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Sandia National Laboratories
Waste Isolation Pilot Plant

Solubility Model for the Fe^{+2} - Mg^{+2} - Na^{+} - Ca^{+2} - H^{+} - Cl^{-} - Oxalate^{-2} - EDTA^{-4} - Citrate^{-3} - H_2O systems

Analysis Report for Data Collected under the Test Plan TP 08-02, Rev.0, "Iron, Lead, Sulfide, and EDTA Solubilities"

Analysis Report Carried Out under the Analysis Plan AP-176, Rev.0, "Analysis Plan for the Development of a Self-Consistent Extension of the WIPP Geochemical Thermodynamic Database that includes Aqueous Ferrous Iron Chemistry"

Supercedes Sections IV.2 to IV.4 of the Analysis Report titled "Derivation of Pitzer Interaction Parameters and Thermodynamic Properties for the Aqueous Species of Ferrous Iron and Their Pairs, Revision 2" by Je-Hun Jang and Sungtae Kim

To be included in the AP-176 records package

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APPROVAL PAGE

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EXECUTIVE SUMMARY

This Analysis Report provides the thermodynamic data required to accurately calculate the dissolved concentration of ferrous iron in the presence of three organic ligands in solutions of higher ionic strength. The organic ligands of concern are $Oxalate^{-2}$, $EDTA^{-4}$, and $Citrate^{-3}$. The Pitzer activity coefficient equation was used to account for the excess free energies of aqueous species. The solubility limiting solids of interest are humboldtine ($FeOxalate.2H_2O(s)$), ferrous iron hydroxide ($Fe(OH)_2(s)$), and hibbingite ($Fe_2Cl(OH)_3(s)$). The solubility of glushinskite ($MgOxalate.2H_2O(s)$) was investigated to provide a clearer picture of the solubility of humboldtine. The background electrolytes were made of the following salts; $MgCl_2$, NaCl, MgHCitrate, and/or Na_2H_2EDTA.

The conclusion of this report is summarized in Table ES- 1 and Table ES- 2. In the two Tables, listed are the 10-based logarithms of equilibrium constants of reactions (Table ES- 1) and the Pitzer interaction parameters (Table ES- 2) related to the solubility model. These two Tables contain the consolidated thermodynamic data captured in Table 1 through Table 10.

Experimental data were sub-divided into five individual systems, and summaries on the five systems are given in the following five sections.

Table ES- 1. Reactions and their 10-based logarithm of equilibrium constants (logKs) for the solubility model for $FeOxalate.2H_2O(s)$, $MgOxalate.2H_2O(s)$, $CaOxalate.H_2O(s)$, $Fe(OH)_2(s)$, and $Fe_2Cl(OH)_3(s)$ in the presence of $Oxalate^{-2}$, $Citrate^{-3}$, and $EDTA^{-4}$ in solutions made of $MgCl_2$ and NaCl.

Reactions	LogK	Source
Aqueous reactions		
$H^+ + OH^- = H_2O$	13.9967	data0.fm1[a], fm2[B]
$H_2Oxalate(aq) = 2H^+ + Oxalate^{2-}$ [b]	-5.6532	data0.fm1, fm2
$HOxalate^- = H^+ + Oxalate^{2-}$	-4.2596	data0.fm1, fm2
$H_4EDTA(aq) = 4H^+ + EDTA^{4-}$ [c]	-23.0393	data0.fm1, fm2
$H_3EDTA^- = 3H^+ + EDTA^{4-}$	-20.5374	data0.fm1, fm2
$H_2EDTA^{2-} = 2H^+ + EDTA^{4-}$	-17.4500	data0.fm1, fm2
$HEDTA^{3-} = H^+ + EDTA^{4-}$	-10.5707	data0.fm1, fm2
$H_3Citrate(aq) = 3H^+ + Citrate^{3-}$ [d]	-14.5098	data0.fm1, fm2
$H_2Citrate^- = 2H^+ + Citrate^{3-}$	-11.2630	data0.fm1, fm2
$HCitrate^{2-} = H^+ + Citrate^{3-}$	-6.4232	data0.fm1, fm2
$MgOH^+ + H^+ = Mg^{2+} + H_2O$	11.8091	data0.fm1, fm2
$MgCitrate^- = Mg^{2+} + Citrate^{3-}$	-5.2997	data0.fm1, fm2
$MgEDTA^{2-} = Mg^{2+} + EDTA^{4-}$	-10.1260	data0.fm1, fm2
$MgOxalate(aq) = Mg^{2+} + Oxalate^{2-}$	-3.7931	data0.fm1, fm2
$Mg(Oxalate)_2^{-2} = Mg^{2+} + 2Oxalate^{2-}$	-5.24	Choppin et al. (2001)
$FeOH^+ + H^+ = Fe^{2+} + H_2O$	9.3148	Shock et al. (1997)
$Fe(OH)_2(aq) + 2H^+ = Fe^{2+} + 2H_2O$	20.8214	Stumm and Morgan (1996)
$Fe(OH)_3^- + 3H^+ = Fe^{2+} + 3H_2O$	31.0000	Baes and Mesmer (1976)
$Fe(OH)_4^{-2} + 4H^+ = Fe^{2+} + 4H_2O$	46.00	Baes and Mesmer (1976)
$FeOxalate(aq) = Fe^{2+} + Oxalate^{2-}$	-3.97	Gustafsson (2015)
$Fe(Oxalate)_2^{-2} = Fe^{+2} + 2Oxalate^{-2}$	-5.90	Gustafsson (2015)
$FeHEDTA^- = Fe^{+2} + H^+ + EDTA^{-4}$	-19.3	Morel and Hering (1993)
$FeEDTA^{2-} = Fe^{2+} + EDTA^{4-}$	-16.1	Morel and Hering (1993)
$FeOHEDTA^{-3} + H^+ = Fe^{+2} + EDTA^{-4} + H_2O$	-6.4	Morel and Hering (1993)
$Fe(OH)_2EDTA^{-4} + 2H^+ = Fe^{+2} + EDTA^{-4} + 2H_2O$	4.3	Morel and Hering (1993)
$FeOHCitrate^{-2} + H^+ = Fe^{2+} + Citrate^{3-} + H_2O$	1.92	This report
$FeCitrate^- = Fe^{2+} + Citrate^{3-}$	-5.7	Morel and Hering (1993)
$FeHCitrate(aq) = Fe^{2+} + H^+ + Citrate^{3-}$	-9.9	Morel and Hering (1993)
$CaOH^+ + H^+ = Ca^{2+} + H_2O$	12.8333	data0.ymp.R2[e]
$CaOxalate(aq) = Ca^{2+} + Oxalate^{2-}$	-3.7931[f]	data0.fm1, fm2
Dissolution		
$Fe(OH)_2(s) + 2H^+ = Fe^{2+} + 2H_2O$	12.83	Jang and Kim (2016)
$Fe_2Cl(OH)_3(s) + 3H^+ = 2Fe^{2+} + Cl^- + 3H_2O$	16.89	Jang and Kim (2016)
$MgOxalate.2H_2O(s) = Mg^{2+} + Oxalate^{2-} + 2H_2O$	-6.50	This report
$FeOxalate.2H_2O(s) = Fe^{2+} + Oxalate^{2-} + 2H_2O$	-8.34	This report
$CaOxalate{\cdot}H_2O(s) = Ca^{2+} + Oxalate^{2-} + H_2O$	-8.7517	data0.fm1, fm2

[A] http://www.solubilityofthings.com/water/ions_solubility/ksp_chart.php, not traceable to original research

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016)

[b] $Oxalate^{2-}$: $C_2O_4^{2-}$

[c] $EDTA^{4-}$: $C_{10}H_{12}O_8N_2^{4-}$

[d] $Citrate^{3-}$: $C_6H_5O_7^{3-}$

[e] data0.ymp.R2 is one of the EQ3/6 databases that comes within the installation package for EQ3/6 v.8.0a.

