J. Millero. Solubility of siderite ( $\text{FeCO}_3$ ) in NaCl solutions. *Journal of Solution Chemistry* 31:97-108.

Smith, R.M. and A.E. Martell. 1976. *Critical Stability Constants, Vol. 4, Inorganic Complexes*. Plenum, New York.

Stauder, S., B. Raue, and F. Sacher. 2005. Thioarsenates in sulfidic waters. *Environmental Science and Technology* 39:5933-5939.

Stefánsson, A. Iron(III) hydrolysis and solubility at 25°C. *Environmental Science and Technology* 41:6117-6123.

Sternbeck, J. 1997. Kinetics of rhodochrosite crystal growth at 25°C: The role of surface speciation. *Geochimica et Cosmochimica Acta* 61:785-793.

Szenknect, S., D.T. Costin, N. Clavier, A. Mesbah, C. Poinssot, P. Vitorge and N. Dacheux. 2013. From uranothorites to coffinite: a solid solution route to the thermodynamic properties of  $\text{USiO}_4$ . *Inorganic Chemistry* 53:6957-6968.

Szenknect, S., A. Mesbah, T. Cordara, N. Clavier, H.-P. Brau, X. Le Goff, C. Poinssot, R.C. Ewing and N. Dacheux. 2016. First experimental determination of the solubility consatnt of coffinite. *Geochimica et Cosmochimica Acta* 181:36-53.

Theberge, S.M. and G.W. Luther. 1997. Determination of the electrochemical properties of a soluble aqueous FeS species present in sulfide solutions. *Aquatic Geochemistry* 3:191-211.

- Torres, J., V. Pintos, L. Gonzatto, S. Dominguez, C. Kremer and E. Kremer. 2011. Selenium chemical speciation in natural waters: protonation and complexation behavior of selenite and selenate in the presence of environmentally relevant cations. *Chemical Geology* 288:32-38.
- Vercouter, T., P. Vitorge, B. Amekraz and C. Moulin. 2008. Stoichiometries and thermodynamic stabilities for aqueous sulfate complexes of U(VI). *Inorganic Chemistry* 47:2180-2189.
- Veselý, V., V. Pekárek, and M. Abbrent. 1965. A study of uranyl phosphates: III. Solubility products of uranyl hydrogen phosphate, uranyl orthophosphate and some alkali uranyl phosphates. *Journal of Inorganic and Nuclear Chemistry* 27:1159-1166.
- Vieillard, P., and Y. Tardy. 1984. Thermodynamic properties of phosphates. In *Phosphate Minerals* (eds. Nriagu, J.D. and P.B. Moore), Springer Verlag, pp. 171-198.
- Wanty, R.B., and M.B. Goldhaber. 1992. Thermodynamics and kinetics of reactions involving vanadium in natural systems: accumulation of vanadium in sedimentary rocks. *Geochimica et Cosmochimica Acta* 56:1471-1483.
- Wilkin, R.T., D. Wallschläger and R.G. Ford. 2003. Speciation of arsenic in sulfidic waters. *Geochemical Transactions* 4:1-7.
- Wolfram, O., and R.E. Krupp. 1996. Hydrothermal solubility of rhodochrosite, Mn(II) speciation, and equilibrium constants. *Geochimica et Cosmochimica Acta* 60:3983-3994.
- Yu, j.-Y., M. Park and J. Kim. 2002. Solubilities of synthetic schwertmannite and ferrihydrite. *Geochemical Journal* 36:119-132.
- Zachara, J.M., J.A. Kittrick, L.S. Dake and J.B. Harsh. 1989. Solubility and surface spectroscopy of zinc precipitates on calcite. *Geochimica et Cosmochimica Acta* 53:9-19.

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference
$UO_2^{+2} + 4H^+ + 2e^- = U^{+4} + 2H_2O$	9.038	-143.860	Guillaumont et al 2003, calculated $\Delta H^0_r$
$UO_2^{+2} + e^- = UO_2^+$	1.484	-6.127	Guillaumont et al 2003, calculated $\Delta H^0_r$
$U^{+4} + H_2O = UOH^{+3} + H^+$	-0.540	46.910	Guillaumont et al 2003
$U^{+4} + 4H_2O = U(OH)_4^0 + 4H^+$	-9.995	109.913	Guillaumont et al 2003, calculated $\Delta H^0_r$
$U^{+4} + F^- = UF^{+3}$	9.420	-5.600	Guillaumont et al 2003
$U^{+4} + 2F^- = UF_2^{+2}$	16.560	-3.500	Guillaumont et al 2003
$U^{+4} + 3F^- = UF_3^+$	21.890	0.500	Guillaumont et al 2003
$U^{+4} + 4F^- = UF_4^0$	26.340	-8.430	Guillaumont et al 2003
$U^{+4} + 5F^- = UF_5^-$	27.730	20.3	Guillaumont et al 2003, $\Delta H^0_r$ Ball and Nordstrom 1991
$U^{+4} + 6F^- = UF_6^{-2}$	29.800	13.8	Guillaumont et al 2003, $\Delta H^0_r$ Ball and Nordstrom 1991
$U^{+4} + Cl^- = UCl^{+3}$	1.720	-19.000	Guillaumont et al 2003
$U^{+4} + Br^- = UBr^{+3}$	1.460	n.a. <sup>1</sup>	Guillaumont et al 2003
$U^{+4} + SO_4^{-2} = USO_4^{+2}$	6.580	8.000	Guillaumont et al 2003
$U^{+4} + 2SO_4^{-2} = U(SO_4)_2^0$	10.510	32.700	Guillaumont et al 2003
$U^{+4} + NO_3^- = UNO_3^{+3}$	1.470	n.a.	Guillaumont et al 2003
$U^{+4} + 2NO_3^- = U(NO_3)_2^{+2}$	2.300	n.a.	Guillaumont et al 2003
$U^{+4} + 4CO_3^{-2} = U(CO_3)_4^{-4}$	35.120	n.a.	Guillaumont et al 2003
$U^{+4} + 5CO_3^{-2} = U(CO_3)_5^{-6}$	34.000	-20.000	Guillaumont et al 2003
$UO_2^{+2} + H_2O = UO_2OH^+ + H^+$	-5.250	43.459	Guillaumont et al 2003, calculated $\Delta H^0_r$
$UO_2^{+2} + 2H_2O = UO_2(OH)_2 + 2H^+$	-12.150	n.a.	Guillaumont et al 2003

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference
$UO_2^{+2} + 3H_2O = UO_2(OH)_3^- + 3H^+$	-20.250	n.a.	Guillaumont et al 2003
$UO_2^{+2} + 4H_2O = UO_2(OH)_4^{-2} + 4H^+$	-32.400	n.a. <sup>1</sup>	Guillaumont et al 2003
$2 UO_2^{+2} + H_2O = (UO_2)_2OH^{+3} + H^+$	-2.700	n.a.	Guillaumont et al 2003
$2UO_2^{+2} + 2H_2O = (UO_2)_2(OH)_2^{+2} + 2H^+$	-5.620	37.595	Guillaumont et al 2003, calculated $\Delta H^0_r$
$3UO_2^{+2} + 4H_2O = (UO_2)_3(OH)_4^{+2} + 4H^+$	-11.900	n.a.	Guillaumont et al 2003
$3UO_2^{+2} + 5H_2O = (UO_2)_3(OH)_5^{+1} + 5H^+$	-15.550	97.064	Guillaumont et al 2003, calculated $\Delta H^0_r$
$3UO_2^{+2} + 7H_2O = (UO_2)_3(OH)_7 + 7H^+$	-32.200	n.a.	Guillaumont et al 2003
$4UO_2^{+2} + 7H_2O = (UO_2)_4(OH)_7^{+1} + 7H^+$	-21.900	n.a.	Guillaumont et al 2003
$UO_2^{+2} + F^- = UO_2F^+$	5.160	1.700	Guillaumont et al 2003
$UO_2^{+2} + 2F^- = UO_2F_2^0$	8.830	2.100	Guillaumont et al 2003
$UO_2^{+2} + 3F^- = UO_2F_3^-$	10.900	2.350	Guillaumont et al 2003
$UO_2^{+2} + 4F^- = UO_2F_4^{-2}$	11.840	0.290	Guillaumont et al 2003
$UO_2^{+2} + Cl^- = UO_2Cl^+$	0.170	8.000	Guillaumont et al 2003
$UO_2^{+2} + 2Cl^- = UO_2Cl_2$	-1.100	15.000	Guillaumont et al 2003
$UO_2^{+2} + Br^- = UO_2Br^+$	0.220	n.a.	Guillaumont et al 2003
$UO_2^{+2} + SO_4^{-2} = UO_2SO_4$	3.290	19.500	Vercouter et al 2008, $\Delta H^0_r$ from Guillaumont et al 2003
$UO_2^{+2} + 2SO_4^{-2} = UO_2(SO_4)_2^{-2}$	4.330	35.100	Vercouter et al 2008, $\Delta H^0_r$ from Guillaumont et al 2003
$UO_2^{+2} + 3SO_4^{-2} = UO_2(SO_4)_3^{-4}$	3.020	n.a.	Guillaumont et al 2003

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference
$UO_2^{+2} + NO_3^- = UO_2NO_3^+$	0.300	n.a.	Guillaumont et al 2003
$UO_2^{+2} + CO_3^{2-} = UO_2CO_3^0$	9.940	5.000	Guillaumont et al 2003
$UO_2^{+2} + 2CO_3^{2-} = UO_2(CO_3)_2^{2-}$	16.610	18.500	Guillaumont et al 2003
$UO_2^{+2} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}$	21.840	-39.200	Guillaumont et al 2003
$3UO_2^{+2} + 6CO_3^{2-} = (UO_2)_3(CO_3)_6^{-6}$	54.000	-62.700	Guillaumont et al 2003
$UO_2^{+2} + CO_3^{2-} + F^- = UO_2CO_3F^-$	13.750	n.a. <sup>1</sup>	Guillaumont et al 2003
$UO_2^{+2} + CO_3^{2-} + 2 F^- = UO_2CO_3F_2^{2-}$	15.570	n.a.	Guillaumont et al 2003
$UO_2^{+2} + CO_3^{2-} + 3 F^- = UO_2CO_3F_3^{3-}$	16.380	n.a.	Guillaumont et al 2003
$UO_2^{+2} + H_4SiO_4^0 = UO_2H_3SiO_4^+ + H^+$	-1.840	n.a.	Guillaumont et al 2003
$Ca^{+2} + UO_2^{+2} + 3CO_3^{2-} = CaUO_2(CO_3)_3^{2-}$	27.180	n.a.	Dong and Brooks 2006
$2Ca^{+2} + UO_2^{+2} + 3CO_3^{2-} = Ca_2UO_2(CO_3)_3^0$	30.700	n.a.	Dong and Brooks 2006
$Sr^{+2} + UO_2^{+2} + 3CO_3^{2-} = SrUO_2(CO_3)_3^{2-}$	26.860	n.a.	Dong and Brooks 2006
$Ba^{+2} + UO_2^{+2} + 3CO_3^{2-} = BaUO_2(CO_3)_3^{2-}$	26.680	n.a.	Dong and Brooks 2006
$2Ba^{+2} + UO_2^{+2} + 3CO_3^{2-} = Ba_2UO_2(CO_3)_3^0$	29.750	n.a.	Dong and Brooks 2006
$Mg^{+2} + UO_2^{+2} + 3CO_3^{2-} = MgUO_2(CO_3)_3^{2-}$	25.800	n.a.	Dong and Brooks 2008
$PO_4^{-3} + 3H^+ = H_3PO_4$	21.702	-9.72	Guillaumont et al. 2003
$PO_4^{-3} + 2H^+ = H_2PO_4^-$	19.562	-18.200	Guillaumont et al. 2003 Table 8-2
$PO_4^{-3} + H^+ = HPO_4^{2-}$	12.350	-14.600	Guillaumont et al. 2003 Table 8-2
$UO_2^{+2} + PO_4^{-3} = UO_2PO_4^-$	11.016	n.a.	Rai et al. 2005
$UO_2^{+2} + H^+ + PO_4^{-3} = UO_2HPO_4^0$	19.59	n.a.	Guillaumont et al. 2003
$UO_2^{+2} + 2H^+ + PO_4^{-3} = UO_2H_2PO_4^+$	22.822	n.a.	Guillaumont et al. 2003
$UO_2^{+2} + 3H^+ + PO_4^{-3} = UO_2H_3PO_4^{+2}$	22.462	n.a.	Guillaumont et al. 2003

