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Experimental Determination of Lead Interactions with Citrate and EDTA in NaCl and $MgCl_2$ Solutions to High Ionic Strength and Its Applications

Yongliang Xiong[1], Leslie Kirkes, Terry Westfall, Cassandra Marrs, Jandi Knox, Heather Burton

Sandia National Laboratories (SNL)
Carlsbad Programs Group
4100 National Parks Highway, Carlsbad, NM 88220, USA

[1] Corresponding author, e-mail: yxiong@sandia.gov.

ABSTRACT

In this study, the interactions of lead with citrate and ethylenediaminetetraacetate (EDTA) are investigated based on solubility measurements as a function of ionic strength at room temperature (22.5 ± 0.5°C) in NaCl and $MgCl_2$ solutions. The formation constants ($\log \beta_1^0$) for $Pb[C_3H_5O(COO)_3]^-$ (abbreviated as $PbCitrate^-$) and $Pb[(CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)]^{2-}$ (abbreviated as $PbEDTA^{2-}$)

$$Pb^{2+} + [C_3H_5O(COO)_3]^{3-} = Pb[C_3H_5O(COO)_3]^- \quad (1)$$

$$Pb^{2+} + (CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)^{4-} = Pb[(CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)]^{2-} \quad (2)$$

are evaluated as 7.28 ± 0.18 (2σ) and 20.00 ± 0.20 (2σ), respectively, with a set of Pitzer parameters describing the specific interactions in NaCl and $MgCl_2$ media. Based on these parameters, the interactions of lead with citrate and EDTA in various low temperature environments can be accurately modelled.

1. INTRODUCTION

Lead is a heavy metal and is known to be toxic to humans under certain conditions. Yet lead has been used throughout history in a number of applications and it therefore persists in our environment. Two chemicals that can strong complexe with lead are citric acid (chemical formula, $C_6H_8O_7$, or structural formula, CH_2COOH-$C(OH)COOH$-CH_2COOH, and its dissociated forms such as $[C_3H_5O(COO)_3]^{3-}$, abbreviated as "Citrate" hereafter), and ethylenediaminetetraacetate, $(CH_2COO)_2N(CH_2)_2N(CH_2COO)_2)^{4-}$, abbreviated as $EDTA^{4-}$. Complexation of citric acid and/or EDTA with lead can either be beneficial or detrimental depending on the chemical system that is being examined. For instance, in UK sewage effluents, the concentrations of EDTA are up to 1.6 mg/L ($\sim 5.5 \times 10^{-6}$ mol•kg^{-1}) (Garland et al., 1985). A concern is that the presence of EDTA in such wastewaters could remobilize toxic heavy metals (like lead) from sediments and sewage sludges because of the strong complexing nature of EDTA. On the other hand, the strong complexing potential of EDTA with heavy metals makes it useful in decontamination of soils polluted by heavy metals such as Pb and Zn (e.g., Theodoratos et al., 2000; Zeng et al., 2005). EDTA forms strong aqueous complexes with targeted heavy metals and can be used for extraction of targeted metal ions presented as contaminants. In addition, EDTA is used medically in chelation therapy to remove lead from the blood stream. The same is true for citric acid which is not used as commonly as EDTA for industrial cleaning purposes but does naturally occur. The strong complexation of lead with citrate renders the important role for citrate in the therapy of lead poisoning (Kety, 1942).

In the field of nuclear waste management, citrate, EDTA, and lead are frequently present together in nuclear waste (Brush and Xiong, 2009), because lead is present as waste as well as a shielding material while citric acid and EDTA are used as decontamination agents (Hummel et al., 2005). Also present in some of the nuclear waste are actinides, especially actinides in the +III oxidation state, which are also known to form relatively strong aqueous complexes with citrate and EDTA. Complexation with the actinides increase their solubilities, a condition adverse to containment of the radioactive material. In this particular example the presence of lead can be seen as beneficial if one notes that complexation of EDTA and/or citric acid occurs simultaneously with lead and with the actinides that are present. Complexation of lead in the nuclear system described above lowers the concentration of EDTA and/or citric acid available for reaction with the actinides creating a geochemical condition that is better for containment of the radioactive elements. This can have real significance for deep geologic disposal of actinide-bearing radioactive waste

There are significant inventories of lead in geological repositories for nuclear waste. For instance, there are sizable inventories of lead in both the waste and in shielding materials in the Waste Isolation Pilot Plant (WIPP) (Xiong et al., 2015). It is estimated that the total inventory for lead including lead in contact handled (CH) and remote handled (RH) waste and lead in waste packaging in the WIPP is on the order of 1,256,796 kg (Xiong et al., 2015). There are also significant inventories of EDTA and citrate in the WIPP. As an example, the 2009 EDTA inventory in the form of NaH_3EDTA for the WIPP was 3.54×10^2 kg for the 2009 Compliance Recertification

Application Performance Assessment Baseline Calculations (CRA-2009 PABC) (Brush and Xiong, 2009). .

Citrate may be naturally occurring in other low temperature aqueous environments, such as metallurgical slags (e.g., Ettler et al., 2004), where it can have an important role enhancing the mobilization of metals including lead.

The objective of this work is to investigate the interactions of lead with citrate and EDTA as a function of ionic strength to 5.0 $mol \cdot kg^{-1}$ in a NaCl medium, and to 6.0 $mol \cdot kg^{-1}$ in $MgCl_2$ medium, as NaCl and $MgCl_2$ are the most common and important components in natural aqueous systems, and they are dominant components in the WIPP brines, Generic Weep Brine (GWB) and Energy Research and Development Administration Well 6 (ERDA-6) (Xiong and Lord, 2008). Based on the measured solubilities, a Pitzer model is developed here for the interactions of lead with citrate and EDTA in NaCl and $MgCl_2$ media. The model enables researchers to estimate with a degree of high precision the interactions of lead with citrate and EDTA in various environments over a wide range of ionic strengths.

2. EXPERIMENTAL SECTION

In these solubility experiments, about 2 grams of litharge—ACS reagent grade lead oxide (CAS 1317-36-8) from MP Biomedicals was weighed out and placed into 150 mL plastic bottles. Then, 100 mL of supporting electrolyte solution were added to those bottles. Once filled, the lids of the bottles were sealed with parafilm.

