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Dissolved Concentration of Iron and Lead in GWB and ERDA-6 in the presence of FeS, PbS, and Organic Ligands

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LIST OF ACRONYMS AND ABBREVIATIONS

Acronym or Abbreviation	Definition
$EDTA^{4-}$	Ethylenediaminetetraacetate anion
ERDA-6	Energy Research and Development Administration (WIPP Well) 6 (synthetic brine representative of fluids in Castile-Fm. brine reservoirs)
FeS(s)	Iron sulfide solid
GWB	Generic Weep Brine (synthetic brine representative of intergranular Salado-Fm. brines at or near the stratigraphic horizon of the repository)
ICP-AES	Inductively Coupled Plasma - Atomic Emission Spectrophotometer
ICP-MS	Inductively Coupled Plasma - Mass Spectrophotometer
PbS(s)	Lead sulfide solid
pHr	Reading on the pH meter and electrode calibrated using commercial pH buffers of low ionic strength
ΣComponent, e.g., ΣFe	Total dissolved concentration of the component
SN	Scientific Notebook
TP	Test Plan
WIPP	Waste Isolation Pilot Plant

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INTRODUCTION

This document summarizes the experimental data that have been collected to date under Test Plan TP 14-05 Revision 0 (Brush and Xiong, 2014).

The following questions are the driver of the work described in the TP above and this document: Do the sulfide minerals of iron and lead resist against the aqueous complexation of the organic ligands with iron and lead? In other words, how far will the following two reactions proceed to the right? Would the WIPP brines have enough ΣFe and ΣPb in the aqueous phase to block the actinides from being solubilized by the organic ligands in the presence of sulfide mineral(s) of iron and lead?

$$FeS(s) + L^{n-} = FeL^{(2-n)} + S^{2-} \quad (1)$$

$$PbS(s) + L^{n-} = PbL^{(2-n)} + S^{2-} \quad (2)$$

, where L^{n-} stands for a fully deprotonated organic ligand, e.g., $EDTA^{4-}$, $Citrate^{3-}$, $Oxalate^{2-}$, $Acetate^{-}$, and FeS(s) and PbS(s) stands for chemical formulae of generic sulfide minerals of iron and lead.

The purpose of the experiments described in this document is to experimentally check how far Reactions (1) and (2) above can proceed to the right in the two WIPP-relevant brines, i.e., GWB (Krumhansl et al., 1991; Snider, 2003) and ERDA-6 (Popielak et al., 1983).

MATERIALS AND METHODS

FeS(s) was purchased from Alfa Aesar (99.98 %, LOT E03X029, No expiration). PbS(s) was purchased from Alfa Aesar (99.9 %, LOT C27T028, No expiration).

GWB was prepared according to the recipe in page 15 of Scientific Notebook (SN) WIPP-FePbS-1. The preparation steps were recorded in page 58 of WIPP-Solubility-41. ERDA-6 was prepared according to the recipe in page 17 of WIPP-FePbS-1. The preparation steps were recorded in page 70 of SN WIPP-Solubility-41. The recipes for both GWB and ERDA-6 represent the brine composition reported in Brush and Domski (2013a, 2013b).

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$Mg(OH)_2$(s), brucite, was purchased from Alfa Aesar (LOT E21T031, no expiration). The preparations of Mg-Phase-5 ($Mg_3Cl(OH)_5.4H_2O$(s)) and hydromagnesite5424 ($Mg_5(CO_3)_4(OH)_2.4H_2O$(s)) were recorded in SNs WIPP-Mineral Fragments-3 (p.44) and WIPP-Mineral Fragments-2 (p.87), respectively.

Experiments of FeS(s) and PbS(s) in GWB and EDRA-6 were prepared in triplicates, thus 12 reactors in total. All twelve reactors received 1.0 gram of brucite, 2.0 gram of Mg-Phase-5, and 2.0 gram of hydromagnesite5424, in addition to 2.0 gram of FeS(s) or PbS(s). Solution volume was 120 mL for all twelve reactors.

The pHr and the concentrations of iron and lead (ΣFe and ΣPb) were measured for the twelve reactors. pHr is reading on the pH meter and electrode calibrated using commercially available pH buffers of low ionic strength (SP 12-14 Revision 4). ΣFe was measured using ICP-MS[1] (SP 12-31 Revision 1), and ΣPb was measured using ICP-AES[2] (SP 12-9 Revision 5).

RESULTS

XRD scan identified the purchased FeS(s) and PbS(s) to be pyrrhotite (Excerpt 1) and galena (Excerpt 2), respectively, both with minor amounts of impurities.

Results are summarized in Table 1 and Table 2. The pHr is well buffered at the values of WIPP interests (8.5 to 9.0). Detectable ΣFe and ΣPb exist in the aqueous phases, which could block the actinides from being solubilized by the organic ligands. The results indicated that ΣFe could be lower than ΣPb in both GWB and ERDA-6.

Detailed thermodynamic interpretation will be performed after more experimental data are collected with better accuracy and precision.

FUTURE WORK

- Complete ΣFe measurement using ICP-MS

[1] Inductively Coupled Plasma - Mass Spectroscopy
[2] Inductively Coupled Plasma - Atomic Emission Spectroscopy

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- Complete XRD for the final solids
- Complete other tasks in TP 14-05 Revision 0

REFERENCES

Brush, L. H. and Domski, P. S., 2013a. Calculation of organic-ligand concentrations for the WIPP CRA-2014 PA. Sandia National Laboratories. ERMS 559005.

Brush, L. H. and Domski, P. S., 2013b. Prediction of baseline actinide solubilities for the WIPP CRA-2014 PA. Sandia National Laboratories. ERMS 559138.