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	log <sub>10</sub> K <sup>0</sup> at 25°C, I = 0	ΔH <sup>0</sup> , at 25°C, I = 0 (kJ/mol)	Reference
UO <sub>2</sub> <sup>+2</sup> + 4H <sup>+</sup> + 2PO <sub>4</sub> <sup>-3</sup> = UO <sub>2</sub> (H <sub>2</sub> PO <sub>4</sub> ) <sub>2</sub> <sup>0</sup>	44.044	n.a.	Guillaumont et al. 2003
UO <sub>2</sub> <sup>+2</sup> + 5H <sup>+</sup> + 2PO <sub>4</sub> <sup>-3</sup> = UO <sub>2</sub> (H <sub>2</sub> PO <sub>4</sub> )(H <sub>3</sub> PO <sub>4</sub> ) <sup>+</sup>	45.054	n.a.	Guillaumont et al. 2003
Ca <sup>+2</sup> + PO <sub>4</sub> <sup>-3</sup> = CaPO <sub>4</sub> <sup>-</sup>	6.46	n.a.	Vieillard and Tardy 1984
Ca <sup>+2</sup> + H <sub>2</sub> PO <sub>4</sub> <sup>-</sup> = CaH <sub>2</sub> PO <sub>4</sub> <sup>+</sup>	1.407	n.a.	Vieillard and Tardy 1984
Ca <sup>+2</sup> + HPO <sub>4</sub> <sup>-2</sup> = CaHPO <sub>4</sub> <sup>0</sup>	2.711	n.a.	Vieillard and Tardy 1984
Fe <sup>+2</sup> + H <sub>2</sub> PO <sub>4</sub> <sup>-</sup> = FeH <sub>2</sub> PO <sub>4</sub> <sup>+</sup>	2.7	n.a.	Vieillard and Tardy 1984
Fe <sup>+2</sup> + HPO <sub>4</sub> <sup>-2</sup> = FeHPO <sub>4</sub> <sup>0</sup>	3.604	n.a.	Vieillard and Tardy 1984
Fe <sup>+3</sup> + HPO <sub>4</sub> <sup>-2</sup> = FeHPO <sub>4</sub> <sup>+</sup>	10.886	n.a.	Vieillard and Tardy 1984
Fe <sup>+3</sup> + H <sub>2</sub> PO <sub>4</sub> <sup>-</sup> = FeH <sub>2</sub> PO <sub>4</sub> <sup>+</sup>	5.43	n.a.	Vieillard and Tardy 1984
K <sup>+</sup> + HPO <sub>4</sub> <sup>-2</sup> = KHPO <sub>4</sub> <sup>-</sup>	1.089	n.a.	Vieillard and Tardy 1984
Mg <sup>+2</sup> + HPO <sub>4</sub> <sup>-2</sup> = MgHPO <sub>4</sub> <sup>0</sup>	2.909	n.a.	Vieillard and Tardy 1984
Mg <sup>+2</sup> + PO <sub>4</sub> <sup>-3</sup> = MgPO <sub>4</sub> <sup>-</sup>	6.586	n.a.	Vieillard and Tardy 1984
Mg <sup>+2</sup> + H <sub>2</sub> PO <sub>4</sub> <sup>-</sup> = MgH <sub>2</sub> PO <sub>4</sub> <sup>+</sup>	1.508	n.a.	Vieillard and Tardy 1984
Na <sup>+</sup> + HPO <sub>4</sub> <sup>-2</sup> = NaHPO <sub>4</sub> <sup>-</sup>	1.2	n.a.	Vieillard and Tardy 1984
Zn <sup>+2</sup> + H <sub>2</sub> PO <sub>4</sub> <sup>-</sup> = ZnH <sub>2</sub> PO <sub>4</sub> <sup>+</sup>	1.6	n.a.	Vieillard and Tardy 1984
Zn <sup>+2</sup> + HPO <sub>4</sub> <sup>-</sup> = ZnHPO <sub>4</sub>	3.3	n.a.	Vieillard and Tardy 1984
V <sup>+3</sup> + H <sub>2</sub> O = VOH <sup>+2</sup> + H <sup>+</sup>	-2.477	30.530	Wanty and Goldhaber 1992
V <sup>+3</sup> + 2H <sub>2</sub> O = V(OH) <sub>2</sub> <sup>+</sup> + 2H <sup>+</sup>	-6.337	n.a.	Wanty and Goldhaber 1992
2V <sup>+3</sup> + 2H <sub>2</sub> O = V <sub>2</sub> (OH) <sub>2</sub> <sup>+4</sup> + 2H <sup>+</sup>	-3.920	46.060	Wanty and Goldhaber 1992
V <sup>+3</sup> + SO <sub>4</sub> <sup>-2</sup> = VSO <sub>4</sub> <sup>+</sup>	3.362	n.a.1	Wanty and Goldhaber 1992

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	log <sub>10</sub> K <sup>0</sup> at 25°C, I = 0	ΔH <sup>0</sup> <sub>r</sub> at 25°C, I = 0 (kJ/mol)	Reference
V <sup>+3</sup> + H <sub>2</sub> O = VO <sup>+2</sup> + e <sup>-</sup> + 2H <sup>+</sup>	-5.700	59.430	Wanty and Goldhaber 1992
VO <sup>+2</sup> + H <sub>2</sub> O = VO(OH) <sup>+</sup> + H <sup>+</sup>	-5.665	55.030	Wanty and Goldhaber 1992
2VO <sup>+2</sup> + 2H <sub>2</sub> O = (VOOH) <sub>2</sub> <sup>+2</sup> + 2H <sup>+</sup>	-6.670	110.860	Wanty and Goldhaber 1992
VO <sup>+2</sup> + F <sup>-</sup> = VOF <sup>+</sup>	2.991	n.a.	Wanty and Goldhaber 1992
VO <sup>+2</sup> + 2F <sup>-</sup> = VOF <sub>2</sub> <sup>0</sup>	4.126	n.a.	Wanty and Goldhaber 1992
VO <sup>+2</sup> + 3F <sup>-</sup> = VOF <sub>3</sub> <sup>-</sup>	5.435	n.a.	Wanty and Goldhaber 1992
VO <sup>+2</sup> + 4F <sup>-</sup> = VOF <sub>4</sub> <sup>-2</sup>	5.519	n.a.	Wanty and Goldhaber 1992
VO <sup>+2</sup> + Cl <sup>-</sup> = VOCl <sup>+</sup>	-0.108	n.a.	Wanty and Goldhaber 1992
VO <sup>+2</sup> + SO <sub>4</sub> <sup>-2</sup> = VOSO <sub>4</sub> <sup>0</sup>	2.557	15.940	Wanty and Goldhaber 1992
V <sup>+3</sup> + 4H <sub>2</sub> O = VO <sub>4</sub> <sup>-3</sup> + 2 e <sup>-</sup> + 8H <sup>+</sup>	-50.912	333.520	Wanty and Goldhaber 1992, Langmuir 1978
VO <sub>4</sub> <sup>-3</sup> + H <sup>+</sup> = HVO <sub>4</sub> <sup>-2</sup>	13.190	-90.000	Wanty and Goldhaber 1992
VO <sub>4</sub> <sup>-3</sup> + 2H <sup>+</sup> = H <sub>2</sub> VO <sub>4</sub> <sup>-</sup>	21.072	-100.000	Wanty and Goldhaber 1992
VO <sub>4</sub> <sup>-3</sup> + 3H <sup>+</sup> = H <sub>3</sub> VO <sub>4</sub> <sup>0</sup>	24.575	-140.000	Wanty and Goldhaber 1992
VO <sub>4</sub> <sup>-3</sup> + 4H <sup>+</sup> = H <sub>4</sub> VO <sub>4</sub> <sup>+</sup>	28.078	-150.000	Wanty and Goldhaber 1992
H <sub>3</sub> AsO <sub>4</sub> <sup>0</sup> + 2H <sup>+</sup> + 2e <sup>-</sup> = H <sub>3</sub> AsO <sub>3</sub> <sup>0</sup> + H <sub>2</sub> O	19.342	-124.740	Nordstrom and Archer 2003
H <sub>3</sub> AsO <sub>3</sub> <sup>0</sup> = H <sub>2</sub> AsO <sub>3</sub> <sup>-</sup> + H <sup>+</sup>	-9.174	27.62	Nordstrom and Archer 2003
H <sub>3</sub> AsO <sub>3</sub> <sup>0</sup> = HAsO <sub>3</sub> <sup>-2</sup> + 2H <sup>+</sup>	-23.233	59.40	Nordstrom and Archer 2003, ΔH <sup>0</sup> <sub>r</sub> from Langmuir et al. 2006

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference
$H_3AsO_3^0 = AsO_3^{-3} + 3H^+$	-38.227	85.80	Nordstrom and Archer 2003, $\Delta H^0_r$ from Langmuir et al. 2006
$H_3AsO_4^0 = H_2AsO_4^- + H^+$	-2.295	-7.970	Nordstrom and Archer 2003
$H_3AsO_4^0 = HAsO_4^{-2} + 2H^+$	-9.288	-4.960	Nordstrom and Archer 2003
$H_3AsO_4^0 = AsO_4^{-3} + 3H^+$	-21.089	13.240	Nordstrom and Archer 2003
$H_3AsO_3^0 + H_2S = H_3AsO_2S^0 + H_2O$	1.700	n.a. <sup>1</sup>	Wilkin et al. 2003
$H_3AsO_3^0 + H_2S = H_2AsO_2S^- + H^+ + H_2O$	-3.450	n.a.	Wilkin et al. 2003
$H_3AsO_3^0 + 2H_2S = HAsOS_2^{-2} + 2H_2O + 2H^+$	-5.380	n.a.	Wilkin et al. 2003
$H_3AsO_3^0 + 3H_2S = HAsS_3^{-2} + 3H_2O + 2H^+$	-0.590	n.a.	Wilkin et al. 2003
$H_3AsO_3^0 + 3H_2S = AsS_3^{-3} + 3H^+ + 3H_2O$	-8.480	n.a.	Wilkin et al. 2003
$H_3AsO_3^0 + 4H_2S = H_4AsS_4^- + 3H_2O + H^+$	8.580	n.a.	Wilkin et al. 2003
$Ca^{+2} + H_3AsO_4^0 = CaH_2AsO_4^+ + H^+$	-1.240	n.a.	Langmuir et al. 2006
$Ca^{+2} + H_3AsO_4^0 = CaHAsO_4^0 + 2H^+$	-6.600	n.a.	Langmuir et al. 2006
$Ca^{+2} + H_3AsO_4^0 = CaAsO_4^- + 3H^+$	-14.870	n.a.	Langmuir et al. 2006
$Mg^{+2} + H_3AsO_4^0 = MgH_2AsO_4^+ + H^+$	-0.780	n.a.	Langmuir et al. 2006
$Mg^{+2} + H_3AsO_4^0 = MgHAsO_4^0 + 2H^+$	-6.430	n.a.	Langmuir et al. 2006
$Mg^{+2} + H_3AsO_4^0 = MgAsO_4^- + 3H^+$	-14.750	n.a.	Langmuir et al. 2006
$Fe^{+3} + H_3AsO_4^0 = FeH_2AsO_4^{+2} + H^+$	1.740	n.a.	Langmuir et al. 2006
$Fe^{+3} + H_3AsO_4^0 = FeHAsO_4^+ + 2H^+$	0.570	n.a.	Langmuir et al. 2006
$Fe^{+3} + H_3AsO_4^0 = FeAsO_4^0 + 3H^+$	-2.190	n.a.	Langmuir et al. 2006
$Fe^{+2} + H_3AsO_4^0 = FeH_2AsO_4^+ + H^+$	0.380	n.a.	Langmuir et al. 2006