[f] By analogy to MgOxalate(aq)

Table ES- 2. The Pitzer interaction parameters related to the Fe^{+2} - Mg^{+2} - Na^{+} - Ca^{+2} - H^{+} - Cl^{-} - Oxalate^{-2} - Citrate^{-3} - EDTA^{-4} - H_2O system.

i	*j*	α_1/α_2[A]	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	Source
Na^{+}	Cl^{-}	2.0/12.0	0.0765	0.2664	0.0	0.00127	data0.fm1[a], fm2[B]
Na^{+}	OH^{-}	2.0/12.0	0.0864	0.253	0.0	0.0044	data0.fm1, fm2
Na^{+}	HOxalate^{-}	2.0/12.0	-0.2448	0.29	0.0	0.068	data0.fm1, fm2
Na^{+}	Oxalate^{2-}[b]	2.0/12.0	-0.2176	1.74	0.0	0.122	data0.fm1, fm2
Na^{+}	H_3EDTA^{-}	2.0/12.0	-0.2345	0.29	0.0	0.059	data0.fm1, fm2
Na^{+}	H_2EDTA^{2-}	2.0/12.0	-0.1262	1.74	0.0	0.054	data0.fm1, fm2
Na^{+}	$HEDTA^{3-}$	2.0/12.0	0.5458	5.22	0.0	-0.048	data0.fm1, fm2
Na^{+}	$EDTA^{4-}$[c]	2.0/12.0	1.016	11.6	0.0	0.001	data0.fm1, fm2
Na^{+}	$H_2Citrate^{-}$	2.0/12.0	-0.1296	0.29	0.0	0.013	data0.fm1, fm2
Na^{+}	$HCitrate^{2-}$	2.0/12.0	-0.0989	1.74	0.0	0.027	data0.fm1, fm2
Na^{+}	$Citrate^{3-}$[d]	2.0/12.0	0.0887	5.22	0.0	0.047	data0.fm1, fm2
Na^{+}	$MgCitrate^{-}$	2.0/12.0	0.1742	0.29	0.0	-0.06923	data0.fm1, fm2
Na^{+}	$MgEDTA^{2-}$	2.0/12.0	0.2134	1.74	0.0	0.00869	data0.fm1, fm2
Mg^{2+}	Cl^{-}	2.0/12.0	0.35235	1.6815	0.0	0.00519	data0.fm1, fm2
$MgOH^{+}$	Cl^{-}	2.0/12.0	-0.1	1.658	0.0	0.0	data0.fm1, fm2
Mg^{2+}	$MgCitrate^{-}$	2.0/12.0	1.0915	1.74	0.0	0.0	New in data0.fm2
Mg^{2+}	$Citrate^{3-}$[d]	1.4/12.0	0.9330	4.4	0.0	0.0	New in data0.fm2
Mg^{2+}	$EDTA^{4-}$[c]	1.4/12.0	-0.01	11.6	0.0	0.30	New in data0.fm2
H^{+}	Cl^{-}	2.0/12.0	0.1775	0.2945	0.0	0.0008	data0.fm1, fm2
Ca^{2+}	Cl^{-}	2.0/12.0	0.3159	1.614	0.0	-0.00034	data0.fm1, fm2
Ca^{2+}	OH^{-}	2.0/12.0	-0.1747	-0.2303	-5.72	0.0	data0.fm1, fm2
Fe^{2+}	Cl^{-}	2.0/12.0	0.37324	1.13499	0.0	-0.02152	Moog et al. (2004)
Mg^{2+}	$Mg(Oxalate)_2^{-2}$	1.4/12.0	0.0434	3.27	-45.74	0.0	This report
Na^{+}	$Fe(OH)_2EDTA^{-4}$	2.0/12.0	0.0256	11.6	-0.05	0.0217	This report
Na^{+}	$FeOHCitrate^{-2}$	2.0/12.0	-0.197	1.74	-0.0912	0.0885	This report
Mg^{+2}	$FeOHCitrate^{-2}$	1.4/12.0	-0.0518	3.27	-45.74	0.134	This report
i	***j***		$\theta_{cc'}$ **or** $\theta_{aa'}$				**Source**
Na^{+}	Mg^{2+}		0.07				data0.fm1, fm2
Na^{+}	Ca^{2+}		0.07				data0.fm1, fm2
Na^{+}	H^{+}		0.036				data0.fm1, fm2
Mg^{2+}	H^{+}		0.10				data0.fm1, fm2
Ca^{2+}	Mg^{2+}		0.007				data0.fm1, fm2
Ca^{2+}	H^{+}		0.092				data0.fm1, fm2
Cl-	OH^{-}		-0.050				data0.fm1, fm2
Na^{+}	Fe^{2+}		0.10945				Moog et al. (2004)
Mg^{2+}	Fe^{2+}		0.14504				Moog et al. (2004)
K^{+}	Fe^{2+}		0.02737				Moog et al. (2004)
Ca^{2+}	Fe^{2+}		0.08112				Moog et al. (2004)
Na^{+}	$FeOH^{+}$		-0.0974				This report
i	***j***		λ_{nc} **or** λ_{na}				
MgOxalate(aq)	Cl^{-}		0.0189				data0.fm1, fm2
CaOxalate(aq)	Cl^{-}		0.0189[A]				data0.fm1, fm2
FeOxalate(aq)	Na^{+}		-0.0407				This report
FeOxalate(aq)	Mg^{2+}		0.0449				This report
FeOxalate(aq)	Cl^{-}		-0.0147				This report

MgOxalate(aq)	Na^+		0.114	This report
MgOxalate(aq)	Mg^{2+}		-0.283	This report
i	***j***	***k***	$\psi_{cc'a}$ **or** $\psi_{aa'c}$	**Source**
Na^+	Ca^{2+}	Cl-	-0.007	data0.fm1, fm2
Na^+	Ca^{2+}	OH^-	-0.0198	New in data0.fm2
Na^+	Mg^{2+}	Cl-	-0.012	data0.fm1, fm2
Na^+	H^+	Cl^-	-0.004	data0.fm1, fm2
Mg^{2+}	$MgOH^+$	Cl-	0.028	data0.fm1, fm2
Mg^{2+}	H^+	Cl-	-0.011	data0.fm1, fm2
Ca^{2+}	Mg^{2+}	Cl-	-0.012	data0.fm1, fm2
Ca^{2+}	H^+	Cl^-	-0.015	data0.fm1, fm2
Cl-	OH^-	Na^+	-0.006	data0.fm1, fm2
Cl-	OH^-	Ca^{2+}	-0.025	data0.fm1, fm2
Fe^{+2}	Na^+	Cl^-	-0.01605	Moog et al. (2004)
Fe^{+2}	K^+	Cl^-	-0.02523	Moog et al. (2004)
Fe^{+2}	Mg^{+2}	Cl^-	-0.02985	Moog et al. (2004)
Fe^{+2}	Ca^{+2}	Cl^-	-0.01599	Moog et al. (2004)

[A] α_1 and α_2 are pre-set constants used in the Pitzer activity coefficient equation. α_1 and α_2 apply for only cation-anion binary pair. α_2 is not applied when $\beta^{(2)}$ is zero or not used. Unit for α_1 and α_2 is $kg^{1/2}\cdot mol^{-1/2}$.

[a] Xiong (2011), [b] Oxalate^{2-}: $C_2O_4^{2-}$, [c] EDTA^{4-}: $C_{10}H_{12}O_8N_2^{4-}$, [d] Citrate^{3-}: $C_6H_5O_7^{3-}$

[B] Common in data0.fm1 (Xiong, 2011) and data0.fm2 (Xiong and Domski, 2016)

Mg^{+2} - Na^+ - H^+ - Cl^- - Oxalate^{-2} - H_2O system

Solubility of glushinskite ($MgOxalate.2H_2O(s)$) was investigated in the $MgCl_2$ and NaCl solutions. No mineral transformation was observed under X-ray diffraction (XRD) (Figure 1). Data and model predictions are illustrated in Figure 2. Thermodynamic data (i.e., equilibrium constants and the related Pitzer interaction parameters) are listed in Table 1 and Table 2. LogK for dissolution of glushinskite, λ of MgOxalate(aq)/Mg^{+2}, λ of MgOxalate(aq)/Na^+, and $\beta^{(0)}$ of Mg^{+2}/$Mg(Oxalate)_2^{-2}$ are determined to be -6.50, -0.283, 0.114, and 0.0434, respectively. $\beta^{(1)}$, $\beta^{(2)}$ and C^ϕ of Mg^{+2}/$Mg(Oxalate)_2^{-2}$ were prescribed at 3.27, -45.74, and 0.0, respectively.

Fe^{+2} - Mg^{+2} - Na^+ - H^+ - Cl^- - Oxalate^{-2} - H_2O system

Solubility of humboldtine ($FeOxalate.2H_2O(s)$) was investigated in the $MgCl_2$ and NaCl solutions. Glushinskite has grown in $MgCl_2$ experiments (Figure 3). Data and model predictions are illustrated in Figure 4. Thermodynamic data (i.e., equilibrium constants and the related Pitzer interaction parameters) are listed in Table 3 and Table 4. LogK of $FeOxalate.2H_2O$ (humboldtine), λ of FeOxalate(aq)/Mg^{+2}, λ of FeOxalate(aq)/Na^+, and λ of FeOxalate(aq)/Cl^- were determined to be -8.34, 0.0449, -0.0407, and -0.0147, respectively.

Ca^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Oxalate^{-2}$ - H_2O system

Solubility of whewellite (CaOxalate.H_2O(s)) was investigated in the $MgCl_2$ and NaCl solutions. Data and model predictions are illustrated in Figure 5. Thermodynamic data (i.e., equilibrium constants and the related Pitzer interaction parameters) are listed in Table 5 and Table 6. No further fitting was required at this time.

Fe^{+2} - Na^{+} - H^{+} - Cl^{-} - $EDTA^{-4}$ - H_2O system

Solubility of $Fe(OH)_2$(s) was investigated in NaCl solutions spiked with Na_2H_2EDTA. Both $Fe(OH)_2$(s) and hibbingite ($Fe_2Cl(OH)_3$(s)) were observed in the XRD scans (Figure 6). Data and model predictions are illustrated in Figure 7. Thermodynamic data (i.e., equilibrium constants and the related Pitzer interaction parameters) are listed in Table 7 and Table 8. $\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(2)}$, and C^{ϕ} of $Na^{+}/Fe(OH)_2EDTA^{-4}$ were determined to be 0.0256, 11.6, -0.05, and 0.0217, respectively.

