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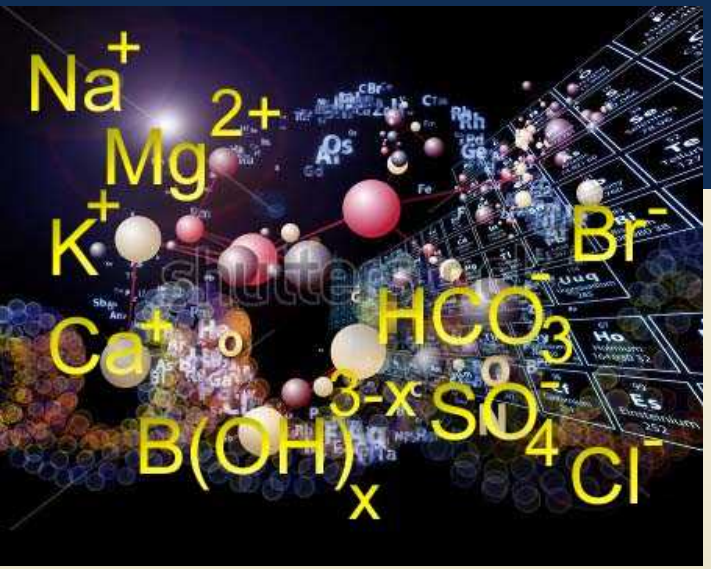

WIPP Thermodynamic Database History and Recent Revisions

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U.S. DEPARTMENT OF ENERGY | NNSA National Nuclear Security Administration

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OUTLINE OF PRESENTATION

- Outline of the WIPP Thermodynamic Database (DATA0.FM1)
- Recent Updates to the WIPP Thermodynamic Database (DATA0.FM2)
- Baseline Solubilities Using DATA0.FM1 and DATA0.FM2
- Uncertainty Distribution for An(IV) using DATA0.FM2
- Uncertainty Distribution for An(III) using DATA0.FM2

WIPP Thermodynamic Database

- The WIPP thermodynamic database
  - at 25°C
  - uses the Pitzer model for calculation of activity coefficients of aqueous species.

- Major Ion Chemistry
  - Harvie-Moller-Weare Model (Harvie et al., 1984): Na-K-H-Ca-Mg-Cl-SO_4-CO_2-H_2O

- Borate Chemistry
  - Felmy-Weare Model (Felmy & Weare, 1986): Na-K-H-Ca-Mg-Cl-SO_4-CO_2-$B(OH)_3$-H_2O

WIPP Thermodynamic Database

- Actinide Chemistry
  - Oxidation State Analogs
    - Am(III) Model: Am(III), Pu(III)
    - Th(IV) Model: Np(IV), Pu(IV), Th(IV)
    - Np(V) Model: Np(V)
    - U(VI) Model: U(VI)
  - Sources
    - Am(III) Model: Pacific Northwest National Laboratory (PNNL), funded by Sandia National Laboratories (SNL)
    - Th(IV) Model: PNNL, funded by SNL
    - Np(V) Model: Lawrence Berkeley National Laboratory (LBNL), funded by SNL
    - Organic Species: Florida State University (FSU), funded by SNL
    - U(VI) Model: Set to 10^{-3} M by EPA
- For CCA, CRA-2004, CRA-2009, DOE used the FMT code for PA calculations. In 2011, EPA approved the migration from the FMT code to EQ3/6 Version 8.0a for PA calculations.

What Do We Mean by a Model?

(I-1)	$H_2O = OH^- + H^+$
(I-2)	$Ca^{2+} + 2H_2O = Ca(OH)_2$(portlandite) $+ 2H^+$
(I-3)	$Na^+ + Cl^- = NaCl$(halite)
(I-4)	$K^+ + Cl^- = KCl$(sylvite)
(I-5)	$Ca^{2+} + 2Cl^- + 4H_2O = CaCl_2 \cdot 4H_2O$(cr)
(I-6)	$4Ca^{2+} + 2Cl^- + 19H_2O = Ca_4Cl_2(OH)_6 \cdot 13H_2O$(CaOxychloride_A) $+ 6H^+$
(I-7)	$2Ca^{2+} + 2Cl^- + 3H_2O = Ca_2Cl_2(OH)_2 \cdot H_2O$(CaOxychloride_B) $+ 2H^+$
(I-8)	$K^+ + Mg^{2+} + 3Cl^- + 6H_2O = KMgCl_3 \cdot 6H_2O$(carnallite)
(I-9)	$H^+ + HCO_3^- = CO_2(aq) + H_2O$
(I-10)	$HCO_3^- = CO_3^{2-} + H^+$
(I-11)	$Na^+ + HCO_3^- = NaHCO_3$(nahcolite)
(I-12)	$3Na^+ + 2HCO_3^- + 2H_2O = Na_3H(CO_3)_2 \cdot 2H_2O$(trona) $+ H^+$
(I-13)	$2Na^+ + HCO_3^- + 10H_2O = Na_2CO_3 \cdot 10H_2O$(natron) $+ H^+$
(I-14)	$2Na^+ + HCO_3^- + 7H_2O = Na_2CO_3 \cdot 7H_2O$(cr) $+ H^+$
(I-15)	$2Na^+ + HCO_3^- + H_2O = Na_2CO_3 \cdot H_2O$(thermonatrite) $+ H^+$
(I-16)	$K^+ + HCO_3^- = KHCO_3$(kalicinite)
(I-17)	$2K^+ + HCO_3^- + 1.5H_2O = K_2CO_3 \cdot 3/2H_2O$(cr) $+ H^+$
(I-18)	$8K^+ + 6HCO_3^- + 3H_2O = K_8H_4(CO_3)_6 \cdot 3H_2O$(cr) $+ 2H^+$
(I-19)	$K^+ + Na^+ + HCO_3^- + 6H_2O = KNaCO_3 \cdot 6H_2O$(cr) $+ H^+$
(I-20)	$2K^+ + Na^+ + 2HCO_3^- + 2H_2O = K_2NaH(CO_3)_2 \cdot 2H_2O$(K-trona) $+ H^+$

What Do We Mean by a Model?

(I-21)	$Ca^{2+} + HCO_3^- = CaCO_3(aragonite) + H^+$
(I-22)	$Ca^{2+} + HCO_3^- = CaCO_3(calcite) + H^+$
(I-23)	$Ca^{2+} + 2Na^+ + 2HCO_3^- + 5H_2O = CaNa_2(CO_3)_2 \cdot 5H_2O(gaylussite) + 2H^+$
(I-24)	$2Na^+ + Ca^{2+} + 2HCO_3^- + 2H_2O = Na_2Ca(CO_3)_2 \cdot 2H_2O(pirssonite) + 2H^+$
(I-25)	$SO_4^{2-} + H^+ = HSO_4^- + H^+$
(I-26)	$Ca^{2+} + SO_4^{2-} = CaSO_4(anhydrite)$
(I-27)	$Ca^{2+} + SO_4^{2-} + 2H_2O = CaSO_4 \cdot 2H_2O(gypsum)$
(I-28)	$2K^+ + SO_4^{2-} = K_2SO_4(arcanite)$
(I-29)	$8K^+ + 6H^+ + 7SO_4^{2-} = K_8H_6(SO_4)_7(misenite)$
(I-30)	$3K^+ + H^+ + 2SO_4^{2-} = K_3H(SO_4)_2(cr)$
(I-31)	$K^+ + Ca^{2+} + 2SO_4^{2-} + H_2O = K_2Ca(SO_4)_2 \cdot H_2O(syngenite)$
(I-32)	$2Na^+ + SO_4^{2-} + 10H_2O = Na_2SO_4 \cdot 10H_2O(mirabilite)$
(I-33)	$2Na^+ + SO_4^{2-} = Na_2SO_4(thenardite)$
(I-34)	$Na^+ + H^+ + 2SO_4^{2-} = Na_3H(SO_4)_2(cr)$
(I-35)	$Na^+ + 3K^+ + 2SO_4^{2-} = NaK_3(SO_4)_2(aphthitalite, galerite)$
(I-36)	$2Na^+ + Ca^{2+} + 2SO_4^{2-} = Na_2Ca(SO_4)_2(glauberite)$
(I-37)	$4Na^+ + Ca^{2+} + 3SO_4^{2-} + 2H_2O = Na_4Ca(SO_4)_3 \cdot 2H_2O(labile\ salt)$
(I-38)	$HPO_4^{2-} + 2H^+ = H_3PO_4(aq)$
(I-39)	$HPO_4^{2-} + H^+ = H_2PO_4^- + H^+$
(I-40)	$HPO_4^{2-} = PO_4^{3-} + H$

What Do We Mean by a Model?

(II-1)	$Mg^{2+} + H_2O = MgOH^+ + H^+$
(II-2)	$Mg^{2+} + 2H_2O = Mg(OH)_2$(brucite) $+ 2H^+$
(II-3)	$Mg^{2+} + 2Cl^- + 6H_2O = MgCl_2 \cdot 6H_2O$(bischofite)
(II-4)	$2Mg^{2+} + Cl^- + 7H_2O = Mg_2Cl(OH)_3 \cdot 4H_2O$(phase 3) $+ 3H^+$
(II-5)	$3Mg^{2+} + Cl^- + 9H_2O = Mg_3Cl(OH)_5 \cdot 4H_2O$(phase 5) $+ 5H^+$
(II-6)	$2Mg^{2+} + Ca^{2+} + 6Cl^- + 12H_2O = Mg_2CaCl_6 \cdot 12H_2O$(tachyhydrite)
(II-7)	$K^+ + Mg^{2+} + 3Cl^- + 6H_2O = KMgCl_3 \cdot 6H_2O$(carnallite)
(II-8)	$Mg^{2+} + HCO_3^- = MgCO_3$(magnesite) $+ H^+$
(II-9)	$Mg^{2+} + HCO_3^- + 3H_2O = MgCO_3 \cdot 3H_2O$(nesquehonite) $+ H^+$
(II-10)	$4Mg^{2+} + 3HCO_3^- + 5H_2O = Mg_4(CO_3)_3(OH)_2 \cdot 3H_2O$(hydromagnesite4323) $+ 5H^+$
(II-11)	$5Mg^{2+} + 4HCO_3^- + 6H_2O = Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$(hydromagnesite5424) $+ 6H^+$
(II-12)	$2K^+ + Mg^{2+} + 2SO_4^{2-} + 4H_2O = K_2Mg(SO_4)_2 \cdot 4H_2O$(leonite)
(II-14)	$2Na^+ + Mg^{2+} + 2SO_4^{2-} + 2H_2O = Na_2Mg(SO_4)_2 \cdot 4H_2O$(boedite)
(II-15)	$Mg^{2+} + SO_4^{2-} + 7H_2O = MgSO_4 \cdot 7H_2O$(epsomite)
(II-16)	$Mg^{2+} + SO_4^{2-} + 6H_2O = MgSO_4 \cdot 6H_2O$(hexahydrite)
(II-17)	$Mg^{2+} + SO_4^{2-} + H_2O = MgSO_4 \cdot H_2O$(kieserite)
(II-18)	$K^+ + Mg^{2+} + Cl^- + SO_4^{2-} + 3H_2O = KMgClSO_4 \cdot 3H_2O$(kainite)
(II-19)	$Mg^{2+} + SO_4^{2-} = MgSO_4$(aq)

What Do We Mean by a Model?

(III-1)	$Mg^{2+} + Acetate^{-} = MgAcetate^{+}$
(III-2)	$Ca^{2+} + Acetate^{-} = CaAcetate^{+}$
(III-3)	$Citrate^{3-} + 3H^{+} = H_3Citrate\ (aq)$
(III-4)	$Citrate^{3-} + 2H^{+} = H_2Citrate^{-}$
(III-5)	$Citrate^{3-} + H^{+} = HCitrate^{2-}$
(III-6)	$Mg^{2+} + Citrate^{3-} = MgCitrate^{-}$
(III-7)	$Ca^{2+} + Citrate^{3-} = CaCitrate^{-}$
(III-8)	$Ca_3(Citrate)_2 \cdot 4H_2O\ (earlandite) = 3Ca^{2+} + 2Citrate^{3-} + 4H_2O$
(III-9)	$EDTA^{4-} + 4H^{+} = H_4EDTA(aq)$
(III-10)	$EDTA^{4-} + 3H^{+} = H_3EDTA^{-}$
(III-11)	$EDTA^{4-} + 2H^{+} = H_2EDTA^{2-}$
(III-12)	$EDTA^{4-} + H^{+} = HEDTA^{3-}$
(III-13)	$Mg^{2+} + EDTA^{4-} = MgEDTA^{2-}$
(III-14)	$Ca^{2+} + EDTA^{4-} = CaEDTA^{2-}$

What Do We Mean by a Model?

(III-15)	$Ca_2EDTA(s) = 2Ca^{2+} + EDTA^{4-}$
(III-16)	$Oxalate^{2-} + 2H^{+} = H_2Oxalate(aq)$
(III-17)	$Oxalate^{2-} + H^{+} = HOxalate^{-}$
(III-18)	$Mg^{2+} + Oxalate^{2-} = MgOxalate(aq)$
(III-19)	$Ca^{2+} + Oxalate^{2-} = CaOxalate(aq)$
(III-20)	$Oxalate^{2-} + 2H^{+} + 2H_2O = H_2Oxalate \cdot 2H_2O$
(III-21)	$Na^{+} + H^{+} + Oxalate^{2-} + H_2O = NaHOxalate \cdot H_2O$
(III-22)	$2Na^{+} + Oxalate^{2-} = Na_2Oxalate$
(III-23)	$Ca^{2+} + Oxalate^{2-} + H_2O = CaOxalate \cdot H_2O$
(III-24)	$Lactate^{-} + H^{+} = HLactate(aq)$

What Do We Mean by a Model?