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ , at 25°C, I = 0 (kJ/mol)	Reference
$\text{Fe}^{+2} + \text{H}_3\text{AsO}_4^0 = \text{FeHAsO}_4^0 + 2\text{H}^+$	-5.750	n.a.	Langmuir et al. 2006
$\text{Fe}^{+2} + \text{H}_3\text{AsO}_4^0 = \text{FeAsO}_4^- + 3\text{H}^+$	-14.030	n.a.	Langmuir et al. 2006
$\text{Mn}^{+2} + \text{H}_3\text{AsO}_4^0 = \text{MnHAsO}_4^0 + 2\text{H}^+$	-5.540	n.a.	Langmuir et al. 2006
$\text{Mn}^{+2} + \text{H}_3\text{AsO}_4^0 = \text{MnAsO}_4^- + 3\text{H}^+$	-14.960	n.a.	Langmuir et al. 2006
$\text{Al}^{+3} + \text{H}_3\text{AsO}_4^0 = \text{AlH}_2\text{AsO}_4^{+2} + \text{H}^+$	0.770	n.a. <sup>1</sup>	Langmuir et al. 2006
$\text{Al}^{+3} + \text{H}_3\text{AsO}_4^0 = \text{AlHAsO}_4^+ + 2\text{H}^+$	-2.000	n.a.	Langmuir et al. 2006
$\text{Al}^{+3} + \text{H}_3\text{AsO}_4^0 = \text{AlAsO}_4^0 + 3\text{H}^+$	-6.990	n.a.	Langmuir et al. 2006
$\text{Zn}^{+2} + \text{H}_3\text{AsO}_4^0 = \text{ZnHAsO}_4^0 + 2\text{H}^+$	-6.080	n.a.	Langmuir et al. 2006
$\text{UO}_2^{+2} + \text{H}_3\text{AsO}_4^0 = \text{UO}_2\text{HAsO}_4^0 + 2\text{H}^+$	-1.981	n.a.	Guillaumont et al. 2003
$\text{UO}_2^{+2} + \text{H}_3\text{AsO}_4^0 = \text{UO}_2\text{H}_2\text{AsO}_4^+ + \text{H}^+$	1.219	n.a.	Guillaumont et al. 2003
$\text{UO}_2^{+2} + 2\text{H}_3\text{AsO}_4^0 = \text{UO}_2(\text{H}_2\text{AsO}_4)_2^0 + 2\text{H}^+$	0.048	n.a.	Guillaumont et al. 2003
$\text{Cu}^{+2} + \text{H}_2\text{O} = \text{CuOH}^+ + \text{H}^+$	-7.95	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + 2\text{H}_2\text{O} = \text{Cu}(\text{OH})_2^0 + 2\text{H}^+$	-16.2	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + 3\text{H}_2\text{O} = \text{Cu}(\text{OH})_3^- + 3\text{H}^+$	-26.6	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + 4\text{H}_2\text{O} = \text{Cu}(\text{OH})_4^{2-} + 4\text{H}^+$	-39.74	n.a.	Powell et al. 2007
$2\text{Cu}^{+2} + \text{H}_2\text{O} = \text{Cu}_2\text{OH}_3^+ + \text{H}^+$	-6.4	n.a.	Powell et al. 2007
$2\text{Cu}^{+2} + 2\text{H}_2\text{O} = \text{Cu}_2(\text{OH})_2^{+2} + 2\text{H}^+$	-10.43	17.539	Powell et al. 2007, $\Delta H^0_r$ Ball and Nordstrom 1991
$3\text{Cu}^{+2} + 4\text{H}_2\text{O} = \text{Cu}_3(\text{OH})_4^{+2} + 4\text{H}^+$	-21.1	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + \text{Cl}^- = \text{CuCl}^+$	0.83	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + 2\text{Cl}^- = \text{CuCl}_2^0$	0.6	n.a.	Powell et al. 2007

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ , at 25°C, I = 0 (kJ/mol)	Reference
$\text{Cu}^{+2} + \text{CO}_3^{-2} = \text{CuCO}_3^0$	6.75	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + 2\text{CO}_3^{-2} = \text{Cu}(\text{CO}_3)_2^{-2}$	10.3	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + \text{H}_2\text{O} + \text{CO}_3^{-2} = \text{Cu}(\text{CO}_3)\text{OH}^- + \text{H}^+$	-2.79	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + \text{HCO}_3^- = \text{CuHCO}_3^+$	1.84	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + \text{SO}_4^{-2} = \text{CuSO}_4^0$	2.35	7.3	Powell et al. 2007
$\text{Cu}^{+2} + \text{HPO}_4^{-2} = \text{CuHPO}_4^0$	3.25	n.a. <sup>1</sup>	Powell et al. 2007
$\text{Cu}^{+2} + 2\text{HPO}_4^{-2} = \text{Cu}(\text{HPO}_4)_2^{-2}$	7.36	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + \text{H}_2\text{PO}_4 = \text{CuH}_2\text{PO}_4^+$	1.14	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + 2\text{H}_2\text{PO}_4 = \text{Cu}(\text{H}_2\text{PO}_4)_2^0$	1.9	n.a.	Powell et al. 2007
$\text{Cu}^{+2} + \text{F}^- = \text{CuF}^+$	1.26	6.78	Ball and Nordstrom 1991
$\text{Cu}^{+2} + 3\text{HS}^- = \text{Cu}(\text{HS})_3^-$	25.9	n.a.	Ball and Nordstrom 1991
$\text{Cu}^{+2} + \text{e}^- = \text{Cu}^+$	2.72	6.90	Ball and Nordstrom 1991
$\text{Cu}^+ + 2\text{Cl}^- = \text{CuCl}_2^-$	5.50	-1.76	Ball and Nordstrom 1991
$\text{Cu}^+ + 3\text{Cl}^- = \text{CuCl}_3^{-2}$	5.70	1.09	Ball and Nordstrom 1991
$\text{Ni}^{+2} + \text{H}_2\text{O} = \text{NiOH}^+ + \text{H}^+$	-9.540	53.800	Gamsjäger et al 2005
$\text{Ni}^{+2} + 3\text{H}_2\text{O} = \text{Ni}(\text{OH})_3^- + 3\text{H}^+$	-29.200	n.a.	Gamsjäger et al 2005
$2\text{Ni}^{+2} + \text{H}_2\text{O} = \text{Ni}_2\text{OH}^{+3} + \text{H}^+$	-10.600	45.900	Gamsjäger et al 2005
$4\text{Ni}^{+2} + 4\text{H}_2\text{O} = \text{Ni}_4(\text{OH})_4^{+4} + 4\text{H}^+$	-27.520	190.000	Gamsjäger et al 2005
$\text{Ni}^{+2} + \text{F}^- = \text{NiF}^+$	1.430	9.500	Gamsjäger et al 2005
$\text{Ni}^{+2} + \text{Cl}^- = \text{NiCl}^+$	0.080	n.a.	Gamsjäger et al 2005
$\text{Ni}^{+2} + \text{HS}^- = \text{NiHS}^+$	5.180	n.a.	Gamsjäger et al 2005
$\text{Ni}^{+2} + \text{SO}_4^{-2} = \text{NiSO}_4$	2.350	5.660	Gamsjäger et al 2005

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ , at 25°C, I = 0 (kJ/mol)	Reference
$\text{Ni}^{+2} + \text{NO}_3^- = \text{NiNO}_3^+$	0.500	n.a.	Gamsjäger et al 2005
$\text{Ni}^{+2} + \text{HPO}_4^{-2} = \text{NiHPO}_4$	3.050	n.a.	Gamsjäger et al 2005
$\text{Ni}^{+2} + \text{CO}_3^{-2} = \text{NiCO}_3$	4.200	n.a.	Gamsjäger et al 2005
$\text{SeO}_4^{-2} + 2\text{H}^+ + 2\text{e}^- = \text{SeO}_3^{-2} + \text{H}_2\text{O}$	28.05	n.a. <sup>1</sup>	Berner 2014
$\text{SeO}_3^{-2} + 7\text{H}^+ + 6\text{e}^- = \text{HSe}^- + 3\text{H}_2\text{O}$	53.55	n.a.	Berner 2014
$\text{SeO}_4^{-2} + \text{H}^+ = \text{HSeO}_4^-$	1.75	20.8	Berner 2014
$\text{SeO}_3^{-2} + \text{H}^+ = \text{HSeO}_3^-$	8.36	n.a.	Berner 2014
$\text{HSeO}_3^- + \text{H}^+ = \text{H}_2\text{SeO}_3$	2.64	n.a.	Berner 2014
$\text{HSe}^- + \text{H}^+ = \text{H}_2\text{Se}$	3.85	n.a.	Berner 2014
$\text{HSe}^- = \text{Se}^{-2} + \text{H}^+$	-14.91	n.a.	Berner 2014
$\text{Ni}^{+2} + \text{SeO}_4^{-2} = \text{NiSeO}_4$	2.67	n.a.	Berner 2014
$\text{Mn}^{+2} + \text{SeO}_4^{-2} = \text{MnSeO}_4$	2.43	n.a.	Berner 2014
$\text{UO}_2^{+2} + \text{SeO}_4^{-2} = \text{UO}_2\text{SeO}_4$	2.74	n.a.	Berner 2014
$\text{UO}_2^{+2} + 2\text{SeO}_4^{-2} = \text{UO}_2(\text{SeO}_4)_2^{-2}$	3.1	n.a.	Berner 2014
$\text{Mg}^{+2} + \text{SeO}_4^{-2} = \text{MgSeO}_4$	2.2	n.a.	Berner 2014
$\text{Ca}^{+2} + \text{SeO}_4^{-2} = \text{CaSeO}_4$	2.0	n.a.	Berner 2014
$\text{Zn}^{+2} + \text{SeO}_4^{-2} = \text{ZnSeO}_4$	2.16	n.a.	Olin et al. 2005
$\text{Fe}^{+2} + \text{SeO}_3^{-2} = \text{FeSeO}_3^+$	11.15	n.a.	Berner 2014