Fe^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Citrate^{-3}$ - H_2O system

Solubility of $Fe(OH)_2$(s) was investigated in NaCl solutions spiked with MgHCitrate and Na_3Citrate. No mineralogical changes were observed from XRD scans (Figure 8 and Figure 9). Data and model predictions are illustrated in Figure 10. Thermodynamic data (i.e., equilibrium constants and the related Pitzer interaction parameters) are listed in Table 9 and Table 10. Fitting variables are logK for dissociation of $FeOHCitrate^{-2}$; $\beta^{(0)}$ and C^{ϕ} of $Mg^{+2}/FeOHCitrate^{-2}$ with $\beta^{(1)}$ and $\beta^{(2)}$ set to 3.27 and -45.74; and $\beta^{(0)}$, $\beta^{(2)}$, and C^{ϕ} of $Na^{+}/FeOHCitrate^{-2}$ with $\beta^{(1)}$ set to 1.74. They are determined to be, 1.92; -0.0518, 0.134; and -0.197, -0.0912, 0.0885, respectively.

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TABLES AND FIGURES

Figure 1. X-ray diffraction (XRD) scans of glushinskite (MgOxalate.2H_2O(s)) aged in $MgCl_2$ and NaCl solutions. No mineralogical change was observed.

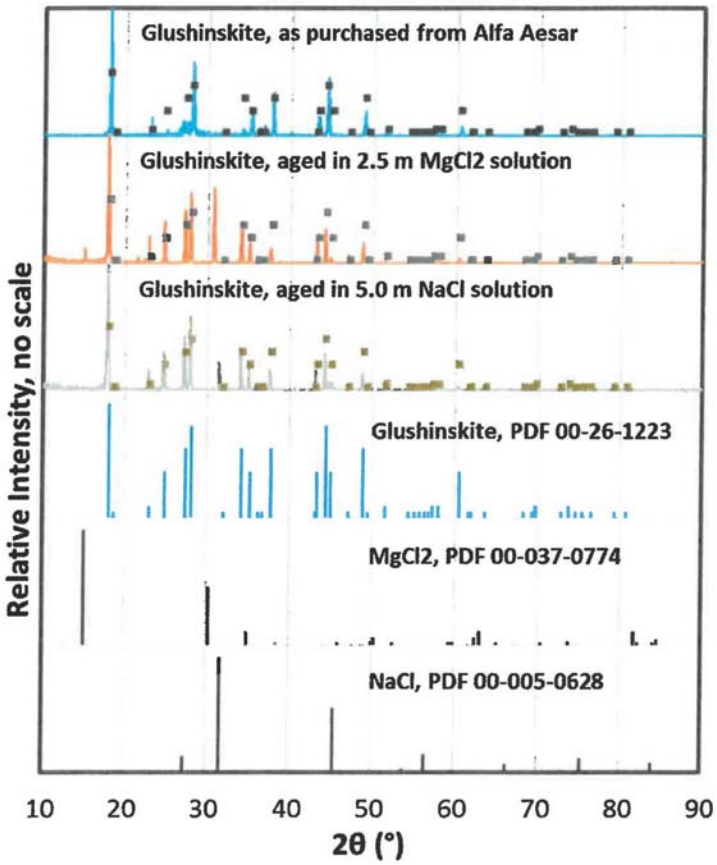

Figure 2. The concentrations (A and B) and activity coefficients (C and D) of relevant Mg^{+2} and $Oxalate^{-2}$ species in equilibrium with glushinskite ($MgOxalate.2H_2O(s)$). The calculated and measured $\Sigma[Oxalate^{-2}]$ (A) and $\Sigma[Mg^{+2}]$ (B) are plotted together. (A, C) Experiments in $MgCl_2$ solutions, (B, D) Experiments in NaCl solutions. Fitting variables are logK of $MgOxalate.2H_2O$ (glushinskite), λ of $MgOxalate(aq)/Mg^{+2}$, λ of $MgOxalate(aq)/Na^{+}$, and $\beta^{(0)}$ of $Mg^{+2}/Mg(Oxalate)_2^{-2}$, and they are determined to be -6.50, -0.283, 0.114, and 0.0434, respectively. $\beta^{(1)}$, $\beta^{(2)}$ and C^{ϕ} of $Mg^{+2}/Mg(Oxalate)_2^{-2}$ were prescribed at 3.27, -45.74, and 0.0. Residuals are 0.085 (A) + 0.089 (B) = 0.174. The residuals prior to fitting those variables were 0.507 (A) + 1.823 (B) = 2.330.

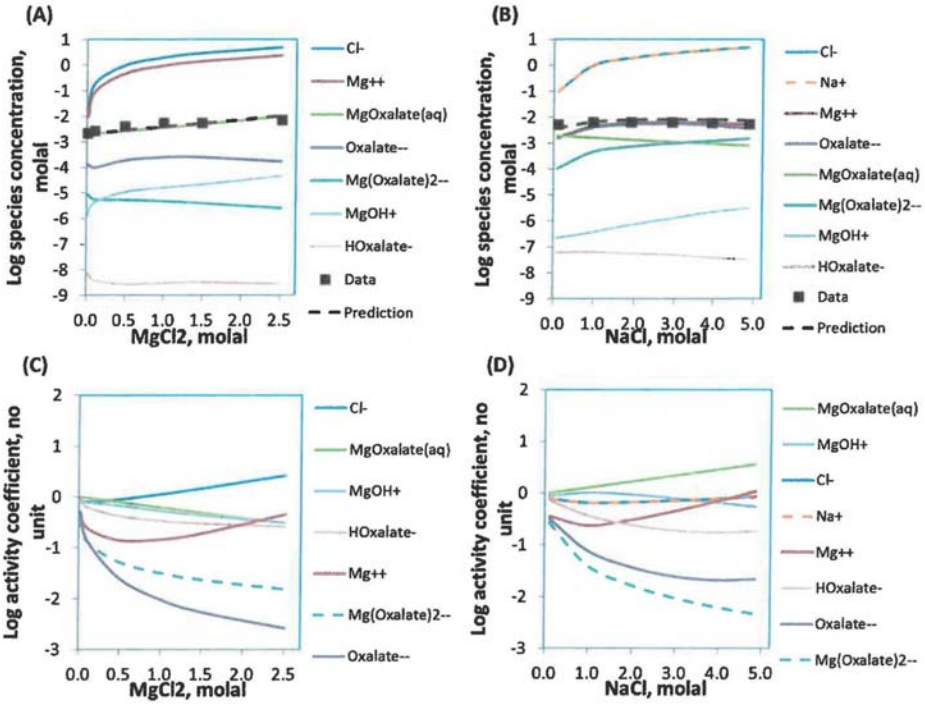

Table 1. Reactions and their 10-based logarithms of equilibrium constants (logKs) for the Mg^{+2} - Na^+ - H^+ - Cl^- - $Oxalate^{-2}$ - H_2O system.

Reactions	LogK	Source
Aqueous reactions		
$H^+ + OH^- = H_2O$	13.9967	data0.fm1[a], fm2[B]
$H_2Oxalate(aq) = 2H^+ + Oxalate^{2-}$ [b]	-5.6532	data0.fm1[a], fm2[B]
$HOxalate^- = H^+ + Oxalate^{2-}$	-4.2596	data0.fm1[a], fm2[B]
$MgOH^+ + H^+ = Mg^{2+} + H_2O$	11.8091	data0.fm1[a], fm2[B]
$MgOxalate(aq) = Mg^{2+} + Oxalate^{2-}$	-3.7931	data0.fm1[a], fm2[B]
$Mg(Oxalate)_2^{-2} = Mg^{+2} + 2Oxalate^{-2}$	-5.24	Choppin et al. (2001)
Dissolution		
$MgOxalate.2H_2O(s) = Mg^{2+} + Oxalate^{2-} + 2H_2O$	-6.50	This report

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016)
[b] $Oxalate^{2-}$: $C_2O_4^{2-}$

Table 2. Pitzer interaction parameters for the Mg^{+2} - Na^+ - H^+ - Cl^- - $Oxalate^{-2}$ - H_2O system.