(IV-1)	$AmCO_3^+ + H^+ = Am^{3+} + HCO_3^-$
(IV-2)	$Am(CO_3)_2^- + 2H^+ = Am^{3+} + 2HCO_3^-$
(IV-3)	$Am(CO_3)_3^{3-} + 3H^+ = Am^{3+} + 3HCO_3^-$
(IV-4)	$Am(CO_3)_4^{5-} + 4H^+ = Am^{3+} + 4HCO_3^-$
(IV-5)	$Am(OH)^{2+} + H^+ = Am^{3+} + H_2O$
(IV-6)	$Am(OH)_2^+ + 2H^+ = Am^{3+} + 2H_2O$
(IV-7)	$Am(OH)_3(aq) + 3H^+ = Am^{3+} + 3H_2O$
(IV-8)	$AmCl^{2+} = Am^{3+} + Cl^-$
(IV-9)	$AmCl_2^+ = Am^{3+} + 2Cl^-$
(IV-10)	$AmSO_4^+ = Am^{3+} + SO_4^{2-}$
(IV-11)	$Am(SO_4)_2^- = Am^{3+} + 2SO_4^{2-}$
(IV-12)	$AmAcetate^{2+} = Am^{3+} + Acetate^-$
(IV-13)	$AmCitrate(aq) = Am^{3+} + Citrate^{3-}$

What Do We Mean by a Model?

(IV-14)	$AmEDTA^{-} = Am^{3+} + EDTA^{4-}$
(IV-15)	$AmLactate^{2+} = Am^{3+} + Lactate^{-}$
(IV-16)	$AmOxalate^{+} = Am^{3+} + Oxalate^{2-}$
(IV-17)	$AmOHCO_3(s) + 2H^{+} = Am^{3+} + H_2O + HCO_3^{-}$
(IV-18)	$Am(OH)_3(s) + 3H^{+} = Am^{3+} + 3H_2O$
(IV-19)	$NaAm(CO_3)_2 \cdot 6H_2O(s) + 2H^{+} = Na^{+} + Am^{3+} + 2HCO_3^{-} + 6H_2O$
(IV-20)	$AmPO_4(cr) + H^{+} = Am^{3+} + HPO_4^{2-}$
(IV-21)	$AmHB_4O_7^{2+} + 9H_2O = Am^{3+} + 4B(OH)_4^{-} + 3H^{+}$

What Do We Mean by a Model?

(VI-1)	$Th(CO_3)_5^{6-} + 5H^+ = Th^{4+} + 5HCO_3^-$
(VI-2)	$Th(OH)_3(CO_3)^- + 4H^+ = Th^{4+} + 3H_2O + HCO_3^-$
(VI-3)	$Th(OH)_4(aq) + 4H^+ = Th^{4+} + 4H_2O$
(VI-4)	$Th(SO_4)_2(aq) = Th^{4+} + 2SO_4^{2-}$
(VI-5)	$Th(SO_4)_3^{2-} = Th^{4+} + 3SO_4^{2-}$
(VI-6)	$ThAcetate^{3+} = Th^{4+} + Acetate^-$
(VI-7)	$Th(Acetate)_2^{2+} = Th^{4+} + 2Acetate^-$
(VI-8)	$ThCitrate^+ = Th^{4+} + Citrate^{3-}$
(VI-9)	$ThEDTA(aq) = Th^{4+} + EDTA^{4-}$
(VI-10)	$ThLactate^{3+} = Th^{4+} + Lactate^-$
(VI-11)	$Th(Lactate)_2^{2+} = Th^{4+} + 2Lactate^-$
(VI-12)	$ThOxalate^{2+} = Th^{4+} + Oxalate^{2-}$
(VI-13)	$ThO_2(am) + 4H^+ = Th^{4+} + 2H_2O$
(VI-14)	$Th(SO_4)_2 \cdot 9H_2O(s) = Th^{4+} + 2SO_4^{2-} + 9H_2O$
(VI-15)	$Th(SO_4)_2 \cdot 8H_2O(s) = Th^{4+} + 2SO_4^{2-} + 8H_2O$
(VI-16)	$Th(SO_4)_2 \cdot Na_2SO_4 \cdot 6H_2O(s) = Th^{4+} + 3SO_4^{2-} + 2Na^+ + 6H_2O$
(VI-17)	$Th(SO_4)_2 \cdot K_2SO_4 \cdot 4H_2O(s) = Th^{4+} + 3SO_4^{2-} + 2K^+ + 4H_2O$
(VI-18)	$Th(SO_4)_2 \cdot 2K_2SO_4 \cdot 2H_2O(s) = Th^{4+} + 4SO_4^{2-} + 4K^+ + 2H_2O$
(VI-19)	$2[Th(SO_4)_2 \cdot 7/2K_2SO_4(s)] = 2Th^{4+} + 11SO_4^{2-} + 14K^+$

What Do We Mean by a Model?

(VII-1)	$NpO_2CO_3^- + H^+ = NpO_2^+ + HCO_3^-$
(VII-2)	$NpO_2(CO_3)_2^{3-} + 2H^+ = NpO_2^+ + 2HCO_3^-$
(VII-3)	$NpO_2(CO_3)_3^{5-} + 3H^+ = NpO_2^+ + 3HCO_3^-$
(VII-4)	$NpO_2(OH)(aq) + H^+ = NpO_2^+ + H_2O$
(VII-5)	$NpO_2(OH)_2^- + 2H^+ = NpO_2^+ + 2H_2O$
(VII-6)	$NpO_2Acetate(aq) = NpO_2^+ + Acetate^-$
(VII-7)	$NpO_2Citrate^{2-} = NpO_2^+ + Citrate^{3-}$
(VII-8)	$NpO_2H_2EDTA^- = NpO_2^+ + EDTA^{4-} + 2H^+$
(VII-9)	$NpO_2HEDTA^{2-} = NpO_2^+ + EDTA^{4-} + H^+$
(VII-10)	$NpO_2EDTA^{3-} = NpO_2^+ + EDTA^{4-}$
(VII-11)	$NpO_2Lactate(aq) = NpO_2^+ + Lactate^-$
(VII-12)	$NpO_2Oxalate^- = NpO_2^+ + Oxalate^{2-}$
(VII-13)	$NpO_2OH(aged) + H^+ = NpO_2^+ + H_2O$
(VII-14)	$NpO_2OH(am) + H^+ = NpO_2^+ + H_2O$
(VII-15)	$2[NaNpO_2CO_3 \cdot 7/2H_2O](s) + 2H^+ = 2NpO_2^+ + 2HCO_3^- + 2Na^+ + 7H_2O$
(VII-16)	$Na_3NpO_2(CO_3)_2(s) + 2H^+ = NpO_2^+ + 2HCO_3^- + 3Na^+$
(VII-17)	$KNpO_2CO_3(s) + H^+ = NpO_2^+ + HCO_3^- + K^+$
(VII-18)	$K_3NpO_2(CO_3)_2(s) + 2H^+ = NpO_2^+ + 2HCO_3^- + 3K^+$

What Do We Mean by a Model?

(VIII-1)	$B(OH)_4^- + H^+ = B(OH)_3(aq) + H_2O$
(VIII-2)	$CaB(OH)_4^+ = Ca^{2+} + B(OH)_4^-$
(VIII-3)	$MgB(OH)_4^+ = Mg^{2+} + B(OH)_4^-$
(VIII-4)	$B_3O_3(OH)_4^- + 5H_2O = 3B(OH)_4^- + 2H^+$
(VIII-5)	$B_4O_5(OH)_4^{2-} + 7H_2O = 4B(OH)_4^- + 2H^+$
(VIII-6)	$NaB(OH)_4(aq) = Na^+ + B(OH)_4^-$
(VIII-7)	$Na_2B_4O_7{\bullet}10H_2O$(Na-tetraborate) $= 2Na^+ + 4B(OH)_4^- + 2H^+ + H_2O$
(VIII-8)	$B(OH)_3(cr) + H_2O = B(OH)_4^- + H^+$
(VIII-9)	$KB_5O_8{\bullet}4H_2O$(K-pentaborate) $+ 8H_2O = K^+ + 5B(OH)_4^- + 4H^+$
(VIII-10)	$K_2B_4O_7{\bullet}4H_2O$(K-tetraborate) $+ 5H_2O = 2K^+ + 4B(OH)_4^- + 2H^+$
(VIII-11)	$NaBO_2{\bullet}4H_2O$(Na-metaborate) $= Na^+ + B(OH)_4^- + 2H_2O$
(VIII-12)	$NaB_5O_8{\bullet}5H_2O$(Na-pentaborate) $+ 7H_2O = Na^+ + 5B(OH)_4^- + 4H^+$
(VIII-13)	$NaBO_2{\bullet}NaCl{\bullet}2H_2O$(teepleite) $= 2Na^+ + B(OH)_4^- + Cl^-$

What Do We Mean Parameterization?

Pitzer interaction parameters involving Am(III) species

Binary interaction parameters				
Species *i*	Species *j*	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
Am^{3+}	Cl^-	0.5856	5.6	-0.0166
Am^{3+}	SO_4^{2-}	1.792	15.04	0.600
$AmCO_3^+$	Cl^-	-0.072	0.403	0.0388
$AmOH^{2+}$	Cl^-	-0.055	1.6	0.05
$Am(OH)_2^+$	Cl^-	-0.616	-0.45	0.05
$AmCl^{2+}$	Cl^-	0.593	3.15	-0.006
$AmCl_2^+$	Cl^-	0.516	1.75	0.010
$AmSO_4^+$	Cl^-	-0.091	-0.39	0.048
$AmAc^{2+}$	Cl^-	0.3088	1.74	-0.132
$AmOx^+$	Cl^-	-0.9374	0.29	0.248
Na^+	$Am(CO_3)_2^-$	-0.240	0.224	0.0284
Na^+	$Am(CO_3)_3^{3-}$	0.125	4.73	0.0007
Na^+	$Am(CO_3)_4^{5-}$	2.022	19.22	-0.305
Na^+	$Am(SO_4)_2^-$	-0.345	0.40	0.051
Na^+	$AmEDTA^-$	-0.2239	0.29	0.095
Interaction parameters for neutral species and mixing parameters				
Species *i*	Species *j*	λ_{ij}	θ_{ij}	
$Am(OH)_3^o$	Na^+	-0.2		
$Am(OH)_3^o$	Cl^-	-0.2		
$AmCit^o$	Cl^-	-0.406		
Am^{3+}	Ca^{2+}		0.2	
$AmCl^{2+}$	Ca^{2+}		-0.014	
$AmCl_2^+$	Ca^{2+}		-0.196	
Am^{3+}	Na^+		0.1	

What Do We Mean by a Parameterization?

Pitzer interaction parameters involving Th(IV) species

Binary interaction parameters				
Species *i*	Species *j*	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
Th^{4+}	Cl^-	1.092	13.7; $\beta^{(2)}$= -160	-0.112
Th^{4+}	SO_4^{2-}	1.56	0	0
Th^{4+}	HSO_4^-	1.44	0	0
$ThAc^{3+}$	Cl^-	1.061	5.22	0.109
$ThCit^+$	Cl^-	-0.7467	0.29	0.319
$ThOx^{2+}$	Cl^-	-0.343	1.74	0.5
Na^+	$Th(CO_3)_5^{-6}$	1.31	30	0
Na^+	$Th(SO_4)_3^{2-}$	0.12	0	0
K^+	$Th(SO_4)_3^{2-}$	0.90	0	0
Interaction parameters for neutral species and mixing parameters				
Species *i*	Species *j*	Species *k*	λ_{ij} or θ_{ij}	Ψ_{ijk}
$Th(SO_4)_2^o$	Cl^-		0.29	
$Th(SO_4)_2^o$	HSO_4^-		0.68	
$ThEDTA^o$	Cl^-		0.1111	
Th^{4+}	Na^+	Cl^-	0.42	0.21
Th^{4+}	Mg^{2+}	Cl^-	0.60	0.21
Th^{4+}	H^+	Cl^-	0.60	0.37
Na^+	$Th(CO_3)_5^{-6}$	Cl^-	2.0	-0.08

What Do We Mean by a Parameterization?

Pitzer interaction parameters involving Np(V) species

Binary interaction parameters				
Species *i*	Species *j*	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}
NpO_2^+	Cl^-	0.1415	0.281	0
Na^+	$NpO_2CO_3^-$	0.1	0.34	0
Na^+	$NpO_2(CO_3)_2^{3-}$	0.48	4.4	0
Na^+	$NpO_2(CO_3)_3^{5-}$	1.80	22.7	0
Na^+	NpO_2Cit^{2-}	-0.4226	1.75	0.142
Na^+	$NpO_2H_2EDTA^-$	-0.8285	0.2575	0.256
Na^+	NpO_2EDTA^{3-}	0.683	5.911	0
Na^+	NpO_2Ox^-	-0.5418	0.29	0.095
K^+	$NpO_2(CO_3)_3^{5-}$	2.34	22.7; $\beta^{(2)}$= -96	-0.22
Mg^{2+}	$NpO_2CO_3^-$	0.1	0.34	0
Mg^{2+}	$NpO_2(CO_3)_2^{3-}$	0.48	4.4	0
Mg^{2+}	$NpO_2(CO_3)_3^{5-}$	2.07	22.7; $\beta^{(2)}$= -48	-0.11
Interaction parameters for neutral species and mixing parameters				
Species *i*	Species *j*	Species *k*	λ_{ij} or θ_{ij}	Ψ_{ijk}
NpO_2OH^o	Cl^-		-0.19	
$NpO_2CO_3^-$	Cl^-	Na^+	-0.21	0
$NpO_2(CO_3)_2^{3-}$	Cl^-	Na^+	-0.26	0
$NpO_2(CO_3)_3^{5-}$	Cl^-	Na^+	-0.26	0
$NpO_2(CO_3)_3^{5-}$	CO_3^{2-}	K^+	-1.9	0

What Supports Database Updates?