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ , at 25°C, I = 0 (kJ/mol)	Reference
$\text{Fe}^{+2} = \text{Fe}^{+3} + \text{e}^-$	-13.02	40.50	Nordstrom et al. 1990
$\text{Fe}^{+2} + \text{H}_2\text{O} = \text{FeOH}^+ + \text{H}^+$	-9.50	55.23	Nordstrom et al. 1990
$\text{Fe}^{+2} + \text{F}^- = \text{FeF}^+$	1.7	n.a. <sup>1</sup>	Lemire et al. 2013
$\text{Fe}^{+2} + \text{Cl}^- = \text{FeCl}^+$	0.14	n.a.	Nordstrom et al. 1990
$\text{Fe}^{+2} + \text{CO}_3^{-2} = \text{FeCO}_3$	5.27	n.a.	Lemire et al. 2013; Bruno et al. 1992
$\text{Fe}^{+2} + 2 \text{CO}_3^{-2} = \text{Fe}(\text{CO}_3)_2^{-2}$	7.03	n.a.	Lemire et al. 2013; Bruno et al. 1992
$\text{Fe}^{+2} + \text{SO}_4^{-2} = \text{FeSO}_4$	2.44	8.4	Lemire et al. 2013
$\text{Fe}^{+3} + \text{H}_2\text{O} = \text{FeOH}^{+2} + \text{H}^+$	-2.19	43.515	Stefánsson 2007, Nordstrom et al. 1990
$\text{Fe}^{+3} + 2 \text{H}_2\text{O} = \text{Fe}(\text{OH})_2^+ + 2\text{H}^+$	-5.76	71.548	Stefánsson 2007, Nordstrom et al. 1990
$\text{Fe}^{+3} + 3 \text{H}_2\text{O} = \text{Fe}(\text{OH})_3 + 3\text{H}^+$	-14.3	103.766	Stefánsson 2007, Nordstrom et al. 1990
$\text{Fe}^{+3} + 4\text{H}_2\text{O} = \text{Fe}(\text{OH})_4^- + 4\text{H}^+$	-21.71	133.473	Stefánsson 2007, Nordstrom et al. 1990
$2\text{Fe}^{+3} + 2\text{H}_2\text{O} = \text{Fe}_2(\text{OH})_2^{+4} + 2\text{H}^+$	-2.92	56.485	Stefánsson 2007, Nordstrom et al. 1990
$\text{Fe}^{+3} + \text{F}^- = \text{FeF}^{+2}$	6.2	11.297	Nordstrom et al. 1990
$\text{Fe}^{+3} + 2\text{F}^- = \text{FeF}_2^+$	10.8	20.084	Nordstrom et al. 1990
$\text{Fe}^{+3} + 3\text{F}^- = \text{FeF}_3$	14.0	22.594	Nordstrom et al. 1990
$\text{Fe}^{+3} + \text{Cl}^- = \text{FeCl}^{+2}$	1.480	23.43	Nordstrom et al. 1990
$\text{Fe}^{+3} + 2\text{Cl}^- = \text{FeCl}_2^+$	2.13	n.a.	Nordstrom et al. 1990
$\text{Fe}^{+3} + 3\text{Cl}^- = \text{FeCl}_3$	1.130	n.a.	Nordstrom et al. 1990
$\text{Fe}^{+3} + \text{SO}_4^{-2} = \text{FeSO}_4^+$	4.250	n.a.	Lemire et al. 2013
$\text{Fe}^{+2} + \text{HS}^- = \text{FeS} + \text{H}^+$	-2.200	n.a.	Rickard 2006

<sup>1</sup> n.a. = not available

Appendix A. Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Sulfide, Manganese, Radium and Molybdenum Aqueous Speciation Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ , at 25°C, I = 0 (kJ/mol)	Reference
$H_2S = HS^- + H^+$	-6.99	22.175	Guillaumont et al. 2003, $\Delta H^0_r$ from Ball and Nordstrom 1991
$HS^- = S^{2-} + H^+$	-12.918	50.626	Ball and Nordstrom 1991
$Mn^{+2} = Mn^{+3} + e^-$	-25.51	107.95	Ball and Nordstrom 1991
$Mn^{+2} + Cl^- = MnCl_+$	0.61	n.a. <sup>1</sup>	Nordstrom et al. 1990
$Mn^{+2} + 2Cl^- = MnCl_2$	0.25	n.a.	Nordstrom et al. 1990
$Mn^{+2} + 3Cl^- = MnCl_3^-$	-0.31	n.a.	Nordstrom et al. 1990
$Mn^{+2} + H_2O = MnOH^+ + H^+$	-10.59	60.25	Ball and Nordstrom 1991
$Mn^{+2} + 3H_2O = Mn(OH)_3^- + 3H^+$	-34.8	n.a.	Ball and Nordstrom 1991
$Mn^{+2} + F^- = MnF^+$	0.84	n.a.	Nordstrom et al. 1990
$Mn^{+2} + SO_4^{2-} = MnSO_4$	2.25	14.1	Nordstrom et al. 1990
$Mn^{+2} + 2NO_3^- = Mn(NO_3)_2$	0.6	-1.66	Ball and Nordstrom 1991
$Mn^{+2} + HCO_3^- = MnHCO_3^+$	1.95	n.a.	Nordstrom et al. 1990
$Mn^{+2} + CO_3^{2-} = MnCO_3$	4.9	n.a.	Nordstrom et al. 1990
$Mn^{+2} + 4H_2O = MnO_4^- + 8H^+ + 5e^-$	-127.824	738.98	Ball and Nordstrom 1991
$Mn^{+2} + 4H_2O = MnO_4^{2-} + 8H^+ + 4e^-$	-118.44	627.68	Ball and Nordstrom 1991
$Ra^{+2} + H_2O = RaOH^+ + H^+$	-13.49	4.6	Sajih et al 2014, Langmuir and Riese 1985
$Ra^{+2} + Cl^- = RaCl^+$	-0.10	2.1	Langmuir and Riese 1985
$Ra^{+2} + CO_3^{2-} = RaCO_3$	2.5	4.5	Langmuir and Riese 1985
$Ra^{+2} + SO_4^{2-} = RaSO_4$	2.75	5.4	Langmuir and Riese 1985
$MoO_4^{2-} + 2H^+ = H_2MoO_4$	8.1636	-26	EPA 1999
$MoO_4^{2-} + H^+ = HMoO_4^-$	4.2988	20	EPA 1999

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data

Reaction	$\log_{10}K_{sp}^0$ at 25°C, I = 0	$\Delta H_r^0$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$UO_2(cr) + 4H^+ = U^{+4} + 2 H_2O$	-4.851	-77.860	Guillaumont et al. 2003, calculated $\Delta H_r^0$	Uraninite
$USiO_4(cr) + 4H^+ = U^{+4} + H_4SiO_4^0$	-8.058	-56.834	Guillaumont et al. 2003 calculated $\Delta H_r^0$	Coffinite
$USiO_4(syn) + 4H^+ = U^{+4} + H_4SiO_4^0$	-5.25	-78.16	Szenknect et al. 2016, Guo et al. (2015)	Synthetic coffinite
$U_4O_9(cr) + 2H^+ = 4UO_2^{+2} + H_2O + 6e^-$	-40.139	150.170	Guillaumont et al. 2003	$U_4O_9(cr)$
$U_3O_8(cr) + 4H^+ = 3UO_2^{+2} + 2H_2O + 2e^-$	-6.575	-53.86	Guillaumont et al. 2003	$\alpha$ - $U_3O_8(cr)$
$UO_2(am,hyd) + 4H^+ = U^{+4} + 2H_2O$	-1.496	n.a. <sup>1</sup>	Guillaumont et al. 2003	Amorphous $UO_2$
$U(HPO_4)_2 \cdot 4H_2O = U^{+4} + 2 H^+ + 2 PO_4^{-3} + 4 H_2O$	-55.194	n.a.	Guillaumont et al. 2003	Hydrous uranous phosphate
$CaU(PO_4)_2 \cdot 2H_2O(cr) = Ca^{+2} + U^{+4} + 2 PO_4^{-3} + 2H_2O$	-52.553	-20.8712	Langmuir 1978	Ningyosite
$U(OH)_2SO_4(cr) = U^{+4} + 2H_2O + SO_4^{-2}$	-3.172	n.a.	Guillaumont et al. 2003	Uranous sulfate
$\gamma$ - $UO_3(cr) + 2H^+ = UO_2^{+2} + H_2O$	7.705	-81.030	Grenthe et al. 1992	$\gamma$ - $UO_3(cr)$
$\beta$ - $UO_2(OH)_2(cr) + 2H^+ = UO_2^{+2} + 2 H_2O$	4.930	-54.460	Guillaumont et al 2003	Schoepite
$UO_3 \cdot 2H_2O(cr) + 2H^+ = UO_2^{+2} + 3 H_2O$	4.808	-85.49	Guillaumont et al. 2003, calculated $\Delta H_r^0$	Metaschoepite(cr)
$UO_3 \cdot 2H_2O(syn) + 2H^+ = UO_2^{+2} + 3 H_2O$	5.600	-85.49	Gorman-Lewis et al. 2008b	Metaschoepite(syn)
$UO_3 \cdot 2H_2O(am) + 2H^+ = UO_2^{+2} + 3 H_2O$	6.330	n.a.	Guillaumont et al. 2003, Sandino and Bruno 1992	Metaschoepite(am)

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{UO}_2\text{CO}_3(\text{cr}) = \text{UO}_2^{+2} + \text{CO}_3^{-2}$	-14.760	-2.928	Guillaumont et al. 2003, calculated $\Delta H^0_r$	Rutherfordine
$\text{UO}_2\text{SO}_4 \cdot 2.5\text{H}_2\text{O}(\text{cr}) = \text{UO}_2^{+2} + \text{SO}_4^{-2} + 2.5\text{H}_2\text{O}$	-1.589	n.a. <sup>1</sup>	Guillaumont et al. 2003	$\text{UO}_2\text{SO}_4 \cdot 2.5\text{H}_2\text{O}(\text{cr})$
$\text{UO}_2\text{SO}_4 \cdot 3\text{H}_2\text{O}(\text{cr}) = \text{UO}_2^{+2} + \text{SO}_4^{-2} + 3\text{H}_2\text{O}$	-0.754	n.a.	Guillaumont et al. 2003	$\text{UO}_2\text{SO}_4 \cdot 3\text{H}_2\text{O}(\text{cr})$
$\text{UO}_2\text{SO}_4 \cdot 3.5\text{H}_2\text{O}(\text{cr}) = \text{UO}_2^{+2} + \text{SO}_4^{-2} + 3.5\text{H}_2\text{O}$	-1.585	n.a.	Guillaumont et al. 2003	$\text{UO}_2\text{SO}_4 \cdot 3.5\text{H}_2\text{O}(\text{cr})$
$\text{Na}(\text{UO}_2)\text{O}(\text{OH})(\text{s}) + 3\text{H}^+ = \text{Na}^+ + \text{UO}_2^{+2} + 2\text{H}_2\text{O}$	9.400	n.a.	Gorman-Lewis et al. 2008b	Clarkeite
$\text{K}_2(\text{UO}_2)_6\text{O}_4(\text{OH})_6 \cdot 7\text{H}_2\text{O}(\text{s}) + 14\text{H}^+ = 2\text{K}^+ + 6\text{UO}_2^{+2} + 17\text{H}_2\text{O}$	35.800	n.a.	Gorman-Lewis et al. 2008b	Compreignacite
$\text{Na}_2(\text{UO}_2)_6\text{O}_4(\text{OH})_6 \cdot 7\text{H}_2\text{O}(\text{s}) + 14\text{H}^+ = 2\text{Na}^+ + 6\text{UO}_2^{+2} + 17\text{H}_2\text{O}$	39.400	-517.39	Gorman-Lewis et al. 2008b	Sodium-compreignacite
$\text{CaU}_6\text{O}_{19} \cdot 11\text{H}_2\text{O}(\text{s}) + 14\text{H}^+ = \text{Ca}^{+2} + 6\text{UO}_2^{+2} + 18\text{H}_2\text{O}$	40.500	-412.740	Guillaumont 2003, Gorman-Lewis et al. 2008b	Becquerelite
$(\text{UO}_2)_2\text{SiO}_4 \cdot 2\text{H}_2\text{O}(\text{s}) + 4\text{H}^+ = 2\text{UO}_2^{+2} + \text{H}_4\text{SiO}_4^0 + 2\text{H}_2\text{O}$	6.430	n.a.	Gorman-Lewis et al. 2007	Soddyite
$\text{Na}(\text{UO}_2)(\text{SiO}_3\text{OH}) \cdot 2\text{H}_2\text{O}(\text{s}) + 3\text{H}^+ = \text{UO}_2^{++} + \text{Na}^+ + \text{H}_4\text{SiO}_4 + 2\text{H}_2\text{O}$	5.85	n.a.	Ilton et al. 2006	Sodium-boltwoodite
$\text{K}(\text{UO}_2)(\text{SiO}_3\text{OH}) \cdot 2\text{H}_2\text{O}(\text{s}) + 3\text{H}^+ = \text{UO}_2^{++} + \text{K}^+ + \text{H}_4\text{SiO}_4 + 2\text{H}_2\text{O}$	4.12	n.a.	Shvareva et al. (2011)	Boltwoodite

