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# SANDIA NATIONAL LABORATORIES
# WASTE ISOLATION PILOT PLANT

## AP-183
## Revision 1

## Analysis Plan to Update the WIPP Geochemical Thermodynamic Database (DATA0.FM1) to Data0.FM4 for CRA-2019

**Task 4.4.2.2.1**

**Effective Date: February 20, 2019**

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# 1 *Introduction and Objectives*

## 1.1 Introduction

This analysis plan guides the addition of thermodynamic parameters to a current quality assured EQ3/6 thermodynamic database with the ultimate creation of a self-consistent new QA EQ3/6 thermodynamic database. The motivation behind creation of this database is to include systems that were posed of interest and recommended by Environmental Protection Agency (EPA) after the submittal of CRA-2014. This analysis plan, as per NP 9-1, is considered a compliance decision analysis.

## 1.2 Objectives

The objective of this analysis plan is to create a single verified self-consistent QA EQ3/6 thermodynamic database which will include parameters outlined in Appendix GEOCHEM 2019, by using the currently verified QA EQ3/6 thermodynamic database, DATA0.FMT.R2, also known as DATA0.FM1 (Xiong, 2011c) as the starting database. The updated QA database resulting from this analysis plan will be called DATA0.FM4.

# *2 Approach*

This AP outlines the procedure for the creation of a single QA EQ3/6 thermodynamic database, which will be built on a previous QA database, DATA0.FM1. The general procedure in the creation of this QA database includes the compilation and testing of an interim database, and final QA approval of the database.

Testing of the preliminary database will consist of running Python minimization scripts (EQ3CodeModule.py) that can be coupled with calculations in the EQ3/6 Version 8.0a (Kirchner, 2012). In this minimization routine, the target thermodynamic parameter can be adjusted to minimize the difference, or to achieve reasonable differences, between the experimental value and predicted value. This minimization routine also can be used to minimize the saturation index by adjusting the target thermodynamic parameters. In this way, the target thermodynamic parameter can be derived. In the User's manual writer by Kirchner (2012), there are several examples showing how to obtain the optimized solubility constant by using the script. The python script coupled with the EQ3/6 Version 8.0a is to be used for evaluation of solubility data to derive Pitzer parameters as well as equilibrium constants. This minimization script has been utilized in previous CRAs and is suitable for achieving objectives in this Analysis Plan.

Running the python scripts from specific CRA-2019 Appendix Geochem equilibrium constants for reactions, Pitzer interaction coefficients or ions implemented in the WIPP chemical conditions process model will be to insure consistency of the derived parameters. If discrepancies are encountered during testing, then the parameters may need to be reevaluated and the pertinent documentation revised. Running of Python scripts will be for those parameters that were fit specifically for the creation of the new thermodynamic database, DATA0.FM4. There will not be running scripts for all the literature values provided in the following sections.

For CRA-2019, the DOE has reviewed the WIPP models for EDTA, citrate, and oxalate. Additional solubility-limiting phases are being added, one for each of the organic ligands, EDTA [$Ca_2EDTA•7H_2O(s)$], citrate (earlandite), and oxalate (glushinskite).

In addition, new to CRA-2019 is the addition of aqueous $Pb^{2+}$ and $Fe^{2+}$ to the WIPP Chemical Conditions Process Model. The DOE is limiting the models for both $Pb^{2+}$ and $Fe^{2+}$ to the inorganic systems. Reactions for lead include four lead-bearing solids: PbO(litharge), $PbCO_3$(cerussite), $Pb(SO_4)(s)$, and $NaPb_2(CO_3)_2(OH)$(abellaite).The aqueous species included are: $Pb^{2+}$, $PbCl^+$, $PbCl_2(aq)$, $PbCl_3^-$, $PbCO_3(aq)$, $Pb(CO_3)_2^{2-}$, $Pb(CO_3)Cl^-$, $PbOH^+$, $Pb(OH)_2(aq)$, $Pb(SO_4)(aq)$, and $Pb(CO_3)(OH)^-$. There is evidence (Xiong 2014a; Xiong 2014b) that $Pb^{2+}$ may form species with EDTA, citrate, and oxalate and that formation of lead-bearing solids with EDTA, citrate, and oxalate may occur (Xiong 2014a; Xiong 2014b). However, development of parameters to support addition of those reactions is still underway and they will not be included in CRA-2019. Addition of interactions between $Pb^{2+}$ and $Fe^{2+}$ and the organic ligands, EDTA, citrate, and oxalate, will be considered for inclusion in CRA-2024. The iron reactions include eight iron-bearing solids: $Fe(OH)_2(s)$ (Ferrous iron hydroxide), $Fe_2Cl(OH)_3(s)$ (hibbingite), $FeCO_3(s)$ (siderite), $Fe_2CO_3(OH)_2(s)$ (chukanovite). It is assumed that these solids are in their crystalline form. The aqueous species included are: $FeOH^+$, $Fe(OH)_2(aq)$, $Fe(OH)_3^-$, $Fe(OH)_4^{2-}$, $FeCO_3(aq)$, $Fe(CO_3)_2^{2-}$.

