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Sandia National Laboratories

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Albuquerque, New Mexico 87185-0101
Livermore, California 94551-0969

date: January 19, 2023

to: SNL WIPP[1] Records Center, SNL WIPP Geochemistry
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subject: Reproduction of log K values for $NaPb_2(CO_3)_2(OH)(s)$ (Abellaite) and $Pb(CO_3)_3^{-4}$, and Pitzer interaction parameter for species pair of Pb^{+2}/H^+

I. Drivers

The log K values and Pitzer interaction parameters (PIPs) in this memorandum were derived in Jang (2022, 2023). The derivations therein were performed using (i) EQ3/6 Version 8.0a (Wolery and Jarek, 2003) (unofficial version), and (ii) Data0.fm1 (Xiong, 2011). Additional softwares used in the derivations are listed in Section III.

This memo details reproduction of the log K values and PIP values of Jang (2022, 2023) using (i) EQ3/6 Version 8.0a *officially installed* on a designated run control coordinator's computer, and (ii) Data0.fm1 *stored in and downloaded from* the CVS repository. This reproduction also uses the same additional softwares in Section III.

Through this memorandum, SNL WIPP Geochemistry will elucidate that (i) the calculation results of *unofficially vs. officially* installed EQ3/6 Version 8.0a *cannot* be different from each other, (ii) the observed differences, if any, *should* be interpreted *from a technical viewpoint*, and (iii) *such observations* of the differences are *useful in improving/strengthening* the technical integrity of SNL WIPP Geochemistry.

[1] SNL WIPP: Sandia National Laboratories at the Waste Isolation Pilot Plant (WIPP). WIPP is located ~26 miles southeast of Carlsbad, New Mexico, U.S.A., built 2,150 feet underground in the Salado Formation, a thick Permian evaporite deposit in the northern Delaware Basin.

WIPP:4.4.2.2.1:PA:QA-L:558083

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II. Roles of Authors

Paul Domski: The designated run control coordinator. P. Domski maintains the qualified versions of SNL WIPP TDB[2] under SNL WIPP QA,[3] which is Data0.fm1 (Xiong, 2011). Domski completed documentation per NP 19-1 Revision 19 to fulfil the requirement of an *official installation* of EQ3/6 Version 8.0a. To run the input files J. Jang provides in Sections V.1 and VI.1, the qualified SNL WIPP TDB requires modifications using the text blocks J. Jang provides in Sections V.2 and VI.2.

Kevins Rhino: K. Rhino organized and completed duplicate runs. He contributed to initiate and handle the review processes under SNL WIPP QA.

Jay Jang: Originator of (i) the EQ3NR input files (*.3i) (Sections V.1 and VI.1), (ii) text blocks to be copied into the qualified SNL WIPP TDB (Sections V.2 and VI.2), and (iii) Python scripts to drive the derivation processes by selectively using modules in EQ3CodeModule.py (Kirchner, 2012) (Sections V.3 and VI.3). J. Jang will provide technical explanations in Section "VIII. Discussion" if differences occur between the results of J. Jang and P. Domski.

III. List of Software

- EQ3/6 Version 8.0a (Wolery and Jarek, 2003)
- EQ3CodeModule.py (Kirchner, 2012)
- Enthought Python Installation Package, epd-7.2-2

IV. Fitting Methodology

This memo reproduces the derivation of the following thermodynamic quantities: (i) the log K values for $NaPb_2(CO_3)_2(OH)$(s) (abellaite) and an aqueous complex $Pb(CO_3)_3^{-4}$ in Jang (2022), and (ii) the Pitzer interaction parameter (θ) for ion pair of Pb^{+2}/H^+ in Jang (2023). The derivations in Jang (2022, 2023) were performed under AP-190, Revision 0 (Jang et al., 2021).

The general steps are as follows:

1. The qualified SNL WIPP TDB, data0.fmt.R2, a.k.a, DATA0.FM1 (Xiong, 2011) is copied to an interim database and renamed DATA0.PB9. DATA0.PB9 was then updated with the text blocks J. Jang provided in Sections V.2 and VI.2. The text blocks contain the parameters that are being derived for the task at hand, and supporting parameters for the chemical system.