97 The supporting electrolytes are a series of NaCl solutions ranging from 0.010
98 mol•kg^{-1} to 5.0 mol•kg^{-1}, and $MgCl_2$ solutions ranging from 0.01 mol•kg^{-1} to 2.0
99 mol•kg^{-1}. The supporting electrolyte solutions were prepared from degassed deionized
100 (DI) water. The degassed DI water was prepared by following a procedure similar to that
101 used by Wood et al. (2002) to remove dissolved CO_2. The Undersaturation experiments
102 are conducted at laboratory room temperature (22.5 ± 0.5°C).
103 The pH readings were measured with an Orion-Ross combination pH glass
104 electrode, coupled with an Orion Research EA 940 pH meter that was calibrated with
105 three pH buffers (pH 4, pH 7, and pH 10). In solutions with an ionic strength higher than
106 0.10 mol•kg^{-1}, hydrogen-ion concentrations on molar scale (pcH) were determined from
107 pH readings by using correction factors for NaCl and $MgCl_2$ solutions determined by Rai
108 et al. (1995) and Hansen (2001), respectively. Based on the equation in Xiong et al.
109 (2010), pcHs are converted to hydrogen-ion concentrations on a molal scale (pmH).
110 Solution samples were periodically withdrawn from experimental runs. Before
111 solution samples were taken, pH readings of experimental runs were measured. The
112 sample size was usually 3 mL. After a solution sample was withdrawn from an
113 experiment and filtered with a 0.2 μm syringe filter, the filtered solution was then
114 weighed, acidified with 0.5 mL of concentrated TraceMetal® grade HNO_3 from Fisher
115 Scientific, and finally diluted to a volume of 10 mL with DI water. If subsequent
116 dilutions were needed, aliquots were taken from the first dilution samples for the second
117 dilution, and aliquots of the second dilution were then taken for further dilution.
118 Lead concentrations of solutions were analyzed with a Perkin Elmer dual-view
119 inductively coupled plasma-atomic emission spectrometer (ICP-AES)

(Perkin Elmer DV 3300). Calibration blanks and standards were precisely matched with experimental matrices. The linear correlation coefficients of calibration curves in all measurements were better than 0.9995. The analytical precision for ICP-AES is better than 1.00% in terms of the relative standard deviation (RSD) based on replicate analyses. Citrate and EDTA concentrations are measured using a DIONEX ion chromatograph (IC) (DIONEX IC 3000).

Solid phases were analyzed using a Bruker D8 Advance X-ray diffractometer with a Sol-X detector, before and after experiments.

3. EXPERIMENTAL RESULTS, AND THERMODYNAMIC MODELING

3.1 Experimental Results

Experimental results for solubilities in NaCl and $MgCl_2$ solutions in the presence of citrate are tabulated in Tables 1 and 2, respectively. Similarly, experimental results for solubilities in NaCl and $MgCl_2$ solutions in the presence of EDTA are tabulated in Tables 3 and 4, respectively.

In Figure 1, total lead concentrations as a function of experimental time in NaCl and $MgCl_2$ solutions with citrate are displayed. The steady concentrations are reached in the second sampling, i.e., which were taken at 785 days (see Tables 1 and 2). From Figure 2, it is clear that steady-state concentrations in NaCl solutions with EDTA are achieved in the third sampling, which was taken at 951 days (Table 3). Lead solubilities as a function of experimental time in $MgCl_2$ solutions with EDTA are displayed in Figure 3. It is apparent from Figure 3 that steady-state lead concentrations in $MgCl_2$ solutions are achieved in the third sampling, which was taken at 937 days (Table 3). It is apparent that steady-state concentrations represent equilibrium concentrations, as the duration of experiments in this work, up to 1,367 days, is significantly longer than previous studies under similar conditions.

3.2 Thermodynamic Modeling

In the following, the experimental data described above are used to derive the thermodynamic parameters. The dissolution reaction for PbO(cr) can be expressed as,

$$PbO(cr) + 2H^+ = Pb^{2+} + H_2O \quad (1)$$

The corresponding solubility product constant of PbO(cr) at infinite dilution can be formulated as follows,

$$K_{s2}^{o} = \frac{a_{Pb^{2+}} \times a_{H_2O}}{a_{PbO(cr)} \times a_{H^+}^2} \quad (2)$$

In this work, the aqueous lead species included are Pb^{2+}, $PbCl^+$, $PbCl_2(aq)$, $PbCl_3^-$, $PbOH^+$, $Pb(OH)_2(aq)$, $Pb(OH)_3^-$, and $PbCitrate^-$. Except for $PbCitrate^-$, other lead species have been covered in several previous publications (Xiong et al., 2013; Xiong, 2015; Xiong et al., 2017).

The formation reaction for $PbCit^-$ is written as,

$$Pb^{2+} + Citrate^{3-} = PbCitrate^- \quad (3)$$

The corresponding formation constant at infinite dilution is,

$$\beta_1^0 = \frac{m_{PbCit^-}}{m_{Pb^{2+}} \times m_{Cit^{3-}}} \times \frac{\gamma_{PbCit^-}}{\gamma_{Pb^{2+}} \times \gamma_{Cit^{3-}}} = \frac{a_{PbCit^-}}{a_{Pb^{2+}} \times a_{Cit^{3-}}} \quad (4)$$

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

$$Pb^{2+} + 2Citrate^{3-} = Pb(Citrate)_2^{4-} \quad (5)$$

The corresponding formation constant at infinite dilution is,

$$\beta_2^0 = \frac{m_{Pb(Cit)_2^{4-}}}{m_{Pb^{2+}} \times m_{Cit^{3-}}^2} \times \frac{\gamma_{Pb(Cit)_2^{4-}}}{\gamma_{Pb^{2+}} \times \gamma_{Cit^{3-}}^2} = \frac{a_{Pb(Cit)_2^{4-}}}{a_{Pb^{2+}} \times a_{Cit^{3-}}^2} \quad (6)$$

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

In this work, the log β_1^0 at infinite dilution was evaluated by using the SIT model (Figure 4) 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 β_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,

194 $$D = \frac{A_\gamma \sqrt{I_m}}{1+1.5\sqrt{I_m}} \quad (8)$$

195

196 where A_γ is the Debye-Hükel slope for activity coefficient, which is 0.5092 at 25°C from

197 Helgeson and Kirkham (1974), and I_m ionic strength on a molal scale.

198 Using experimental solubility data of PbO(cr) in NaCl solutions and $MgCl_2$

199 solutions from this work, the Pitzer parameters associated with $PbCit^-$ are modeled

200 (Table 5), based on thermodynamic modeling with the Pitzer equations. The computer

201 code, EQ3/6 Version 8.0a (Wolery et al., 2010; Xiong, 2011), is used as the modeling

202 platform, which was also used in previous modeling work for obtaining thermodynamic

203 properties including the Pitzer parameters (e.g., Xiong et al., 2013, 2017; Xiong, 2013,

204 2015). The database containing all parameters necessary including thermodynamic

205 properties for the modeling, is data0.fm1 (Xiong, 2011). In the database, data0.fm1, the

206 interaction parameters for major ions are from Harvie et al. (1984), and the interaction

207 parameters for organic ligands are from Choppin et al. (2001).

208 In the model fitting, the experimental data were first used to generate EQ3/6 input

209 files. Then, a script such as a Python script was generated to call the targeted parameters,

210 and call EQ3/6. The minimization subroutine in the script automatically compares

211 differences between experimental values [i.e., total calcium concentrations,

212 ΣPb(II)/mol•kg^{-1}] and model-predicted values produced by a set of inputted parameters

213 in each iteration. The iteration is completed when the difference is finally minimized.