There is evidence (Jang 2017) that $Fe^{2+}$ may form species such as FeOxalate(aq), $Fe(Oxalate)_2^{2-}$, $FeEDTA^2$, $FeHEDTA^-$, $FeOHEDTA^{3-}$, $Fe(OH)_2EDTA^{4-}$, $FeOHCitrate^{2-}$, $FeCitrate^-$, and FeHCitrate(aq). However, development of parameters to support addition of those reactions is still underway and will not be included in CRA-2019.

A listing of the proposed equilibrium constants and Pitzer interaction coefficients from CRA-2019 Appendix Geochem to be included DATA0.FM4 may be found in the following sections.

## 2.1 Thermodynamic Data and Pitzer Parameters

The thermodynamic database including additional data for CRA-2019 interactions with organic ligands, magnesium oxide, lead, and iron are planning to be changed. Those changes are described in this section. The database that will be used to incorporate those changes is DATA0.FM4.

In the last two recertification cycles, the presence of a tetraborate species in WIPP brines has become of interest because of its potential effect on AN(III) solubility. Discussions between the DOE and the EPA have focused on the possible formation of a tetraborate complex with +III actinides. The conclusion of those discussions is that while addition of a tetraborate complex may be considered for inclusion in CRA-2024, the complex will not be included in CRA-2019.

The borate model remains unchanged for CRA-2019. No additional borate-containing solids or aqueous species have been added to the WIPP Chemical Conditions Process Model since the CCA.

### 2.1.1 Thermodynamic and Pitzer Data for Organic Ligand and Solids

The changes that are being proposed are the addition of solubility-limiting phases for EDTA, citrate, and oxalate. Without solubility-limiting phases, these ligands are present in the aqueous phase to the extent that the inventory allows.

For CRA-2019, the DOE has reviewed the WIPP thermodynamic and Pitzer data for EDTA, citrate, and oxalate. For consistency with the NEA database project, the DOE decided to align the WIPP Process models and parameters accordingly and therefore values for EDTA, citrate and oxalate from Hummel et al. (2005) are used. The values from Hummel et al. (2005) for $EDTA^{4-}$, $HEDTA^{3-}$, $H_2EDTA^{2-}$, $H_3EDTA^{-}$, $NaEDTA^{3-}$ , MgHEDTA are shown in Table 1.

Solubility-limiting phases are being added, one for each of the organic ligands, EDTA [$Ca_2EDTA{\bullet}7H_2O(s)$], citrate (earlandite), and oxalate (glushinskite). Then, using project-specific solubility data for the three solids, $Ca_2EDTA{\bullet}7H_2O$ (s), earlandite, and glushinskite, the DOE derives appropriate Pitzer interaction coefficients for the EDTA, citrate, and oxalate systems.

A new aqueous species, $NaEDTA^{3-}$ , is being added for CRA-2019. However, Hummel et al. (2005) did not provide Pitzer coefficients while Felmy and Mason (2003) did provide Pitzer coefficients. Because the log K values from both sources are similar, the DOE decided to use interaction coefficients from Felmy and Mason (2003) at the same time they are using the log K value from Hummel et al. (2005). The Pitzer interaction coefficient values are shown in Table 2. Because the interaction parameters for $K^+$ and $EDTA^-$ were originally set using an analogy to $Na^+$, that analogy is being extended by changing the $K^+$ interaction parameters to match the $Na^+$ interaction parameters here.

The species, $MgHEDTA^-$, does not exist in DATA0.FM1 and will be added to DATA0.FM4 with a formation log K value of -4.5 from Hummel et al. (2005).

**Table 1. Equilibrium Constants for Reactions with EDTA Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| Reaction | log K | Source |
|---|---|---|
| Solid Dissolution Reactions | | |
| $Ca_2EDTA{\bullet}7H_2O(s) \leftrightarrow 2Ca^{2+} + EDTA^{4-} + 7H_2O$ | -16.81 | Domski (2018a) |
| Aqueous Dissociation Reactions | | |
| $H_4EDTA(aq) \leftrightarrow EDTA^{4-} + 4H^+$ | -23.42 | Hummel et al. (2005) |
| $H_3EDTA^- \leftrightarrow EDTA^{4-} + 3H^+$ | -21.19 | Hummel et al. (2005) |
| $H_2EDTA^{2-} \leftrightarrow EDTA^{4-} + 2H^+$ | -18.04 | Hummel et al. (2005) |
| $HEDTA^{3-} \leftrightarrow EDTA^{4-} + H^+$ | -11.24 | Hummel et al. (2005) |
| $NaEDTA^{3-} \leftrightarrow Na^+ + EDTA^{4-}$ | -2.80 | Hummel et al. (2005) |
| $MgEDTA^{2-} \leftrightarrow Mg^{2+} + EDTA^{4-}$ | -10.90 | Hummel et al. (2005) |

| Reaction | log K | Source |
|---|---|---|
| $MgHEDTA^{1-} \leftrightarrow H^+ + MgEDTA^{2-}$ | -4.50 | Hummel et al. (2005) |
| $CaEDTA^{2-} \leftrightarrow Ca^{2+} + EDTA^{4-}$ | -12.69 | Hummel et al. (2005) |
| $CaHEDTA^{1-} \leftrightarrow H^+ + CaEDTA^{2-}$ | -3.54 | Hummel et al. (2005) |