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ , at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{Ca}(\text{UO}_2)_2(\text{SiO}_3\text{OH})_2 \cdot 5\text{H}_2\text{O}(\text{s}) + 6\text{H}^+ = \text{Ca}^{+2} + 2\text{UO}_2^{+2} + 2\text{H}_4\text{SiO}_4 + 5\text{H}_2\text{O}$	11.01	n.a.	Prikryl (2008)	Uranophane
$\text{Na}_2(\text{UO}_2)_2(\text{Si}_2\text{O}_5)_3 \cdot 4\text{H}_2\text{O}(\text{s}) + 6\text{H}^+ + 5\text{H}_2\text{O} = 2\text{Na}^+ + 2\text{UO}_2^{+2} + 6\text{H}_4\text{SiO}_4$	1.500	n.a.	Guillaumont et al. 2003, Nguyen et al. 1992	Sodium-weeksite
$(\text{UO}_2)_3(\text{PO}_4)_2(\text{cr}) = 3\text{UO}_2^{+2} + 2\text{PO}_4^{-3}$	-36.318	-134.500	Guillaumont et al 2003, calculated $\Delta H^0_r$	Uranyl orthophosphate, anhydrous
$\text{UO}_2\text{HPO}_4 \cdot 4\text{H}_2\text{O}(\text{cr}) = \text{UO}_2^{+2} + \text{H}^+ + \text{PO}_4^{-3} + 4\text{H}_2\text{O}$	-24.202	23.248	Guillaumont et al 2003, calculated $\Delta H^0_r$	Hydrogen autunite
$\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 3\text{H}_2\text{O}(\text{cr}) = \text{Ca}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + 3\text{H}_2\text{O}$	-48.360	n.a. <sup>1</sup>	Gorman-Lewis et al. 2009	Autunite
$\text{UO}_2\text{HPO}_4 \cdot 3\text{H}_2\text{O}(\text{cr}) = \text{UO}_2^{+2} + \text{H}^+ + \text{PO}_4^{-3} + 3\text{H}_2\text{O}$	-13.170	62.330	Gorman-Lewis et al. 2009, calculated $\Delta H^0_r$	Uranyl hydrogen phosphate
$(\text{UO}_2)_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}(\text{cr}) = 3\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + 4\text{H}_2\text{O}$	-49.080	-30.015	Rai et al 2005, calculated $\Delta H^0_r$	Uranyl orthophosphate
$\text{Mg}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Mg}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + x\text{H}_2\text{O}$	-42.052	-95.695	Langmuir 1978, calculated $\Delta H^0_r$	Saleeite
$\text{Sr}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Sr}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + x\text{H}_2\text{O}$	-42.116	-70.738	Langmuir 1978, calculated $\Delta H^0_r$	Strontium autunite
$\text{Ba}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Ba}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + x\text{H}_2\text{O}$	-43.204	-50.451	Langmuir 1978, calculated $\Delta H^0_r$	Uranocircite

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium,  
Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{Fe}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Fe}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + x\text{H}_2\text{O}$	-42.722	-94.686	Langmuir 1978, calculated $\Delta H^0_r$	Bassetite
$\text{Pb}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Pb}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + x\text{H}_2\text{O}$	-42.767	-54.8284	Langmuir 1978, calculated $\Delta H^0_r$	Przhevalskite
$\text{Cu}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Cu}^{+2} + 2\text{UO}_2^{+2} + 2\text{PO}_4^{-3} + x\text{H}_2\text{O}$	-43.004	-78.771	Langmuir 1978, calculated $\Delta H^0_r$	Torbernite
$\text{Cu}[(\text{UO}_2)(\text{PO}_4)]_2 \cdot 8\text{H}_2\text{O}(\text{cr}) + 2\text{H}^+ = \text{Cu}^{+2} + 2\text{UO}_2^{+2} + 2\text{HPO}_4^{-2} + 8\text{H}_2\text{O}$	-28.0	n.a.	Ilton et al. 2010	Metatorbernite
$\text{CsUO}_2\text{PO}_4 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Cs}^+ + \text{UO}_2^{+2} + \text{PO}_4^{-3} + x\text{H}_2\text{O}$	-24.882	n.a.	Grenthe et al. 1992, Vesely et al. 1965	Cesium autunite
$\text{NaUO}_2\text{PO}_4 \cdot x\text{H}_2\text{O}(\text{cr}) = \text{Na}^+ + \text{UO}_2^{+2} + \text{PO}_4^{-3} + x\text{H}_2\text{O}$	-23.642	-13.078	Grenthe et al. 1992, Vesely et al. 1965, calculated $\Delta H^0_r$ from Langmuir 1978	Sodium autunite
$\text{KUO}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}(\text{cr}) = \text{K}^+ + \text{UO}_2^{+2} + \text{PO}_4^{-3} + 3\text{H}_2\text{O}$	-26.280	13.563	Gorman-Lewis et al. 2008a, Pavkovic et al. 1982, calculated $\Delta H^0_r$ from Langmuir 1978	Potassium autunite
$\text{NH}_4\text{UO}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}(\text{cr}) = \text{NH}_4^+ + \text{UO}_2^{+2} + \text{PO}_4^{-3} + 3\text{H}_2\text{O}$	-26.500	29.423	Gorman-Lewis et al. 2008a, Markovic et al. 1988, calculated $\Delta H^0_r$ from Langmuir 1978	Uramphite

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$K_2(UO_2)_2(VO_4)_2(cr) = 2K^+ + 2UO_2^{+2} + 2VO_4^{-3}$	-56.730	233.023	calculated from Hostetler and Garrels (1962) data, calculated $\Delta H^0_r$	Carnotite
$Ca(UO_2)_2(VO_4)_2(cr) = Ca^{+2} + 2UO_2^{+2} + 2VO_4^{-3}$	-53.006	152.435	recalculated from carnotite solubility, calculated $\Delta H^0_r$	Tyuyamunite
$Mn_3(PO_4)_2(cr) = 3Mn^{+2} + 2PO_4^{-3}$	-23.827	2.120 kcal	retained from wateq4f.dat	$Mn_3(PO_4)_2$
$MnHPO_4 = Mn^{+2} + HPO_4^{-2}$	-12.947	n.a. <sup>1</sup>	retained from wateq4f.dat	$MnHPO_4$
$MnPO_4 \cdot 1.5H_2O(cr) = Mn^{+3} + PO_4^{-3} + 1.5H_2O$	-34.82	n.a.	Boyle and Lindsay 1985a	$MnPO_4 \cdot 1.5H_2O(cr)$
$MnHPO_4 \cdot 3H_2O(cr) = Mn^{+2} + PO_4^{-3} + H^+ + 3H_2O$	-12.585	n.a.	Boyle and Lindsay 1985b	$MnHPO_4 \cdot 3H_2O(cr)$
$Mn_3(PO_4)_2 \cdot 3H_2O(cr) = 3Mn^{+2} + 2PO_4^{-3} + 3H_2O$	-27.32	n.a.	Boyle and Lindsay 1985b	$Mn_3(PO_4)_2 \cdot 3H_2O(cr)$
$Mn_5H_2(PO_4)_4 \cdot 4H_2O(cr) = 5Mn^{+2} + 2H^+ + 4PO_4^{-3} + 4H_2O$	-73.44	n.a. <sup>1</sup>	Boyle and Lindsay 1985b	Mn-Hureaulite
$Zn_3(PO_4)_2 \cdot 4H_2O(cr) = 3Zn^{+2} + 2PO_4^{-3} + 4H_2O$	-35.324	n.a.	Viellard and Tardy 1984	$\alpha$ -Hopeite
$Zn_3(PO_4)_2 = 3Zn^{+2} + 2PO_4^{-3}$	-32.064	n.a.	Viellard and Tardy 1984	$Zn_3(PO_4)_2(cr)$
$Cu_3(PO_4)_2 = 3Cu^{+2} + 2PO_4^{-3}$	-36.914	n.a.	Viellard and Tardy 1984	$Cu_3(PO_4)_2$
$Cu_3(PO_4)_2 \cdot 3H_2O = 3Cu^{+2} + 2PO_4^{-3} + 3H_2O$	-35.144	n.a.	Viellard and Tardy 1984	$Cu_3(PO_4)_2 \cdot 3H_2O$
$Ni_3(PO_4)_2 = 3Ni^{+2} + 2PO_4^{-3}$	-30.304	n.a.	Viellard and Tardy 1984	$Ni_3(PO_4)_2(cr)$
$Fe_3(PO_4)_2 \cdot 8H_2O = 3Fe^{+2} + 2PO_4^{-3} + 8H_2O$	-36.024	n.a.	Viellard and Tardy 1984	Vivianite
$FePO_4 \cdot 2H_2O = Fe^{+3} + PO_4^{-3} + 2H_2O$	-26.572	-2.030 kcal from wateq4f	Viellard and Tardy 1984	Strengite
$Fe_3(PO_4)_2 = 3Fe^{+2} + 2PO_4^{-3}$	-46.134	n.a.	Viellard and Tardy 1984	$Fe_3(PO_4)_2(cr)$
$Mg_3(PO_4)_2 = 3Mg^{+2} + 2PO_4^{-3}$	-23.304	n.a.	Viellard and Tardy 1984	Farringtonite