[2] TDB: Thermodynamic Database
[3] QA: Quality Assurance

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2. The Python scripts specific to the problems and which calls the parameter optimization modules in "EQ3CodeModule.py" (Kirchner, 2012) are executed and the specified parameters are derived. The parameters were derived by use of the inverse method(s) implemented in the Python Module "EQ3CodeModule.py" (Kirchner, 2012). The Python scripts are written and provided by J. Jang in Sections V.3 and VI.3.

3. The output of the Python script, a text file named "results.txt", which consists of the derived parameter(s) and the calculated residual at each iteration, is evaluated in Excel to determine the parameter(s) which corresponds to the smallest value of the residual. The residual is defined in Equation (1).

$$\text{Residual} = \Sigma_{i=1}^{n}[\log \Sigma Pb_{meas,\,i} - \log \Sigma Pb_{calc,\,i}]^2 \quad (1)$$

where n is the number of measurements being simulated; $\Sigma Pb_{meas,\,i}$ and $\Sigma Pb_{calc,\,i}$ correspond to the measured and calculated total dissolved concentrations of Pb, respectively, at a given experimental condition, i.

In Section V, the log K values for $NaPb_2(CO_3)_2(OH)(s)$ (abellaite) and the aqueous complex $Pb(CO_3)_3^{-4}$ were fit simultaneously to the experimental data presented in Jang (2022). In Section VI, the Pitzer interaction parameter (θ) for the Pb^{+2}/H^+ pair was fit to the experimental data presented in Jang (2023). Justification for the use of the selected parameters can be found in Jang (2022, 2023). See Jang (2022, 2023) for the specifics regarding the experimental conditions and measurements used for this fitting procedure. The parameters of Jang (2022, 2023) that are reproduced by the run control coordinator are documented in this memorandum to SNL WIPP Records Center.

V. Log K values in Jang (2022)

The best fit values obtained by J. Jang are: log K(Abellaite) = 2.5988.. ≡ 2.59 and log K($Pb(CO_3)_3^{-4}$) = 27.1916.. ≡ 27.19 with a residual = 0.04133.. ≡ 0.041 (Jang, 2022). Underlined numbers are truncated, not rounded off, to keep the traceability to the output file of the derivation process. The residual is computed using Equation (1).

V.1. EQ3NR Input files

Following six *.3i files are archived in "abell_3i.zip".[4]

- Carb01.3i through Carb06.3i (6 files)

[4] Stored in "cvs/CVSLIB/WIPP_EXTERNAL/verif_abellaite"

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V.2. Text Blocks to be copied and pasted into Data0.PB9

Add the following into the "elements" section of Data0.PB9. This block can be placed in any location within the "elements" section.

```
Pb          207.19000
```

Add the following into the "basis species" section of Data0.PB9. Be cautious to add this block in the same order of "elements" section. If not the database and the Python script will not work.

```
Pb++
     charge  =   2.0
****
     1 element(s):
      1.0000 Pb
```

Add the following into the "aqueous species" section of Data0.PB9. These blocks can be placed in any location within the "aqueous species" section.

```
+------------------------------------------------------------
PbHCO3+                    PbHCO3+
     charge  =   1.0
****
     4 element(s):
      1.0000 C              1.0000 Pb             3.0000 O
      1.0000 H
****
     3 species in aqueous dissociation reaction:
    -1.0000  PbHCO3+                  1.0000  Pb++
     1.0000  HCO3-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data    -1.8600    No_Data    No_Data
        No_Data    No_Data    No_Data    No_Data
* Source: Pb+2 + HCO3- = PbHCO3+; 1.86 +/- 0.1 <- Powell et al. (2009), p.2429,
Provisional
+------------------------------------------------------------
PbCO3(aq)                  PbCO3(0)
     charge  =   0.0
****
     3 element(s):
      1.0000 C              1.0000 Pb             3.0000 O
****
     4 species in aqueous dissociation reaction:
    -1.0000  PbCO3(aq)               -1.0000  H+
     1.0000  Pb++                     1.0000  HCO3-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data     3.88     No_Data    No_Data
        No_Data    No_Data    No_Data    No_Data
* Source: 3.88 = 10.33 + (-6.45 +/- 0.72 <- Powell et al. (2009), p.2429),
indicative
```