214 With regard to the interactions of lead with EDTA, the formation reaction for

215 $PbEDTA^{2-}$ is written as,

$$Pb^{2+} + EDTA^{4-} = PbEDTA^{2-} \quad (9)$$

The corresponding formation constant at infinite dilution is,

$$\beta_1^0 = \frac{m_{PbEDTA^{2-}}}{m_{Pb^{2+}} \times m_{EDTA^{4-}}} \times \frac{\gamma_{PbEDTA^{2-}}}{\gamma_{Pb^{2+}} \times \gamma_{EDTA^{4-}}} = \frac{a_{PbEDTA^{2-}}}{a_{Pb^{2+}} \times a_{EDTA^{4-}}} \quad (10)$$

In the review performed by Andregg (1977), the author recommended a value of 18.30 ± 0.21 in logarithmic unit at an ionic strength of 0.1 M and 20°C for the formation constant for Reaction (3). In the work of Uhler and Helz (1984), they obtained a value of 17.42 ± 0.06 at an ionic strength of 0.16 M and 25°C. In this work, the value recommended by Andregg (1977) is extrapolated to infinite dilution by using the Davies equation,

$$\log \gamma_i = -A_\gamma z_i^2 \left(\frac{\sqrt{I_m}}{1+\sqrt{I_m}} + 0.2 I_m\right) \quad (10)$$

where γ_i is the activity coefficient of species i, A_γ the Debye-Hükel slope for activity coefficient, which is 0.5092 at 25°C from Helgeson and Kirkham (1974), z_i the charge of species i, and I_m ionic strength on molal scale.

According to Eq. (10), a value of 20.42 ± 0.21 (2σ) for $\log \beta_1^o$ is obtained based on the calculation for the value of Andregg (1977). Similarly, based on the value of Uhler and Helz (1984), the value for $\log \beta_1^o$ is 20.01 ± 0.20 (2σ). Using these two values

237 as baselines, the $\log \beta_1^o$ is varied from 20.42 to 20.00 in the simulations for deciding
238 which value should be selected. The simulations indicate that the residuals slightly
239 decrease when $\log \beta_1^o$ changes to 20.00, when other parameters remain constant.
240 Therefore, $\log \beta_1^o$ of 20.00 is selected, and it is identical to the value calculated from that
241 of Uhler and Helz (1984) when the uncertainties are taken into consideration.
242 Using experimental solubility data of PbO(cr) in NaCl and $MgCl_2$ solutions from
243 this work, the Pitzer parameters associated with $PbEDTA^{2-}$ are modeled.
244 In Table 5, the formation constant for $PbCitrate^-$ and $PbEDTA^{2-}$, and a set of
245 Pitzer parameters describing the specific interactions of lead with citrate and EDTA in
246 NaCl and $MgCl_2$ media are listed. These Pitzer parameters are similar to those found in
247 the literature for the similar interactions in terms of magnitude. For example, $\beta^{(0)}$ for
248 Na^+—$PbCit^-$ is 0.535, in comparison to –0.2239 and –0.5418 for Na^+—$AmEDTA^-$ and
249 Na^+—$NpO_2Oxalate^-$ in data0.fm1, respectively, in magnitude. The C^ϕ for Na^+—$PbCit^-$ is
250 0.0196, in comparison to 0.002 and 0.095 for Na^+—$AmEDTA^-$ and Na^+—$NpO_2Oxalate^-$
251 in data0.fm1, respectively. As other examples, $\beta^{(0)}$ for Na^+—$PbEDTA^{2-}$ is 0.25, in
252 comparison of 0.4733 and 0.2134 for Na^+—NpO_2HEDTA^{2-} and Na^+—$MgEDTA^{2-}$ in
253 data0.fm1, respectively. The C^ϕ for Na^+—$PbEDTA^{2-}$ is 0.0525, in comparison of
254 0.00869 and 0.142 for Na^+—$MgEDTA^{2-}$ and Na^+—$NpO_2Citrate^{2-}$ in data0.fm1,
255 respectively. The $\beta^{(0)}$ for Mg^{2+}—$PbEDTA^{2-}$ is 0.85, in comparison of –1.42258 from
256 Yin et al. (2007) for the Mg^{2+}—$B_4O_2(OH)_4^{2-}$ interaction. The C^ϕ for Mg^{2+}—$PbEDTA^{2-}$
257 is 0.1, in comparison of 0.025 for the Mg^{2+}—SO_4^{2-} interaction in data0.fm1.

It should be noted that the $\beta^{(1)}$'s in Table 5 were not calculated. Instead, they are pre-set to the average values for the respective interactions, following the paradigm of Choppin et al. (2001). In Choppin et al. (2001), they calculated and recommended a set of average values of $\beta^{(1)}$'s for various interactions (e.g., 1:1, 1:2/2:1, 1:3/3:1, etc., interactions), based on the $\beta^{(1)}$ values for respective interactions from literature.

4. CONCLUSIONS

In this study, we investigate the interactions of lead with citrate and EDTA in NaCl and $MgCl_2$ solutions via a series of long-term solubility measurements. A Pitzer model is developed based on these solubility measurements. This model would provide accurate descriptions about the interactions of lead with citrate and EDTA in NaCl and $MgCl_2$ matrixes under various conditions with applications to many fields such as nuclear waste management and environmental remediation of heavy metal contamination.

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Table 1. Experimental results for solubility of PbO(cr) in NaCl solutions in the presence of citrate produced at SNL at 22.5 ± 0.5 ºC.

Experimental number	day	ΣPb, mol•kg^{-1}	ΣNa, mol•kg^{-1}	ΣCl, mol•kg^{-1}	pmH [A]	ΣCitrate, mol•kg^{-1}	ΣMg, mol•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

[A] Values of pmH reported are calculated by using the correction factors (A_M) from Rai et al. (1995) for pH readings, and conversion factors (Θ) from molarity to molality, $pmH = pH_{ob} + A_M - \log \Theta$ (Xiong et al., 2010). The conversion factors are calculated from densities for NaCl solutions, which are from Sőhnel and Novotný (1985).

Table 2. Experimental results for solubility of PbO(cr) in $MgCl_2$ solutions in the presence of citrate produced at SNL at 22.5 ± 0.5 °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

[A] Values of pmH reported are calculated by using the correction factors (A_M) from Hansen (2001) for pH readings, and conversion factors (Θ) from molarity to molality, $pmH = pH_{ob} + A_M - \log \Theta$ (Xiong et al., 2010). The conversion factors are from the EQ3 output files with the respective $MgCl_2$ concentrations.