- Data that can be used to parameterize an additional reaction becomes available
- Data specific to a reaction that can be used to replace an analog assumption becomes available

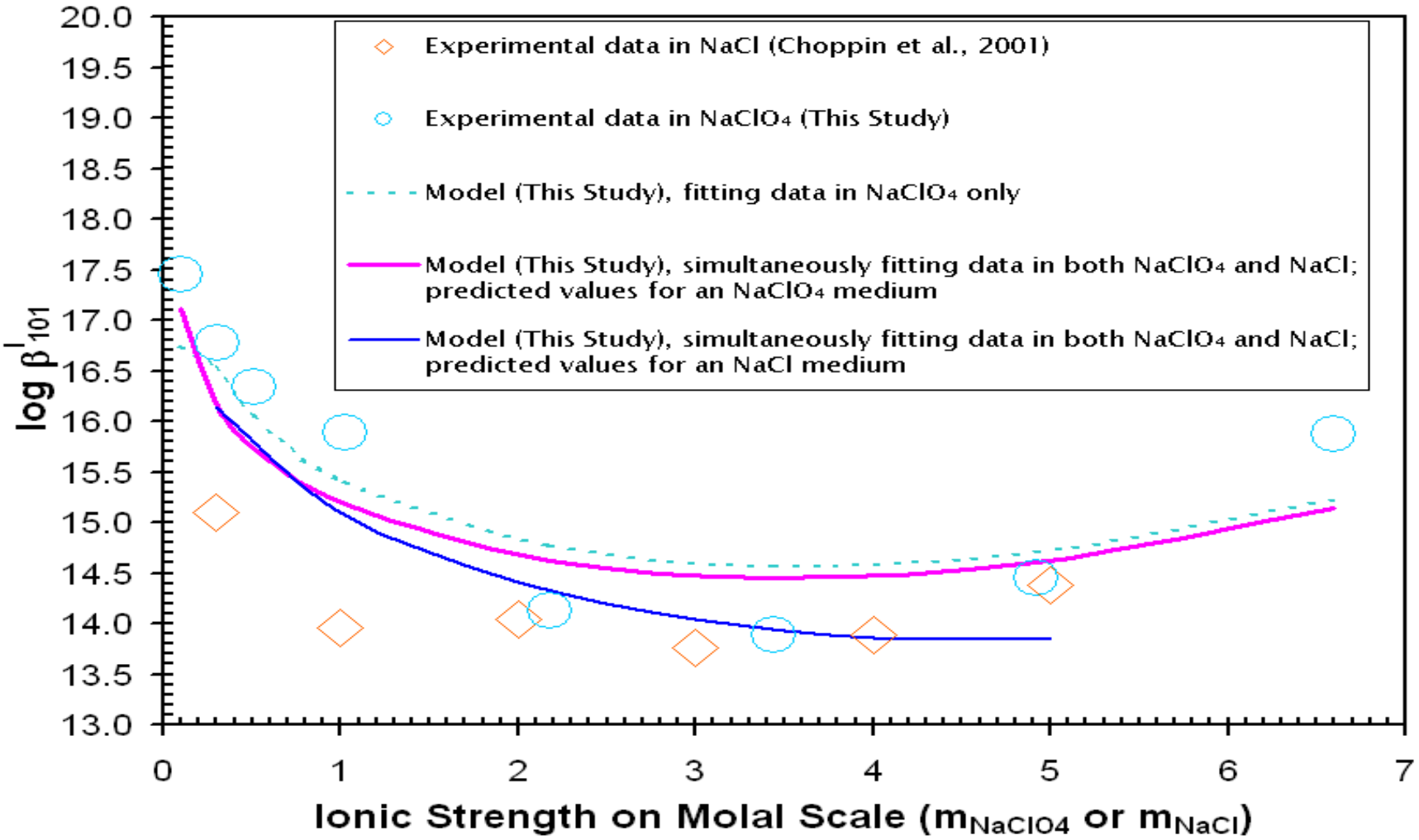
Experimental data used to parameterize a reaction must be in simple brines and must vary over ionic strength because we use the Pitzer model.

Parameterization of Reaction IV-14

$$AmEDTA^{-} = Am^{3+} + EDTA^{4-}$$

- The value in DATA0.FM1 ($\log \beta^0_1 = 18.97$) was derived from data in the ionic strength range from 0.3 m to 5.0 m in NaCl solutions.
- Since the 2009 Recertification of WIPP, new experimental data in the ionic strength range from 0.10 m to 6.60 m in $NaClO_4$ solutions have been produced at the Choppin Group at Florida State University.
- A revised value ($\log \beta^0_1 = 20.55$) derived using the new data is valid to higher ionic strengths (e.g., Thakur, Xiong, Borkowski, and Choppin, 2014).
- $\log \beta^0_1 = 20.55$ was incorporated in DATA0.FM2

Parameterization of Reaction IV-14



Parameterization of Reaction II-11

$$Mg_5(CO_3)_4(OH)_2 \bullet 4H_2O(hydromagnesite5424) + 6H^+ = 5Mg^{2+} + 4HCO_3^- + 6H_2O$$

- The value in DATA0.FM1 (log K^0 = 32.2529) was from Robie and Hemingway (1973)
- They calculated it from the Gibbs free energy of formation for hydromagnesite
- The Gibbs free energy of formation was calculated using calorimetric measurements of hydromagnesite.
- This does not represent a direct equilibrium measurement.

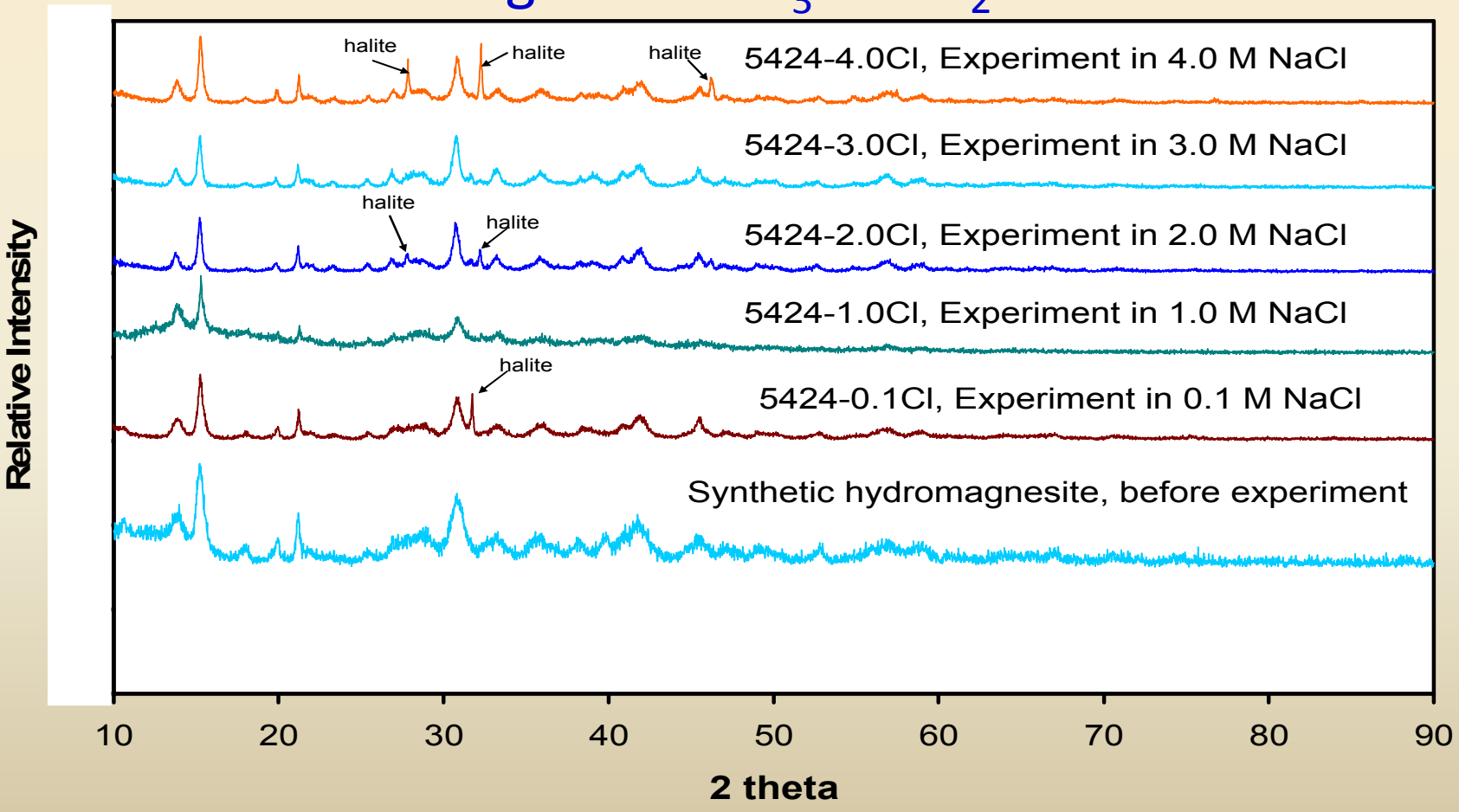
Parameterization of Reaction II-11

$$Mg_5(CO_3)_4(OH)_2 \bullet 4H_2O(hydromagnesite5424) + 6H^+ = 5Mg^{2+} + 4HCO_3^- + 6H_2O$$

- Xiong (2011) performed equilibrium solubility measurements on synthetic and natural hydromagnesite
- He derived a value of log K^0 = 29.3328 for synthetic hydromagnesite
- This value is in excellent agreement with the value provided by Konigsberger et al. (1992)
- Konigsberger et al. (1992) derived their value by evaluation of the solubility data of Riesen (1969) in 3.0 m $NaClO_4$.
- log K^0 = 29.3328 was incorporated in DATA0.FM2

Parameterization of Reaction II-11

$$Mg_5(CO_3)_4(OH)_2 \bullet 4H_2O(hydromagnesite5424) + 6H^+ = 5Mg^{2+} + 4HCO_3^- + 6H_2O$$



Parameterization of Reaction IV-21

$$AmHB_4O_7^{2+} + 9H_2O = Am^{3+} + 4B(OH)_4^- + 3H^+$$

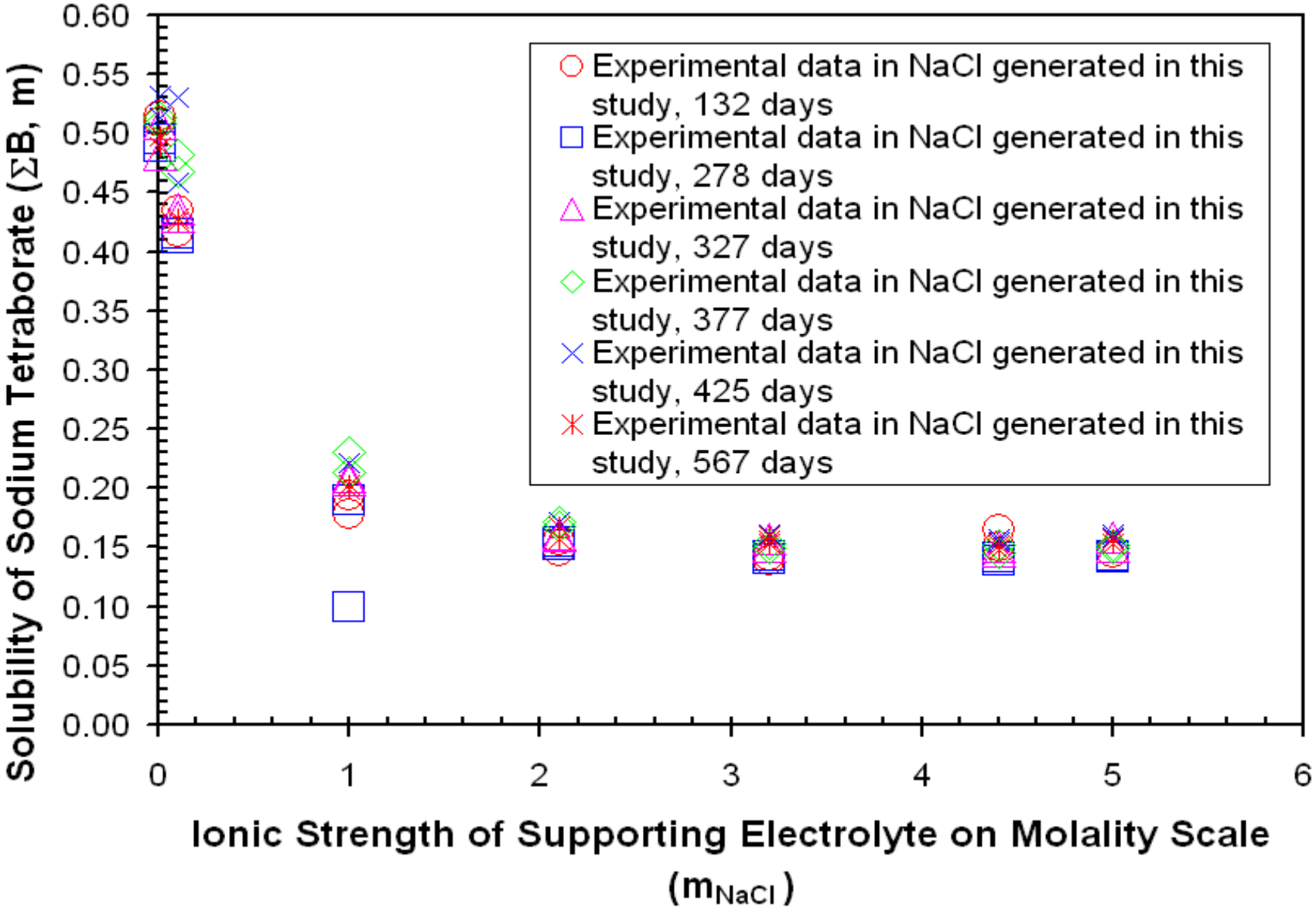
- This is a new reaction that has no parameters in DATA0.FM1
- Borkowski et al. (2010) produced solubility data for Nd(III)
- Xiong (2015) used that data to derive the equilibrium constant for above reaction and the Pitzer parameters associated with $AmHB_4O_7^{2+}$
- The Xiong (2015) values were incorporated in DATA0.FM2