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```
* Pb+2 + CO3-2 = PbCO3(aq); 6.45 +/- 0.72 <- Powell et al. (2009), p.2429),
indicative
+------------------------------------------------------------------
Pb(CO3)2-2                Pb(CO3)2--
     charge  =  -2.0
****
     3 element(s):
      2.0000 C              1.0000 Pb             6.0000 O
****
     4 species in aqueous dissociation reaction:
    -1.0000  Pb(CO3)2-2              -2.0000  H+
     1.0000  Pb++                     2.0000  HCO3-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data    10.53    No_Data   No_Data
        No_Data   No_Data   No_Data   No_Data
* Source: 10.53 = 2*10.33 + (-10.13 +/- 0.24 <- Powell et al. (2009), p.2429),
provisional
* Pb+2 + 2CO3-2 = Pb(CO3)2-2; 10.13 +/- 0.24 <- Powell et al. (2009), p.2429),
provisional
+------------------------------------------------------------------
Pb(CO3)OH-                Pb(CO3)OH(-1)
     charge  =  -1.0
****
     4 element(s):
      1.0000 C              1.0000 Pb             4.0000 O
      1.0000 H
****
     5 species in aqueous dissociation reaction:
    -1.0000  Pb(CO3)OH-              -2.0000  H+
     1.0000  Pb++                     1.0000  HCO3-
     1.0000  H2O
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data    13.42    No_Data   No_Data
        No_Data   No_Data   No_Data   No_Data
* Source: 13.42 = 13.99 + 10.33 + (-10.9 +/- 0.2 <- Powell et al. (2009), p.2441),
Statistically predicted
* Pb+2 + CO3-2 + OH- = Pb(CO3)OH-; 10.9 +/- 0.2 <- Powell et al. (2009), p.2441),
Statistically predicted
+------------------------------------------------------------------
Pb(CO3)3-4                Pb(CO3)3(-4)
     charge  =  -4.0
****
     3 element(s):
      3.0000 C              1.0000 Pb             9.0000 O
****
     4 species in aqueous dissociation reaction:
    -1.0000  Pb(CO3)3-4              -3.0000  H+
     1.0000  Pb++                     3.0000  HCO3-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data    27.19    No_Data   No_Data
        No_Data   No_Data   No_Data   No_Data
* Source: Jang (2022)
+------------------------------------------------------------------
```

Add the following into the "solids" section of Data0.PB9. This block can be placed in any location within this section.

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```
+------------------------------------------------------------------

NaPb2(CO3)2(OH)(s)          Abellaite
     V0PrTr   =    000.000 cm**3/mol [source:          ]
****
     5 element(s):
      2.0000 Pb              2.0000 C              7.0000 O
      1.0000 Na              1.0000 H
****
     6 species in aqueous dissociation reaction:
    -1.0000  NaPb2(CO3)2(OH)(s)          -3.0000  H+
     2.0000  Pb++                         2.0000  HCO3-
     1.0000  Na+                          1.0000  H2O
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data      2.59     No_Data    No_Data
        No_Data    No_Data    No_Data    No_Data
* Source: Jang (2022)
+------------------------------------------------------------------
```

V.3. Python Scripts driving the Estimation Process

Below is the copy of the Python script J. Jang used in his derivation. The run control coordinator uses an interim database named Data0.PB9 (Section IV), therefore, the "fmj" in red font below needs to be replaced with "pb9" (letter case independent).

```
#!/opt/epd-7.2-2/bin/python
from EQ3CodeModule import *
import numpy
#Un-comment SetVerbose() to see step by step results
SetVerbose()

#name the location of the code and database.
if os.name<>"posix":
    EQ3='c:\\Eq3_6v8.0a\\RunEQ3_eqpt.bat'
    DB="c:\\eq3_6v8.0a\\db\\data0.fmj"
else:
    EQ3="./run_all.sh fmj"
    DB="./data0.fmj"

#For Log-K values, you must specify one or more [temperature,value] pairs
LogK_Data1=[[25,2.77]]
LogK_Data2=[[25,27.00]]
#Format: [[[species set],[identifier(s)],value | [temp,value]],[...]]
#To do one-by-one
DB_values=[
           [["NaPb2(CO3)2(OH)(s)","Abellaite"],"LogK",LogK_Data1],    \
           [["Pb(CO3)3-4","Pb(CO3)3(-4)"],"LogK",LogK_Data2]    \
           ]
#Create the DB_Data object, which stores information for changing the database
DB_params=DB_Data(DB_values)

#Create a DataSet object listing the input and output files. The output files
#are assumed to have the same names but with a .3o extension. Because the Log K
#values in the input files will not be changed, Update must be set to False.
```