i	j	α_1/α_2[A]	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^Φ	Source
Na^+	Cl^-	2.0/12.0	0.0765	0.2664	0.0	0.00127	data0.fm1[a], fm2[B]
Na^+	OH^-	2.0/12.0	0.0864	0.253	0.0	0.0044	data0.fm1[a], fm2[B]
Na^+	$HOxalate^-$	2.0/12.0	-0.2448	0.29	0.0	0.068	data0.fm1[a], fm2[B]
Na^+	$Oxalate^{2-}$ [b]	2.0/12.0	-0.2176	1.74	0.0	0.122	data0.fm1[a], fm2[B]
Mg^{2+}	Cl^-	2.0/12.0	0.35235	1.6815	0.0	0.00519	data0.fm1[a], fm2[B]
$MgOH^+$	Cl^-	2.0/12.0	-0.1	1.658	0.0	0.0	data0.fm1[a], fm2[B]
H^+	Cl^-	2.0/12.0	0.1775	0.2945	0.0	0.0008	data0.fm1[a], fm2[B]
Mg^{2+}	$Mg(Oxalate)_2^{-2}$	1.4/12.0	0.0434	3.27	-45.74	0.0	This report
i	**j**		**$\theta_{cc'}$ or $\theta_{aa'}$**				**Source**
Na^+	Mg^{2+}		0.07				data0.fm1[a], fm2[B]
Na^+	H^+		0.036				data0.fm1[a], fm2[B]
Mg^{2+}	H^+		0.10				data0.fm1[a], fm2[B]
Cl-	OH^-		-0.050				data0.fm1[a], fm2[B]
i	**j**		**λ_{nc} or λ_{na}**				**Source**
MgOxalate(aq)	Cl^-		0.0189				data0.fm1[a], fm2[B]
MgOxalate(aq)	Na^+		0.114				This report
MgOxalate(aq)	Mg^{2+}		-0.283				This report
i	**j**	**k**	**$\psi_{cc'a}$ or $\psi_{aa'c}$**				**Source**
Na^+	Mg^{2+}	Cl-	-0.012				data0.fm1[a], fm2[B]
Na^+	H^+	Cl^-	-0.004				data0.fm1[a], fm2[B]
Mg^{2+}	$MgOH^+$	Cl-	0.028				data0.fm1[a], fm2[B]
Mg^{2+}	H^+	Cl-	-0.011				data0.fm1[a], fm2[B]
Cl-	OH^-	Na^+	-0.006				data0.fm1[a], fm2[B]

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016)
[b] $Oxalate^{2-}$: $C_2O_4^{2-}$

Figure 3. X-ray diffraction (XRD) scans of humboldtine (FeOxalate.2H_2O(s)) aged in $MgCl_2$ solutions. Glushinskite (MgOxalate.2H_2O(s)) formed under these experimental conditions.

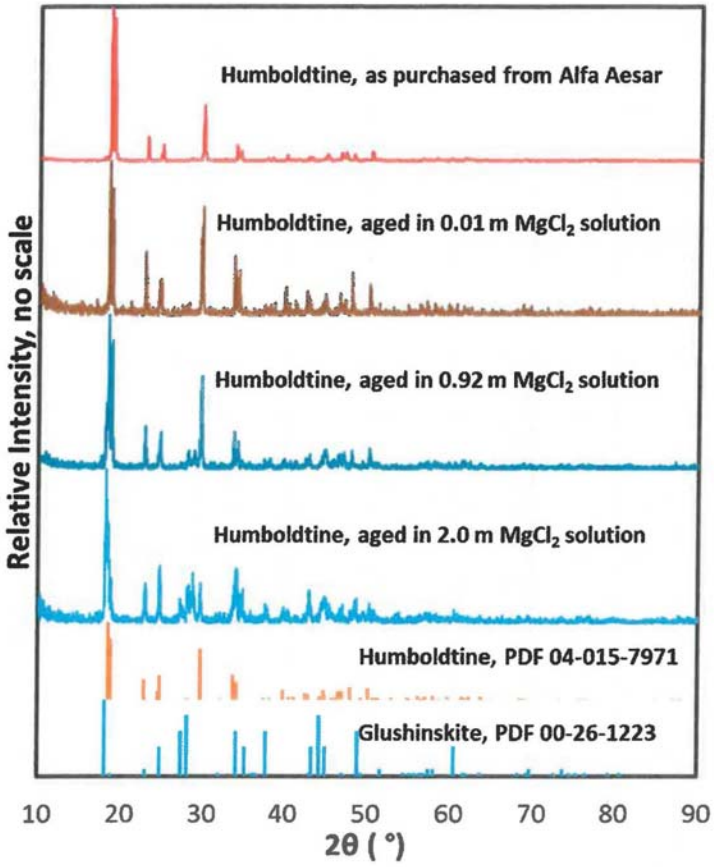

Figure 4. The concentrations (A and B) and activity coefficients (C and D) of relevant Fe^{+2} and $Oxalate^{-2}$ species in equilibrium with humboldtine ($FeOxalate.2H_2O(s)$). The calculated and measured $\Sigma[Fe^{+2}]$ are plotted together. (A, C) Experiments in $MgCl_2$ solutions, (B, D) Experiments in NaCl solutions. Fitting variables are logK of $FeOxalate.2H_2O$ (humboldtine), λ of $FeOxalate(aq)/Mg^{+2}$, λ of $FeOxalate(aq)/Na^{+}$, and λ of $FeOxalate(aq)/Cl^{-}$. They are determined to be 0.0449, -0.0407, and -0.0147, respectively. Residuals are 0.004 (A) + 0.020 (B) = 0.024. The residuals prior to the fitting those variables were 3.087 (A) + 8.132 (B) = 11.218.

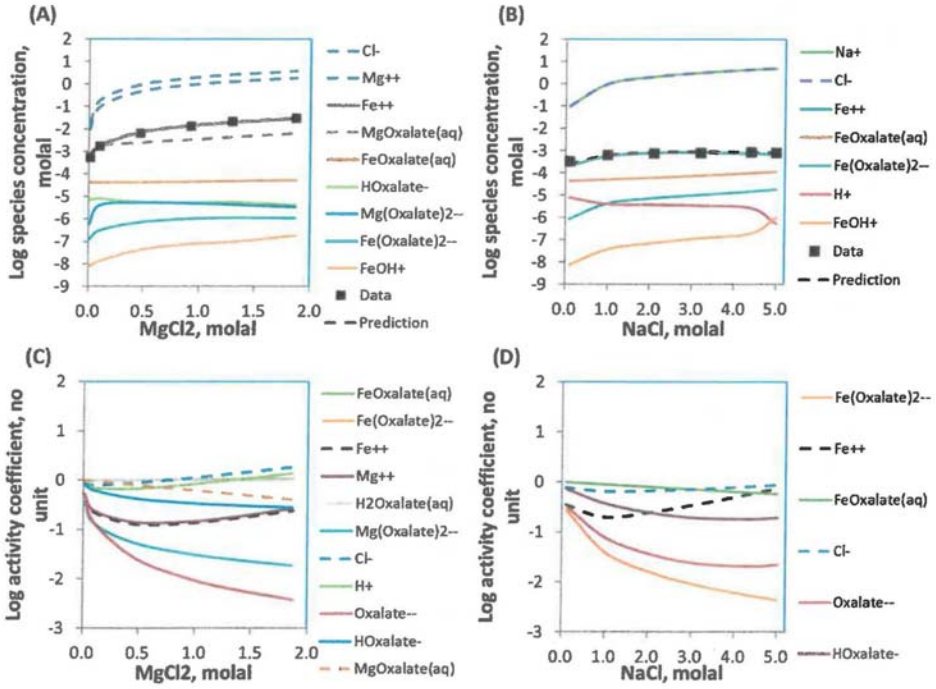

Table 3. Reactions and their 10-based logarithms of equilibrium constants (logKs) for the Fe^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Oxalate^{-2}$ - H_2O system

Reactions	LogK	Source
Aqueous reactions		
$H^+ + OH^- = H_2O$	13.9967	data0.fm1[a], fm2[B]
$H_2Oxalate(aq) = 2H^+ + Oxalate^{2-}$ [b]	-5.6532	data0.fm1[a], fm2[B]
$HOxalate^- = H^+ + Oxalate^{2-}$	-4.2596	data0.fm1[a], fm2[B]
$MgOH^+ + H^+ = Mg^{2+} + H_2O$	11.8091	data0.fm1[a], fm2[B]
$MgOxalate(aq) = Mg^{2+} + Oxalate^{2-}$	-3.7931	data0.fm1[a], fm2[B]
$Mg(Oxalate)_2^{-2} = Mg^{+2} + 2Oxalate^{-2}$	-5.24	Choppin et al. (2001)
$FeOH^+ + H^+ = Fe^{2+} + H_2O$	9.3148	data0.ymp.R2[e]
$Fe(OH)_2(aq) + 2H^+ = Fe^{2+} + 2H_2O$	20.8214	Stumm and Morgan (1996)
$Fe(OH)_3^- + 3H^+ = Fe^{2+} + 3H_2O$	31.0000	data0.ymp.R2[e]
$FeOxalate(aq) = Fe^{2+} + Oxalate^{2-}$	-3.97	Gustafsson (2015)
$Fe(Oxalate)_2^{-2} = Fe^{+2} + 2Oxalate^{-2}$	-5.90	Gustafsson (2015)
Dissolution		
$MgOxalate.2H_2O(s) = Mg^{2+} + Oxalate^{2-} + 2H_2O$	-6.50	This report
$FeOxalate.2H_2O(s) = Fe^{2+} + Oxalate^{2-} + 2H_2O$	-8.34	This report

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016)

[b] $Oxalate^{2-}$: $C_2O_4^{2-}$

[e] data0.ymp.R2 is one of the EQ3/6 databases that comes within the installation package for EQ3/6 v.8.0a.