Parameterization of Reaction VIII-6

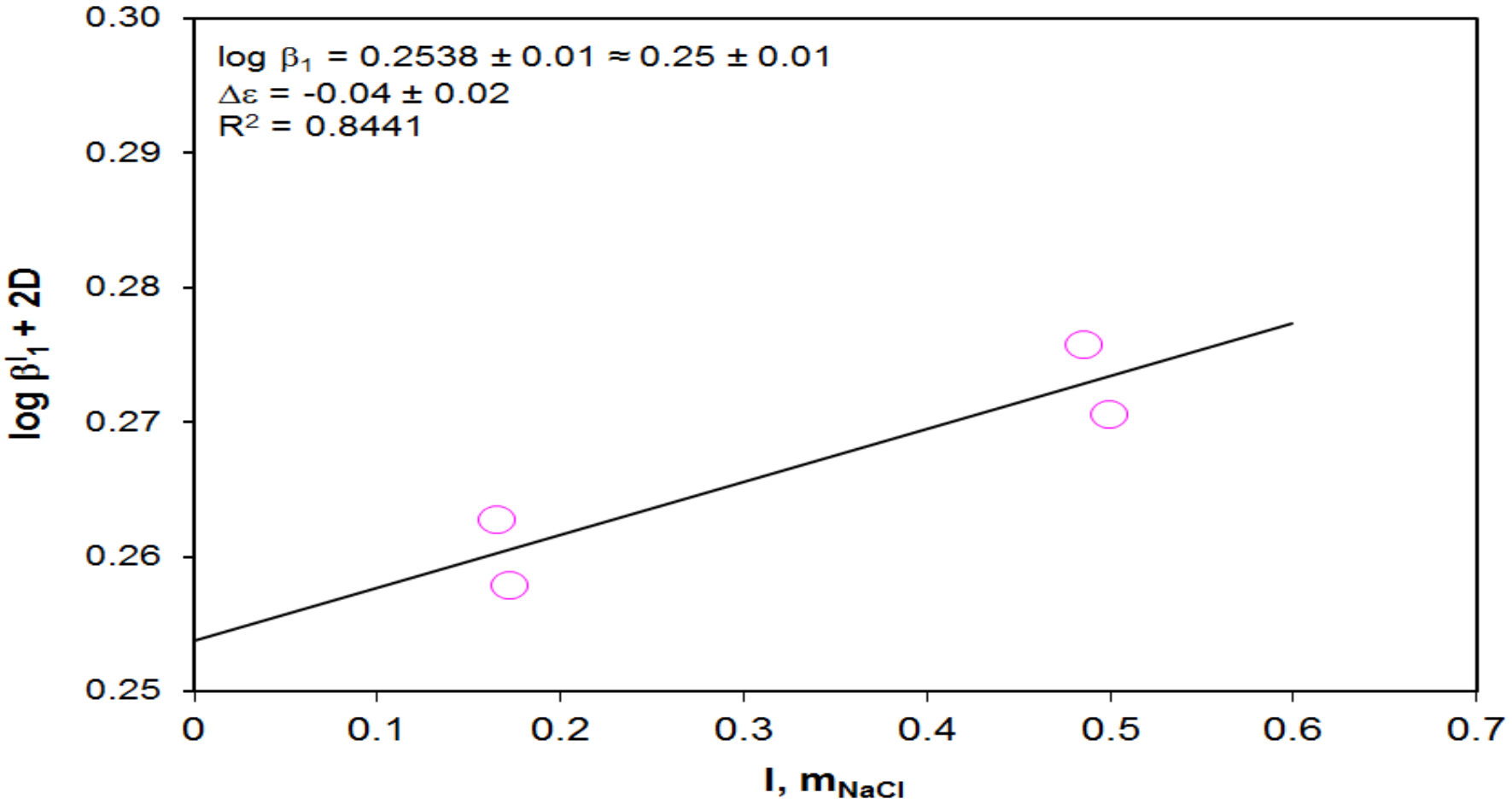
$$NaB(OH)_4(aq) = Na^+ + B(OH)_4^-$$

- This is a new reaction that has no parameters in DATA0.FM1
- Numerous researchers have suggested the existence of this complex in solutions containing sodium (e.g., Reardon, 1976; Corti et al., 1980; Rowe et al., 1989; Pokrovski et al., 1995; Akinfiev et al., 2006).
- Experimental solubility data for sodium tetraborate in NaCl solutions was produced by Xiong et al. (2012)
- Also available was literature solubility data for sodium tetraborate in Na_2SO_4 solutions

Parameterization of Reaction VIII-6



Parameterization of Reaction VIII-6



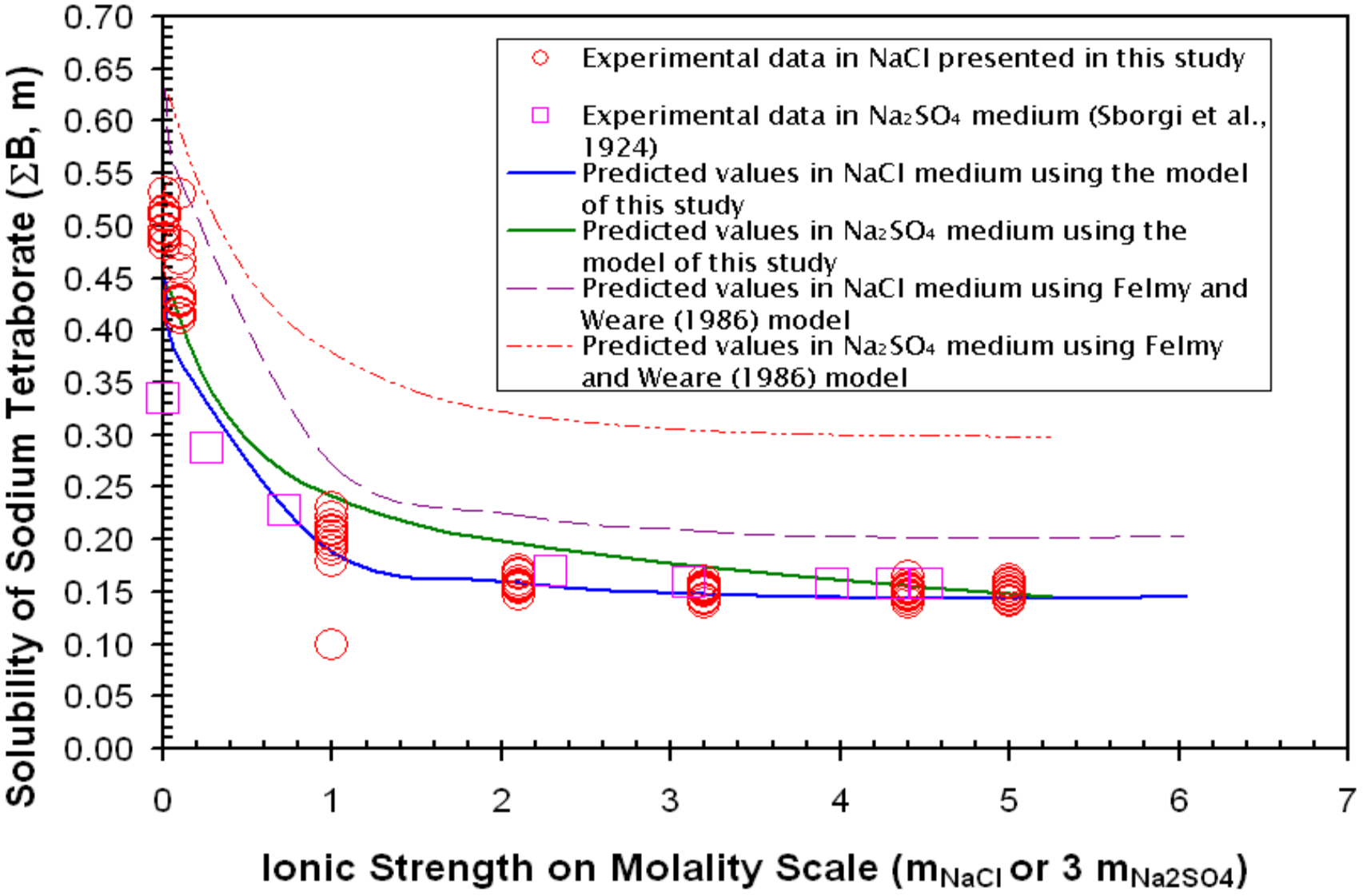
Parameterization of Reaction VIII-6

Table 2. The revised thermodynamic model for the Na–$B(OH)_3$–Cl–SO_4 system developed in this study*.

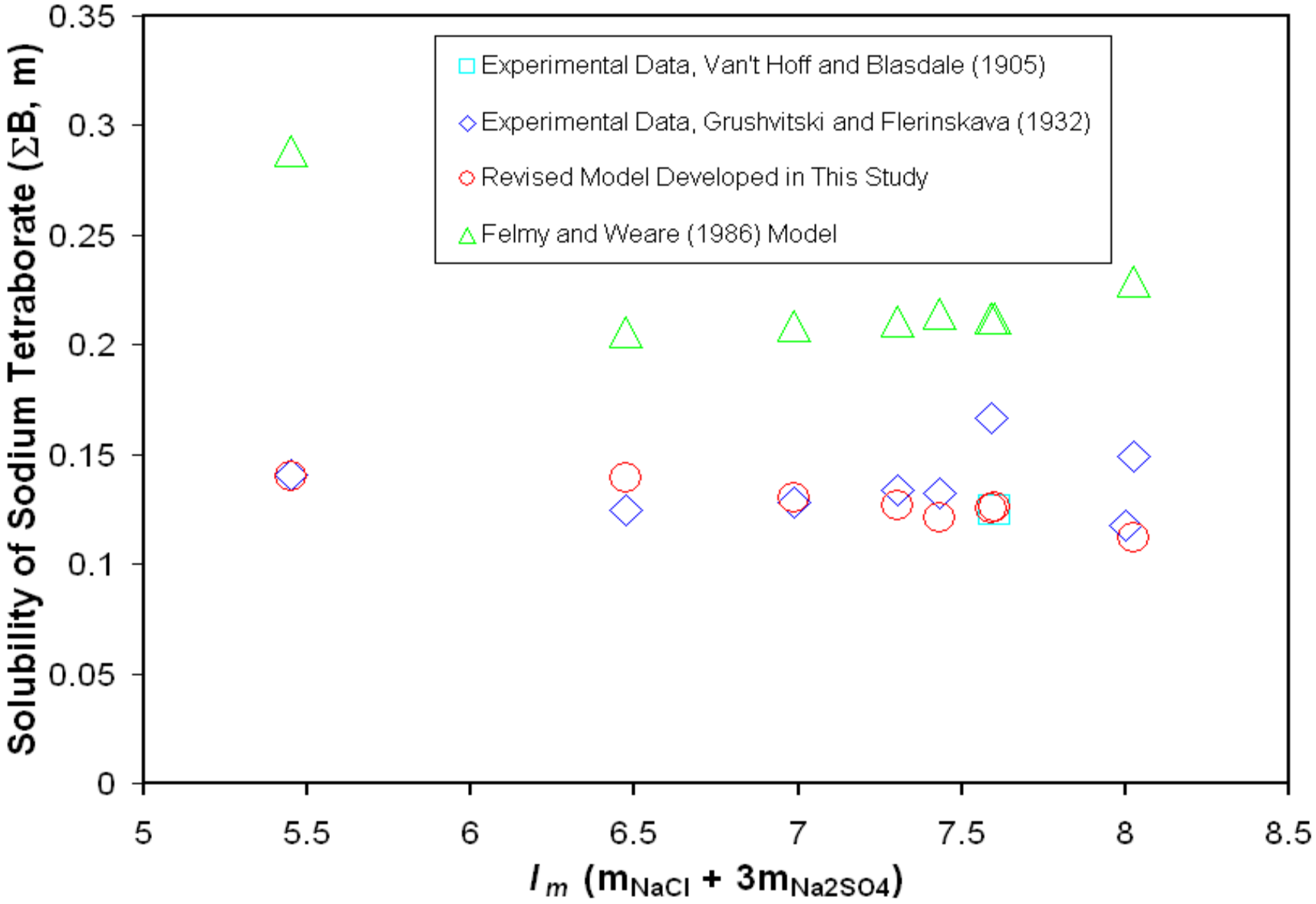
Pitzer Mixing Parameters and Interaction Parameters Involving Neutral Species				
Species, *i*	Species, *j*	Species, *k*	θ_{ij} or λ_{ij}	Ψ_{ijk} or ζ_{ijk}
$B(OH)_4^-$	SO_4^{-2}		0.1697	
$NaB(OH)_4(aq)$	Na^+		0.09192	
$B_4O_5(OH)_4^{-2}$	SO_4^{-2}	Na^+		0.096
Equilibrium Constants for Solubility and Complex Formation Reactions				
Reaction			log *K* or log β_1 at 25 °C unless otherwise noted	
$Na_2B_4O_7 \cdot 10H_2O = 2Na^+ + 4B(OH)_4^- + 2H^+ + H_2O$			-24.88 ± 0.10 (2σ)	
$Na^+ + B(OH)_4^- = NaB(OH)_4(aq)$			0.25 ± 0.01 (25 °C)	

*Unless otherwise noted, other parameters, which are not listed, are the same as those in Felmy and Weare (1986) model.