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```
DS1 = DataSet("Carb%2.2i", 6, ["NaPb2(CO3)2(OH)(s)"], Stoich={"Pb++":1},
Update=False)

Solve = Estimate([DS1],EQ3,DB,"Sensible",DB_params)

print Solve.Minimize()
```

VI. PIP in Jang (2023)

J. Jang obtained the PIP for the cation-cation pair of Pb^{+2}/H^{+}, denoted $\theta(Pb^{+2}/H^{+})$ hereafter, to be 0.34350.. ≡ 0.343 using the information below (Jang, 2023) with Residual = 1.14565.. ≡ 1.145. Residual is defined in Equation (1) in page 3. Underlined numbers are truncated, not rounded off, to keep the traceability to the output file of the derivation process.

VI.1. EQ3NR Input files

The following twenty five *.3i files are archived in "anglesite_3i.zip".[5]

- h2so4_01.3i through h2so4_09.3i (9 files)
- kolt_01.3i through kolt_10.3i (10 files)
- pbso4_01.3i through pbso4_06.3i (6 files)

VI.2. Text Blocks to be copied and pasted into Data0.PB9

Add the following into the "ca combinations: beta(n)(ca) and Cphi(ca) [optional: alpha(n)(ca)]" section of Data0.PB9. These blocks can be placed in any location within this section.

```
+--------------------------------------------------------------------
H+                          PbCl3-
  alpha(1) = 2.0
  alpha(2) = 12.0
  beta(0)
    a1 = 0.1077
    a2 = 0.
    a3 = 0.
    a4 = 0.
  beta(1)
    a1 = 0.0425
    a2 = 0.
    a3 = 0.
    a4 = 0.
  beta(2)
    a1 = 0.00
    a2 = 0.
    a3 = 0.
    a4 = 0.
  Cphi:
    a1 = -0.0035
    a2 = 0.
```

[5] Stored in "cvs/CVSLIB/WIPP_EXTERNAL/verif_abellaite"

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```
    a3 = 0.
    a4 = 0.0
* Source: Jang (2021, Revisit), Jang et al. (2021, GEOC-21-11)
+------------------------------------------------------------
Na+                         PbCl3-
  alpha(1) = 2.0
  alpha(2) = 12.0
  beta(0)
    a1 = -0.10025
    a2 = 0.
    a3 = 0.
    a4 = 0.
  beta(1)
    a1 = 0.
    a2 = 0.
    a3 = 0.
    a4 = 0.
  beta(2)
    a1 = 0.0
    a2 = 0.
    a3 = 0.
    a4 = 0.
  Cphi:
    a1 = 0.0
    a2 = 0.
    a3 = 0.
    a4 = 0.
* Source: Jang et al. (2021, GEOC-21-11)
+------------------------------------------------------------
```

Add the following into the "cc' and aa' combinations: theta(cc') and theta(aa')" section of Data0.PB9. This block can be placed in any location within this section.

```
+------------------------------------------------------------
Pb++                        H+
  theta:
    a1 = 0.343
    a2 = 0.
    a3 = 0.
    a4 = 0.
* Source: Jang (2023)
+------------------------------------------------------------
```

Add the following into the "elements" section of Data0.PB9

```
Pb        207.19000
```

Add the following into the "basis species" section of Data0.PB9. Be cautious to add this block in the same order of "elements" section. If not the database and the Python script will not work.

```
Pb++
     charge   =    2.0
****
     1 element(s):
      1.0000 Pb
```