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{MgHPO}_4 \cdot 3\text{H}_2\text{O} = \text{Mg}^{+2} + \text{H}^+ + \text{PO}_4^{-3} + 3\text{H}_2\text{O}$	-18.182	n.a.	Viellard and Tardy 1984	Newberryite
$\text{MgHPO}_4 = \text{Mg}^{+2} + \text{H}^+ + \text{PO}_4^{-3}$	-18.172	n.a.	Viellard and Tardy 1984	$\text{MgHPO}_4(\text{cr})$
$\text{Mg}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O} = 3 \text{Mg}^{+2} + 2 \text{PO}_4^{-3} + 8 \text{H}_2\text{O}$	-25.224	n.a.	Viellard and Tardy 1984	Bobierite
$\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O} = 3\text{Mg}^{+2} + 2\text{PO}_4^{-3} + 22 \text{H}_2\text{O}$	-23.124	n.a.	Viellard and Tardy 1984	$\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O}(\text{cr})$
$\text{CaHPO}_4 \cdot 2\text{H}_2\text{O} = \text{Ca}^{+2} + \text{H}^+ + \text{PO}_4^{-3} + 2\text{H}_2\text{O}$	-18.91	n.a.	Viellard and Tardy 1984	Brushite
$\text{CaHPO}_4 = \text{Ca}^{+2} + \text{H}^+ + \text{PO}_4^{-3}$	-19.01	n.a.	Viellard and Tardy 1984	Monetite
$\text{Ca}_3(\text{PO}_4)_2 = 3\text{Ca}^{+2} + 2\text{PO}_4^{-3}$	-28.94	n.a.	Viellard and Tardy 1984	$\beta$ -Whitlockite
$\text{Ca}_8\text{H}_2(\text{PO}_4)_6 \cdot 5\text{H}_2\text{O} = 8\text{Ca}^{+2} + 6\text{PO}_4^{-3} + 2\text{H}^+ + 5\text{H}_2\text{O}$	-93.892	n.a.	Viellard and Tardy 1984	Octacalcium phosphate
$\text{SrHPO}_4 = \text{Sr}^{+2} + \text{H}^+ + \text{PO}_4^{-3}$	-19.272	n.a. <sup>1</sup>	Viellard and Tardy 1984	$\text{SrHPO}_4(\text{cr})$
$\text{BaHPO}_4 = \text{Ba}^{+2} + \text{H}^+ + \text{PO}_4^{-3}$	-19.762	n.a.	Viellard and Tardy 1984	$\text{BaHPO}_4(\text{cr})$
$\text{AlPO}_4 \cdot 2\text{H}_2\text{O} = \text{Al}^{+3} + \text{PO}_4^{-3} + 2\text{H}_2\text{O}$	-22.082	n.a.	Viellard and Tardy 1984	Variscite
$\text{FePO}_4 = \text{Fe}^{+3} + \text{PO}_4^{-3}$	-21.902	n.a.	Viellard and Tardy 1984	$\text{FePO}_4(\text{cr})$
$\text{KMgPO}_4 \cdot 6\text{H}_2\text{O} = \text{K}^+ + \text{Mg}^{+2} + \text{PO}_4^{-3} + 6 \text{H}_2\text{O}$	-10.632	n.a.	Viellard and Tardy 1984	$\text{KMgPO}_4 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{CaAlH}(\text{PO}_4)_2 \cdot 6\text{H}_2\text{O} = \text{Ca}^{+2} + \text{Al}^{+3} + 2\text{PO}_4^{-3} + \text{H}^+ + 6 \text{H}_2\text{O}$	-39.024	n.a.	Viellard and Tardy 1984	$\text{CaAlH}(\text{PO}_4)_2 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{K}_3\text{Al}_5(\text{PO}_4)_8\text{H}_6 \cdot 18\text{H}_2\text{O} = 3 \text{K}^+ + 5 \text{Al}^{+3} + 8 \text{PO}_4^{-3} + 6 \text{H}^+ + 18 \text{H}_2\text{O}$	-178.8	n.a.	Viellard and Tardy 1984	$\text{K}_3\text{Al}_5(\text{PO}_4)_8\text{H}_6 \cdot 18\text{H}_2\text{O}(\text{cr})$
$\text{KAl}_2(\text{PO}_4)_2\text{OH} \cdot 2\text{H}_2\text{O} + \text{H}^+ = \text{K}^+ + 2 \text{Al}^{+3} + 2 \text{PO}_4^{-3} + 3 \text{H}_2\text{O}$	-41.024	n.a.	Viellard and Tardy 1984	Minyulite
$\text{Ni}_3(\text{PO}_4)_2 = 3\text{Ni}^{+2} + 2\text{PO}_4^{-3}$	-31.3	n.a.	wateq4f.dat database	$\text{Ni}_3(\text{PO}_4)_2$

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium,  
Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{Ca}_5(\text{PO}_4)_3\text{OH} + \text{H}^+ = 5 \text{Ca}^{+2} + 3 \text{PO}_4^{-3} + \text{H}_2\text{O}$	-42.886	n.a.	Viellard and Tardy 1984 from Lindsay and Moreno 1960	Hydroxyapatite
$\text{Ca}_5(\text{PO}_4)_3\text{OH} + \text{H}^+ = 5 \text{Ca}^{+2} + 3 \text{PO}_4^{-3} + \text{H}_2\text{O}$	-45.531	n.a.	McDowell et al. 1977	Hydroxyapatite
$\text{Ca}_5(\text{PO}_4)_3\text{OH} + \text{H}^+ = 5 \text{Ca}^{+2} + 3 \text{PO}_4^{-3} + \text{H}_2\text{O}$	-38.15	n.a.	Eighmy et al. 1997	Hydroxyapatite
$\text{Ca}_5(\text{PO}_4)_3\text{F} = 5\text{Ca}^{+2} + 3\text{PO}_4^{-3} + \text{F}^-$	-59.636	n.a.	Viellard and Tardy 1984	Fluorapatite(syn)
$\text{Ca}_5(\text{PO}_4)_3\text{Cl} = 5\text{Ca}^{+2} + 3\text{PO}_4^{-3} + \text{Cl}^-$	-53.116	n.a.	Viellard and Tardy 1984	Chlorapatite(syn)
$\text{Ca}_{9.316}\text{Na}_{0.36}\text{Mg}_{0.144}(\text{PO}_4)_{4.8}(\text{CO}_3)_{1.2}\text{F}_{2.48} =$ $9.316\text{Ca}^{+2} + 0.36\text{Na}^+ + 0.144\text{Mg}^{+2} + 4.8\text{PO}_4^{-3}$ $+ 1.2\text{CO}_3^{-2} + 2.48\text{F}^-$	-59.636	n.a.	wateq4f.dat, based on Chien and Black 1976	FCO3-apatite
$\text{VOOH}(\text{s}) + 3\text{H}^+ = \text{V}^{+3} + 2 \text{H}_2\text{O}$	4.950	n.a. <sup>1</sup>	Wanty and Goldhaber 1992, Langmuir 1979	Montroseite
$\text{V}(\text{OH})_3(\text{ppt}) + 3\text{H}^+ = \text{V}^{+3} + 3\text{H}_2\text{O}$	10.128	n.a.	Wanty and Goldhaber 1992	V(OH)3(ppt)
$\text{V}_2\text{O}_3(\text{cr}) + 6\text{H}^+ = 2 \text{V}^{+3} + 3 \text{H}_2\text{O}$	9.900	-158.890	Wanty and Goldhaber 1992	V <sub>2</sub> O <sub>3</sub> (cr)
$\text{VO}(\text{OH})_2(\text{ppt}) + 2\text{H}^+ = \text{VO}^{+2} + 2\text{H}_2\text{O}$	6.110	n.a.	Wanty and Goldhaber 1992	VO(OH) <sub>2</sub> (ppt)
$\text{FeAsO}_4(\text{cr}) + 3\text{H}^+ = \text{Fe}^{+3} + \text{H}_3\text{AsO}_4$	-5.031	n.a.	Langmuir et al. 2006	Scorodite
$\text{FeAsO}_4(\text{am}) + 3\text{H}^+ = \text{Fe}^{+3} + \text{H}_3\text{AsO}_4$	-2.211	n.a.	Langmuir et al. 2006	Amorphous ferric arsenate
$\text{As}_2\text{O}_3(\text{cr}) + 3\text{H}_2\text{O} = 2\text{H}_3\text{AsO}_3$	-1.349	30.040	Nordstrom and Archer 2003	Arsenolite
$\text{As}_2\text{O}_3(\text{cr}) + 3\text{H}_2\text{O} = 2\text{H}_3\text{AsO}_3$	-1.382	28.440	Nordstrom and Archer 2003	Claudetite
$\text{As}_2\text{S}_3(\text{cr}) + 6\text{H}_2\text{O} = 2 \text{H}_3\text{AsO}_3 + 3\text{H}_2\text{S}$	-25.353	200.260	Nordstrom and Archer 2003	Orpiment
$\text{AsS}(\text{cr}) + 3\text{H}_2\text{O} = \text{H}_3\text{AsO}_3 + \text{H}_2\text{S} + \text{H}^+ + \text{e}^-$	-13.145	108.330	Nordstrom and Archer 2003	Realgar

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{As}(\text{cr}) + 3\text{H}_2\text{O} = \text{H}_3\text{AsO}_3 + 3\text{H}^+ + 3\text{e}^-$	-12.505	115.130	Nordstrom and Archer 2003	As(cr)
$\text{As}_2\text{S}_3(\text{am}) + 6\text{H}_2\text{O} = 2\text{H}_3\text{AsO}_3 + 3\text{H}_2\text{S}$	-23.935	181.360	Nordstrom and Archer 2003	As <sub>2</sub> S <sub>3</sub> (am)
$\text{CuO}(\text{s}) + 2\text{H}^+ = \text{Cu}^{+2} + \text{H}_2\text{O}$	7.64	-15.240	Powell et al. 2007, $\Delta H^0_r$ from Ball and Nordstrom 1991	Tenorite
$\text{Cu}(\text{OH})_2(\text{s}) + 2\text{H}^+ = \text{Cu}^{+2} + 2\text{H}_2\text{O}$	8.67	-15.250	Powell et al. 2007, $\Delta H^0_r$ from Ball and Nordstrom 1991	Cu(OH) <sub>2</sub> (s)
$\text{CuF}_2 \cdot 2\text{H}_2\text{O}(\text{s}) = \text{Cu}^{+2} + 2\text{F}^- + 2\text{H}_2\text{O}$	-4.55	-15.3	Ball and Nordstrom 1991	CuF <sub>2</sub> •2H <sub>2</sub> O(s)
$\text{Cu}_3(\text{OH})_4\text{SO}_4(\text{s}) + 4\text{H}^+ = 3\text{Cu}^{+2} + 4\text{H}_2\text{O} + \text{SO}_4^{-2}$	8.29	n.a. <sup>1</sup>	Ball and Nordstrom 1991	Antlerite
$\text{Cu}_4(\text{OH})_6\text{SO}_4(\text{s}) + 6\text{H}^+ = 4\text{Cu}^{+2} + 6\text{H}_2\text{O} + \text{SO}_4^{-2}$	15.34	n.a.	Ball and Nordstrom 1991	Brochantite
$\text{Cu}_3(\text{OH})_4\text{SO}_4(\text{s}) + 4\text{H}^+ = 3\text{Cu}^{+2} + 4\text{H}_2\text{O} + \text{SO}_4^{-2}$	8.29	n.a.	Ball and Nordstrom 1991	Antlerite
$\text{Cu}_4(\text{OH})_6\text{SO}_4(\text{s}) + 6\text{H}^+ = 4\text{Cu}^{+2} + 6\text{H}_2\text{O} + \text{SO}_4^{-2}$	15.34	n.a.	Ball and Nordstrom 1991	Brochantite
$\text{Cu}_4(\text{OH})_6\text{SO}_4 \cdot \text{H}_2\text{O}(\text{s}) + 6\text{H}^+ = 4\text{Cu}^{+2} + 7\text{H}_2\text{O} + \text{SO}_4^{-2}$	16.79	n.a.	Ball and Nordstrom 1991	Langite
$\text{Cu}(\text{s}) = \text{Cu}^+ + \text{e}^-$	-8.76	71.67	Ball and Nordstrom 1991	Copper metal
$\text{CuCl}(\text{s}) = \text{Cu}^+ + \text{Cl}^-$	-6.76	41.76	Ball and Nordstrom 1991	Nantokite
$\text{CuF}(\text{s}) = \text{Cu}^+ + \text{F}^-$	7.08	-51.76	Ball and Nordstrom 1991	CuF(s)
$\text{Cu}_2\text{O}(\text{s}) + 2\text{H}^+ = 2\text{Cu}^+ + \text{H}_2\text{O}$	-1.55	26.13	Ball and Nordstrom 1991	Cuprite
$\text{Cu}_2\text{S}(\text{s}) + \text{H}^+ = 2\text{Cu}^+ + \text{HS}^-$	-34.619	206.5	Ball and Nordstrom 1991	Chalcocite
$\text{Cu}_2\text{SO}_4(\text{s}) = 2\text{Cu}^+ + \text{SO}_4^{-2}$	-1.95	-19.08	Ball and Nordstrom 1991	Cu <sub>2</sub> SO <sub>4</sub> (s)
$\text{NiO}(\text{cr}) + 2\text{H}^+ = \text{Ni}^{+2} + \text{H}_2\text{O}$	12.48	n.a.	Gamsjäger et al 2005	Bunsenite
$\beta\text{-Ni}(\text{OH})_2(\mu\text{c}) + 2\text{H}^+ = \text{Ni}^{+2} + 2\text{H}_2\text{O}$	11.9	n.a.	Gamsjäger et al 2005	β-Ni(OH) <sub>2</sub> (μc)