Parameterization of Reaction VIII-6



Parameterization of Reaction VIII-6



Parameterization of Reaction VIII-6

Table 4. Comparison of independent, experimental equilibrium compositions for multiple equilibrium assemblages containing sodium tetraborate ($Na_2B_4O_7 \cdot 10H_2O$) (borax) with predicted compositions in mixtures of NaCl and Na_2SO_4 at 25 °C

Experimental data for equilibrium compositions					
m_{Na}	m_{Cl}	m_{SO4}	$m_{\Sigma B}$	Equilibrium Assemblage*	References
6.967	5.516	0.694	0.125	BRX+HLT+THNDT	Van't Hoff and Blasdale (1905)
6.355	5.925	0.183	0.128	BRX+HLT	Grushvitski and Flerinskava (1932)
6.527	5.408	0.526	0.134	BRX+HLT	*ibid.*
6.716	5.286	0.673	0.167	BRX+HLT	*ibid.*
6.939	5.441	0.716	0.132	BRX+HLT+THNDT	*ibid.*
6.603	4.723	0.903	0.149	BRX	*ibid.*
6.477	3.168	1.619	0.141	BRX+MRBLT+THNDT	*ibid.*
3.774	0.248	1.734	0.117	BRX+MRBLT	*ibid.*

Parameterization of Reaction VIII-6

Equilibrium compositions predicted by the Felmy and Weare (1986) model				
m_{Na}	m_{Cl}	m_{SO4}	$m_{\Sigma B}$	Equilibrium Assemblage*
6.971	5.484	0.691	0.212	BRX+HLT+THNDT
6.364	5.896	0.182	0.206	BRX+HLT
6.532	5.381	0.523	0.209	BRX+HLT
6.704	5.260	0.670	0.211	BRX+HLT
6.943	5.413	0.712	0.212	BRX+HLT+THNDT
6.602	4.699	0.898	0.214	BRX
6.367	3.229	1.512	0.229	BRX+MRBLT+THNDT
3.833	0.246	1.721	0.289	BRX+MRBLT
Equilibrium compositions predicted by the model developed in this study				
m_{Na}	m_{Cl}	m_{SO4}	$m_{\Sigma B}$	Equilibrium Assemblage*
6.947	5.494	0.692	0.136	BRX+HLT+THNDT
6.347	5.906	0.183	0.151	BRX+HLT
6.512	5.393	0.524	0.141	BRX+HLT
6.683	5.272	0.671	0.137	BRX+HLT
6.922	5.426	0.714	0.136	BRX+HLT+THNDT
6.577	4.711	0.900	0.131	BRX
6.284	3.273	1.475	0.120	BRX+MRBLT+THNDT
3.780	0.247	1.728	0.151	BRX+MRBLT

Parameterization of Reaction VIII-6

Difference in %** between experimental values and those predicted by the FW86 model				
ΔNa in %	ΔCl in %	ΔSO_4 in %	ΔΣB in %	Equilibrium Assemblage*
0.060	-0.586	-0.507	69.788	BRX+HLT+THNDT
0.133	-0.483	-0.482	60.851	BRX+HLT
0.084	-0.496	-0.497	55.925	BRX+HLT
-0.170	-0.504	-0.503	26.362	BRX+HLT
0.072	-0.508	-0.507	60.305	BRX+HLT+THNDT
-0.016	-0.514	-0.515	43.776	BRX
-1.701	1.941	-6.656	62.343	BRX+MRBLT+THNDT
1.555	-0.725	-0.725	145.848	BRX+MRBLT
Difference in %** between experimental values and those predicted by the model developed in this study				
ΔNa in %	ΔCl in %	ΔSO_4 in %	ΔΣB in %	Equilibrium Assemblage*
-0.290	-0.402	-0.274	9.240	BRX+HLT+THNDT
-0.121	-0.309	-0.308	18.448	BRX+HLT
-0.227	-0.284	-0.285	5.249	BRX+HLT
-0.495	-0.275	-0.274	-18.006	BRX+HLT
-0.245	-0.273	-0.272	2.587	BRX+HLT+THNDT
-0.392	-0.259	-0.260	-11.967	BRX
-2.974	3.327	-8.884	-14.637	BRX+MRBLT+THNDT
0.145	-0.307	-0.307	28.758	BRX+MRBLT

*Abbreviations for minerals: BRX, borax ($Na_2B_4O_7 \cdot 10H_2O$); HLT, halite (NaCl); MRBLT, mirabilite ($Na_2SO_4 \cdot 10H_2O$); THNDT, thenardite (Na_2SO_4).

**Difference in % is defined as, using concentrations of sodium on molal scale as an example,

$$\Delta\text{Na in \%} = 100 \times \frac{m_{Na,Model} - m_{Na,Experimental}}{m_{Na,Experimental}}$$

Parameterization of the Lead Reactions

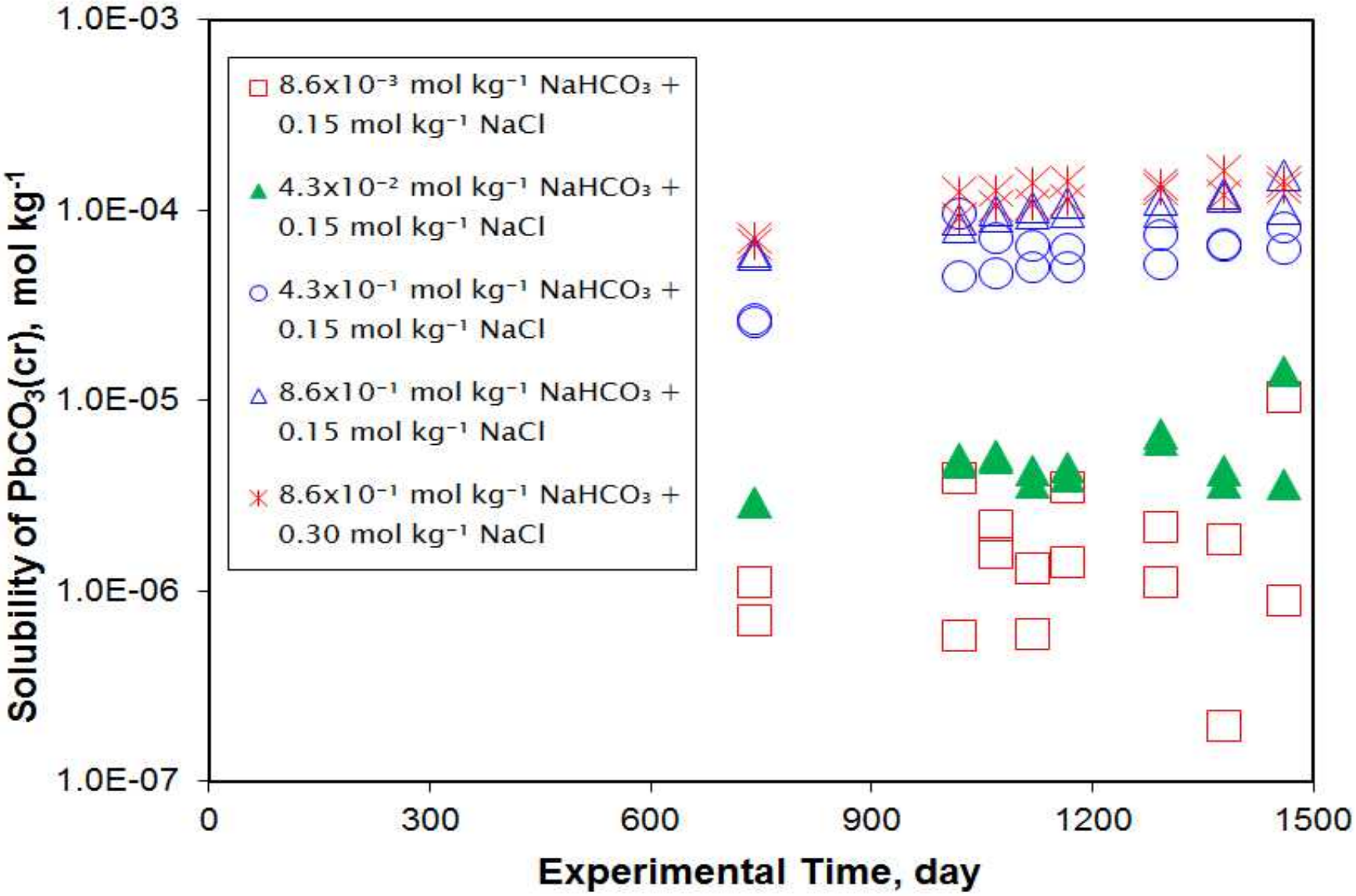
Appendix A, Table 8. Reactions Included in DATA0.FM2 for Pb Species

(VIII-1)	$Pb^{2+} + Cl^{-} = PbCl^{+}$
(VIII-2)	$Pb^{2+} + 2Cl^{-} = PbCl_2(aq)$
(VIII-3)	$Pb^{2+} + 3Cl^{-} = PbCl_3^{-}$
(VIII-4)	$Pb^{2+} + Oxalate^{2-} = PbOxalate(aq)$
(VIII-5)	$Pb^{2+} + 2Oxalate^{2-} = Pb(Oxalate)_2^{2-}$
(VIII-6)	$Pb^{2+} + CO_3^{2-} = PbCO_3(aq)$
(VIII-7)	$Pb^{2+} + 2CO_3^{2-} = Pb(CO_3)_2^{2-}$
(VIII-8)	$Pb^{2+} + CO_3^{2-} + Cl^{-} = Pb(CO_3)Cl^{-}$
(VIII-9)	$Pb^{2+} + H_2O(l) = PbOH^{+} + H^{+}$
(VIII-10)	$Pb^{2+} + 2H_2O(l) = Pb(OH)_2(aq) + 2H^{+}$
(VIII-11)	$Pb^{2+} + 3H_2O(l) = Pb(OH)_3^{-} + 3H^{+}$
(VIII-12)	$PbB_4O_7(aq) + 9H_2O(l) = Pb^{2+} + 4B(OH)_4^{-} + 2H^{+}$
(VIII-13)	$PbB(OH)_4^{+} = Pb^{2+} + B(OH)_4^{-}$
(VIII-14)	$Pb[B(OH)_4]_2(aq) = Pb^{2+} + 2B(OH)_4^{-}$
(VIII-15)	$Pb^{2+} + EDTA^{4-} = PbEDTA^{2-}$
(VIII-16)	$Pb^{2+} + Citrate^{3-} = PbCitrate^{-}$
(VIII-17)	$PbOxalate(cr) = Pb^{2+} + Oxalate^{2-}$
(VIII-18)	$PbCO_3(cerussite) = Pb^{2+} + CO_3^{2-}$
(VIII-19)	$PbO(litharge) + 2H^{+} = Pb^{2+} + H_2O$
(VIII-20)	$Pb(BO_2)_2 \bullet H_2O + 3H_2O(l) = Pb^{2+} + 2B(OH)_4^{-}$

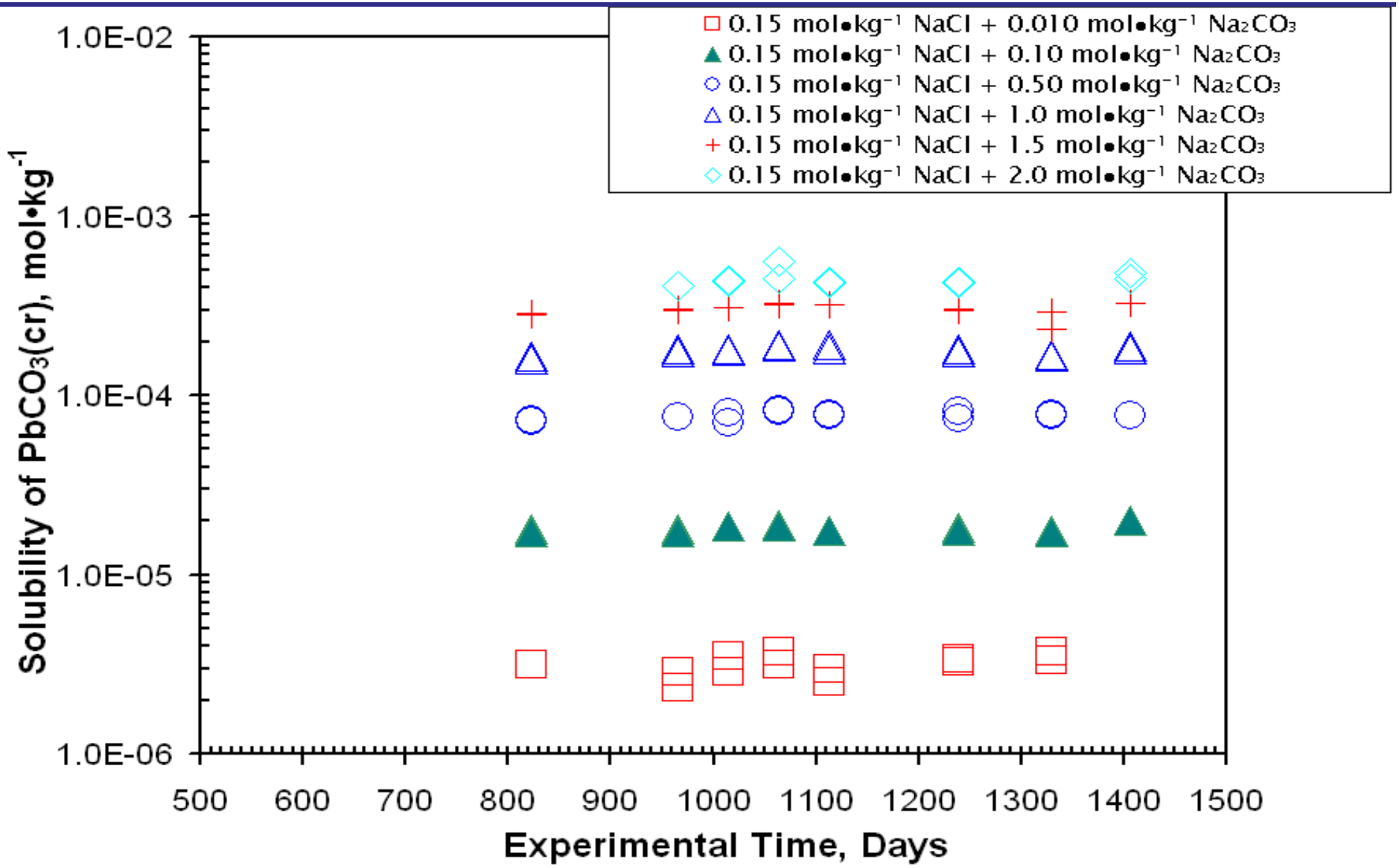
Parameterization of the Lead Carbonate Reactions