Add the following into the "aqueous species" section of Data0.PB9

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```
+---------------------------------------------------------------
PbCl+                    PbCl(+)
     charge  =   1.0
****
     2 element(s):
      1.0000 Pb             1.0000 Cl
****
     3 species in aqueous dissociation reaction:
    -1.0000  PbCl+                        1.0000  Pb++
     1.0000  Cl-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data   -1.4800   No_Data   No_Data
        No_Data   No_Data   No_Data   No_Data
* Source: Jang (2021, Revisit), Millero and Byrne (1984)
+---------------------------------------------------------------
PbCl2(aq)                PbCl2(0)
     charge  =   0.0
****
     2 element(s):
      1.0000 Pb             2.0000 Cl
****
     3 species in aqueous dissociation reaction:
    -1.0000  PbCl2(aq)                    1.0000  Pb++
     2.0000  Cl-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data   -2.0300   No_Data   No_Data
        No_Data   No_Data   No_Data   No_Data
* Source: Jang (2021, Revisit), Millero and Byrne (1984)
+---------------------------------------------------------------
PbCl3-                   PbCl3(-)
     charge  =  -1.0
****
     2 element(s):
      1.0000 Pb             3.0000 Cl
****
     3 species in aqueous dissociation reaction:
    -1.0000  PbCl3-                       1.0000  Pb++
     3.0000  Cl-
*
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data   -1.8600  No_Data   No_Data
        No_Data   No_Data   No_Data   No_Data
* Source: Jang (2021, Revisit), Millero and Byrne (1984)
+---------------------------------------------------------------
```

Add the following into the "solids" section of Data0.PB9

```
+---------------------------------------------------------------------
PbSO4(s)                 PbSO4(c)
     V0PrTr  =   000.000 cm**3/mol [source:         ]
****
     3 element(s):
      1.0000 Pb             1.0000 S            4.0000 O
****
     3 species in aqueous dissociation reaction:
    -1.0000  PbSO4(s)                     1.0000  Pb++
     1.0000  SO4--
*
```


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```
**** logK grid [0-25-60-100C @1.0132bar; 150-200-250-300C @Psat-H2O]:
        No_Data    -7.77    No_Data    No_Data
        No_Data   No_Data   No_Data    No_Data
* Source: Jang (2023). In agreement with Powell et al. (2009)
+--------------------------------------------------------------------
```

VI.3. Python Scripts driving the Estimation Process

Below is the copy of the Python script J. Jang used in his derivation. The run control coordinator uses the modified SNL WIPP TDB named Data0.PB9 (Section IV), therefore, the "fmj" in red font below needs to be replaced with "pb9" (letter case independent).

```
#!/opt/epd-7.2-2/bin/python
from EQ3CodeModule import *
import numpy
#Un-comment SetVerbose() to see step by step results
SetVerbose()

#name the location of the code and database.
if os.name<>"posix":
    EQ3='c:\\Eq3_6v8.0a\\RunEQ3_eqpt.bat'
    DB="c:\\eq3_6v8.0a\\db\\data0.fmj"
else:
    EQ3="./run_all.sh fmj"
    DB="./data0.fmj"

DB_values=[
           [["Pb++","H+"],["theta","a1"], 0.0]  \
           ]
#Create the DB_Data object, which stores information for changing the database
DB_params=DB_Data(DB_values)

DS1 = DataSet("h2so4_%2.2i", 9, ["PbSO4(s)"], Stoich={"Pb++":1}, Update=False)
DS2 = DataSet("kolt_%2.2i", 10, ["PbSO4(s)"], Stoich={"Pb++":1}, Update=False)
DS3 = DataSet("pbso4_%2.2i", 6, ["PbSO4(s)"], Stoich={"Pb++":1}, Update=False)

Solve = Estimate([DS1, DS2, DS3],EQ3,DB,"Sensible",DB_params)

print Solve.Minimize()
```

VII. Summary

Paul Domski, as the run control coordinator, reproduced the log K values and PIP values in Jang (2022, 2023) using the components provided by J. Jang in Sections V and VI using EQ3/6 Version 8.0a *officially installed* per the requirements of NP 19-1 Revision 19. In Table VII-1, the derivations in Jang (2022, 2023) are compared with the results of the run control coordinator.

The two log K values showed agreement down to the second digit below the decimal point (Table VII-1). The residual also showed agreement down to the third digit below the decimal point. SNL WIPP Geochemistry will accept the log K values as in Jang (2022).

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The $\theta(Pb^{+2}/H^{+})$ showed difference at the fourth digit below the decimal point (Table VII-1). The residual is different at the second digit below the decimal point. J. Jang provided an explanation on the difference in Section "VIII. Discussion".