Table 4. Pitzer interaction parameters for the Fe^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Oxalate^{-2}$ - H_2O system.

i	j	α_1/α_2[A]	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{Φ}	Source
Na^+	Cl^-	2.0/12.0	0.0765	0.2664	0.0	0.00127	data0.fm1[a], fm2[B]
Na^+	OH^-	2.0/12.0	0.0864	0.253	0.0	0.0044	data0.fm1[a], fm2[B]
Na^+	$HOxalate^-$	2.0/12.0	-0.2448	0.29	0.0	0.068	data0.fm1[a], fm2[B]
Na^+	$Oxalate^{2-}$, [b]	2.0/12.0	-0.2176	1.74	0.0	0.122	data0.fm1[a], fm2[B]
Mg^{2+}	Cl^-	2.0/12.0	0.35235	1.6815	0.0	0.00519	data0.fm1[a], fm2[B]
$MgOH^+$	Cl^-	2.0/12.0	-0.1	1.658	0.0	0.0	data0.fm1[a], fm2[B]
H^+	Cl^-	2.0/12.0	0.1775	0.2945	0.0	0.0008	data0.fm1[a], fm2[B]
Fe^{2+}	Cl^-	2.0/12.0	0.37324	1.13499	0.0	-0.02152	Moog et al. (2004)
Mg^{2+}	$Mg(Oxalate)_2^{-2}$	1.4/12.0	0.0434	3.27	-45.74	0.0	This report
i	j		$\theta_{cc'}$ or $\theta_{aa'}$				Source
Na^+	Mg^{2+}		0.07				data0.fm1[a], fm2[B]
Na^+	H^+		0.036				data0.fm1[a], fm2[B]
Na^+	Fe^{2+}		0.10945				Moog et al. (2004)
Na^+	$FeOH^+$		-0.0974				Jang and Kim (2016)
Mg^{2+}	H^+		0.10				data0.fm1[a], fm2[B]
Mg^{2+}	Fe^{2+}		0.14504				Moog et al. (2004)
Cl-	OH^-		-0.050				data0.fm1[a], fm2[B]
i	j		λ_{nc} or λ_{na}				Source

MgOxalate(aq)	Cl^-		0.0189	data0.fm1[a], fm2[B]
MgOxalate(aq)	Na^+		0.114	This report
MgOxalate(aq)	Mg^{2+}		-0.283	This report
FeOxalate(aq)	Na^+		-0.0407	This report
FeOxalate(aq)	Mg^{2+}		0.0449	This report
FeOxalate(aq)	Cl^-		-0.0147	This report
i	**j**	**k**	**$\psi_{cc'a}$ or $\psi_{aa'c}$**	**Source**
Na^+	Mg^{2+}	Cl-	-0.012	data0.fm1[a], fm2[B]
Na^+	H^+	Cl^-	-0.004	data0.fm1[a], fm2[B]
Mg^{2+}	$MgOH^+$	Cl-	0.028	data0.fm1[a], fm2[B]
Mg^{2+}	H^+	Cl-	-0.011	data0.fm1[a], fm2[B]
Fe^{+2}	Na^+	Cl^-	-0.01605	Moog et al. (2004)
Fe^{+2}	Mg^{+2}	Cl^-	-0.02985	Moog et al. (2004)
Cl-	OH^-	Na^+	-0.006	data0.fm1[a], fm2[B]

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016)
[b] Oxalate^{2-}: $C_2O_4^{2-}$

Figure 5. The concentrations (A and B) and activity coefficients (C and D) of relevant Ca^{+2} and $Oxalate^{-2}$ species in equilibrium with whewellite ($CaOxalate.H_2O(s)$). The calculated and measured $\Sigma[Ca^{+2}]$ are plotted together. (A, C) Experiments in $MgCl_2$ solutions, (B, D) Experiments in NaCl solutions. No further fitting was required.

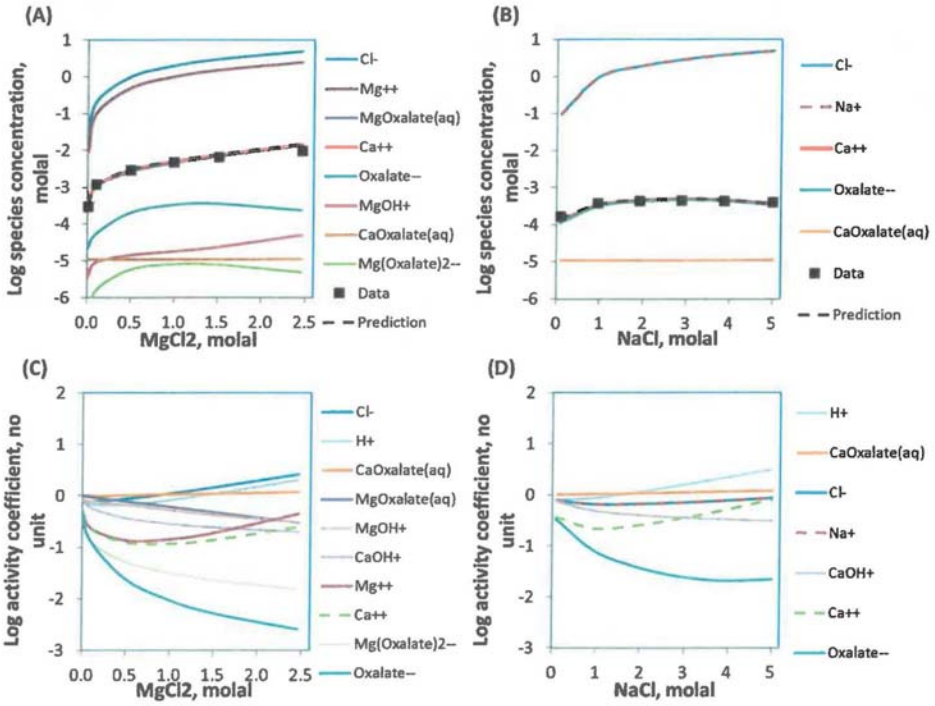

Table 5. Reactions and their 10-based logarithms of equilibrium constants (logKs) for the Ca^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Oxalate^{-2}$ - H_2O system

Reactions	LogK	Source
Aqueous reactions		
$H^+ + OH^- = H_2O$	13.9967	data0.fm1[a], fm2[B]
$H_2Oxalate(aq) = 2H^+ + Oxalate^{2-}$ [b]	-5.6532	data0.fm1[a], fm2[B]
$HOxalate^- = H^+ + Oxalate^{2-}$	-4.2596	data0.fm1[a], fm2[B]
$MgOH^+ + H^+ = Mg^{2+} + H_2O$	11.8091	data0.fm1[a], fm2[B]
$MgOxalate(aq) = Mg^{2+} + Oxalate^{2-}$	-3.7931	data0.fm1[a], fm2[B]
$Mg(Oxalate)_2^{-2} = Mg^{+2} + 2Oxalate^{-2}$	-5.24	Choppin et al. (2001)
$CaOH^+ + H^+ = Ca^{2+} + H_2O$	12.8333	data0.ymp.R2[e]
$CaOxalate(aq) = Ca^{2+} + Oxalate^{2-}$	-3.7931[f]	data0.fm1[a], fm2[B]
Dissolution		
$MgOxalate.2H_2O(s) = Mg^{2+} + Oxalate^{2-} + 2H_2O$	-6.50	This report
$CaOxalate.H_2O(s) = Fe^{2+} + Oxalate^{2-} + 2H_2O$	-8.7517	data0.fm1[a], fm2[B]

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domsky, 2016)

[b] $Oxalate^{2-}$: $C_2O_4^{2-}$

[e] data0.ymp.R2 is one of the EQ3/6 databases that comes within the installation package for EQ3/6 v.8.0a.