**Table 2. Pitzer Interaction Coefficients (Cation-Anion Interactions) for Ions Containing EDTA Implemented in the WIPP Chemical Conditions Process Model**

| *I* | *j* | $\alpha_1/\alpha_2^A$ | $\beta^{(0)}$ | $\beta^{(1)}$ | $C^{\varphi}$ | Source |
|---|---|---|---|---|---|---|
| Cation-Anion Interactions | | | | | | |
| $EDTA^{4-}$ | $Na^+$ | 2.0/12.0 | 1.10 | 15.6 | 0.001 | Felmy and Mason (2003) |
| $EDTA^{4-}$ | $K^+$ | 2.0/12.0 | 1.10 | 15.6 | 0.001 | Felmy and Mason (2003) |
| $HEDTA^{3-}$ | $Na^+$ | 2.0/12.0 | 0.546 | 5.22 | -0.048 | Giambalvo (2002) |
| $HEDTA^{3-}$ | $K^+$ | 2.0/12.0 | 0.546 | 5.22 | -0.048 | Giambalvo (2002) |
| $H_2EDTA^{2-}$ | $Na^+$ | 2.0/12.0 | -0.126 | 1.74 | 0.054 | Giambalvo (2002) |
| $H_2EDTA^{2-}$ | $K^+$ | 2.0/12.0 | -0.126 | 1.74 | 0.054 | Giambalvo (2002) |
| $H_3EDTA^-$ | $Na^+$ | 2.0/12.0 | -0.235 | 0.29 | 0.059 | Giambalvo (2002) |
| $H_3EDTA^-$ | $K^+$ | 2.0/12.0 | -0.235 | 0.29 | 0.059 | Giambalvo (2002) |
| $NaEDTA^{3-}$ | $Na^+$ | 2.0/12.0 | 0.59 | 5.39 | 0 | Felmy and Mason (2003) |

All values for $\beta^{(2)}$ are 0.

Table 3 contains the thermodynamic parameters for $Ca_2EDTA{\bullet}7H_2O(s)$ and $CaEDTA^{2-}$, and the Pitzer parameters for the $Na^+$—$CaEDTA^{2-}$ ion pair that will be included in DATA0.FM4.

**Table 3. The Pitzer Model for the $Na^+$—$Mg^{2+}$—$Ca^{2+}$—$Cl^-$—$EDTA^{4-}$—$H_2O$ System at 25°C with $Ca_2EDTA{\bullet}7H_2O(s)$ From Domski (2018a) and Domski (2018b).**

| Equilibrium Constants at infinite dilution for Dissolution Reaction of $Ca_2EDTA{\bullet}7H_2O(s)$ and Formation Reaction of $CaEDTA^{2-}$ | | | | |
|---|---|---|---|---|
| Reactions | | | log K | |
| $Ca_2EDTA{\bullet}7H_2O(s) \leftrightarrow 2Ca^{2+} + EDTA^{4-} + 7H_2O(l)$ | | | -16.81 | |
| $Ca^{2+} + EDTA^{4-} \leftrightarrow CaEDTA^{2-}$ | | | 12.69 | |
| Pitzer Parameters[A] | | | | |
| Species, *i* | Species, *j* | $\beta^{(0)}$ | $\beta^{(1)}$ | $C^{\phi}$ |
| $Na^+$ | $CaEDTA^{2-}$ | 0.185 | 2.405 | 0 |

There are no changes recommended for $Na^+$ and citrate. There are no dissolution or dissociation reactions for $Na^+$ and citrate species or solids in DATA0.FM1, and there are no new data that indicate species or solids should be added. DATA0.FM1 contains Pitzer interaction coefficients for $Na^+$ and $H_2Citrate^-$, $Na^+$ and $HCitrate^{2-}$, and $Na^+$ and $Citrate^{3-}$, which will not change for CRA-2019. The Pitzer interaction coefficients are based on Mizera et al. (1999).

The DOE is planning the addition of earlandite, $Ca_3(Citrate)_2 \bullet 4H_2O(s)$ to the EQ3/6 database for CRA-2019. Earlandite would act as a solubility-limiting phase for citrate in the baseline solubility calculations. Hummel et al. (2005) gives values for log K for the reactions shown in Table 4. The Pitzer interaction coefficients for ions containing citrate ligands implemented in the WIPP Chemical Conditions Process Model are shown in Table 5.

