

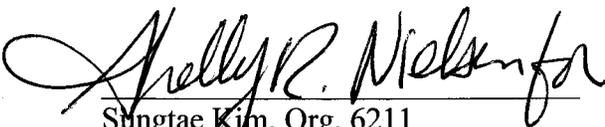
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Sandia National Laboratories
Waste Isolation Pilot Plant

Experimental and Thermodynamic Modeling of PbCit^- Interactions in NaCl and MgCl_2 Solutions

Work Carried Out under Tasks 20 and 21 of AP-154: Analysis Plan for Derivation of Thermodynamic Properties Including Pitzer Parameters for Solubility Studies of Iron, Lead and EDTA.

To be included in the AP-154 records package

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Information Only

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1 INTRODUCTION

The Waste Isolation Pilot Plant (WIPP) is a U.S. Department of Energy (DOE) repository in southeast New Mexico for defense-related transuranic (TRU) waste. The repository, which opened in March 1999, is located at a subsurface depth of 655 m in the Salado Fm., a Permian bedded-salt formation. It is planned to use a significant amount of lead as a radiation-shielding material for wastes with significant γ -radiation placed in the WIPP.

Citrate, $(\text{CH}_2\text{COO})_2\text{C}(\text{OH})(\text{COO})^{3-}$ or $\text{C}_6\text{H}_5\text{O}_7^{3-}$, abbreviated as Cit^{3-} , is one organic ligand present in the waste to be disposed in the WIPP that has an impact to the Performance Assessment (PA). When it is dissolved in WIPP brines such as Generic Weep Brine (GWB) from the Salado Fm. at the stratigraphic horizon of the repository, and Energy Research, or Development Administration (WIPP Well) 6 (ERDA-6) from the underlying Castile Fm., it can potentially form strong complexes with actinides, increasing their mobile fraction of actinides. On the other hand, lead also forms a strong complex with Cit^{3-} . Consequently, lead would compete with actinides for soluble Cit^{3-} to form PbCit^- , which would decrease the mobile fraction of actinides. Therefore, knowledge of the interactions of PbCit^{3-} with major ions in the WIPP brines over a range of ionic strengths important for the WIPP fortifies the near-field geochemical model for the WIPP.

This analysis report (AR) provides the derivation of thermodynamic properties and Pitzer parameters for the Pb-Cit^{3-} system. Derivations use solubility data of litharge (PbO) in NaCl and MgCl_2 solutions in the presence of Cit^{3-} reported in Kirkes, Olivas, Jang, Kim, and Xiong (2014)

The computer code EQ3/6 Version 8.0a (Wolery, 2008; Wolery et al., 2010; Xiong, 2011b) was used for this analysis. Wolery (2008), Wolery et al. (2010) and Xiong (2011b) completed the qualification of Version 8.0a of EQ3/6 according to Sandia National Laboratories' (SNL's) WIPP quality assurance (QA) procedures for WIPP compliance-related actinide solubility calculations, and US EPA approved it on September 27, 2011 (US EPA, 2011).

This analysis was carried out under Tasks 20 and 21 of AP-154, Revision 2 (Xiong, 2013b).

Table 1 (see next page) defines the generic abbreviations, acronyms, and initialisms used in this report and other analysis reports.

Table 1. Abbreviations, acronyms, and initialisms.

Abbreviation, Acronym, or Initialism	Definition
a	activity
acetate	CH_3COO^- or CH_3CO_2^-
Am, Am(III)	americium, americium in the +III oxidation state
am	amorphous
anhydrite	CaSO_4
AP	analysis plan
aq	aqueous
aragonite	CaCO_3 , a polymorph of CaCO_3 that is metastable with respect to calcite
atm	atmosphere(s)
B, B(III)	boron, boron in the +III oxidation state
β_{mn}	cumulative formation constant at infinite dilution with “m” number of the complexing ligand <i>A</i> and “n” number of the complexing ligand <i>B</i> in case that there are two complexing ligands
β_n	cumulative formation constant at infinite dilution with “n” number of the complexing ligand <i>A</i> in case that there is only one complexing ligand
Br, Br(-I)	bromine, bromine in the -I oxidation state
brucite	$\text{Mg}(\text{OH})_2$
C	carbon
Ca, Ca(II), Ca^{2+}	calcium, calcium in the +II oxidation state, calcium ion
calcite	CaCO_3 , the thermodynamically stable polymorph of CaCO_3
cerussite	$\text{PbCO}_3(\text{cr})$
citrate	$(\text{CH}_2\text{COO})_2\text{C}(\text{OH})(\text{COO})^{3-}$ or $(\text{CH}_2\text{CO}_2)_2\text{C}(\text{OH})(\text{CO}_2)^{3-}$
Cl, Cl(-I), Cl^-	chlorine, chlorine in the -I oxidation state, chloride ion
CMS	(Sandia/WIPP software) Configuration Management System
CO_2	carbon dioxide
CO_3^{2-}	carbonate
CRA-2009	the second WIPP Compliance Recertification Application, submitted to the EPA in March 2009
DB	(thermodynamic) database
DOE	(U.S.) Department of Energy