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\beta\text{-Ni(OH)}_2(\text{cr}) + 2\text{H}^+ = \text{Ni}^{+2} + 2\text{H}_2\text{O}$	11.02	-84.36	Gamsjäger et al 2005	$\beta\text{-Ni(OH)}_2(\text{cr})$
$\text{NiCl}_2 \cdot 6\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + 2\text{Cl}^- + 6\text{H}_2\text{O}$	3.045	n.a.	Gamsjäger et al 2005	$\text{NiCl}_2 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{NiCl}_2 \cdot 4\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + 2\text{Cl}^- + 4\text{H}_2\text{O}$	3.802	n.a.	Gamsjäger et al 2005	$\text{NiCl}_2 \cdot 4\text{H}_2\text{O}(\text{cr})$
$\text{NiCl}_2 \cdot 2\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + 2\text{Cl}^- + 2\text{H}_2\text{O}$	4.881	n.a.	Gamsjäger et al 2005	$\text{NiCl}_2 \cdot 2\text{H}_2\text{O}(\text{cr})$
$\text{NiSO}_4 \cdot 7\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + \text{SO}_4^{-2} + 7\text{H}_2\text{O}$	-2.267	12.167	Gamsjäger et al 2005	Morenosite
$\text{NiCO}_3(\text{cr}) = \text{Ni}^{+2} + \text{CO}_3^{-2}$	-10.982	n.a.	Gamsjäger et al 2005	$\text{NiCO}_3(\text{cr})$
$\text{NiCO}_3 \cdot 5.5\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + \text{CO}_3^{-2} + 5.5\text{H}_2\text{O}$	-7.519	n.a.	Gamsjäger et al 2005	$\text{NiCO}_3 \cdot 5.5\text{H}_2\text{O}(\text{cr})$
$\text{Ni}_4(\text{OH})_6\text{SO}_4(\text{s}) + 6\text{H}^+ = 4\text{Ni}^{+2} + \text{SO}_4^{-2} + 6\text{H}_2\text{O}$	32	n.a. <sup>1</sup>	Ball and Nordstrom 1991	$\text{Ni}_4(\text{OH})_6\text{SO}_4(\text{s})$
$\text{NiS}(\text{s}) + \text{H}^+ = \text{Ni}^{+2} + \text{HS}^-$	-8.042	2.5 kcal/mol	Ball and Nordstrom 1991	Millerite
$\text{NiSO}_4 \cdot 6\text{H}_2\text{O}(\text{s}) = \text{Ni}^{+2} + \text{SO}_4^{-2} + 6\text{H}_2\text{O}$	-2.04	1.1 kcal/mol	Ball and Nordstrom 1991	Retgersite
$\text{Ni}_2\text{SiO}_4(\text{s}) + 4\text{H}^+ = 2\text{Ni}^{+2} + \text{H}_4\text{SiO}_4$	14.54	-33.360 kcal/mol	Ball and Nordstrom 1991	$\text{Ni}_2\text{SiO}_4(\text{s})$
$\text{Ni}_3(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}(\text{s}) = 3\text{Ni}^{+2} + 2\text{AsO}_4^{-3} + 8\text{H}_2\text{O}$	-25.511	n.a.	Ball and Nordstrom 1991	$\text{Ni}_3(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}(\text{s})$
$\text{Al(OH)}_3(\mu\text{c}) + 3\text{H}^+ = \text{Al}^{+3} + 3\text{H}_2\text{O}$	9.35	-24.5 kcal/mol	Nordstrom et al. 1990	Microcrystalline gibbsite
$\text{Al}_4(\text{OH})_{10}\text{SO}_4(\text{s}) + 10\text{H}^+ = 4\text{Al}^{+3} + \text{SO}_4^{-2} + 10\text{H}_2\text{O}$	22.7	n.a.	Ball and Nordstrom 1991	Basaluminite
$\text{Al}_4(\text{OH})_{10}\text{SO}_4(\text{s}) + 10\text{H}^+ = 4\text{Al}^{+3} + \text{SO}_4^{-2} + 10\text{H}_2\text{O}$	23.9	n.a.	Sánchez-España et al. 2011	Hydrobasaluminite
$\text{Zn}_5(\text{OH})_6(\text{CO}_3)_2 + 6\text{H}^+ = 5\text{Zn}^{+2} + 6\text{H}_2\text{O} + 2\text{CO}_3^{-2}$	9.65	n.a.	Schindler et al. 1969 as reported by Zachara et al. 1989	Hydrozincite

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{Fe(OH)}_3(\text{am}) + 3\text{H}^+ = \text{Fe}^{+3} + 3\text{H}_2\text{O}$	4.891	n.a.	Ball and Nordstrom 1991	Amorphous ferric hydroxide
$\text{Fe(OH)}_3(\text{s}) + 3\text{H}^+ = \text{Fe}^{+3} + 3\text{H}_2\text{O}$	3.7	n.a.	Majzlan et al. 2004	Ferrihydrite (2-line)
$\text{Fe(OH)}_3(\text{s}) + 3\text{H}^+ = \text{Fe}^{+3} + 3\text{H}_2\text{O}$	3.2	n.a.	Majzlan et al. 2004	Ferrihydrite (6-line)
$\text{Fe}_8\text{O}_8(\text{OH})_{4.8}(\text{SO}_4)_{1.6}(\text{s}) + 20.8\text{H}^+ = 8\text{Fe}^{+3} + 1.6\text{SO}_4^{-2} + 12.8\text{H}_2\text{O}$	18.0	n.a. <sup>1</sup>	Bigham et al. 1996	Schwertmannite
$\text{Cu}_2\text{CO}_3(\text{OH})_2(\text{s}) = 2\text{Cu}^{+2} + \text{CO}_3^{-2} + 2\text{OH}^-$	-33.16	n.a.	Powell et al. 2007	Malachite
$\text{Cu}_3(\text{CO}_3)_2(\text{OH})_2(\text{s}) = 3\text{Cu}^{+2} + 2\text{CO}_3^{-2} + 2\text{OH}^-$	-44.9	n.a.	Powell et al. 2007	Azurite
$\text{CuS}(\text{s}) + \text{H}^+ = \text{Cu}^{+2} + \text{HS}^-$	-22.27	101	Ball and Nordstrom 1991	Covellite
$\text{CuF}_2 = \text{Cu}^{+2} + 2\text{F}^-$	-0.620	-55.8	Ball and Nordstrom 1991	$\text{CuF}_2(\text{s})$
$\text{BaSeO}_4(\text{cr}) = \text{Ba}^{+2} + \text{SeO}_4^{-2}$	-7.25	n.a.	Berner 2014	$\text{BaSeO}_4(\text{cr})$
$\text{CaSeO}_4 \cdot 2\text{H}_2\text{O}(\text{cr}) = \text{Ca}^{+2} + \text{SeO}_4^{-2} + 2\text{H}_2\text{O}$	-2.68	n.a.	Olin et al. 2005	$\text{CaSeO}_4 \cdot 2\text{H}_2\text{O}(\text{cr})$
$\text{MgSeO}_4 \cdot 6\text{H}_2\text{O}(\text{cr}) = \text{Mg}^{+2} + \text{SeO}_4^{-2} + 6\text{H}_2\text{O}$	-1.133	n.a.	Olin et al. 2005	$\text{MgSeO}_4 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{NiSeO}_4 \cdot 6\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + \text{SeO}_4^{-2} + 6\text{H}_2\text{O}$	-1.381	n.a.	Olin et al. 2005	$\text{NiSeO}_4 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{CuSeO}_4 \cdot 5\text{H}_2\text{O}(\text{cr}) = \text{Cu}^{+2} + \text{SeO}_4^{-2} + 5\text{H}_2\text{O}$	-2.44	n.a.	Olin et al. 2005	$\text{CuSeO}_4 \cdot 5\text{H}_2\text{O}(\text{cr})$
$\text{ZnSeO}_4 \cdot 6\text{H}_2\text{O}(\text{cr}) = \text{Zn}^{+2} + \text{SeO}_4^{-2} + 6\text{H}_2\text{O}$	-1.21	n.a.	Olin et al. 2005	$\text{ZnSeO}_4 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{NiSeO}_3 \cdot 2\text{H}_2\text{O}(\text{cr}) = \text{Ni}^{+2} + \text{SeO}_3^{-2} + 2\text{H}_2\text{O}$	-5.8	n.a.	Berner 2014	$\text{NiSeO}_3 \cdot 2\text{H}_2\text{O}(\text{cr})$
$\text{MnSeO}_3 \cdot 2\text{H}_2\text{O}(\text{cr}) = \text{Mn}^{+2} + \text{SeO}_3^{-2} + 2\text{H}_2\text{O}$	-7.6	n.a.	Berner 2014	$\text{MnSeO}_3 \cdot 2\text{H}_2\text{O}(\text{cr})$
$\text{MgSeO}_3 \cdot 6\text{H}_2\text{O}(\text{cr}) = \text{Mg}^{+2} + \text{SeO}_3^{-2} + 6\text{H}_2\text{O}$	-5.82	n.a.	Berner 2014	$\text{MgSeO}_3 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{CaSeO}_3 \cdot \text{H}_2\text{O}(\text{cr}) = \text{Ca}^{+2} + \text{SeO}_3^{-2} + \text{H}_2\text{O}$	-6.4	n.a.	Berner 2014	$\text{CaSeO}_3 \cdot \text{H}_2\text{O}(\text{cr})$
$\text{SrSeO}_3(\text{cr}) = \text{Sr}^{+2} + \text{SeO}_3^{-2}$	-6.3	n.a.	Berner 2014	$\text{SrSeO}_3(\text{cr})$
$\text{BaSeO}_3(\text{cr}) = \text{Ba}^{+2} + \text{SeO}_3^{-2}$	-6.5	n.a.	Berner 2014	$\text{BaSeO}_3(\text{cr})$