**Table 4. Equilibrium Constants for Reactions with Citrate Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| Reaction | log K | Source |
|---|---|---|
| Solid Dissociation Reactions | | |
| $Ca_3(Citrate)_2 \bullet 4H_2O(s) \leftrightarrow 3Ca^+ + 2Citrate^{3-} + 4H_2O$ | -17.90 | Hummel et al. (2005)[A] |
| Aqueous Dissociation Reactions | | |
| $H_3Citrate\ (aq) \leftrightarrow H_2Citrate^- + H^+$ | -3.13 | Hummel et al. (2005) |
| $H_3Citrate\ (aq) \leftrightarrow Citrate^{3-} + 3H^+$ | -14.27 | Hummel et al. (2005) |
| $H_2Citrate^- \leftrightarrow HCitrate^{2-} + H^+$ | -4.78 | Hummel et al. (2005) |
| $H_2Citrate^- \leftrightarrow Citrate^{3-} + 2H^+$ | -11.14 | Hummel et al. (2005) |
| $HCitrate^{2-} \leftrightarrow Citrate^{3-} + H^+$ | -6.36 | Hummel et al. (2005) |
| $MgHCitrate\ (aq) \leftrightarrow Mg^{2+} + HCitrate^{2-}$ | -2.60 | Hummel et al. (2005) |
| $MgHCitrate\ (aq) \leftrightarrow Mg^{2+} + H^+ + Citrate^{3-}$ | -8.96 | Hummel et al. (2005) |
| $Mg(H_2Citrate)^+ \leftrightarrow Mg^{2+} + H_2Citrate^-$ | -1.31 | Hummel et al. (2005) |
| $Mg(H_2Citrate)^+ \leftrightarrow Mg^{2+} + 2H^+ + Citrate^{3-}$ | -12.45 | Hummel et al. (2005) |
| $MgCitrate^- \leftrightarrow Mg^{2+} + Citrate^{3-}$ | -4.81 | Hummel et al. (2005) |
| $CaCitrate^- \leftrightarrow Ca^{2+} + Citrate^{3-}$ | -4.80 | Hummel et al. (2005) |
| $CaHCitrate(aq) \leftrightarrow Ca^{2+} + H^+ + Citrate^{3-}$ | -9.28 | Hummel et al. (2005) |
| $CaHCitrate(aq) = Ca^{2+} + HCitrate^{2-}$ | -2.92 | Hummel et al. (2005) |
| $Ca(H_2Citrate)^+ = Ca^{2+} + H_2Citrate^{1-}$ | -1.53 | Hummel et al. (2005) |
| $Ca(H_2Citrate)^+ \leftrightarrow Ca^{2+} + 2H^+ + Citrate^{3-}$ | -12.67 | Hummel et al. (2005) |

[A]Value from Hummel et al. (2005) converted to the EQ3/6 basis species reaction by adding a value of -11.14 from $H_2Citrate^-$ value.

**Table 5. Pitzer Interaction Coefficients (Cation-Anion Interactions) for Ions Containing Citrate Implemented in the WIPP Chemical Conditions Process Model**

| *I* | *j* | $\alpha_1/\alpha_2^A$ | $\beta^{(0)}$ | $\beta^{(1)}$ | $C^{\varphi}$ | Source |
|---|---|---|---|---|---|---|
| Cation-Anion Interactions | | | | | | |
| $Citrate^{3-}$ | $Na^+$ | 2.0/12.0 | 0.0887 | 5.22 | 0.047 | Giambalvo (2002) |
| $Citrate^{3-}$ | $K^+$ | 2.0/12.0 | 0.0887 | 5.22 | 0.047 | Giambalvo (2002) |
| $H_2Citrate^-$ | $Na^+$ | 2.0/12.0 | -0.127 | 0.29 | 0.013 | Giambalvo (2002) |
| $H_2Citrate^-$ | $K^+$ | 2.0/12.0 | -0.130 | 0.29 | 0.013 | Giambalvo (2002) |
| $HCitrate^{2-}$ | $Na^+$ | 2.0/12.0 | -0.0989 | 1.74 | 0.027 | Giambalvo (2002) |
| $HCitrate^{2-}$ | $K^+$ | 2.0/12.0 | -0.0989 | 1.74 | 0.027 | Giambalvo (2002) |
| $CaCitrate^-$ | $Na^+$ | 2.0/12.0 | 0.174 | 0.29 | -0.069 | Giambalvo (2002) |
| $CaCitrate^-$ | $K^+$ | 2.0/12.0 | 0.174 | 0.29 | -0.069 | Giambalvo (2002) |
| $MgCitrate^-$ | $Na^+$ | 2.0/12.0 | 0.174 | 0.29 | -0.069 | Giambalvo (2002) |
| $MgCitrate^-$ | $K^+$ | 2.0/12.0 | 0.174 | 0.29 | -0.069 | Giambalvo (2002) |

All values for $\beta^{(2)}$ are 0.