[f] By analogy to MgOxalate(aq)

Table 6. Pitzer interaction parameters for the Ca^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Oxalate^{-2}$ - H_2O system

i	j	α_1/α_2[A]	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	Source
Na^+	Cl^-	2.0/12.0	0.0765	0.2664	0.0	0.00127	data0.fm1[a], fm2[B]
Na^+	OH^-	2.0/12.0	0.0864	0.253	0.0	0.0044	data0.fm1[a], fm2[B]
Na^+	$HOxalate^-$	2.0/12.0	-0.2448	0.29	0.0	0.068	data0.fm1[a], fm2[B]
Na^+	$Oxalate^{2-}$[b]	2.0/12.0	-0.2176	1.74	0.0	0.122	data0.fm1[a], fm2[B]
Mg^{2+}	Cl^-	2.0/12.0	0.35235	1.6815	0.0	0.00519	data0.fm1[a], fm2[B]
$MgOH^+$	Cl^-	2.0/12.0	-0.1	1.658	0.0	0.0	data0.fm1[a], fm2[B]
H^+	Cl^-	2.0/12.0	0.1775	0.2945	0.0	0.0008	data0.fm1[a], fm2[B]
Ca^{2+}	Cl^-	2.0/12.0	0.3159	1.614	0.0	-0.00034	data0.fm1[a], fm2[B]
Ca^{2+}	OH^-	2.0/12.0	-0.1747	-0.2303	-5.72	0.0	data0.fm1[a], fm2[B]
Mg^{2+}	$Mg(Oxalate)_2^{-2}$	1.4/12.0	0.0434	3.27	-45.74	0.0	This report
i	**j**		**$\theta_{cc'}$ or $\theta_{aa'}$**				**Source**
Na^+	Mg^{2+}		0.07				data0.fm1[a], fm2[B]
Na^+	H^+		0.036				data0.fm1[a], fm2[B]
Na^+	Ca^{2+}		0.07				data0.fm1[a], fm2[B]
Mg^{2+}	H^+		0.10				data0.fm1[a], fm2[B]
Ca^{2+}	Mg^{2+}		0.007				data0.fm1[a], fm2[B]
Ca^{2+}	H^+		0.092				data0.fm1[a], fm2[B]
Cl-	OH^-		-0.050				data0.fm1[a], fm2[B]
i	**j**		**λ_{nc} or λ_{na}**				**Source**
MgOxalate(aq)	Cl^-		0.0189				data0.fm1[a], fm2[B]
CaOxalate(aq)	Cl^-		0.0189[A]				data0.fm1[a], fm2[B]
MgOxalate(aq)	Na^+		0.114				This report
MgOxalate(aq)	Mg^{2+}		-0.283				This report
i	**j**	**k**	**$\psi_{cc'a}$ or $\psi_{aa'c}$**				**Source**
Na^+	Ca^{2+}	Cl-	-0.007				data0.fm1[a], fm2[B]
Na^+	Ca^{2+}	OH-	-0.0198				New in data0.fm2
Na^+	Mg^{2+}	Cl-	-0.012				data0.fm1[a], fm2[B]
Na^+	H^+	Cl^-	-0.004				data0.fm1[a], fm2[B]
Mg^{2+}	$MgOH^+$	Cl-	0.028				data0.fm1[a], fm2[B]
Mg^{2+}	H^+	Cl-	-0.011				data0.fm1[a], fm2[B]
Ca^{2+}	Mg^{2+}	Cl-	-0.012				data0.fm1[a], fm2[B]
Ca^{2+}	H^+	Cl^-	-0.015				data0.fm1[a], fm2[B]
Cl-	OH^-	Na^+	-0.006				data0.fm1[a], fm2[B]
Cl-	OH^-	Ca^{2+}	-0.025				data0.fm1[a], fm2[B]

[A] By analogy to MgOxalate(aq)

[a] data0.fm1: See Xiong (2011) , [B] data0.fm2 (Xiong and Domsky, 2016)

[b] $Oxalate^{2-}$: $C_2O_4^{2-}$

Figure 6. XRD scans of $Fe(OH)_2(s)$ and hibbingite aged in NaCl + Na_2H_2EDTA solutions. Both $Fe(OH)_2(s)$ and hibbingite ($Fe_2Cl(OH)_3(s)$) persist for experiments with NaCl > 0.7 m.

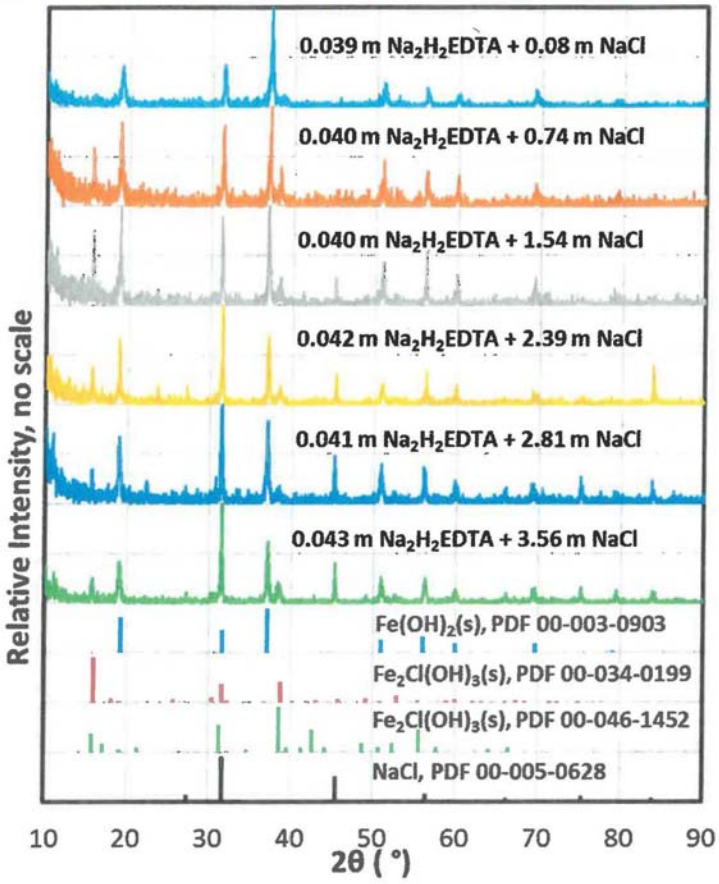

Figure 7. The concentrations (A) and activity coefficients (B) of relevant Fe^{+2} and $EDTA^{-4}$ species in equilibrium with Fe(OH)2(s) in NaCl + Na2H2EDTA solutions. The calculated and measured $\Sigma[Fe^{+2}]$ are plotted together.

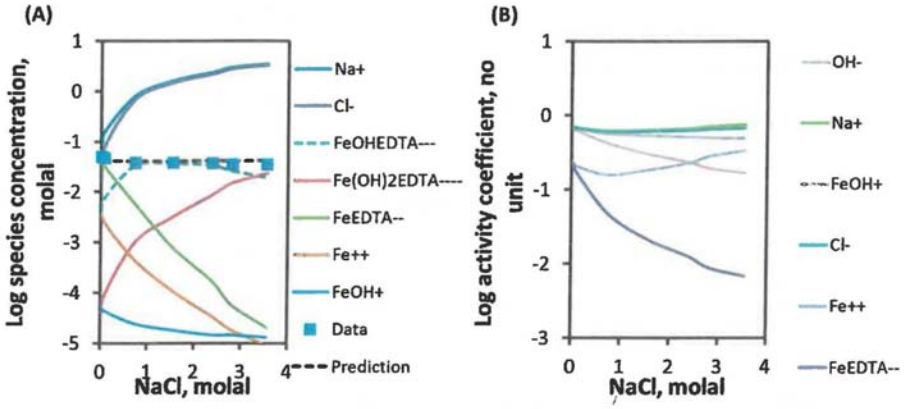

Table 7. Reactions and their 10-based logarithms of equilibrium constants (logKs) for the Fe^{+2} - Na^+ - H^+ - Cl^- - $EDTA^{-4}$ - H_2O system.

Reactions	LogK	Source
Aqueous reactions		
$H^+ + OH^- = H_2O$	13.9967	data0.fm1[a], fm2[B]
$H_4EDTA(aq) = 4H^+ + EDTA^{4-}$[c]	-23.0393	data0.fm1, fm2
$H_3EDTA^- = 3H^+ + EDTA^{4-}$	-20.5374	data0.fm1, fm2
$H_2EDTA^{2-} = 2H^+ + EDTA^{4-}$	-17.4500	data0.fm1, fm2
$HEDTA^{3-} = H^+ + EDTA^{4-}$	-10.5707	data0.fm1, fm2
$MgOH^+ + H^+ = Mg^{2+} + H_2O$	11.8091	data0.fm1, fm2
$MgEDTA^{2-} = Mg^{2+} + EDTA^{4-}$	-10.1260	data0.fm1, fm2
$FeOH^+ + H^+ = Fe^{2+} + H_2O$	9.3148	data0.ymp.R2[e]
$Fe(OH)_2(aq) + 2H^+ = Fe^{2+} + 2H_2O$	20.8214	Stumm and Morgan (1996)
$Fe(OH)_3^- + 3H^+ = Fe^{2+} + 3H_2O$	31.0000	data0.ymp.R2[e]
$FeHEDTA^- = Fe^{+2} + H^+ + EDTA^{-4}$	-19.3	Morel and Hering (1993)
$FeEDTA^{2-} = Fe^{2+} + EDTA^{4-}$	-16.1	Morel and Hering (1993)
$FeOHEDTA^{-3} + H^+ = Fe^{+2} + EDTA^{-4} + H_2O$	-6.4	Morel and Hering (1993)
$Fe(OH)_2EDTA^{-4} + 2H^+ = Fe^{+2} + EDTA^{-4} + 2H_2O$	4.3	Morel and Hering (1993)
Dissolution		
$Fe(OH)_2(s) + 2H^+ = Fe^{2+} + 2H_2O$	12.83	This report
$Fe_2Cl(OH)_3(s) + 3H^+ = 2Fe^{2+} + Cl^- + 3H_2O$	16.89	This report

[a] data0.fm1: See Xiong (2011) , [B] data0.fm2 (Xiong and Domsky, 2016)

[c] $EDTA^{4-}$: $C_{10}H_{12}O_8N_2^{4-}$

[e] data0.ymp.R2 is one of the EQ3/6 databases that comes within the installation package for EQ3/6 v.8.0a.