435
436 Table 3. Experimental results concerning solubility of PbO(cr) in NaCl solutions in the
437 presence of EDTA produced at SNL at 22.5 ± 0.5 ºC.
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Experimental number	day	ΣPb, mol•kg^{-1}	ΣNa, mol•kg^{-1}	ΣCl, mol•kg^{-1}	pmH [A]	ΣEDTA, mol•kg^{-1}	ΣMg, mol•kg^{-1}
Mg2EDTA-0.01-2	848	1.07E-01	0.01	0.01	10.63	0.15	0.29
Mg2EDTA-0.1-1	848	1.01E-01	0.09	0.09	10.83	0.15	0.29
Mg2EDTA-0.1-2	848	9.71E-02	0.09	0.09	10.75	0.15	0.29
Mg2EDTA-1.0-1	848	9.38E-02	0.94	0.94	10.80	0.15	0.29
Mg2EDTA-1.0-2	848	1.03E-01	0.94	0.94	10.72	0.15	0.29
Mg2EDTA-2.0-1	848	1.06E-01	1.87	1.87	10.78	0.15	0.29
Mg2EDTA-2.0-2	848	1.01E-01	1.87	1.87	10.72	0.15	0.29
Mg2EDTA-3.0-1	848	9.62E-02	2.80	2.80	10.83	0.15	0.29
Mg2EDTA-3.0-2	848	9.24E-02	2.80	2.80	10.77	0.15	0.29
Mg2EDTA-4.0-1	848	9.03E-02	3.74	3.74	11.01	0.15	0.29
Mg2EDTA-4.0-2	848	9.27E-02	3.74	3.74	10.83	0.15	0.29
Mg2EDTA-5.0-1	848	9.65E-02	4.67	4.67	10.71	0.15	0.29
Mg2EDTA-5.0-2	848	9.69E-02	4.67	4.67	11.01	0.15	0.29
Mg2EDTA-0.01-2	905	9.89E-02	0.01	0.01	10.56	0.15	0.29
Mg2EDTA-0.1-1	905	1.02E-01	0.09	0.09	10.75	0.15	0.29
Mg2EDTA-0.1-2	905	1.01E-01	0.09	0.09	10.62	0.15	0.29
Mg2EDTA-1.0-1	905	1.00E-01	0.94	0.94	10.68	0.15	0.29
Mg2EDTA-1.0-2	905	9.55E-02	0.94	0.94	10.67	0.15	0.29
Mg2EDTA-2.0-1	905	1.13E-01	1.87	1.87	10.71	0.15	0.29
Mg2EDTA-2.0-2	905	8.88E-02	1.87	1.87	10.66	0.15	0.29
Mg2EDTA-3.0-1	905	1.01E-01	2.80	2.80	10.77	0.15	0.29
Mg2EDTA-3.0-2	905	9.18E-02	2.80	2.80	10.64	0.15	0.29
Mg2EDTA-4.0-1	905	9.88E-02	3.74	3.74	10.92	0.15	0.29
Mg2EDTA-4.0-2	905	9.03E-02	3.74	3.74	10.75	0.15	0.29
Mg2EDTA-5.0-1	905	9.75E-02	4.67	4.67	10.62	0.15	0.29
Mg2EDTA-5.0-2	905	9.96E-02	4.67	4.67	10.96	0.15	0.29
Mg2EDTA-0.01-2	951	1.14E-01	0.01	0.01	10.52	0.15	0.29
Mg2EDTA-0.1-1	951	1.03E-01	0.09	0.09	10.66	0.15	0.29
Mg2EDTA-0.1-2	951	9.36E-02	0.09	0.09	10.56	0.15	0.29
Mg2EDTA-1.0-1	951	9.16E-02	0.94	0.94	10.61	0.15	0.29
Mg2EDTA-1.0-2	951	9.82E-02	0.94	0.94	10.58	0.15	0.29
Mg2EDTA-2.0-1	951	1.04E-01	1.87	1.87	10.60	0.15	0.29

Mg2EDTA-2.0-2	951	1.02E-01	1.87	1.87	10.55	0.15	0.29
Mg2EDTA-3.0-1	951	9.35E-02	2.80	2.80	10.64	0.15	0.29
Mg2EDTA-3.0-2	951	8.86E-02	2.80	2.80	10.47	0.15	0.29
Mg2EDTA-4.0-1	951	8.48E-02	3.74	3.74	10.77	0.15	0.29
Mg2EDTA-4.0-2	951	9.34E-02	3.74	3.74	10.63	0.15	0.29
Mg2EDTA-5.0-1	951	9.68E-02	4.67	4.67	10.45	0.15	0.29
Mg2EDTA-5.0-2	951	9.20E-02	4.67	4.67	10.80	0.15	0.29
Mg2EDTA-0.01-2	1000	1.11E-01	0.01	0.01	10.47	0.15	0.29
Mg2EDTA-0.1-1	1000	1.02E-01	0.09	0.09	10.62	0.15	0.29
Mg2EDTA-0.1-2	1000	9.92E-02	0.09	0.09	10.52	0.15	0.29
Mg2EDTA-1.0-1	1000	9.30E-02	0.94	0.94	10.59	0.15	0.29
Mg2EDTA-1.0-2	1000	1.03E-01	0.94	0.94	10.56	0.15	0.29
Mg2EDTA-2.0-1	1000	1.04E-01	1.87	1.87	10.55	0.15	0.29
Mg2EDTA-2.0-2	1000	1.04E-01	1.87	1.87	10.52	0.15	0.29
Mg2EDTA-3.0-1	1000	9.21E-02	2.80	2.80	10.62	0.15	0.29
Mg2EDTA-3.0-2	1000	9.64E-02	2.80	2.80	10.45	0.15	0.29
Mg2EDTA-4.0-1	1000	9.60E-02	3.74	3.74	10.73	0.15	0.29
Mg2EDTA-4.0-2	1000	9.19E-02	3.74	3.74	10.59	0.15	0.29
Mg2EDTA-5.0-1	1000	9.58E-02	4.67	4.67	10.43	0.15	0.29
Mg2EDTA-5.0-2	1000	8.87E-02	4.67	4.67	10.80	0.15	0.29
Mg2EDTA-0.01-2	1056	1.11E-01	0.01	0.01	10.50	0.15	0.29
Mg2EDTA-0.1-1	1056	1.06E-01	0.09	0.09	10.66	0.15	0.29
Mg2EDTA-0.1-2	1056	1.01E-01	0.09	0.09	10.54	0.15	0.29
Mg2EDTA-1.0-1	1056	8.97E-02	0.94	0.94	10.63	0.15	0.29
Mg2EDTA-1.0-2	1056	9.92E-02	0.94	0.94	10.61	0.15	0.29
Mg2EDTA-2.0-1	1056	1.02E-01	1.87	1.87	10.60	0.15	0.29
Mg2EDTA-2.0-2	1056	9.97E-02	1.87	1.87	10.56	0.15	0.29
Mg2EDTA-3.0-1	1056	9.45E-02	2.80	2.80	10.63	0.15	0.29
Mg2EDTA-3.0-2	1056	9.23E-02	2.80	2.80	10.45	0.15	0.29
Mg2EDTA-4.0-1	1056	8.90E-02	3.74	3.74	10.73	0.15	0.29
Mg2EDTA-4.0-2	1056	9.38E-02	3.74	3.74	10.61	0.15	0.29
Mg2EDTA-5.0-1	1056	9.16E-02	4.67	4.67	10.80	0.15	0.29
Mg2EDTA-5.0-2	1056	9.77E-02	4.67	4.67	10.46	0.15	0.29
Mg2EDTA-0.01-2	1213	1.10E-01	0.01	0.01	10.51	0.15	0.29
Mg2EDTA-0.1-1	1213	1.04E-01	0.09	0.09	10.66	0.15	0.29
Mg2EDTA-0.1-2	1213	9.92E-02	0.09	0.09	10.54	0.15	0.29
Mg2EDTA-1.0-1	1213	9.12E-02	0.94	0.94	10.64	0.15	0.29
Mg2EDTA-1.0-2	1213	9.95E-02	0.94	0.94	10.62	0.15	0.29