The DOE is not updating the H-oxalic and Na-oxalic acid parameters at this time. Table 6 gives log K values from DATA0.FM1.Tables 7 and 8 shows the Pitzer interaction parameters. DATA0.FM1 contains Pitzer interaction coefficients for $Oxalate^{2-}$ and $Na^+$. The DOE will continue to use the values from DATA0.FM1 for CRA-2019.

The DOE is planning the addition of glushinskite [MgOxalate•$2H_2O$(s)], a solid phase containing oxalate, to the EQ3/6 database for CRA-2019. Glushinskite would act as an additional solubility-limiting phase for oxalate in the baseline solubility calculations.

Table 6 shows log K values for solids and species containing $Mg^{2+}$ and Oxalate. Table 7 shows Pitzer interaction coefficients for $Na^+$ and $(Oxalate)_2^{2-}$ for CRA-2019. Table 8 shows Pitzer interaction coefficients for the aqueous species MgOxalate(aq). One Pitzer coefficient was added to the parameters to complete the fit to the data. The coefficients added are for the interaction between MgOxalate(aq) and $Na^+$ and $Mg^{2+}$. Interactions between these two ions are not included in DATA0.FM1.

The DOE proposed the addition of whewellite for CRA-2004. The value shown in Table 6 from Xiong (2004) will be used in CRA-2019. Hummel et al. (2005) provides log k values for two other solids that could form from $Ca^{2+}$ and $Oxalate^{2-}$. These two solids will not be added to the WIPP model for CRA-2019 because they are not expected to exist in the WIPP environment. The value of 0.0189 for $\lambda_{na}$ for CaOxalate(aq) and Cl- interactions that was included in DATA0.FM1 will remain for CRA-2019.

**Table 6. Equilibrium Constants for Reactions with Oxalate Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| Reaction | log K | Source |
|---|---|---|
| Solid Dissolution Reactions | | |
| CaOxalate•$H_2O$(s) (whewellite)↔ $Ca^{2+}$ + $Oxalate^{2-}$ + $H_2O$ | -8.75 | Xiong (2004) |
| MgOxalate•2$H_2O$(s) (glushinskite) ↔ $Mg^{2+}$ + $Oxalate^{2-}$ + 2$H_2O$ | -6.41 | AP-182 (2019) |
| $H_2$Oxalate•2$H_2O$(s) ↔ $Oxalate^{2-}$ + 2$H^+$ + 2$H_2O$ | -5.66 | Giambalvo (2002) |
| NaHOxalate•$H_2O$(s) ↔ $Na^+$ + $H^+$ + $Oxalate^{2-}$ +$H_2O$ | -6.06 | Giambalvo (2002) |
| $Na_2$Oxalate(s) ↔ 2$Na^+$ + $Oxalate^{2-}$ | -2.41 | Giambalvo (2002) |
| Aqueous Dissociation Reactions | | |
| CaOxalate(aq) ↔ $Ca^{2+}$ + $Oxalate^{2-}$ | -3.79 | Giambalvo (2002) |
| MgOxalate(aq) ↔ $Mg^{2+}$ + $Oxalate^{2-}$ | -3.79 | Giambalvo (2002) |
| Mg(Oxalate)$_2^{2-}$ ↔ $Mg^{2+}$ + 2$Oxalate^{2-}$ | -5.24 | Jang and Kim (2016) |
| $H_2$Oxalate(aq) ↔ $Oxalate^{2-}$ + 2$H^+$ | -5.65 | Giambalvo (2002) |
| $HOxalate^-$↔ $Oxalate^{2-}$ + $H^+$ | -4.26 | Giambalvo (2002) |

**Table 7. Pitzer Interaction Coefficients (Cation-Anion Interactions) for Ions Containing Oxalate Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| *I* | *j* | $\alpha_1/\alpha_2$ | $\beta^{(0)}$ | $\beta^{(1)}$ | $C^{\varphi}$ | Source |
|---|---|---|---|---|---|---|
| Cation-Anion Interactions | | | | | | |
| $Oxalate^{2-}$ | $Na^+$ | 2.0/12.0 | -0.2176 | 1.74 | 0.122 | Giambalvo (2002) |
| $Oxalate^{2-}$ | $K^+$ | 2.0/12.0 | -0.218 | 1.74 | 0.122 | Giambalvo (2002f) |
| Mg(Oxalate)$_2^{2-}$ | $Na^+$ | 1.4/12.0 | 0.07 | 0 | 0 | AP-182 (2019) |
| $HOxalate^-$ | $Na^+$ | 2.0/12.0 | -0.245 | 0.29 | 0.068 | Giambalvo (2002) |
| $HOxalate^-$ | $K^+$ | 2.0/12.0 | -0.245 | 0.29 | 0.068 | Giambalvo (2002) |

All values for $\beta^{(2)}$ are 0.