Table 8. Pitzer interaction parameters for the Fe^{+2} - Na^+ - H^+ - Cl^- - $EDTA^{-4}$ - H_2O system.

i	j	α_1/α_2[A]	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^ϕ	Source
Na^+	Cl^-	2.0/12.0	0.0765	0.2664	0.0	0.00127	data0.fm1[a], fm2
Na^+	OH^-	2.0/12.0	0.0864	0.253	0.0	0.0044	data0.fm1, fm2
Na^+	H_3EDTA^-	2.0/12.0	-0.2345	0.29	0.0	0.059	data0.fm1, fm2
Na^+	H_2EDTA^{2-}	2.0/12.0	-0.1262	1.74	0.0	0.054	data0.fm1, fm2
Na^+	$HEDTA^{3-}$	2.0/12.0	0.5458	5.22	0.0	-0.048	data0.fm1, fm2
Na^+	$EDTA^{4-}$,[c]	2.0/12.0	1.016	11.6	0.0	0.001	data0.fm1, fm2
Na^+	$MgEDTA^{2-}$	2.0/12.0	0.2134	1.74	0.0	0.00869	data0.fm1, fm2
Mg^{2+}	Cl^-	2.0/12.0	0.35235	1.6815	0.0	0.00519	data0.fm1, fm2
$MgOH^+$	Cl^-	2.0/12.0	-0.1	1.658	0.0	0.0	data0.fm1, fm2
Mg^{2+}	$EDTA^{4-}$,[c]	1.4/12.0	-0.01	11.6	0.0	0.30	new in fm2, to be tested
H^+	Cl^-	2.0/12.0	0.1775	0.2945	0.0	0.0008	data0.fm1, fm2
Fe^{2+}	Cl^-	2.0/12.0	0.37324	1.13499	0.0	-0.02152	Moog et al. (2004)
Na^+	$Fe(OH)_2EDTA^{-4}$	2.0/12.0	0.0256	11.6	-0.05	0.0217	This report

i	j	$\theta_{cc'}$ or $\theta_{aa'}$	Source
Na^+	Mg^{2+}	0.07	data0.fm1, fm2
Na^+	H^+	0.036	data0.fm1, fm2
Mg^{2+}	H^+	0.10	data0.fm1, fm2
Cl-	OH^-	-0.050	data0.fm1, fm2
Na^+	Fe^{2+}	0.10945	Moog et al. (2004)
Na^+	$FeOH^+$	-0.0974	Jang and Kim (2016)
Mg^{2+}	Fe^{2+}	0.14504	Moog et al. (2004)

i	j	k	$\psi_{cc'a}$ or $\psi_{aa'c}$	Source
Na^+	Mg^{2+}	Cl-	-0.012	data0.fm1, fm2
Na^+	H^+	Cl^-	-0.004	data0.fm1, fm2
Mg^{2+}	$MgOH^+$	Cl-	0.028	data0.fm1, fm2
Mg^{2+}	H^+	Cl-	-0.011	data0.fm1, fm2
Cl-	OH^-	Na^+	-0.006	data0.fm1, fm2
Fe^{+2}	Na^+	Cl^-	-0.01605	Moog et al. (2004)
Fe^{+2}	Mg^{+2}	Cl^-	-0.02985	Moog et al. (2004)

[A] α_1 and α_2 are pre-set constants used in the Pitzer activity coefficient equation. α_1 and α_2 apply for only cation-anion binary pair. α_2 is not applied when $\beta^{(2)}$ is zero or not used. Unit for α_1 and α_2 is $kg^{1/2}\cdot mol^{-1/2}$.

[a] Xiong (2011), [b] Oxalate^{2-}: $C_2O_4^{2-}$, [c] $EDTA^{4-}$: $C_{10}H_{12}O_8N_2^{4-}$, [d] Citrate^{3-}: $C_6H_5O_7^{3-}$

Figure 8. XRD scans of $Fe(OH)_2(s)$ aged in MgHCitrate + NaCl solutions. No mineralogical changes observed.

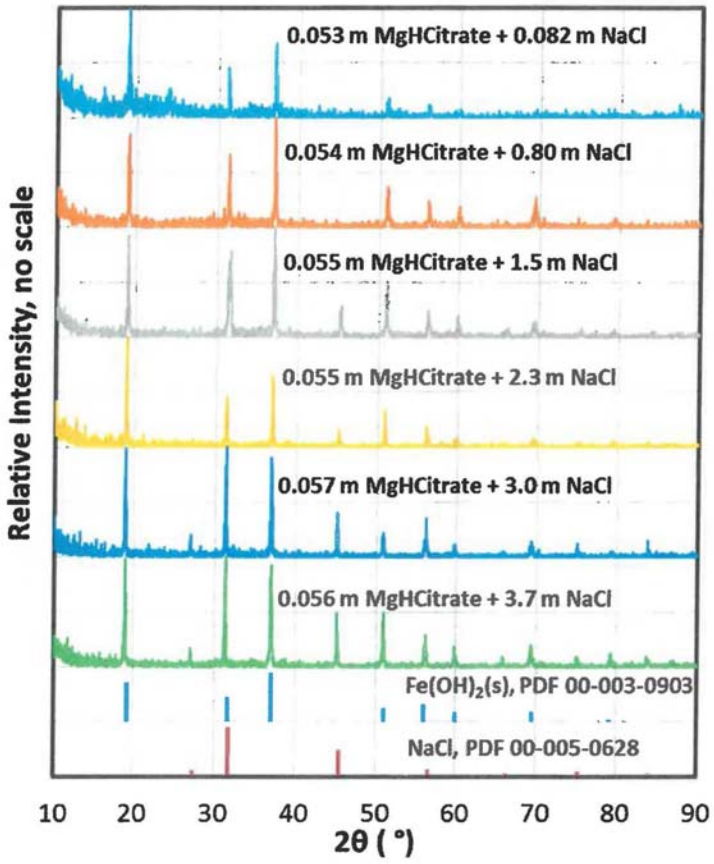

Figure 9. XRD scans of $Fe(OH)_2(s)$ aged in $Na_3Citrate$ + NaCl solutions. No mineralogical change was observed.

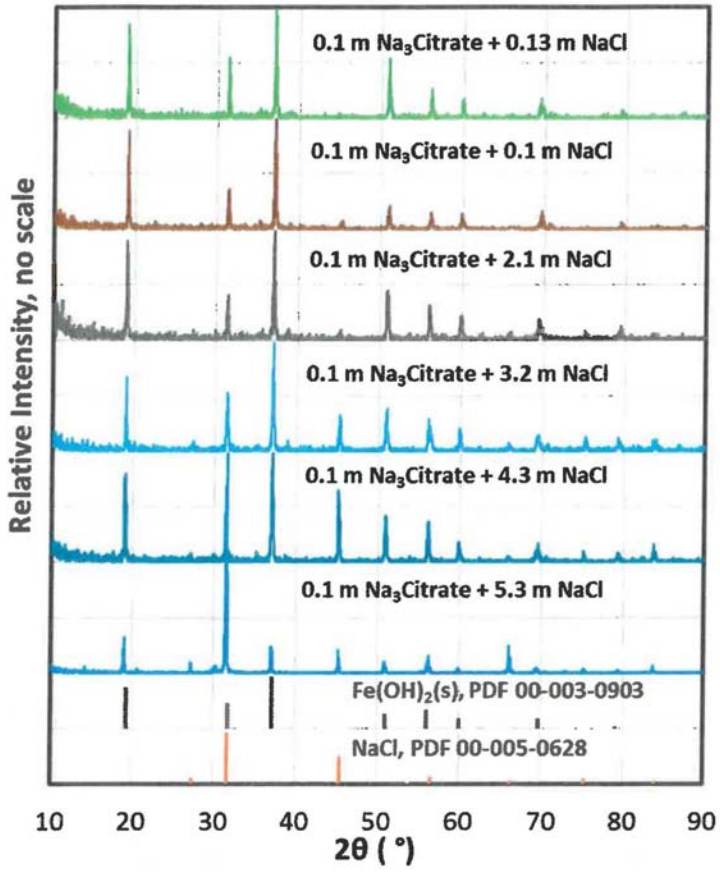

Figure 10. The concentrations (A and B) and activity coefficients (C and D) of relevant Fe^{+2} and $Citrate^{-3}$ species in equilibrium with $Fe(OH)_2(s)$. The calculated and measured $\Sigma[Fe^{+2}]$ are plotted together. (A, C) Experiments in MgHCitrate + NaCl solutions, (B, D) Experiments in Na3Citrate + NaCl solutions. Fitting variables are logK of $FeOHCitrate^{-2}$; $\beta^{(0)}$ and C^{Φ} of $Mg^{+2}/FeOHCitrate^{-2}$ with $\beta^{(1)}$ and $\beta^{(2)}$ set to 3.27 and -45.74; $\beta^{(0)}$, $\beta^{(2)}$, and C^{Φ} of $Na^{+}/FeOHCitrate^{-2}$ with $\beta^{(1)}$ set to 1.74. They are determined to be, 1.92; -0.0518, 0.134; -0.197, -0.0912, 0.0885, respectively. Residuals are 0.115 (A) + 0.016 (B) = 0.131. The residuals prior to the fitting those variables were 0.182 (A) + 0.075 (B) = 0.258.