Mg2EDTA-2.0-1	1213	1.03E-01	1.87	1.87	10.60	0.15	0.29
Mg2EDTA-2.0-2	1213	9.90E-02	1.87	1.87	10.55	0.15	0.29
Mg2EDTA-3.0-1	1213	9.40E-02	2.80	2.80	10.60	0.15	0.29
Mg2EDTA-3.0-2	1213	8.63E-02	2.80	2.80	10.38	0.15	0.29
Mg2EDTA-4.0-1	1213	9.08E-02	3.74	3.74	10.68	0.15	0.29
Mg2EDTA-4.0-2	1213	8.98E-02	3.74	3.74	10.60	0.15	0.29
Mg2EDTA-5.0-1	1213	9.88E-02	4.67	4.67	10.45	0.15	0.29
Mg2EDTA-5.0-2	1213	9.33E-02	4.67	4.67	10.79	0.15	0.29
Mg2EDTA-0.01-2	1302	1.15E-01	0.01	0.01	10.43	0.15	0.29
Mg2EDTA-0.1-1	1302	1.07E-01	0.09	0.09	10.57	0.15	0.29
Mg2EDTA-0.1-2	1302	9.96E-02	0.09	0.09	10.45	0.15	0.29
Mg2EDTA-1.0-1	1302	9.41E-02	0.94	0.94	10.56	0.15	0.29
Mg2EDTA-1.0-2	1302	1.03E-01	0.94	0.94	10.54	0.15	0.29
Mg2EDTA-2.0-1	1302	1.06E-01	1.87	1.87	10.51	0.15	0.29
Mg2EDTA-2.0-2	1302	1.03E-01	1.87	1.87	10.48	0.15	0.29
Mg2EDTA-3.0-1	1302	9.70E-02	2.80	2.80	10.52	0.15	0.29
Mg2EDTA-3.0-2	1302	9.42E-02	2.80	2.80	10.28	0.15	0.29
Mg2EDTA-4.0-1	1302	9.72E-02	3.74	3.74	10.60	0.15	0.29
Mg2EDTA-4.0-2	1302	9.48E-02	3.74	3.74	10.51	0.15	0.29
Mg2EDTA-5.0-1	1302	9.78E-02	4.67	4.67	10.39	0.15	0.29
Mg2EDTA-5.0-2	1302	9.49E-02	4.67	4.67	10.69	0.15	0.29
Mg2EDTA-0.01-2	1345	1.07E-01	0.01	0.01	10.43	0.15	0.29
Mg2EDTA-0.1-1	1345	1.03E-01	0.09	0.09	10.53	0.15	0.29
Mg2EDTA-0.1-2	1345	1.01E-01	0.09	0.09	10.43	0.15	0.29
Mg2EDTA-1.0-1	1345	9.19E-02	0.94	0.94	10.54	0.15	0.29
Mg2EDTA-1.0-2	1345	9.61E-02	0.94	0.94	10.51	0.15	0.29
Mg2EDTA-2.0-1	1345	1.02E-01	1.87	1.87	10.49	0.15	0.29
Mg2EDTA-2.0-2	1345	9.83E-02	1.87	1.87	10.44	0.15	0.29
Mg2EDTA-3.0-1	1345	9.98E-02	2.80	2.80	10.48	0.15	0.29
Mg2EDTA-3.0-2	1345	9.63E-02	2.80	2.80	10.24	0.15	0.29
Mg2EDTA-4.0-1	1345	9.28E-02	3.74	3.74	10.55	0.15	0.29
Mg2EDTA-4.0-2	1345	9.32E-02	3.74	3.74	10.46	0.15	0.29
Mg2EDTA-5.0-1	1345	1.00E-01	4.67	4.67	10.35	0.15	0.29
Mg2EDTA-5.0-2	1345	9.19E-02	4.67	4.67	10.64	0.15	0.29

[A] Values of pmH reported are calculated by using the correction factors (A_M) from Rai et al. (1995) for pH readings, and conversion factors (Θ) from molarity to molality, $pmH = pH_{ob} + A_M - \log \Theta$ (Xiong et al., 2010). The conversion factors are calculated from densities for NaCl solutions, which are from Sőhnel and Novotný (1985).

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Table 4. Experimental results concerning solubility of PbO(cr) in $MgCl_2$ solutions in the presence of EDTA produced at SNL at 22.5 ± 0.5 ºC.

Experimental number	Day	ΣPb, mol•kg^{-1}	ΣMg, mol•kg^{-1}	ΣCl, mol•kg^{-1}	pmH [A]	ΣEDTA, mol•kg^{-1}
PbO-0.01ED-1	328	3.58E-02	8.00E-03	0.0160	11.70	0.0418
PbO-0.01ED-2	328	3.52E-02	8.00E-03	0.0160	11.64	0.0418
PbO-0.1ED-1	328	3.44E-02	0.0800	0.160	9.61	0.0418
PbO-0.1ED-2	328	3.39E-02	0.0800	0.160	9.62	0.0418
PbO-1.0ED-1	328	2.50E-02	0.800	1.60	9.25	0.0418
PbO-1.0ED-2	328	2.31E-02	0.800	1.60	9.25	0.0418
PbO-1.5ED-1	328	1.95E-02	1.20	2.40	9.20	0.0418
PbO-1.5ED-2	328	1.89E-02	1.20	2.40	9.20	0.0418
PbO-2.0ED-1	328	1.59E-02	1.60	3.20	9.16	0.0418
PbO-2.0ED-2	328	1.69E-02	1.60	3.20	9.15	0.0418
PbO-2.5ED-1	328	1.63E-02	2.00	4.00	9.11	0.0418
PbO-2.5ED-2	328	1.58E-02	2.00	4.00	9.10	0.0418
PbO-0.01ED-1	407	3.61E-02	8.00E-03	0.0160	11.67	0.0418
PbO-0.01ED-2	407	3.58E-02	8.00E-03	0.0160	11.65	0.0418
PbO-0.1ED-1	407	3.36E-02	0.0800	0.160	9.59	0.0418
PbO-0.1ED-2	407	3.50E-02	0.0800	0.160	9.64	0.0418
PbO-1.0ED-1	407	3.15E-02	0.800	1.60	9.19	0.0418
PbO-1.0ED-2	407	3.19E-02	0.800	1.60	9.16	0.0418
PbO-1.5ED-1	407	3.13E-02	1.20	2.40	9.09	0.0418
PbO-1.5ED-2	407	3.10E-02	1.20	2.40	9.16	0.0418
PbO-2.0ED-1	407	2.97E-02	1.60	3.20	9.03	0.0418
PbO-2.0ED-2	407	3.01E-02	1.60	3.20	9.05	0.0418
PbO-2.5ED-1	407	2.73E-02	2.00	4.00	8.98	0.0418
PbO-2.5ED-2	407	3.08E-02	2.00	4.00	8.97	0.0418
PbO-0.01ED-1	937	4.14E-02	8.00E-03	0.0160	11.72	0.0418
PbO-0.01ED-2	937	3.92E-02	8.00E-03	0.0160	11.70	0.0418
PbO-0.1ED-1	937	3.81E-02	0.0800	0.160	9.59	0.0418
PbO-0.1ED-2	937	3.91E-02	0.0800	0.160	9.58	0.0418
PbO-1.0ED-1	937	3.80E-02	0.800	1.60	9.20	0.0418
PbO-1.0ED-2	937	3.72E-02	0.800	1.60	9.19	0.0418