Table 1 continued on next page

Table 1. Abbreviations, acronyms, and initialisms (continued).

Abbreviation, Acronym, or Initialism	Definition
dolomite	$\text{CaMg}(\text{CO}_3)_2$, a carbonate mineral that is nucleates and grows slowly under low-temperature conditions and is often suppressed (prevented from forming) in geochemical modeling calculations
EDTA	ethylenediaminetetraacetate, $(\text{CH}_2\text{COO})_2\text{N}(\text{CH}_2)_2\text{N}(\text{CH}_2\text{COO})_2^{4-}$ or $(\text{CH}_2\text{CO}_2)_2\text{N}(\text{CH}_2)_2\text{N}(\text{CH}_2\text{CO}_2)^{4-}$
EPA	(U.S.) Environmental Protection Agency
EQ3/6	a geochemical software package for speciation and solubility calculations (EQ3NR) and reaction-path calculations (EQ6)
GWB	Generic Weep Brine, a synthetic brine representative of intergranular Salado brines at or near the stratigraphic horizon of the repository
gypsum	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$
H or H_2 , H^+	hydrogen or hydrogen ion
halite	NaCl
H_2O	water (aq, g, or contained in solid phases)
hydromagnesite	$\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$
I	ionic strength
K, K(I)	potassium, potassium in the +I oxidation state
kg	kilogram(s)
K_s^o	solubility constant at infinite dilution
Litharge	$\text{PbO}(\text{cr})$
M	molar
m	meter(s) or molal
magnesite	MgCO_3
Mg, Mg(II)	magnesium, magnesium in the +II oxidation state
MgO	magnesium oxide, used to refer to the WIPP engineered barrier, which includes periclase as the primary constituent and various impurities
mM	millimolar
Na, Na(I), Na^+	sodium, sodium in the +I oxidation state, sodium ion
nesquehonite	$\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$
Np, Np(V)	neptunium, neptunium in the +V oxidation state
O or O_2	oxygen
OH, OH^-	hydroxide or hydroxide ion
oxalate	$(\text{COO})_2^{2-}$ or $\text{C}_2\text{O}_4^{2-}$

Table 1 continued on next page

Table 1. Abbreviations, acronyms, and initialisms (continued).

Abbreviation, Acronym, or Initialism	Definition
PA	performance assessment
PABC	Performance Assessment Baseline Calculations
Pb, Pb(II), Pb ²⁺	lead, lead in the +II oxidation state, lead ion
periclase	pure, crystalline MgO, the primary constituent of the WIPP engineered barrier
pH	the negative, common logarithm of the activity of H ⁺
pCH	the negative, common logarithm of the molar concentration of H ⁺
phase 3	Mg ₂ Cl(OH) ₃ ·4H ₂ O
phase 5	Mg ₃ (OH) ₅ Cl·4H ₂ O
polyhalite	K ₂ MgCa ₂ (SO ₄) ₄ ·2H ₂ O
QA	quality assurance
Rev.	revision
RH	relative humidity
S, S(VI), SO ₄ ²⁻	sulfur, sulfur in the +VI oxidation state, sulfate ion
s	solid
SCA	S. Cohen and Associates
SNL	Sandia National Laboratories
Th, Th(IV)	thorium, thorium in the +IV oxidation state
TIC	total inorganic C
WIPP	(U.S. DOE) Waste Isolation Pilot Plant
wt %	weight percent
μ ⁰ /RT	dimensionless standard chemical potential

2 METHODS

The objective of this analysis is to derive thermodynamic properties for the $\text{Na}^+ - \text{Mg}^{2+} - \text{Pb}^{2+} - \text{Cl}^- - \text{Cit}^{3-}$ system based on solubility data for litharge ($\text{PbO}(\text{cr})$) in NaCl and MgCl_2 solutions produced at SNL (Kirkes, Olivas, Jang, Kim, and Xiong, 2014). Tables 2 and 3 list experimental data in NaCl , and in MgCl_2 solutions from the above report, respectively.