Table VII-1. Reproduction of log K values and PIP values in Jang (2022, 2023) by P. Domski. Underlined numbers are truncated.

	Jang (2022)				
	I. Before this memo	II. After this memo[A]	III. Percentage (%) between I and IV	IV. This memo	Note (if any)
Log K of $Pb(CO_3)_3^{-4}$	27.1916..	*27.19*	0.0004	27.19170	See "*Results_Pb_param.xlsx*" tab "Abellaite"
Log K of Abellaite	2.5988..	*2.59*	0.004	2.598902	
Residual:	0.04133..	0.041	0.007	0.041327	
	Jang (2023)				
	I. Before this memo	II. After this memo[A]	III. Percentage (%) between I and IV	IV. This memo	Note (if any)
$\theta(Pb^{+2}/H^{+})$	0.34350..	*0.343*	0.07	0.34375	See "*Results_Pb_param.xlsx*" tab "Anglesite"
Residual:	1.14565..	1.174	3	1.17450	

[A] *Italic numbers* in these columns are the values to be used in CRA-related calculations.

VIII. Discussion

The difference of $\theta(Pb^{+2}/H^{+})$ (Table VII-1) originates from the difference in the databases used by J. Jang and P. Domski. J. Jang used a database containing Pb-hydrolysis reactions and their log K values adopted from Powell et al. (2009) with a purpose of gauging the influence of Pb-hydrolysis on the chemical affinity of Pb^{+2} - SO_4^{-2} (Jang, 2023) and Pb^{+2} - CO_3^{-2} (Jang, 2022). The hypothesis being tested is:

> "The influence of Pb-hydrolysis would be minimal and non-identifiable in both carbonate and sulfate systems."

When J. Jang removed the Pb-hydrolysis reactions and their log K values from his database, J. Jang exactly[6] reproduced Domski's results using the EQ3/6 Version 8.0a *unofficially installed* on his computer, proving that there is no difference between *unofficially vs. officially installed* EQ3/6 Version 8.0a.

In case of the log K values for $NaPb_2(CO_3)_2(OH)(s)$ (abellaite) and $Pb(CO_3)_3^{-4}$ (Jang, 2022), the differences are found at the digits to be truncated (i.e., underlined numbers in Table VII-1). As a result of the truncation, the influence of the Pb-hydrolysis reaction does not impact in the derivation of the log

[6] To the last digit of the numbers

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K values of $NaPb_2(CO_3)_2(OH)(s)$ (abellaite) and the aqueous complex $Pb(CO_3)_3^{-4}$. The difference of $\theta(Pb^{+2}/H^+)$ also did not impact when the numbers are truncated as shown in Table VII-1.

The observations above indicate that the speciation of Pb^{+2} in the presence of CO_3^{-2} and SO_4^{-2} can be dealt with independently of the Pb-hydrolysis model of Powell et al. (2009) under the chemical conditions employed in Jang (2022, 2023). The value of this memo 0.343 will be included in Jang (2023) because SNL WIPP Geochemistry is in the process of securing sets of measurements amended with uncertainty to verify the accuracy of Pb-hydrolysis model in Powell et al. (2009). Thus, SNL WIPP Geochemistry is not in the position to accept or reject the Pb-hydrolysis model of Powell et al. (2009) for now.

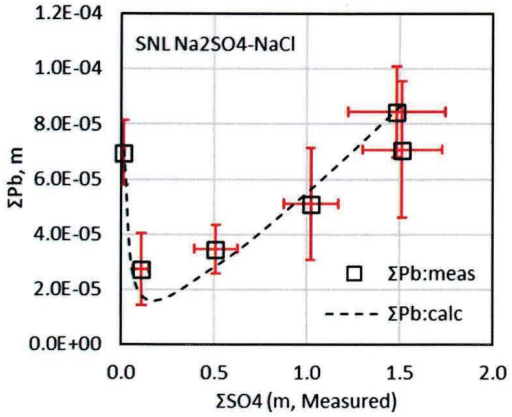
Therefore, J. Jang updated Jang (2023) in accordance with Table VII-1. Note that the difference of $\theta(Pb^{+2}/H^+)$ was confined within 4% difference in the predicted ΣPb, which would be considered non-identifiable difference compared to the uncertainty of the measurements (Figure VIII-1), until SNL WIPP Geochemistry secure additional measurements of better precision.