PbO-1.5ED-1	937	3.73E-02	1.20	2.40	9.16	0.0418
PbO-1.5ED-2	937	3.91E-02	1.20	2.40	9.17	0.0418
PbO-2.0ED-1	937	3.47E-02	1.60	3.20	9.09	0.0418
PbO-2.0ED-2	937	4.14E-02	1.60	3.20	9.12	0.0418
PbO-2.5ED-1	937	3.71E-02	2.00	4.00	8.95	0.0418
PbO-2.5ED-2	937	3.70E-02	2.00	4.00	8.95	0.0418
PbO-0.01ED-1	986	4.01E-02	8.00E-03	0.0160	11.66	0.0418
PbO-0.01ED-2	986	3.97E-02	8.00E-03	0.0160	11.63	0.0418
PbO-0.1ED-1	986	3.89E-02	0.0800	0.160	9.51	0.0418
PbO-0.1ED-2	986	3.96E-02	0.0800	0.160	9.47	0.0418
PbO-1.0ED-1	986	3.79E-02	0.800	1.60	9.16	0.0418
PbO-1.0ED-2	986	3.73E-02	0.800	1.60	9.20	0.0418
PbO-1.5ED-1	986	3.76E-02	1.20	2.40	9.15	0.0418
PbO-1.5ED-2	986	3.74E-02	1.20	2.40	9.16	0.0418
PbO-2.0ED-1	986	3.71E-02	1.60	3.20	9.12	0.0418
PbO-2.0ED-2	986	3.64E-02	1.60	3.20	9.12	0.0418
PbO-2.5ED-1	986	3.76E-02	2.00	4.00	8.96	0.0418
PbO-2.5ED-2	986	3.79E-02	2.00	4.00	8.97	0.0418
PbO-0.01ED-1	1035	4.06E-02	8.00E-03	0.0160	11.77	0.0418
PbO-0.01ED-2	1035	4.14E-02	8.00E-03	0.0160	11.76	0.0418
PbO-0.1ED-1	1035	4.09E-02	0.0800	0.160	9.60	0.0418
PbO-0.1ED-2	1035	3.97E-02	0.0800	0.160	9.62	0.0418
PbO-1.0ED-1	1035	4.29E-02	0.800	1.60	9.21	0.0418
PbO-1.0ED-2	1035	4.07E-02	0.800	1.60	9.14	0.0418
PbO-1.5ED-1	1035	4.09E-02	1.20	2.40	9.15	0.0418
PbO-1.5ED-2	1035	4.11E-02	1.20	2.40	9.18	0.0418
PbO-2.0ED-1	1035	3.98E-02	1.60	3.20	9.03	0.0418
PbO-2.0ED-2	1035	3.94E-02	1.60	3.20	9.16	0.0418
PbO-2.5ED-1	1035	3.82E-02	2.00	4.00	9.04	0.0418
PbO-2.5ED-2	1035	3.79E-02	2.00	4.00	9.05	0.0418
PbO-0.01ED-1	1084	4.01E-02	8.00E-03	0.0160	11.70	0.0418
PbO-0.01ED-2	1084	3.91E-02	8.00E-03	0.0160	11.70	0.0418
PbO-0.1ED-1	1084	3.96E-02	0.0800	0.160	9.56	0.0418
PbO-0.1ED-2	1084	3.96E-02	0.0800	0.160	9.55	0.0418

PbO-1.0ED-1	1084	3.84E-02	0.800	1.60	9.16	0.0418
PbO-1.0ED-2	1084	3.40E-02	0.800	1.60	9.19	0.0418
PbO-1.5ED-1	1084	3.78E-02	1.20	2.40	9.15	0.0418
PbO-1.5ED-2	1084	3.75E-02	1.20	2.40	9.16	0.0418
PbO-2.0ED-1	1084	3.80E-02	1.60	3.20	9.09	0.0418
PbO-2.0ED-2	1084	3.74E-02	1.60	3.20	9.10	0.0418
PbO-2.5ED-1	1084	3.73E-02	2.00	4.00	8.93	0.0418
PbO-2.5ED-2	1084	3.71E-02	2.00	4.00	8.93	0.0418
PbO-0.01ED-1	1236	3.99E-02	8.00E-03	0.0160	11.75	0.0418
PbO-0.01ED-2	1236	3.94E-02	8.00E-03	0.0160	11.74	0.0418
PbO-0.1ED-1	1236	3.88E-02	0.0800	0.160	9.62	0.0418
PbO-0.1ED-2	1236	3.90E-02	0.0800	0.160	9.58	0.0418
PbO-1.0ED-1	1236	3.64E-02	0.800	1.60	9.27	0.0418
PbO-1.0ED-2	1236	3.79E-02	0.800	1.60	9.28	0.0418
PbO-1.5ED-1	1236	3.67E-02	1.20	2.40	9.23	0.0418
PbO-1.5ED-2	1236	3.68E-02	1.20	2.40	9.22	0.0418
PbO-2.0ED-1	1236	3.67E-02	1.60	3.20	9.18	0.0418
PbO-2.0ED-2	1236	4.02E-02	1.60	3.20	9.17	0.0418
PbO-2.5ED-1	1236	3.61E-02	2.00	4.00	8.99	0.0418
PbO-2.5ED-2	1236	3.59E-02	2.00	4.00	8.99	0.0418
PbO-0.01ED-1	1344	3.99E-02	8.00E-03	0.0160	11.71	0.0418
PbO-0.01ED-2	1344	3.99E-02	8.00E-03	0.0160	11.70	0.0418
PbO-0.1ED-1	1344	3.94E-02	0.0800	0.160	9.55	0.0418
PbO-0.1ED-2	1344	3.93E-02	0.0800	0.160	9.56	0.0418
PbO-1.0ED-1	1344	3.67E-02	0.800	1.60	9.22	0.0418
PbO-1.0ED-2	1344	3.71E-02	0.800	1.60	9.22	0.0418
PbO-1.5ED-1	1344	3.63E-02	1.20	2.40	9.17	0.0418
PbO-1.5ED-2	1344	3.61E-02	1.20	2.40	9.17	0.0418
PbO-2.0ED-1	1344	3.55E-02	1.60	3.20	9.12	0.0418
PbO-2.0ED-2	1344	3.55E-02	1.60	3.20	9.11	0.0418
PbO-2.5ED-1	1344	3.68E-02	2.00	4.00	8.94	0.0418
PbO-2.5ED-2	1344	3.70E-02	2.00	4.00	8.94	0.0418