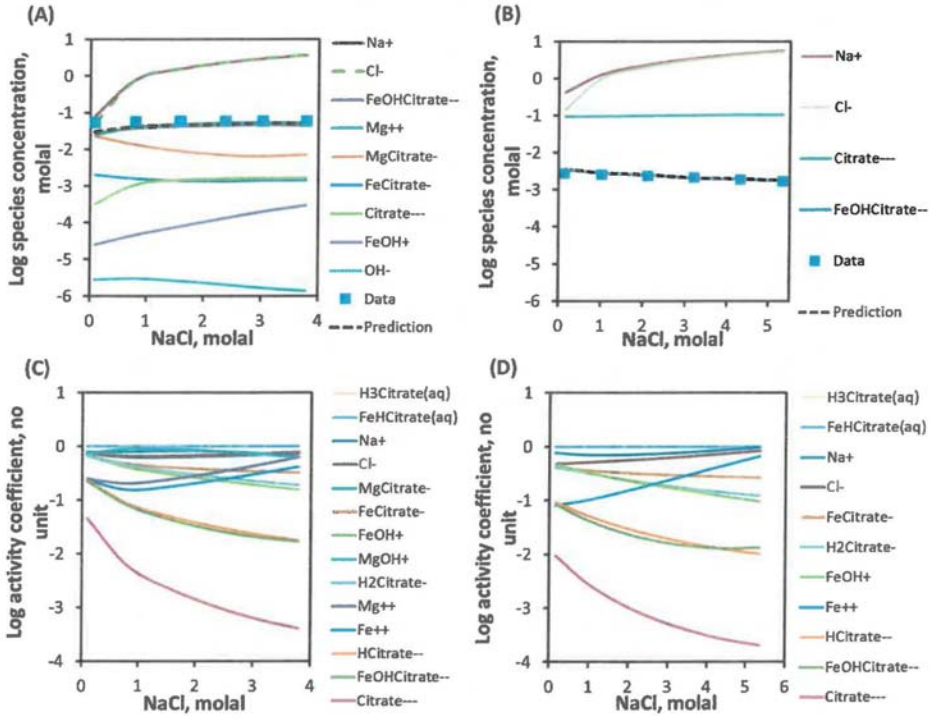

Table 9. Reactions and 10-based logarithms of their equilibrium constants (logKs) for the Fe^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Citrate^{-3}$ - H_2O system

Reactions	LogK	Source
Aqueous reactions		
$H^+ + OH^- = H_2O$	13.9967	data0.fm1[a], fm2[B]
$H_3Citrate(aq) = 3H^+ + Citrate^{3-}$ [d]	-14.5098	data0.fm1[a], fm2[B]
$H_2Citrate^- = 2H^+ + Citrate^{3-}$	-11.2630	data0.fm1[a], fm2[B]
$HCitrate^{2-} = H^+ + Citrate^{3-}$	-6.4232	data0.fm1[a], fm2[B]
$MgOH^+ + H^+ = Mg^{2+} + H_2O$	11.8091	data0.fm1[a], fm2[B]
$MgCitrate^- = Mg^{2+} + Citrate^{3-}$	-5.2997	data0.fm1[a], fm2[B]
$FeOH^+ + H^+ = Fe^{2+} + H_2O$	9.3148	data0.ymp.R2[e]
$Fe(OH)_2(aq) + 2H^+ = Fe^{2+} + 2H_2O$	20.8214	Stumm and Morgan (1996)
$Fe(OH)_3^- + 3H^+ = Fe^{2+} + 3H_2O$	31.0000	data0.ymp.R2[e]
$FeOHCitrate^{-2} + H^+ = Fe^{2+} + Citrate^{3-} + H_2O$	1.92	This report
$FeCitrate^- = Fe^{2+} + Citrate^{3-}$	-5.7	Morel and Hering (1993)
$FeHCitrate(aq) = Fe^{2+} + H^+ + Citrate^{3-}$	-9.9	Morel and Hering (1993)
Dissolution		
$Fe(OH)_2(s) + 2H^+ = Fe^{2+} + 2H_2O$	12.83	This report
$Fe_2Cl(OH)_3(s) + 3H^+ = 2Fe^{2+} + Cl^- + 3H_2O$	16.89	This report

[a] data0.fm1: See Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016)

[d] $Citrate^{3-}$: $C_6H_5O_7^{3-}$

[e] data0.ymp.R2 is one of the EQ3/6 databases that comes within the installation package for EQ3/6 v.8.0a.

Table 10. Pitzer interaction parameters for the Fe^{+2} - Mg^{+2} - Na^{+} - H^{+} - Cl^{-} - $Citrate^{-3}$ - H_2O system

i	j	α_1/α_2[A]	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	Source
Na^{+}	Cl^{-}	2.0/12.0	0.0765	0.2664	0.0	0.00127	data0.fm1[a], fm2[B]
Na^{+}	OH^{-}	2.0/12.0	0.0864	0.253	0.0	0.0044	data0.fm1[a], fm2[B]
Na^{+}	$H_2Citrate^{-}$	2.0/12.0	-0.1296	0.29	0.0	0.013	data0.fm1[a], fm2[B]
Na^{+}	$HCitrate^{2-}$	2.0/12.0	-0.0989	1.74	0.0	0.027	data0.fm1[a], fm2[B]
Na^{+}	$Citrate^{3-}$[,d]	2.0/12.0	0.0887	5.22	0.0	0.047	data0.fm1[a], fm2[B]
Na^{+}	$MgCitrate^{-}$	2.0/12.0	0.1742	0.29	0.0	-0.06923	data0.fm1[a], fm2[B]
Mg^{2+}	Cl^{-}	2.0/12.0	0.35235	1.6815	0.0	0.00519	data0.fm1[a], fm2[B]
$MgOH^{+}$	Cl^{-}	2.0/12.0	-0.1	1.658	0.0	0.0	data0.fm1[a], fm2[B]
Mg^{2+}	$MgCitrate^{-}$	2.0/12.0	1.0915	1.74	0.0	0.0	New in data0.fm2[B]
Mg^{2+}	$Citrate^{3-}$[,d]	1.4/12.0	0.9330	4.4	0.0	0.0	New in data0.fm2[B]
H^{+}	Cl^{-}	2.0/12.0	0.1775	0.2945	0.0	0.0008	data0.fm1[a], fm2[B]
Fe^{2+}	Cl^{-}	2.0/12.0	0.37324	1.13499	0.0	-0.02152	Moog et al. (2004)
Na^{+}	$FeOHCitrate^{-2}$	2.0/12.0	-0.197	1.74	-0.0912	0.0885	This report
Mg^{+2}	$FeOHCitrate^{-2}$	1.4/12.0	-0.0518	3.27	-45.74	0.134	This report

i	j	$\theta_{cc'}$ or $\theta_{aa'}$	Source
Na^{+}	Mg^{2+}	0.07	data0.fm1[a], fm2[B]
Na^{+}	H^{+}	0.036	data0.fm1[a], fm2[B]
Mg^{2+}	H^{+}	0.10	data0.fm1[a], fm2[B]
Cl-	OH^{-}	-0.050	data0.fm1[a], fm2[B]
Na^{+}	Fe^{2+}	0.10945	Moog et al. (2004)
Na^{+}	$FeOH^{+}$	-0.0974	This report
Mg^{2+}	Fe^{2+}	0.14504	Moog et al. (2004)

i	j	k	$\psi_{cc'a}$ or $\psi_{aa'c}$	Source
Na^{+}	Mg^{2+}	Cl-	-0.012	data0.fm1[a], fm2[B]
Na^{+}	H^{+}	Cl^{-}	-0.004	data0.fm1[a], fm2[B]
Mg^{2+}	$MgOH^{+}$	Cl-	0.028	data0.fm1[a], fm2[B]
Mg^{2+}	H^{+}	Cl-	-0.011	data0.fm1[a], fm2[B]
Cl-	OH^{-}	Na^{+}	-0.006	data0.fm1[a], fm2[B]
Fe^{+2}	Na^{+}	Cl^{-}	-0.01605	Moog et al. (2004)
Fe^{+2}	Mg^{+2}	Cl^{-}	-0.02985	Moog et al. (2004)

[A] α_1 and α_2 are pre-set constants used in the Pitzer activity coefficient equation. α_1 and α_2 apply for only cation-anion binary pair. α_2 is not applied when $\beta^{(2)}$ is zero or not used. Unit for α_1 and α_2 is $kg^{1/2}\cdot mol^{-1/2}$.

[a] Xiong (2011), [B] data0.fm2 (Xiong and Domski, 2016), [b] Oxalate^{2-}: $C_2O_4^{2-}$, [c] EDTA^{4-}: $C_{10}H_{12}O_8N_2^{4-}$, [d] Citrate^{3-}: $C_6H_5O_7^{3-}$