The dissolution reaction for $\text{PbO}(\text{cr})$ can be expressed as,



The corresponding solubility product constant of $\text{PbO}(\text{cr})$ at infinite dilution can be formulated as follows,

$$K_{s2}^o = \frac{a_{\text{Pb}^{2+}} \times a_{\text{H}_2\text{O}}}{a_{\text{PbO}(\text{cr})} \times a_{\text{H}^+}^2} \quad (2)$$

In this analysis, the aqueous lead species included are Pb^{2+} , PbCl^+ , $\text{PbCl}_2(\text{aq})$, PbCl_3^- , and PbEDTA^{2-} . In the Analysis Report for lead oxalate, Pb^{2+} , PbCl^+ , $\text{PbCl}_2(\text{aq})$, PbCl_3^- have been incorporated and the sources for them have been cited (Xiong, 2013a). Therefore, their citations will not be repeated in this Analysis Report.

The formation reaction for PbCit^- is written as,



The corresponding formation constant at infinite dilution is,

$$\beta_1^o = \frac{m_{\text{PbCit}^-}}{m_{\text{Pb}^{2+}} \times m_{\text{Cit}^{3-}}} \times \frac{\gamma_{\text{PbCit}^-}}{\gamma_{\text{Pb}^{2+}} \times \gamma_{\text{Cit}^{3-}}} = \frac{a_{\text{PbCit}^-}}{a_{\text{Pb}^{2+}} \times a_{\text{Cit}^{3-}}} \quad (4)$$

Similarly, the second complex, $\text{Pb}(\text{Cit})_2^{4-}$ could be formed. The formation reaction for $\text{Pb}(\text{Cit})_2^{4-}$ is written as,



The corresponding formation constant at infinite dilution is,

$$\beta_2^o = \frac{m_{\text{Pb}(\text{Cit})_2^{4-}}}{m_{\text{Pb}^{2+}} \times m_{\text{Cit}^{3-}}^2} \times \frac{\gamma_{\text{Pb}(\text{Cit})_2^{4-}}}{\gamma_{\text{Pb}^{2+}} \times \gamma_{\text{Cit}^{3-}}^2} = \frac{a_{\text{Pb}(\text{Cit})_2^{4-}}}{a_{\text{Pb}^{2+}} \times a_{\text{Cit}^{3-}}^2} \quad (6)$$

In this analysis report, the $\log K_{s2}^o$ for Reaction (1) is taken from ymp.R2/ymp.R0 (Table 4). In the literature, it has been demonstrated that the strength of $\text{Ni}(\text{Cit})_2^{4-}$ is much weaker than that of NiCit^- (Still and Wikberg, 1980). Therefore, $\text{Pb}(\text{Cit})_2^{4-}$ is not considered in this AR based on the analog to $\text{Ni}(\text{Cit})_2^{4-}$.

In this work, the $\log \beta_1^0$ at infinite dilution was evaluated by using the SIT model (Figure 1) based on the conditional stability constants at certain ionic strengths from the literature (Kety, 1942; Katano et al., 1999; Martell and Smith, 2004). For the detailed descriptions about using the SIT model for extrapolation to infinite dilution, please see Xiong (2012). The $\log \beta_1^0$ is given by,

$$\log \beta_1^I + 12D = \log \beta_1^0 - \Delta\varepsilon \times I_m \quad (7)$$

In Eq. (7), the Debye-Huckel term, D , is defined as,

$$D = \frac{A_\gamma \sqrt{I_m}}{1 + 1.5\sqrt{I_m}} \quad (8)$$

where A_γ is the Debye-Hückel slope for activity coefficient, which is 0.5092 at 25°C from Helgeson and Kirkham (1974), and I_m ionic strength on molal scale.