Given the uncertainty associated with the measurements reported in Jang (2022, 2023) and the accuracy of Pb-Hydrolysis model of Powell et al. (2009), the hypothesis above can be accepted. SNL WIPP Geochemistry will evaluate the accuracy of the Pb-hydrolysis model of Powell et al. (2009) in near future.

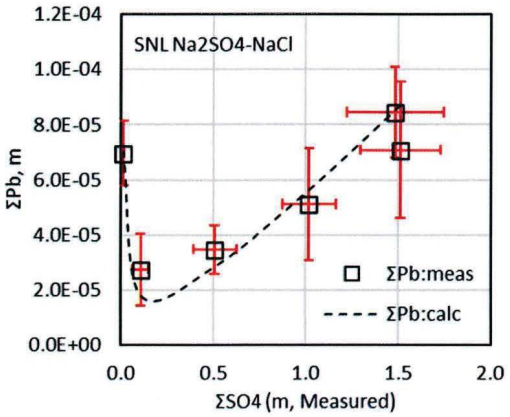
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Figure VIII-1. A-1,2,3. Prediction of ΣPb with $\theta(Pb^{+2}/H^{+}) = 0.3435$ as in Figure VIII-5 of Jang (2023). B-1,2,3. Prediction of ΣPb with $\theta(Pb^{+2}/H^{+}) = 0.3437$ compared with the same measurements as in A-1,2,3. The predicted ΣPb in both A and B are identical within the reported uncertainty.