[A] Values of pmH reported are calculated by using the correction factors (A_M) from Hansen (2001) for pH readings, and conversion factors (Θ) from molarity to molality,

454 pmH = $pH_{ob} + A_M - \log \Theta$ (Xiong et al., 2010). The conversion factors are from the EQ3
455 output files with the respective $MgCl_2$ concentrations.
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Table 5. Equilibrium constants at infinite dilution, 25°C and 1 bar, Pitzer interaction parameters in the Na^+—Mg^{2+}—Pb^{2+}—Cl^-—$[C_3H_5O(COO)_3]^{3-}$ (or $Citrate^{3-}$)—$EDTA^{4-}$ system

Pitzer Parameters					
Species, *i*	Species, *j*	$\beta^{(0)}$	$\beta^{(1)}$	C^{ϕ}	References
Na^+	$Pb[C_3H_5O(COO)_3]^-$	0.535	0.29 [A]	0.0196	This work
Mg^{2+}	$Pb[C_3H_5O(COO)_3]^-$	1.97	1.74 [A]	0.0771	This work
Na^+	$PbEDTA^{2-}$	0.65	1.74 [A]	0.065	This work
Mg^{2+}	$PbEDTA^{2-}$	1.84	3.27	–0.15	This work [C]
Equilibrium Constants for Dissolution Reaction of PbO(cr) and Formation Reactions for $Pb[C_3H_5O(COO)_3]^-$ and $PbEDTA^{2-}$					
Reaction			log K_{sp} and log β_1 at 25 °C		References
$PbO(cr) + 2H^+ = Pb^{2+} + H_2O(l)$			12.59		ymp.R2/ymp.R0
$Pb^{2+} + [C_3H_5O(COO)_3]^{3-} = Pb[C_3H_5O(COO)_3]^-$			7.28 ± 0.18 (2σ)		This work
$Pb^{2+} + EDTA^{4-} = PbEDTA^{2-}$			20.00 ± 0.20 (2σ)		This work

[A] Values are set according to AP-154, Revision 2 (Xiong, 2013b).

Figure Captions

Figure 1. A plot showing experimental total lead concentrations in equilibrium with litharge, α-PbO(cr), in NaCl and $MgCl_2$ solutions with citrate as a function of experimental time produced in this study.

Figure 2. A plot showing experimental total lead concentrations in equilibrium with litharge, α-PbO(cr), in NaCl solutions with EDTA as a function of experimental time produced in this study.

Figure 3. A plot showing experimental total lead concentrations in equilibrium with litharge, α-PbO(cr), in $MgCl_2$ solutions with EDTA as a function of experimental time produced in this study.

Figure 4. A plot showing [log β_1^I + 12D] as a function of ionic strengths, where log β_1^I is a stability constant for $PbCitrate^-$ at a certain ionic strength.

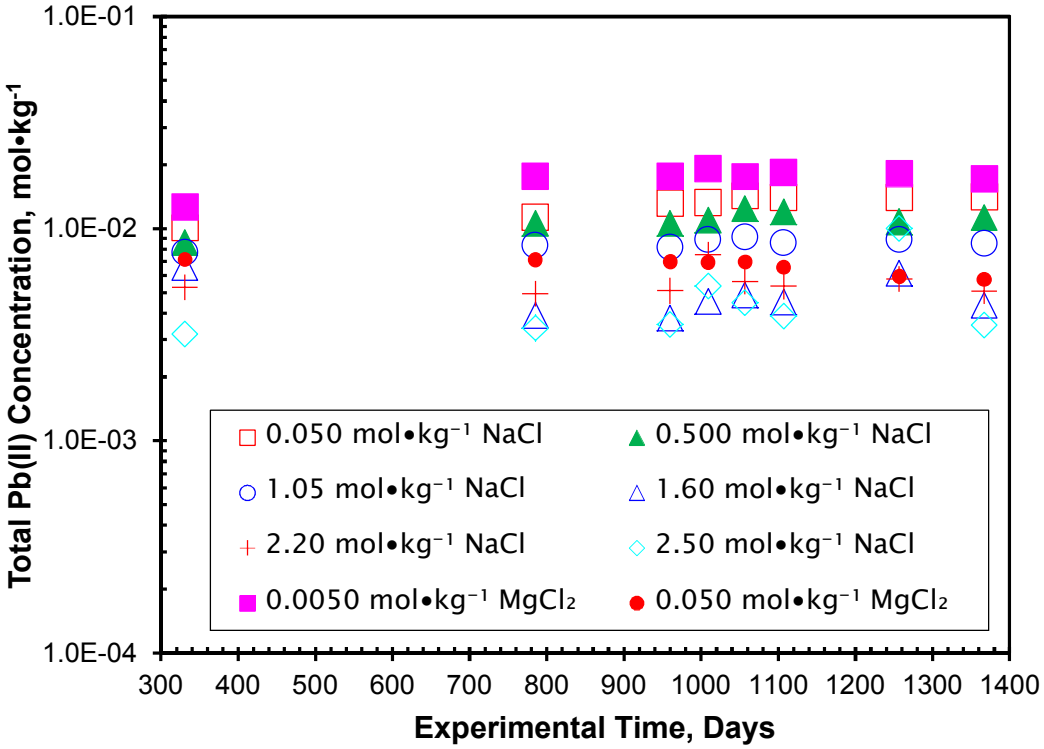

Figure 1. A plot showing experimental total lead concentrations in equilibrium with litharge, α-PbO(cr), in NaCl and $MgCl_2$ solutions with citrate as a function of experimental time produced in this study.