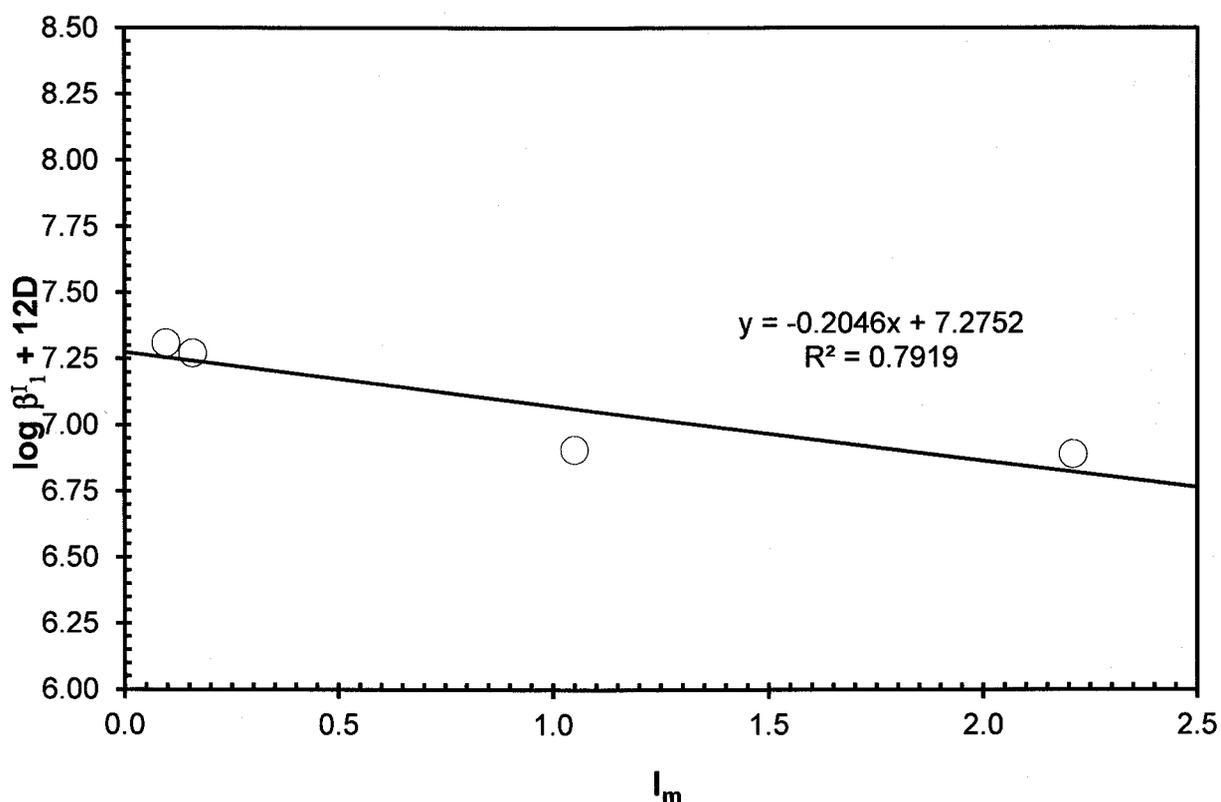


Figure 1. A plot showing $[\log \beta_1' + 12D]$ as a function of ionic strengths, where $\log \beta_1'$ is a stability constant at a certain ionic strength.

Using experimental solubility data of $\text{PbO}(\text{cr})$ in NaCl solutions and MgCl_2 solutions from this work, the Pitzer parameters associated with PbCit^- are modeled using EQ3/6 Version 8.0a (Tables 4 and 5) with the Python script (Kirchner, 2012).

In the modeling, the experimental data in NaCl solutions are employed to generate EQ3 files from CiNa-1.3i through CiNa-48.3i. Similarly, the experimental data in MgCl_2 solutions are used to produce EQ3 input files from CiMg-1.3i through CiMg-16.3i. The parameters obtained in this analysis are tabulated in Tables 4 and 5. These parameters correspond to those at Iteration 26 (see Task20-21_Results_PSD.txt and Results_07-16-2014.txt). In the data0.fm1, the $\beta^{(0)}$'s for most of the 1:1 interaction pairs are between 0.0068 and 0.5418. Therefore, the parameters at Iteration 26 are selected, because the parameters at that iteration are consistent with literature values in the data0.fm1, and are reproducible on Paul Domski's computer (Task20-21_Results_PSD.txt) and my computer (Results_07-16-2014.txt). The parameters including $\beta^{(0)}$'s and C^ϕ 's for 1:2 and 2:1 interaction pairs at other iterations are too large to be consistent with the known parameters. Notice that the above selection criterion has to be applied based on the following constraint and consideration. In this suite of EQ3 input files, when the

Python script changes the targeted parameters to certain values, some of input files create a convergence problem before the optimization is completed. In addition, we have also tried to set boundaries for the targeted parameters by using the script generated by Sungtae Kim. However, none of the combinations that have been chosen resulted in realistic parameter values.