**Table 8. Pitzer Interaction Coefficients (Neutral-Cation; Neutral-Anion Interactions) for Ions Containing Organic Ligands Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| *i* | *j* | $\lambda_{nc'}$ or $\lambda_{na'}$ | Source |
|---|---|---|---|
| Neutral-Cation Interactions | | | |
| MgOxalate(aq) | $Na^+$ | 0.08 | AP-182 (2019) |
| MgOxalate(aq) | $Mg^{2+}$ | -0.20 | AP-182 (2019) |
| Neutral-Anion Interactions | | | |
| CaOxalate(aq) | $Cl^-$ | 0.0189 | Giambalvo (2002) |
| MgOxalate(aq) | $Cl^-$ | 0.0189 | Giambalvo (2002) |

### 2.1.2 Interactions with MgO Solids

Recently, an error in transcription was discovered when the data from Robie and Hemingway (1973) was compared to that in DATA0.FM1. The error is in the FMT thermodynamic database fmt_050405.chemdat (Xiong et al. 2005) where the $\mu^0$/RT value attributed to Robie and Hemingway (1973) does not match what was reported by Robie and Hemingway (1973).

Using either the $\mu^0$/RT value derived from Robie and Hemingway (1973; -2365.82) or the standard free energy of formation (-5,864.74 kJ/mol), the log K values for the reaction are: 62.77 and 31.49, respectively.

Per a September 2018 Technical Exchange meeting (2018), DOE agreed to use EPA's recommended corrected log K value of 31.49 from Robie and Hemingway (1973) in the CRA-2019.

**Table 9. Equilibrium Constants for Dissolution Reactions for MgO Solids Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| Reaction | log K | Source |
|---|---|---|
| Solid Dissolution Reactions | | |
| $Mg_5(CO_3)_4(OH)_2{\bullet}4H_2O$(hydromagnesite5424) + $6H^+ \leftrightarrow 5Mg^{2+} + 4HCO_3^- + 6H_2O$ | 31.49 | Robie and Hemingway (1973) |

### 2.1.3 Interactions with Lead Systems

Lead is being added to the WIPP Chemical Conditions Process Model for CRA-2019. A summary of the parameters and their log K values are presented in Table 10. The lead system was initially parameterized primarily with data gathered by the DOE as part of their geochemistry research program for the WIPP (Xiong 2013, 2014a, 2014b, 2015a, 2015b). The DOE decided to use a lead model based on a critical review of the chemical speciation of $Pb^{2+}$ with inorganic ligands published as an International Union of Pure and Applied Chemists (IUPAC) technical document (Powell et al. 2009) for CRA-2019.

Experiments for cerussite [$PbCO_3$(s)] solubility in $NaHCO_3$ + NaCl and $NaHCO_3$ + $Na_2CO_3$ solutions were conducted by the DOE geochemistry research program for the WIPP (Xiong 2015). During examination of the solid phase at the end of the experiments, abellaite, $NaPb_2(CO_3)_2(OH)$, was identified. Therefore, the data are for a dual salt system of cerussite and abellaite. The experimental protocols for the data collected for this work are described in Kirkes et al. (2014). The recent discovery that cerussite transformed to abellaite in some of Xiong's (2015) experiments has led the DOE to use log K values from Powell et al. (2009) until abellaite can be added to the model. The values to be used in CRA-2019 are shown in Table 11. Xiong (2015a, 2015b) experiments pertaining to $PbCO_3$(aq), $Pb(CO_3)_2^{2-}$, $Pb(CO_3)Cl^-$ , $Pb(OH)_2$(aq) and $Pb(OH)_3^-$ systems require further characterization, therefore equilibrium constants for these dissociation systems and $PbCl_2$ and $Pb(OH)_2$ were found in the literature.

Recent XRD analysis of the solids from Xiong (2015b) indicates the presence of multiple solid phases in addition to α-PbO. This has led the DOE to use log K values from Powell et al. (2009).

Once the solid phases are identified, the DOE will revisit the experiments to determine if additional parameters are required. The values to be used in CRA-2019 are shown in Table 10.

Powell et al. (2009) contains information about lead reactions with sulfate that are germane to the WIPP. That information is shown in Table 10. There will be no Pitzer interaction coefficients derived for the lead systems.