- This is a new reaction set that has no parameters in DATA0.FM1
- Xiong has performed lead solubility experiments at the SNL Carlsbad facility(Xiong, 2015a and 2015b)
- Xiong produced experimental solubility data for lead carbonate, $PbCO_3$(cr), and cerussite, in mixtures of $NaCl+NaHCO_3$ and $NaHCO_3+Na_2CO_3$ at the Carlsbad Facility of SNL

Parameterization of the Lead Reactions



Parameterization of the Lead Reactions



Parameterization of the Lead Reactions

Table 1. Equilibrium constants at infinite dilution at 25°C and 1 bar for the Na^+—Pb^{2+}—Cl^-—HCO_3^-—CO_3^{2-} system

Reactions	log K_s^o, log β_1^o, log β_2^o or log β_{11}^o	Reference and Remarks
$PbCO_3$(cerussite, cr) = Pb^{2+} + CO_3^{2-}	–13.65 ± 0.15 (2σ) [A]	This study, based on solubility of $PbCO_3$(cr) in the mixtures of $NaHCO_3$ and NaCl and in the mixtures of $NaHCO_3$ and Na_2CO_3
Pb^{2+} + CO_3^{2-} = $PbCO_3$(aq)	6.87 ± 0.09 (2σ)	Woosley and Millero (2013)
Pb^{2+} + $2CO_3^{2-}$ = $Pb(CO_3)_2^{2-}$	10.41 ± 0.18 (2σ)	Easley and Byrne (2011)
Pb^{2+} + CO_3^{2-} + Cl^- = $Pb(CO_3)Cl^-$	7.23 ± 0.74 (2σ)	Woosley and Millero (2013)

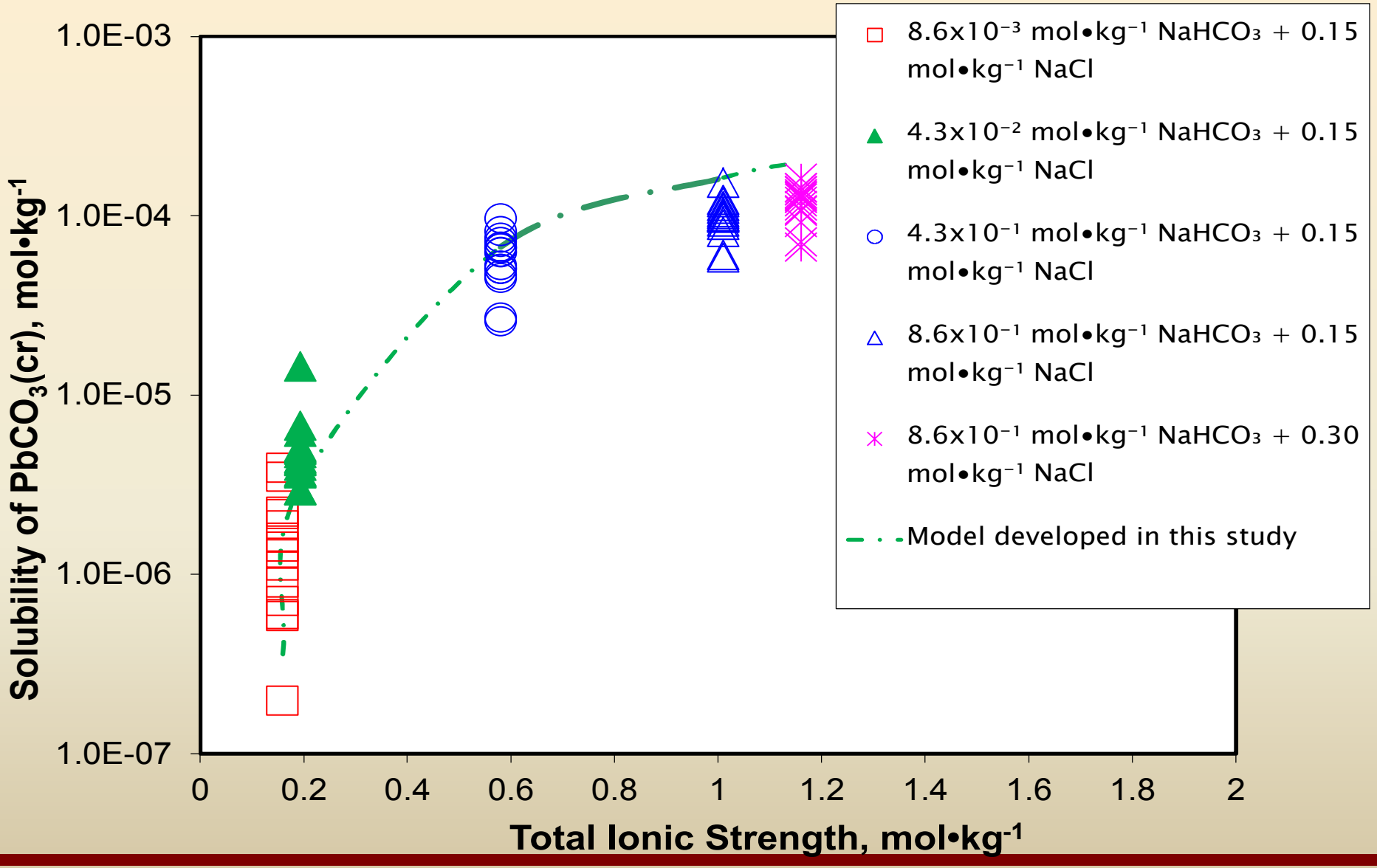
[A] In the modeling, the log K modeled is for the reaction $PbCO_3$(cr) + H^+ = Pb^{2+} + HCO_3^-, which can be converted to log K_s^o listed in Table 4 with the log K_2 in the database for the reaction HCO_3^- = H^+ + CO_3^{2-}, which is –10.3392.

Parameterization of the Lead Reactions

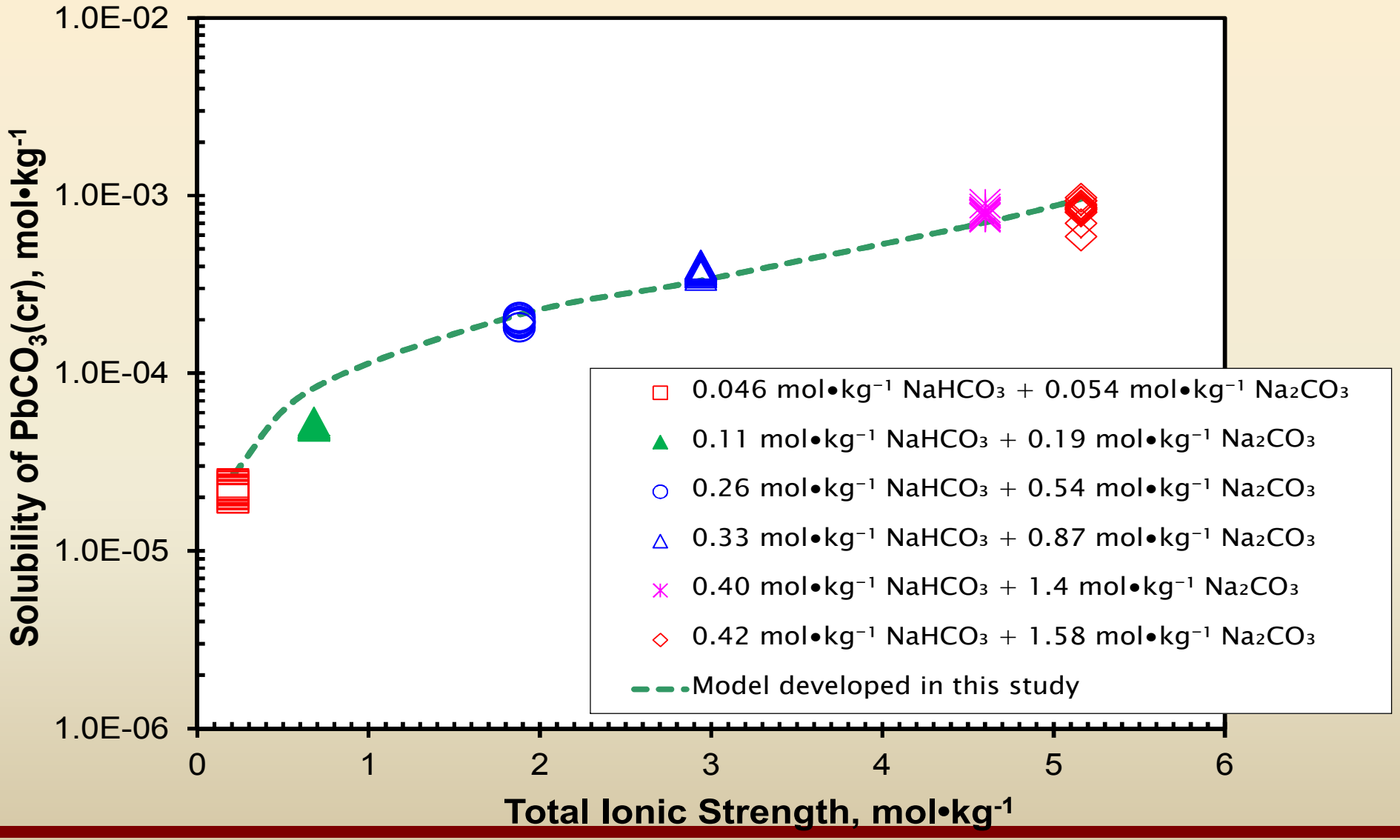
Table 2. Pitzer interaction parameters at 25°C and 1 bar for the Na^+—Pb^{2+}—Cl^-—HCO_3^-—CO_3^{2-} system

Pitzer Binary Interaction Parameters					
Species *i*	Species *j*	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}	Reference
Na^+	$Pb(CO_3)_2^{2-}$	0.1975	1.74	–0.2105	This study
Na^+	$Pb(CO_3)Cl^-$	0.3799	0.29	0.1921	This study
Pitzer Mixing Interaction Parameters (theta parameter) and Interaction Parameters Involving Neutral Species (lambda and zeta parameters)					
Species *i*	Species *j*	Species *k*	λ_{ij} or θ_{ij}	ζ_{ijk}	Reference
HCO_3^-	$Pb(CO_3)_2^{2-}$		0.1476		This study
CO_3^{2-}	$Pb(CO_3)_2^{2-}$		0.2223		This study
Cl^-	$PbCO_3(aq)$		–0.02		Woosley and Millero (2013)
Na^+	$PbCO_3(aq)$	Cl^-	0	–0.145	Woosley and Millero (2013)