It should be noted that the EQ3/6 run was officially performed by Paul Domski. The run on my computer mentioned above is for comparison to show reproducibility.

Table 2. Experimental results for solubility of PbO(cr) in NaCl solutions produced at SNL at 22.5 ± 1.5 °C*.

Experimental number	day	$\Sigma\text{Pb, mol}\cdot\text{kg}^{-1}$	$\Sigma\text{Na, mol}\cdot\text{kg}^{-1}$	$\Sigma\text{Cl, mol}\cdot\text{kg}^{-1}$	pmH ^A	$\Sigma\text{Citrate, mol}\cdot\text{kg}^{-1}$	$\Sigma\text{Mg, mol}\cdot\text{kg}^{-1}$
PbO-0.1-1	331	1.01E-02	0.05	0.05	8.25	0.08	0.08
PbO-1.0-1	331	8.61E-03	0.50	0.50	7.52	0.10	0.10
PbO-2.0-1	331	7.72E-03	1.05	1.05	7.32	0.10	0.10
PbO-3.0-1	331	6.55E-03	1.60	1.60	7.67	0.10	0.10
PbO-4.0-1	331	5.27E-03	2.20	2.20	7.54	0.10	0.10
PbO-5.0-1	331	3.19E-03	2.50	2.50	7.18	0.09	0.09
PbO-0.1-1	785	1.14E-02	0.05	0.05	8.13	0.08	0.08
PbO-1.0-1	785	1.06E-02	0.50	0.50	7.57	0.10	0.10
PbO-2.0-1	785	8.39E-03	1.05	1.05	7.18	0.10	0.10
PbO-3.0-1	785	3.91E-03	1.60	1.60	7.26	0.09	0.09
PbO-4.0-1	785	4.92E-03	2.20	2.20	7.31	0.10	0.10
PbO-5.0-1	785	3.40E-03	2.50	2.50	6.99	0.09	0.09
PbO-0.1-1	960	1.32E-02	0.05	0.05	8.50	0.09	0.09
PbO-1.0-1	960	1.06E-02	0.50	0.50	7.61	0.11	0.11
PbO-2.0-1	960	8.20E-03	1.05	1.05	7.29	0.11	0.11
PbO-3.0-1	960	3.79E-03	1.60	1.60	7.40	0.12	0.12
PbO-4.0-1	960	5.10E-03	2.20	2.20	7.48	0.13	0.13
PbO-5.0-1	960	3.54E-03	2.50	2.50	7.17	0.12	0.12
PbO-0.1-1	1009	1.33E-02	0.05	0.05	8.51	0.08	0.08
PbO-1.0-1	1009	1.10E-02	0.50	0.50	7.60	0.10	0.10
PbO-2.0-1	1009	8.89E-03	1.05	1.05	7.27	0.11	0.11
PbO-3.0-1	1009	4.53E-03	1.60	1.60	7.43	0.11	0.11
PbO-4.0-1	1009	7.54E-03	2.20	2.20	7.49	0.12	0.12
PbO-5.0-1	1009	5.36E-03	2.50	2.50	7.18	0.11	0.11
PbO-0.1-1	1057	1.43E-02	0.05	0.05	8.45	0.09	0.09
PbO-1.0-1	1057	1.24E-02	0.50	0.50	7.52	0.10	0.10
PbO-2.0-1	1057	9.14E-03	1.05	1.05	7.20	0.11	0.11
PbO-3.0-1	1057	4.88E-03	1.60	1.60	7.35	0.10	0.10
PbO-4.0-1	1057	5.63E-03	2.20	2.20	7.40	0.11	0.11
PbO-5.0-1	1057	4.48E-03	2.50	2.50	7.12	0.11	0.11