Brush, L. H., and Y. Xiong, 2014. Test Plan for the Experimental Determination of the Solubilities of Iron and Lead in the presence of Dissolved Hydrogen Sulfide Species. Test Plan (TP) 14-05, Revision 0. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A., ERMS 562861.

Krumhansl, J.L., K.M. Kimball, and C.L. Stein. 1991. *Intergranular Fluid Compositions from the Waste Isolation Pilot Plant (WIPP), Southeastern New Mexico*. SAND90-0584. Albuquerque, NM: Sandia National Laboratories.

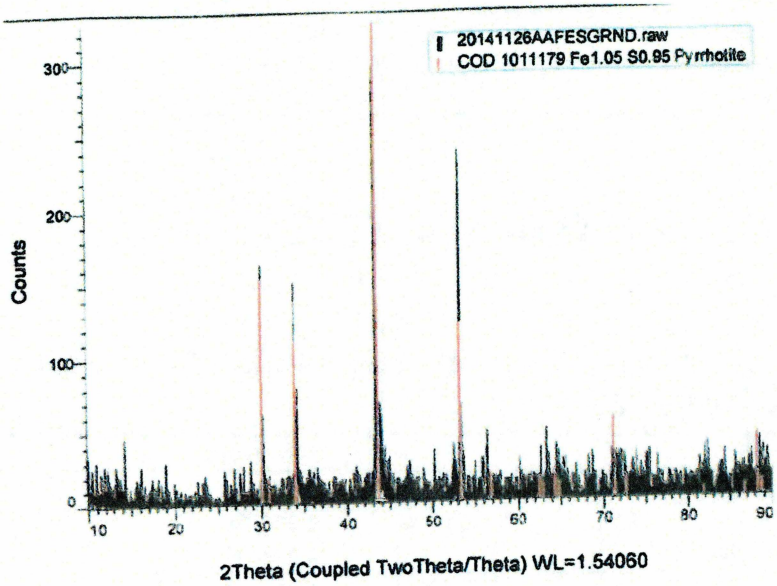
Popielak, R.S., R.L. Beauheim, S.R. Black, W.E. Coons, C.T. Ellingson, and R.L. Olsen, 1983. *Brine Reservoirs in the Castile Formation, Waste Isolation Pilot Plant (WIPP) Project, Southeastern New Mexico*. TME 3153. Carlsbad, NM, U.S. Department of Energy WIPP Project Office.

Snider, A.C. 2003. "Verification of the Definition of Generic Weep Brine and the Development of a Recipe for This Brine." Analysis report, April 8, 2003. Carlsbad, NM: Sandia National Laboratories. ERMS 527505.

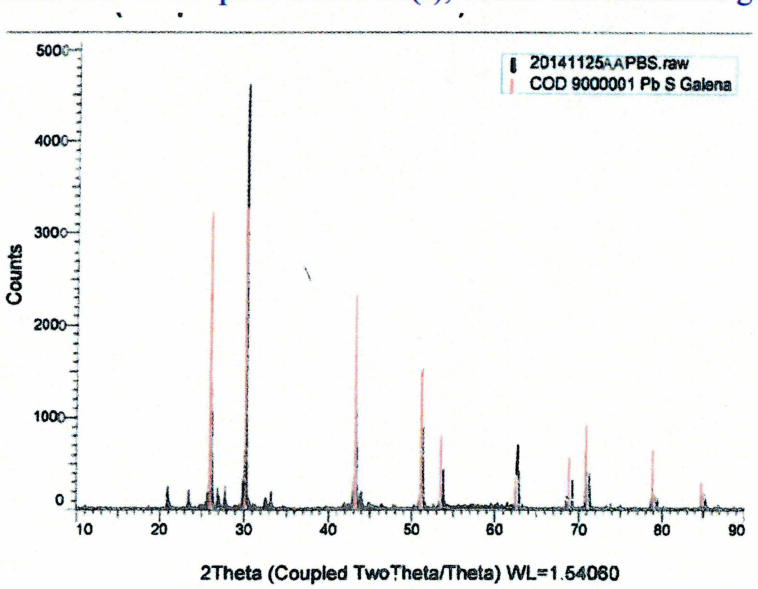
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INSERTS: TABLES AND FIGURES

Excerpt 1. XRD scan of the purchased FeS(s), which was identified pyrrhotite (WIPP-FePbS-1, page 11).



Excerpt 2. XRD scan of the purchased PbS(s), which was identified galena (WIPP-FePbS-1, page 13).



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106 Table 1. Dissolved concentration of Fe (ΣFe) and pHr measured from GWB and ERDA-6 in the presence
107 of pyrrhotite and organic ligands of WIPP interests.

Brine	pHr	ΣFe, mol/L
GWB	8.501	1.73E-06*
	8.480	1.58E-06*
	8.483	9.26E-06
ERDA-6	8.877	1.62E-06*
	8.835	1.75E-06*
	8.828	1.67E-06*

108 * Flagged by Lab: Data source is the Scientific Notebook (SN) WIPP-FePbS-1, pages 32 and 37.
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110 Table 2. Dissolved concentration of Pb (ΣPb) and pHr measured from GWB and ERDA-6 in the presence
111 of galena and organic ligands of WIPP interests.

Brine	pHr	ΣPb, mol/L
GWB	8.455	5.57E-04
	8.478	5.35E-04
	8.488	5.32E-04
ERDA-6	8.837	1.85E-04
	8.842	1.79E-04
	8.823	1.91E-04

112 Data sources is the SN WIPP-FePbS-1, pages 32 and 36.
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