**Table 10. Equilibrium Constants for Reactions with Lead Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| Reaction | log K | Source |
|---|---|---|
| Solid Dissolution Reactions | | |
| α-PbO(litharge) + $2H^+ \leftrightarrow Pb^{2+} + H_2O(l)$ | 12.62 | Powell et al. (2009) |
| $PbCO_3$(cerussite) + $H^+ \leftrightarrow Pb^{2+} + HCO_3^-$ | -2.84 | Powell et al. (2009) |
| $Pb_2CO_3Cl_2(s) + H^+ \leftrightarrow 2Pb^{2+} + HCO_3^{1-} + 2Cl^-$ | 0.41 | Powell et al. (2009) |
| $PbSO_4(s) + H^+ \leftrightarrow Pb^{2+} + HSO_4^{1-}$ | -5.82 | Powell et al. (2009) |
| Aqueous Dissociation Reactions | | |
| $PbCl^+ \leftrightarrow Pb^{2+} + Cl^-$ | -1.50 | Powell et al. (2009) |
| $PbCl_2(aq) \leftrightarrow Pb^{2+} + 2Cl^-$ | -2.10 | Powell et al. (2009) |
| $PbCl_3^- \leftrightarrow Pb^{2+} + 3Cl^-$ | -2.00 | Powell et al. (2009) |
| $PbOH^+ + H^+ \leftrightarrow Pb^{2+} + H_2O$ | 7.46 | Powell et al. (2009) |
| $Pb(OH)_2(aq) + 2H^+ \leftrightarrow Pb^{2+} + 2H_2O(l)$ | 16.94 | Powell et al. (2009) |
| $Pb(CO_3)(OH)^- + 2H^+ \leftrightarrow Pb^{2+} + HCO_3^{1-} + H_2O(l)$ | 13.44 | Powell et al. (2009) |
| $Pb(CO_3)Cl^- + H^+ \leftrightarrow Pb^{2+} + HCO_3^- + Cl^-$ | 3.87 | Powell et al. (2009) |
| $PbCO_3(aq) + H^+ \leftrightarrow Pb^{2+} + HCO_3^-$ | 3.89 | Powell et al. (2009) |
| $Pb(CO_3)_2^{2-} + 2H^+ \leftrightarrow Pb^{2+} + 2HCO_3^-$ | 10.55 | Powell et al. (2009) |
| $PbSO_4(aq) + H^+ \leftrightarrow Pb^{2+} + HSO_4^{1-}$ | - 0.74 | Powell et al. (2009) |

### 2.1.4 Interactions with Iron Systems

The model developed for $Fe(OH)_2(s)$ solubility is shown in Table 12. The first four reactions considered are: 1) dissolution of $Fe(OH)_2(s)$, 2) dissociation of $Fe(OH)_2(aq)$, 3) dissociation of $FeOH^+$, and 4) dissociation of $Fe(OH)_3^-$. A fifth reaction for dissociation of $Fe(OH)_4^{2-}$ was examined but rejected because speciation diagrams [using log K values from Baes and Mesmer (1976)] indicate that $Fe(OH)_4^{2-}$ is not likely to exist under WIPP conditions. These four reactions will be added to DATA0.FM4 for CRA-2019. Experiments covered two basic iron systems, 1) Fe-Na-Cl-$H_2O$, and 2) Fe-Na-Cl-$CO_3$-$H_2O$, which are the systems that will be parameterized for CRA-2019.

The DOE has two data sets collected since 2007. The first data set was published in Nemer et al. (2011). The second data set was entered into the WIPP records center as part of Jang (2016). Kim et al. (2017) combined the two data sets and published a model for aqueous $Fe_2Cl(OH)_3(s)$ solubility. The data shown in Table 11 is from references named Kim et al. (2017).

Kim et al. (2017) adopted a log K value for siderite dissolution of 0.12 from Stumm and Morgan (1996).

Kim et al. (2017) performed a data fit to obtain values for $\beta^{(0)}$ and $\beta^{(1)}$ for the $Na^+$ and $Fe(CO_3)_2^{2-}$ interaction of -0.23 and 6.26, respectively. The values are shown in Table 12.

Kim et al. (2017) surveyed a number of references to choose a value for $\Delta G_f^0$ for dissolution of chukanovite. Ultimately, Kim et al. (2017) chose to use an average value of 1171.7 kJ/mol from a number of sources. They then calculated a log K value for dissolution of 12.32. The values are shown in Table 11.

**Table 11. Equilibrium Constants for Reactions with Iron Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| Reaction | log K | Source |
|---|---|---|
| Solid Dissolution Reactions | | |
| $Fe(OH)_2(s) + 2H^+$ (ferrous hydroxide) $\leftrightarrow Fe^{2+} + 2H_2O$ | 12.89 | Kim et al. (2017) |
| $Fe_2Cl(OH)_3(s) + 3H^+$ (hibbingite) $\leftrightarrow 2Fe^{2+} + Cl^- + 3H_2O$ | 17.08 | Kim et al. (2017) |
| $FeCO_3(s) + H^+$ (siderite) $\leftrightarrow Fe^{2+} + HCO_3^-$ | -0.12 | Stumm and Morgan (1996) |
| $Fe_2CO_3(OH)_2(s) + 3H^+$ (chukanovite) $\leftrightarrow 2Fe^{2+} + HCO_3^- + 2H_2O$ | 12.32 | Kim et al. (2017) |
| Aqueous Dissociation Reactions | | |
| $FeOH^+ + H^+ \leftrightarrow Fe^{2+} + H_2O$ | 9.31 | Shock et al. (1997) |
| $Fe(OH)_2(aq) + 2H^+ \leftrightarrow Fe^{2+} + 2H_2O$ | 20.82 | Stumm and Morgan (1996) |
| $Fe(OH)_3^- + 3H^+ \leftrightarrow Fe^{2+} + 3H_2O$ | 31.0 | Baes and Mesmer (1976) |
| $FeCO_3(aq) + H^+ \leftrightarrow Fe^{2+} + HCO_3^-$ | 4.83 | Bruno et al. (1992) |
| $Fe(CO_3)_2^{2-} + 2H^+ \leftrightarrow Fe^{2+} + 2HCO_3^{1-}$ | 13.89 | Kim et al. (2017) |