PbO-0.1-1	1107	1.40E-02	0.05	0.05	8.52	0.08	0.08
PbO-1.0-1	1107	1.20E-02	0.50	0.50	7.56	0.09	0.09
PbO-2.0-1	1107	8.59E-03	1.05	1.05	7.26	0.10	0.10
PbO-3.0-1	1107	4.50E-03	1.60	1.60	7.43	0.09	0.09
PbO-4.0-1	1107	5.35E-03	2.20	2.20	7.45	0.10	0.10
PbO-5.0-1	1107	3.86E-03	2.50	2.50	7.16	0.09	0.09
PbO-0.1-1	1257	1.41E-02	0.05	0.05	8.55	0.08	0.08
PbO-1.0-1	1257	1.08E-02	0.50	0.50	7.59	0.09	0.09
PbO-2.0-1	1257	8.88E-03	1.05	1.05	7.34	0.10	0.10
PbO-3.0-1	1257	6.15E-03	1.60	1.60	7.44	0.09	0.09
PbO-4.0-1	1257	5.80E-03	2.20	2.20	7.46	0.10	0.10
PbO-5.0-1	1257	1.00E-02	2.50	2.50	7.17	0.09	0.09
PbO-0.1-1	1357	1.41E-02	0.05	0.05	8.56	0.09	0.09
PbO-1.0-1	1357	1.13E-02	0.50	0.50	7.59	0.10	0.10
PbO-2.0-1	1357	8.54E-03	1.05	1.05	7.34	0.10	0.10
PbO-3.0-1	1357	4.36E-03	1.60	1.60	7.45	0.10	0.10
PbO-4.0-1	1357	5.08E-03	2.20	2.20	7.50	0.11	0.11
PbO-5.0-1	1357	3.51E-03	2.50	2.50	7.20	0.10	0.10

*Notice that the experimental data are compiled in Kirkes, Olivas, Jang, Kim, Xiong (2014). Citrate concentrations are based on the corresponding magnesium concentrations.

^A pH readings obtained by using a pH electrode were first converted to hydrogen ion concentrations on molar scale (pCH) by applying correction factors from Rai et al. (1995), and then converted to hydrogen ion concentrations on molal scale (pmH) using the equation of Xiong et al. (2010) based on molality/molarity ratios listed under the tab “MolalityToMolarityRatios” in the Spreadsheet AR_AP154_Tasks20-21_Modeling.xls.

Table 3. Experimental results for solubility of PbO(cr) in MgCl₂ solutions produced at SNL at 22.5 ± 1.5 °C*.

Experimental number	Day	ΣPb, mol•kg ⁻¹	ΣMg, mol•kg ⁻¹	ΣCl, mol•kg ⁻¹	pmH ^A	ΣCitrate, mol•kg ⁻¹
PbO-0.01Mg-1	331	1.27E-02	1.25E-01	1.00E-02	8.69	1.20E-01
PbO-0.1Mg-1	331	7.12E-03	1.70E-01	1.00E-01	7.96	1.20E-01
PbO-0.01Mg-1	785	1.77E-02	1.25E-01	1.00E-02	8.87	1.20E-01
PbO-0.1Mg-1	785	7.15E-03	1.70E-01	1.00E-01	7.87	1.20E-01
PbO-0.01Mg-1	960	1.77E-02	1.25E-01	1.00E-02	8.88	1.20E-01
PbO-0.1Mg-1	960	6.98E-03	1.70E-01	1.00E-01	8.00	1.20E-01
PbO-0.01Mg-1	1009	1.92E-02	1.25E-01	1.00E-02	8.88	1.20E-01
PbO-0.1Mg-1	1009	6.94E-03	1.70E-01	1.00E-01	8.00	1.20E-01
PbO-0.01Mg-1	1057	1.77E-02	1.25E-01	1.00E-02	8.79	1.20E-01
PbO-0.1Mg-1	1057	6.97E-03	1.70E-01	1.00E-01	7.87	1.20E-01
PbO-0.01Mg-1	1107	1.85E-02	1.25E-01	1.00E-02	8.86	1.20E-01
PbO-0.1Mg-1	1107	6.59E-03	1.70E-01	1.00E-01	7.92	1.20E-01
PbO-0.01Mg-1	1257	1.82E-02	1.25E-01	1.00E-02	8.86	1.20E-01
PbO-0.1Mg-1	1257	5.95E-03	1.70E-01	1.00E-01	7.80	1.20E-01
PbO-0.01Mg-1	1367	1.72E-02	1.25E-01	1.00E-02	8.86	1.20E-01
PbO-0.1Mg-1	1367	5.77E-03	1.70E-01	1.00E-01	7.74	1.20E-01

* Notice that this AR only analyses the experimental data at pmH above neutrality. In the lower pmH range, PbO(cr) may not be the solubility-controlling phase. The initial MgHCitrate concentration is used.