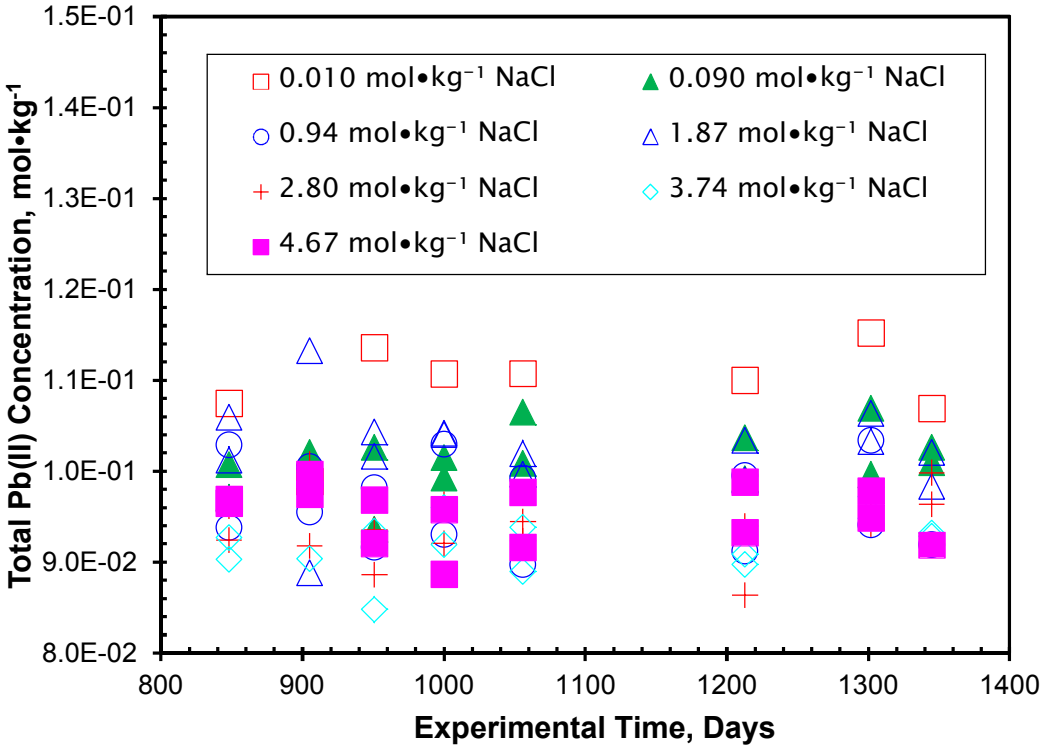

Figure 2. A plot showing experimental total lead concentrations in equilibrium with litharge, α-PbO(cr), in NaCl solutions with EDTA as a function of experimental time produced in this study.

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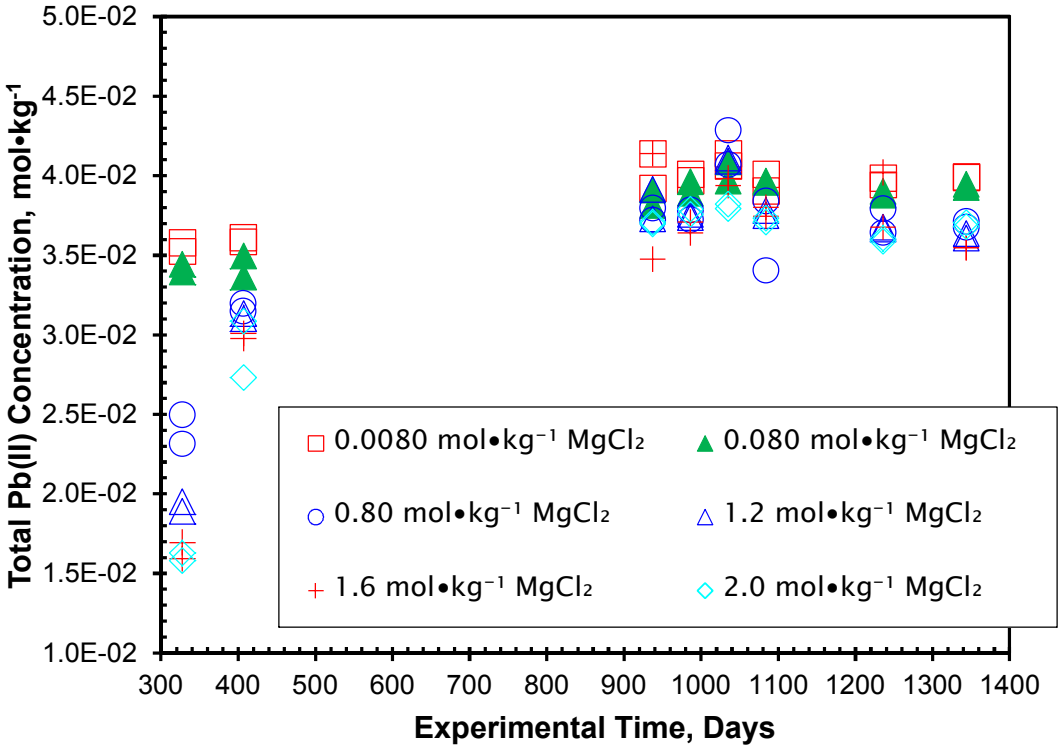

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513 Figure 3. A plot showing experimental total lead concentrations in equilibrium with
514 litharge, α-PbO(cr), in $MgCl_2$ solutions with EDTA as a function of experimental time
515 produced in this study.
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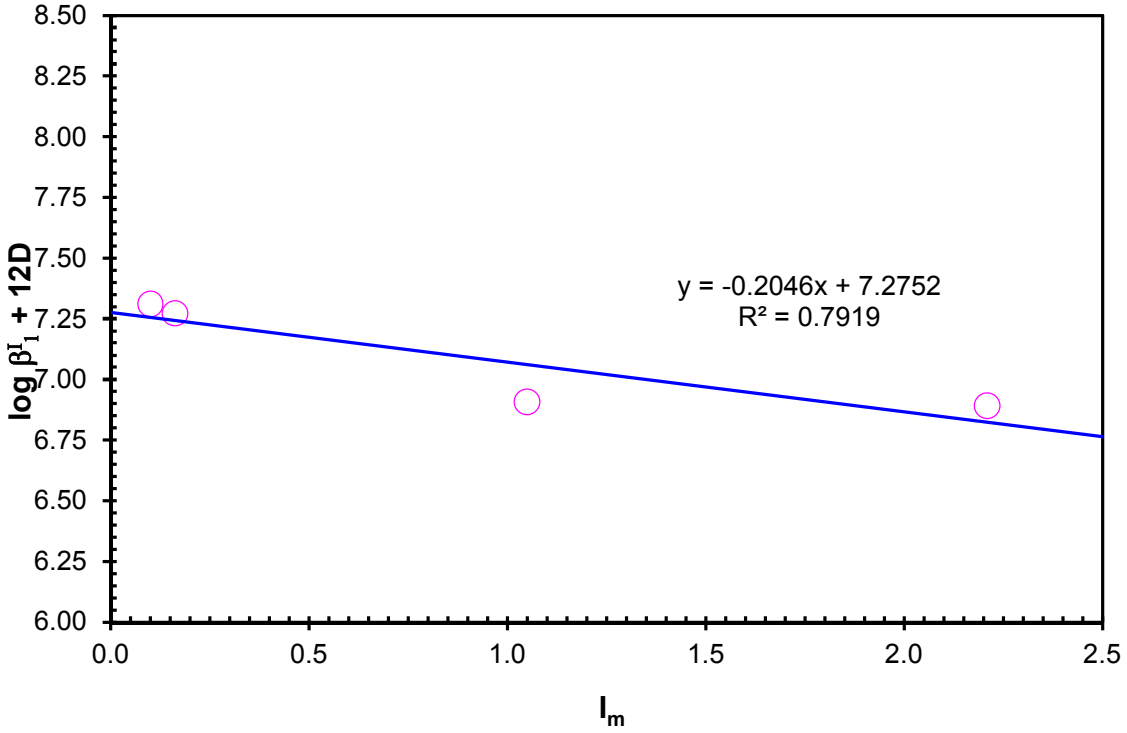

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521 Figure 4. A plot showing [log β_1^I + 12D] as a function of ionic strengths, where log β_1^I
522 is a stability constant for $PbCitrate^-$ at a certain ionic strength.
523