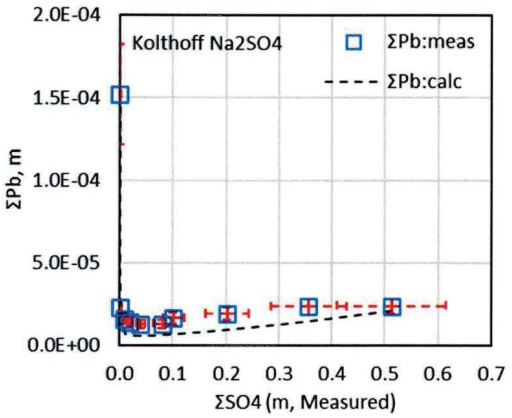
A-1



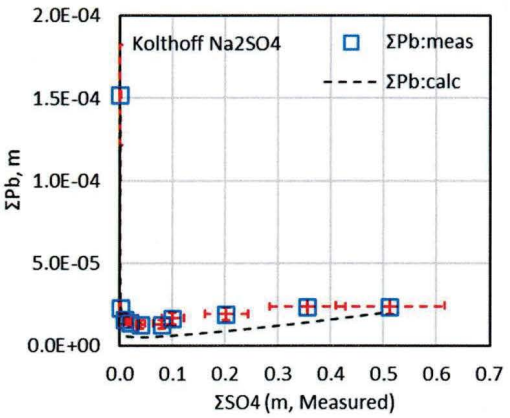
B-1



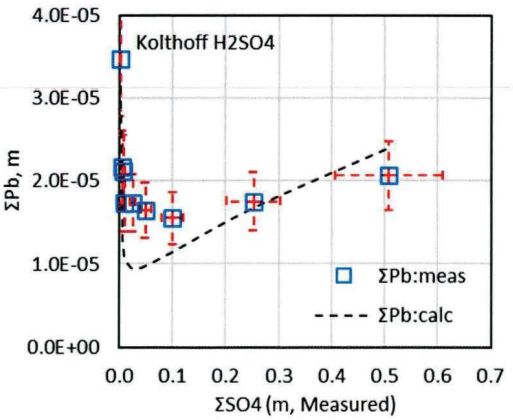
A-2



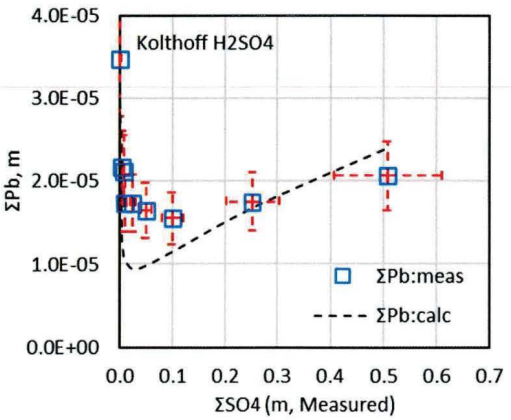
B-2



A-3



B-3



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IX. Acknowledgment

Technical review was conducted by Mahawa-Essa Mabossani "Romeo" Akara. During the technical review, Akara exactly reproduced the run control coordinator's results using *unofficially installed* EQ3/6 Version 8.0a obtained from the path in Section III. QA review by Shelly Nielsen; Management review by Chammi Miller. Comments and resolutions are archived under SNL WIPP QA.

X. REFERENCES

Jang, J., 2021. Revisit to the Pitzer Geochemical Thermodynamic Model for the $PbCl_2$(s) - HCl - H_2O System. Memo to SNL WIPP Records Center and SNL WIPP Geochemistry, May 25, 2021. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A. ERMS 575309.

Jang, J., P. Hora, L. Kirkes, C. Miller, and L. Zhang, 2021. Analysis Plan for Analysis Reports documenting Solubility and Complexation of Iron, Lead, Magnesium, Neodymium, and Boron in WIPP-Relevant Brines under Test Plans 00-07, 06-03, 08-02, 10-01, 12-02, 14-03, 14-05, 16-02, 19-01, and 20-01. Analysis Plan AP-190 Revision 0. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A. ERMS 575624.

Jang, J., P. Hora, L. Kirkes, C. Miller, and L. Zhang, 2021. Analysis Report documenting Solubility and Complexation of Iron, Lead, Magnesium, Neodymium, and Boron in the WIPP-Relevant Brines under TPs 06-03, 08-02, 12-02, 14-03, 14-05, 16-02, 19-01, and 20-01. Revision 0. GEOC-21-11. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A. ERMS 576381. SAND2022-13723R.

Jang, J., 2022. The $PbCO_3$(s) - $NaHCO_3$ - Na_2CO_3 - H_2O Experiment (Revision 0): Addendum to Xiong (2015a,b). Memo to SNL WIPP Records Center, SNL WIPP Geochemistry. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A. ERMS pending. Submittal to SNL WIPP Records made on 1/12/2023.

Jang, J., 2023. The $PbSO_4$(s) - Na_2SO_4 - NaCl - H_2O Experiment (Revision 0). Memo to SNL WIPP Records Center, SNL WIPP Geochemistry. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A. ERMS pending. In QA Review as of 1/19/2023.

Kirchner, T. B., 2012. User's Manual for The EQ3CodeModule Version 1.00. Sandia National Laboratories, Carlsbad, New Mexico, U.S.A. ERMS 557360.

Millero, F. J. and Byrne, R. H., 1984. Use of Pitzer's equations to determine the media effect on the formation of lead chloro complexes. *Geochimica et Cosmochimica Acta* 48, 1145-1150..

Powell, K. J., P. L. Brown, R. H. Byrne, T. Gajda, G. Hefter, A.-K. Leuz, S. Sjoberg, and H. Wanner. 2009. Chemical speciation of environmentally significant metals with inorganic ligands. Part 3: The Pb^{2+} + OH^-, Cl^-, CO_3^{2-}, SO_4^{2-}, and PO_4^{3-} systems (IUPAC Technical Report), Pure Appl. Chem., Vol. 81, No. 12, 2425-2476..

Information Only

543 Wolery, T., and R. L. Jarek, 2003. Software User's Manual, EQ3/6, Version 8.0, 01/01/2003. ERMS
544 548926.[7]

545 Xiong, Y., 2011. Release of EQ3/6 Database DATA0.FM1. Email to Jennifer Long, March 9, 2011.
546 Sandia National Laboratories, Carlsbad, New Mexico. ERMS 555152.

547

548 Line number up to this line = 548; no broken in-text link found as of 1/19/2023 by J. Jang.[8]

[7] User's manual for Version 8.0, and also applicable to Version 8.0a.
[8] If the underlined line number does not match with the line number in the left margin, please let J. Jang know to fix the format of this document.

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