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{Fe}_2(\text{SeO}_3)_3 \cdot 6\text{H}_2\text{O}(\text{cr}) = 2\text{Fe}^{+2} + 3\text{SeO}_3^{-2} + 6\text{H}_2\text{O}$	-41.58	n.a.	Berner 2014	$\text{Fe}_2(\text{SeO}_3)_3 \cdot 6\text{H}_2\text{O}(\text{cr})$
$\text{FeSe}(\text{cr}) + \text{H}^+ = \text{Fe}^{+2} + \text{HSe}^-$	-11	n.a.	Elrashidi et al. 1987, Smith and Martell 1976	$\text{FeSe}(\text{cr})$
$\text{Se}(\text{trig,black}) + \text{H}^+ + \text{e}^- = \text{HSe}^-$	-7.6	n.a. <sup>1</sup>	Berner 2014	Elemental selenium
$\text{Se}(\text{mono,red}) + \text{H}^+ + \text{e}^- = \text{HSe}^-$	-7.4	n.a.	Olin et al. 2005	Elemental selenium
$\text{Se}(\text{am}) + \text{H}^+ + 2\text{e}^- = \text{HSe}^-$	-6.57	n.a.	Iida et al. 2010	Amorphous elemental selenium
$\text{Fe}(\text{OH})_3 + 3\text{H}^+ = \text{Fe}^{+3} + 3\text{H}_2\text{O}$	4.891	n.a.	Ball and Nordstrom 1991	$\text{Fe}(\text{OH})_3(\text{a})$
$\text{Fe}(\text{OH})_3 + 3\text{H}^+ = \text{Fe}^{+3} + 3\text{H}_2\text{O}$	3.5	n.a.	Majzlan et al 2004 and Stefánsson 2007	Ferrihydrite-2L
$\text{Fe}(\text{OH})_3 + 3\text{H}^+ = \text{Fe}^{+3} + 3\text{H}_2\text{O}$	3.2	n.a.	Majzlan et al 2004	Ferrihydrite-6L
$\text{Fe}_8\text{O}_8(\text{OH})_{4.8}(\text{SO}_4)_{1.6}(\text{s}) + 20.8\text{H}^+ = 8\text{Fe}^{+3} + 1.6\text{SO}_4^{-2} + 12.8\text{H}_2\text{O}$	18	n.a.	Bigham et al. 1996	Schwertmannite
$\text{FeOOH} + 3\text{H}^+ = \text{Fe}^{+3} + 2\text{H}_2\text{O}$	0.17	-60.59	Lemire et al. 2013, Stefánsson 2007, Ball and Nordstrom 1991 ( $\Delta H^0_r$ )	Goethite
$\text{Fe}_2\text{O}_3 + 6\text{H}^+ = 2\text{Fe}^{+3} + 3\text{H}_2\text{O}$	-4.008	-129.06	Ball and Nordstrom 1991	Hematite
$\text{Fe}_2\text{O}_3 + 6\text{H}^+ = 2\text{Fe}^{+3} + 3\text{H}_2\text{O}$	6.386	n.a.	Ball and Nordstrom 1991	Maghemite
$\text{Fe}_3\text{O}_4 + 8\text{H}^+ = 2\text{Fe}^{+3} + \text{Fe}^{+2} + 4\text{H}_2\text{O}$	3.737	-211.13	Ball and Nordstrom 1991	Magnetite
$\text{FeSO}_4 \cdot 7\text{H}_2\text{O} = \text{Fe}^{+2} + \text{SO}_4^{-2} + 7\text{H}_2\text{O}$	-2.209	20.53	Nordstrom et al. 1990	Melanterite
$\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6 + 6\text{H}^+ = \text{K}^+ + 3\text{Fe}^{+3} + 2\text{SO}_4^{-2} + 6\text{H}_2\text{O}$	-11	-45	Baron and Palmer 1995	Jarosite-K

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data in the Database (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{NaFe}_3(\text{SO}_4)_2(\text{OH})_6 + 6\text{H}^+ = \text{Na}^+ + 3\text{Fe}^{+3} + 2\text{SO}_4^{-2} + 6\text{H}_2\text{O}$	-5.28	-151.38	Alpers et al. 1989	Jarosite-Na
$(\text{K}_{0.77}\text{Na}_{0.03}\text{H}_{0.2})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6 + 5.8\text{H}^+ = 0.77\text{K}^+ + 0.03\text{Na}^+ + 3\text{Fe}^{+3} + 2\text{SO}_4^{-2} + 6\text{H}_2\text{O}$	-9.83	n.a.	Alpers et al. 1989	Jarosite(ss)
$(\text{H}_3\text{O})\text{Fe}_3(\text{SO}_4)_2(\text{OH})_6 + 5\text{H}^+ = 3\text{Fe}^{+3} + 2\text{SO}_4^{-2} + 7\text{H}_2\text{O}$	-5.21	0.542	Majzlan et al. 2004 and Lemire et al. 2013	Jarosite-H
$\text{FeS}(\text{s}) + 2\text{H}^+ = \text{Fe}^{+2} + \text{H}_2\text{S}(\text{aq})$	3.5	n.a. <sup>1</sup>	Rickard 2006, Lemire et al. (2013)	Mackinawite
$\text{FeS}(\text{s}) + 2\text{H}^+ = \text{Fe}^{+2} + \text{H}_2\text{S}(\text{aq})$	4.1	n.a.	Lemire et al. 2013	FeS(ppt)
$\text{FeS}_2 + 2\text{H}^+ + 2\text{e}^- = \text{Fe}^{+2} + 2\text{HS}^-$	-18.479	47.28	Ball and Nordstrom 1991	Pyrite
$\text{Fe}_3\text{S}_4 + 4\text{H}^+ = 2\text{Fe}^{+3} + \text{Fe}^{+2} + 4\text{HS}^-$	-45.035	n.a.	Ball and Nordstrom 1991	Greigite
$\text{FeCO}_3 = \text{Fe}^{+2} + \text{CO}_3^{-2}$	-10.90	-12.986	Bénézech et al 2009, $\Delta H^0_r$ from Lemire et al. 2013	Siderite(cr)
$\text{FeCO}_3 = \text{Fe}^{+2} + \text{CO}_3^{-2}$	-10.4	n.a.	Jensen et al. 2002	Siderite(ppt)
$\text{MnSO}_4 = \text{Mn}^{+2} + \text{SO}_4^{-2}$	2.669	-54.768	Ball and Nordstrom 1991	MnSO4(s)
$\text{MnO}_2 + 4\text{H}^+ + 2\text{e}^- = \text{Mn}^{+2} + 2\text{H}_2\text{O}$	41.38	-272.42	Ball and Nordstrom 1991	Pyrolusite
$\text{MnO}_2 + 4\text{H}^+ + 2\text{e}^- = \text{Mn}^{+2} + 2\text{H}_2\text{O}$	43.601	n.a.	Ball and Nordstrom 1991	Birnessite
$\text{MnO}_2 + 4\text{H}^+ + 2\text{e}^- = \text{Mn}^{+2} + 2\text{H}_2\text{O}$	42.564	n.a.	Ball and Nordstrom 1991	Nsutite
$\text{Mn}_2\text{O}_3 + 6\text{H}^+ = 2\text{Mn}^{+3} + 3\text{H}_2\text{O}$	-0.611	-63.785	Ball and Nordstrom 1991	Bixbyite
$\text{Mn}_3\text{O}_4 + 8\text{H}^+ + 2\text{e}^- = 3\text{Mn}^{+2} + 4\text{H}_2\text{O}$	61.03	-421.08	Ball and Nordstrom 1991	Hausmannite
$\text{Mn}(\text{OH})_2 + 2\text{H}^+ = \text{Mn}^{+2} + 2\text{H}_2\text{O}$	15.2	n.a.	Ball and Nordstrom 1991	Pyrochroite
$\text{MnOOH} + 3\text{H}^+ + \text{e}^- = \text{Mn}^{+2} + 2\text{H}_2\text{O}$	25.340	n.a.	Ball and Nordstrom 1991	Manganite

<sup>1</sup> n.a. = not available

Appendix B. Selected Uranium, Phosphate, Vanadium, Arsenic, Copper, Nickel, Selenium, Iron, Manganese, Radium, Molybdenum and Other Solid Phase Data in the Database (contd)

Reaction	$\log_{10}K^0$ at 25°C, I = 0	$\Delta H^0_r$ at 25°C, I = 0 (kJ/mol)	Reference	Phase name
$\text{MnCO}_3 = \text{Mn}^{+2} + \text{CO}_3^{-2}$	-10.39	n.a.	Nordstrom et al. 1990	Rhodochrosite(d)
$\text{MnCO}_3 = \text{Mn}^{+2} + \text{CO}_3^{-2}$	-11.13	-5.98	Nordstrom et al. 1990	Rhodochrosite(cr)
$\text{MnS} + \text{H}^+ = \text{Mn}^{+2} + \text{HS}^-$	3.8	-24.23	Ball and Nordstrom 1991	MnS(Green)
$\text{RaCO}_3 = \text{Ra}^{+2} + \text{CO}_3^{-2}$	-8.3	-11.7	Langmuir and Riese 1985	Radium carbonate
$\text{RaSO}_4 = \text{Ra}^{+2} + \text{SO}_4^{-2}$	-10.26	-39.3	Langmuir and Riese 1985	Radium sulfate
$\text{MoS}_2 + 4\text{H}_2\text{O} = \text{MoO}_4^{-2} + 6\text{H}^+ + 2\text{HS}^- + 2\text{e}^-$	-70.2596	389.02	EPA 1999	MoS <sub>2</sub> (s)
$\text{MoO}_3 + \text{H}_2\text{O} = \text{MoO}_4^{-2} + 2\text{H}^+$	-8	n.a. <sup>1</sup>	EPA 1999	MoO <sub>3</sub> (s)

<sup>1</sup> n.a. = not available

Appendix C. Surface Complexation Data for Uranyl, Carbonate, Bicarbonate, Radium and Molybdenum Adsorption on Hydrous Ferric Oxide

Surface Complexation Reaction	$\log_{10}K$	Reference
$\text{Hfo\_sOH} + \text{UO}_2^{+2} = \text{Hfo\_sOUO}_2^+ + \text{H}^+$	3.792	Mahoney et al. 2009
$\text{Hfo\_wOH} + \text{UO}_2^{+2} = \text{Hfo\_wOUO}_2^+ + \text{H}^+$	2.507	Mahoney et al. 2009
$\text{Hfo\_wOH} + \text{UO}_2^{+2} + \text{CO}_3^{-2} = \text{Hfo\_wOUO}_2\text{CO}_3^- + \text{H}^+$	9.150	Mahoney et al. 2009
$\text{Hfo\_wOH} + \text{UO}_2^{+2} + 2\text{CO}_3^{-2} = \text{Hfo\_wOUO}_2(\text{CO}_3)_2^{-3} + \text{H}^+$	15.28	Mahoney et al. 2009
$\text{Hfo\_wOH} + \text{CO}_3^{-2} + \text{H}^+ = \text{Hfo\_wCO}_3^- + \text{H}_2\text{O}$	12.78	Mahoney et al. 2009 from Appelo et al. 2002
$\text{Hfo\_wOH} + \text{CO}_3^{-2} + 2\text{H}^+ = \text{Hfo\_wHCO}_3 + \text{H}_2\text{O}$	20.37	Mahoney et al. 2009 from Appelo et al. 2002
$\text{Hfo\_sOH} + \text{Ra}^{+2} = \text{Hfo\_sOHRa}^{+2}$	6.66	Sajih et al. 2014
$\text{Hfo\_wOH} + \text{Ra}^{+2} = \text{Hfo\_wOHRa}^{+2}$	-5.67	Sajih et al. 2014
$\text{Hfo\_sOH} + \text{MoO}_4^{-2} + \text{H}^+ = \text{Hfo\_sMoO}_4^- + \text{H}_2\text{O}$	9.5	EPA 1999 from Dzombak and Morel 1990
$\text{Hfo\_wOH} + \text{MoO}_4^{-2} + \text{H}^+ = \text{Hfo\_wMoO}_4^- + \text{H}_2\text{O}$	9.5	EPA 1999 from Dzombak and Morel 1990
$\text{Hfo\_sOH} + \text{MoO}_4^{-2} = \text{Hfo\_sOHMoO}_4^{-2}$	2.4	EPA 1999 from Dzombak and Morel 1990
$\text{Hfo\_wOH} + \text{MoO}_4^{-2} = \text{Hfo\_wOHMoO}_4^{-2}$	2.4	EPA 1999 from Dzombak and Morel 1990