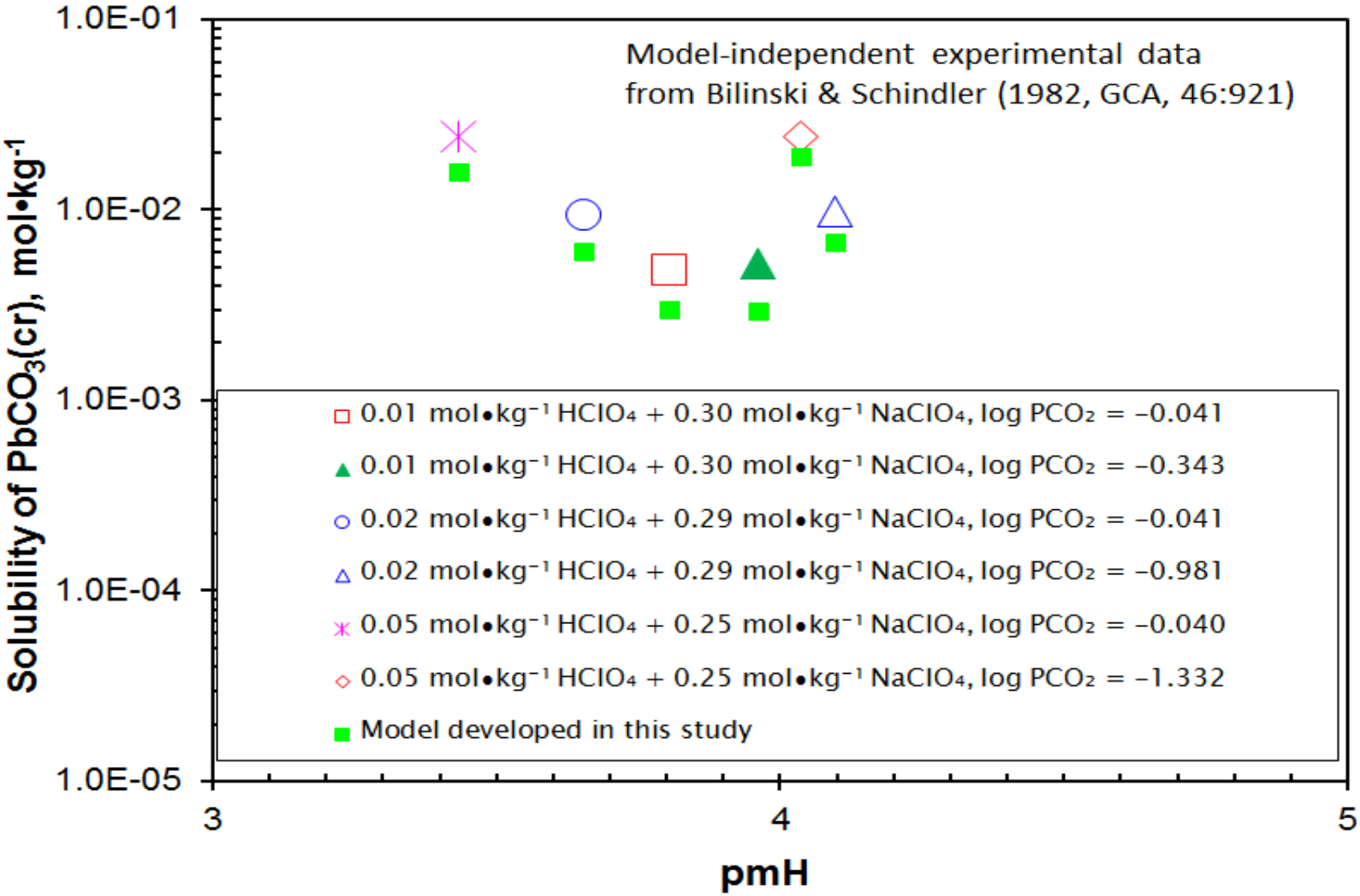
Parameterization of the Lead Reactions



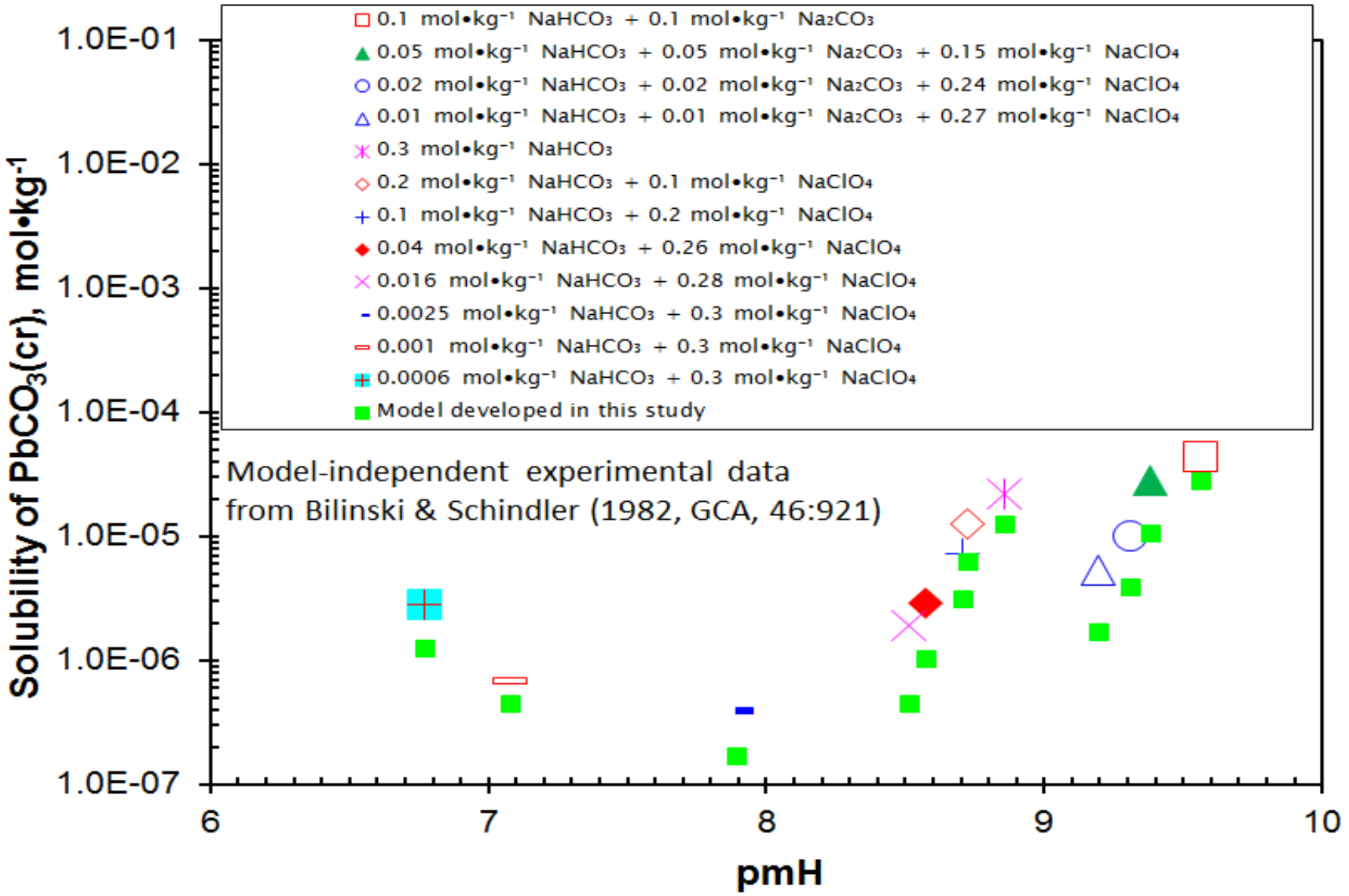
Parameterization of the Lead Reactions



Parameterization of the Lead Reactions



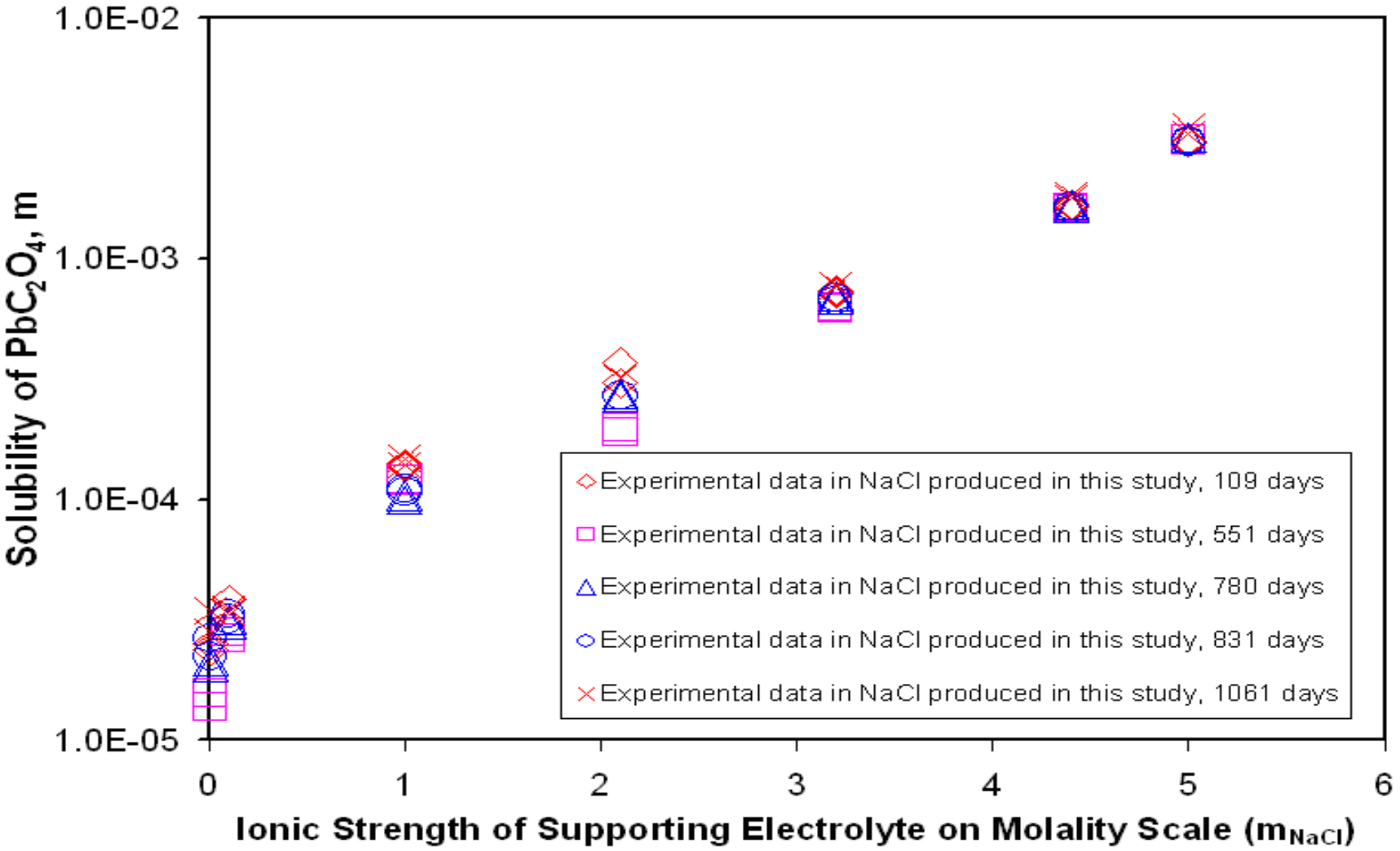
Parameterization of the Lead Reactions



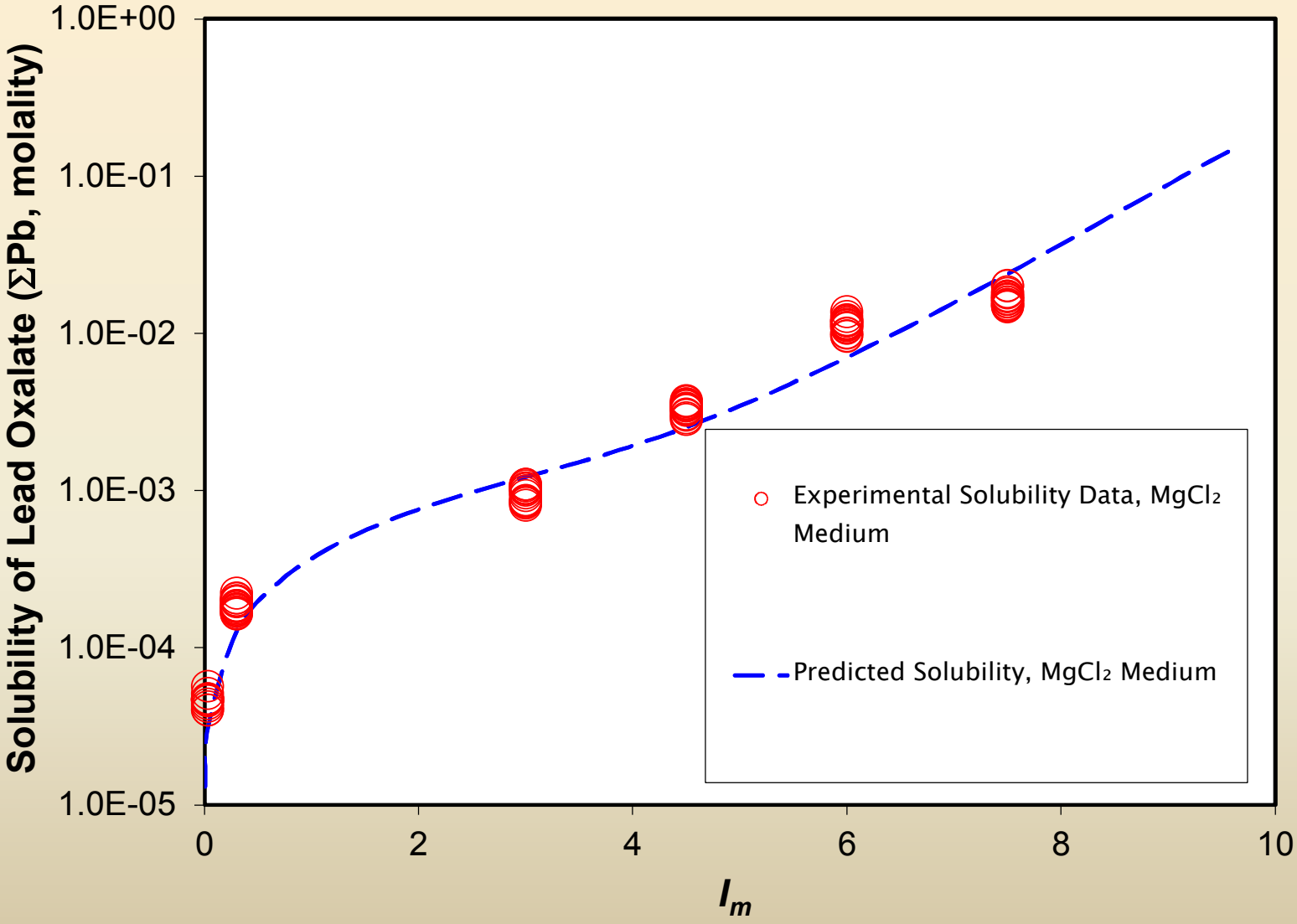
Parameterization of the Lead Oxalate Reactions

- This is a new reaction set that has no parameters in DATA0.FM1
- Xiong produced experimental solubility data for lead oxalate, $PbC_2O_4(cr)$, in NaCl solutions (Xiong et al., 2011)
- Literature solubility data for lead oxalate in $K_2C_2O_4$ solutions was available.