^A pH readings obtained by using a pH electrode were first converted to hydrogen ion concentrations on molar scale (pcH) by applying correction factors (Hansen, 2001), and then converted to hydrogen ion concentrations on molal scale (pmH) using the equation of Xiong et al. (2010) assuming unity value for molality/molarity ratios because of low MgCl₂ concentrations.

Table 4. Equilibrium constants at infinite dilution at 25°C and 1 bar for the Na⁺—Mg²⁺—Pb²⁺—Cl⁻—Cit³⁻ system

Reactions	$\log K_{s2}^o, \log \beta_1^o$	Reference and Remarks
$\text{PbO}(\text{cr}) + 2\text{H}^+ = \text{Pb}^{2+} + \text{H}_2\text{O}$	12.59	ymp.R2/ymp.R0
$\text{Pb}^{2+} + \text{Cit}^{3-} = \text{PbCit}^-$	$7.28 \pm 0.18 (2\sigma)$	This work, based on the values extrapolated to infinite dilution using the SIT model.

Table 5. Pitzer interaction parameters at 25°C and 1 bar for the Na⁺—Mg²⁺—Pb²⁺—Cl⁻—Cit³⁻ system

Pitzer Binary Interaction Parameters					
Species <i>i</i>	Species <i>j</i>	$\beta^{(0)}$	$\beta^{(1)}$	C^ϕ	Reference
Na ⁺	PbCit ⁻	0.535	0.29	0.0196	This study
Mg ²⁺	PbCit ⁻	1.97	1.74	0.0771	This study

Table 6. Locations of the Excel spreadsheets, EQ3/6 I/O files associated with calculations for this analysis*.

Description or Title of File(s)	Location of File(s)
Spreadsheet AR_AP154_Tasks20-21_Modeling.xls	In zip file AP154_Tasks20-21_DataPackage.zip, library LIBAP154_FILES
EQ3/6 DB DATA0.p19	In zip file AP154_Tasks20-21_DataPackage.zip, library LIBAP154_FILES
EQ3/6 I/O files: CiNa.3i through CiNa-48.3i CiMg-1.3i through CiMg-16.3i	In zip file AP154_Tasks20-21_DataPackage.zip, library LIBAP154_FILES
Python input script for modeling: PbCitrate_NaCl_MgCl2_logK_subroutine_QA.py	In zip file AP154_Tasks20-21_DataPackage.zip, library LIBAP154_FILES
Modeling Results: Results_07-16-2014.txt; Task20-21_Results_PSD.txt	In zip file AP154_Tasks20-21_DataPackage.zip, library LIBAP154_FILES

*The path for where the files listed in this table are located should be:/nfs/data/CVSLIB/WIPP_EXTERNAL/ap154_files/Files.

3 RESULTS

Tables 4 and 5 provide the $\log K_{s,2}^o$, and Pitzer parameter associated with $\log \beta_1^o$ in the $\text{Na}^+ - \text{Mg}^{2+} - \text{Pb}^{2+} - \text{Cl}^- - \text{Cit}^{3-}$ system derived in this study.

The Pitzer parameters evaluated by this study are similar to literature values for similar interactions in terms of magnitude. For example, $\beta^{(0)}$ for $\text{Na}^+ - \text{PbCit}^-$ is 0.535, in comparison to -0.2239 and -0.5418 for $\text{Na}^+ - \text{AmEDTA}^-$ and $\text{Na}^+ - \text{NpO}_2\text{Oxalate}^-$ in data0.fm1, respectively. The C^ϕ for $\text{Na}^+ - \text{PbCit}^-$ is 0.0196, in comparison to 0.002 and 0.095 for $\text{Na}^+ - \text{AmEDTA}^-$ and $\text{Na}^+ - \text{NpO}_2\text{Oxalate}^-$ in data0.fm1, respectively.

4 CONCLUSIONS

In this analysis report, the Pitzer parameter associated with $\log \beta_1^o$ for PbCit^- in the $\text{Na}^+ - \text{Mg}^{2+} - \text{Pb}^{2+} - \text{Cl}^- - \text{Cit}^{3-}$ system are obtained. With these parameters, the interactions of lead as radiation shielding material with Cit^{3-} in the brines can be modeled, and would have an impact on PA.

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