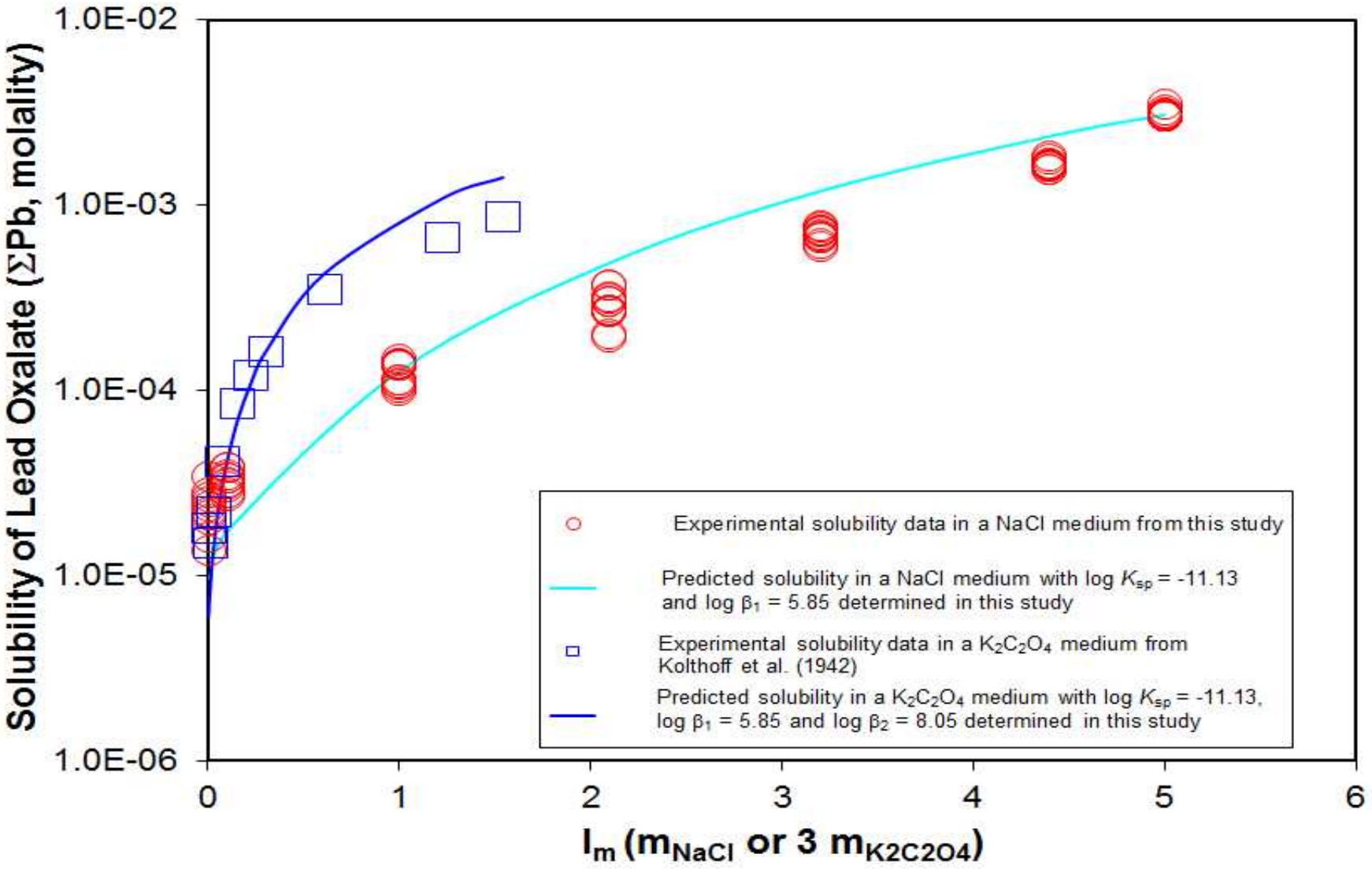
Parameterization of the Lead Reactions



Parameterization of the Lead Reactions



Parameterization of the Lead Reactions



Parameterization of the Lead Reactions

Table 4. Equilibrium constants at infinite dilution, 25°C and 1 bar, Pitzer interaction parameters, in Na^+—K^+—Pb^{2+}—Cl^-—$C_2O_4^{2-}$ system

Reactions	log K_{sp} or log β_n	Reference and Remarks
$PbC_2O_4(cr) = Pb^{2+} + C_2O_4^{2-}$	-11.13 ± 0.15 (2σ)	This study, based on solubility of $PbC_2O_4(cr)$ in NaCl medium presented in the present work, Pitzer model
$Pb^{2+} + C_2O_4^{2-} = PbC_2O_4(aq)$	5.85 ± 0.10 (2σ)	This study, based on solubility of $PbC_2O_4(cr)$ in NaCl medium presented in the present work, Pitzer model
$Pb^{2+} + 2C_2O_4^{2-} = Pb(C_2O_4)_2^{2-}$	8.05 ± 0.15 (2σ)	This study, evaluated from solubilities of $PbC_2O_4(cr)$ in $K_2C_2O_4$ medium from Kolthoff et al. (1942), Pitzer model

Parameterization of the Lead Reactions

Table 4. Equilibrium constants at infinite dilution, 25°C and 1 bar, Pitzer interaction parameters, in Na^+—K^+—Pb^{2+}—Cl^-—$C_2O_4^{2-}$ system (cont.)

$Pb^{2+} + Cl^- = PbCl^+$	1.48	Millero and Byrne (1984), evaluated by using Pitzer model; uncertainty not given
$Pb^{2+} + 2Cl^- = PbCl_2(aq)$	2.03	Millero and Byrne (1984), evaluated by using Pitzer model; uncertainty not given
$Pb^{2+} + 3Cl^- = PbCl_3^-$	1.86	Millero and Byrne (1984), evaluated by using Pitzer model; uncertainty not given

Parameterization of the Lead Reactions

Sandia National Laboratories

Pitzer Binary Interaction Parameters					
Species *i*	Species *j*	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}	
Pb^{2+}	Cl^-	0.26	1.64	0.088	Millero and Byrne (1984)
$PbCl^+$	Cl^-	0.15	0	0	Millero and Byrne (1984)
Na^+	$PbCl_3^-$	–0.0605	0	0.091	This study
K^+	$Pb(C_2O_4)_2^{2-}$	0	–1.86	0.198	This study
Na^+	$Pb(C_2O_4)_2^{2-}$	0	–1.86	0.198	This study, in analog to K^+—$Pb(C_2O_4)_2^{2-}$
Pitzer Mixing Interaction Parameters and Interaction Parameters Involving Neutral Species					
Species *i*	Species *j*	Species *k*	λ_{ij} or θ_{ij}	ζ_{ijk}	
Cl^-	$PbCl_2(aq)$		–0.14		This study
Na^+	$PbCl_2(aq)$		–0.11		Felmy et al. (2000)
Na^+	Pb^{2+}		0.10		Felmy et al. (2000)
Na^+	$PbCl_2(aq)$	Cl^-		0	This study
Na^+	$PbC_2O_4(aq)$	Cl^-	0	0	This study

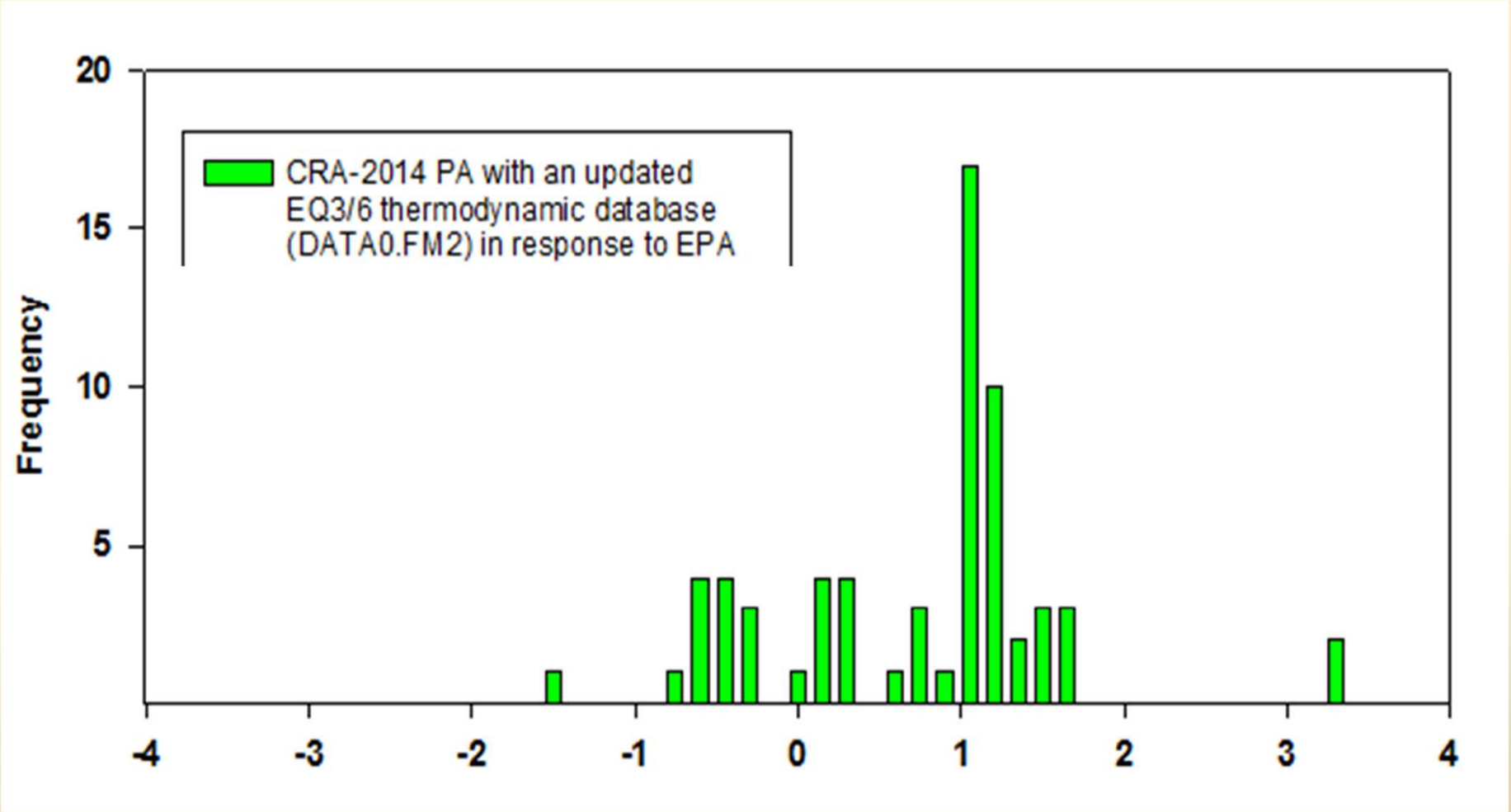
Base Solubilities Using DATA0.FM1 and DATA0.FM2

	GWB (FM1)	GWB (FM2)	ERDA-6 (FM1)	ERDA-6 (FM2)	
Th(IV)	6.05E-08	4.78E-08	7.02E-08	5.46E-08	M
Np(V)	2.77E-7	9.98E-07	8.76E-7	1.45E-06	M
Am(III)	2.59E-6	2.32E-07	1.48E-6	6.03E-08	M
f_{CO2}	3.14E-6	5.84E-07	3.14E-6	5.84E-07	atm
pH	8.82	8.84	8.99	9.22	Pitzer scale

Base Solubilities Using DATA0.FM1 and DATA0.FM2

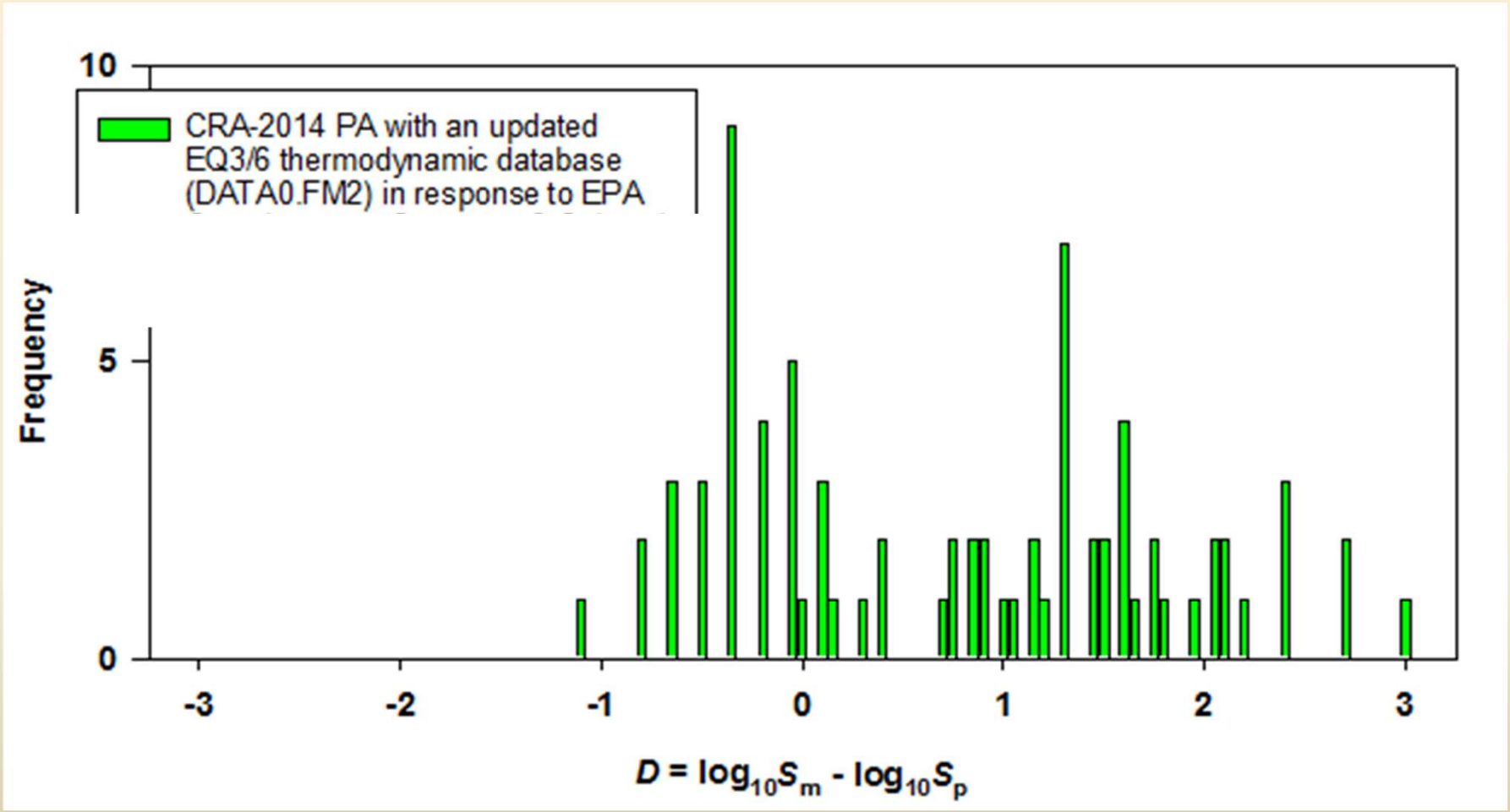
- Th(IV) solubility goes down
- Np(V) solubility goes up
- Am(III) solubility goes down
- f_{CO_2} goes down
- pH goes up

An (IV) Uncertainty Distribution Using DATA0.FM2



$$D = \log_{10} S_m - \log_{10} S_p$$

An (III) Uncertainty Distribution Using DATA0.FM2



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