**Table 12. Pitzer Interaction Coefficients (Cation-Anion Interactions) for Ions Containing Iron Implemented in the WIPP Chemical Conditions Process Model for CRA-2019**

| *I* | *j* | $\alpha_1/\alpha_2^A$ | $\beta^{(0)}$ | $\beta^{(1)}$ | $\beta^{(2)}$ | $C^{\varphi}$ | Source |
|---|---|---|---|---|---|---|---|
| $Fe^{2+}$ | $Cl^-$ | 2.0/12.0 | 0.373 | 1.135 | 0 | -0.022 | Moog et al. (2004) |
| $Na^+$ | $Fe(CO_3)_2^{-2}$ | 2.0/12.0 | -0.23 | 6.26 | 0 | 0 | Kim et al. (2017) |

**Table 13. Pitzer Interaction Coefficients (Cation-Cation Interactions) for Ions Containing Iron Implemented in the WIPP Chemical Conditions Process Model**

| ***i*** | ***j*** | **$\theta_{cc'}$ or $\theta_{aa'}$** | **Source** |
|---|---|---|---|
| $Na^+$ | $Fe^{2+}$ | 0.110 | Moog et al. (2004) |
| $Mg^{2+}$ | $Fe^{2+}$ | 0.145 | Moog et al. (2004) |
| $K^+$ | $Fe^{2+}$ | 0.0274 | Moog et al. (2004) |
| $Ca^{2+}$ | $Fe^{2+}$ | 0.0811 | Moog et al. (2004) |

**Table 14. Pitzer Interaction Coefficients (Cation-Cation-Anion Interactions) for Ions Containing Iron Implemented in the WIPP Chemical Conditions Process Model**

| *I* | *j* | *k* | $\psi_{ijk}$ | Source |
|---|---|---|---|---|
| $Fe^{+2}$ | $Na^+$ | $Cl^-$ | -0.0161 | Moog et al. (2004) |
| $Fe^{+2}$ | $K^+$ | $Cl^-$ | -0.0252 | Moog et al. (2004) |
| $Fe^{+2}$ | $Mg^{+2}$ | $Cl^-$ | -0.0299 | Moog et al. (2004) |
| $Fe^{+2}$ | $Ca^{+2}$ | $Cl^-$ | -0.0160 | Moog et al. (2004) |

## 2.2 Software List

The software to be used is EQ3/6 Version 8.0a (Wolery and Jarek, 2003; Wolery, 2008; Wolery et al., 2010; Xiong, 2011b), and DATA0.FM1 (Xiong, 2011a). In addition, off the shelf software such as Microsoft Excel, and the Excel macro *GetEQData_v101f.xls* will be used to extract data from the EQ3/6 output files, and the open source text editor Notepad++ may be used to edit the preliminary and final databases.

### 2.3 Acceptance Criteria

During the testing phases of the preliminary and interim databases comparisons between fitted parameters and EQ3/6 output will be made in Microsoft Excel. Parameters and simulation using the final interim database will be considered acceptable if they agree to within 10%. If the percent difference exceeds 10%, a new value for lambda will be sought in the interim database. Also, if the 10% criteria is exceeded it will be refitted or the discrepancy will be documented for acceptability. If the percent difference is less than or equal to 10%, the value is acceptable. In addition, it should be pointed out that comparisons between fitted values will only be from Domski and Xiong sources. The literature source values will be directly put into the datablocks of the EQ3/6 database and there will be no comparison.

## 3 Tasks

There are two tasks under this AP, which will result in the creation and documentation of a QA thermodynamic database. The subtasks for this task includes the creation of a preliminary database, testing of the preliminary database, evaluation of the test results, evaluation of errors and re-testing as necessary, and issuance of the final QA database(s).

There are three major steps that go into the task of creating DATA0.FM4 in this AP: (1) computation of solubility constants from experimental data; (2) computation of formation constants from experimental data; and (3) derivation of Pitzer parameters from experimental data.

The final QA databases will be documented in an analysis report which will accompany the issuance of the QA database, DATA0.FM4.

Table 15 lists the task, database name, and completion date for AP-183.

**Table 15. Task list and completion date**

| Task | Database | Completion Date |
|---|---|---|
| 1 | DATA0.FM4 | 02/28/19 |
| 2 | Analysis Report for Creation of DATA0.FM4 | 02/28/19 |

Paul Domski will be the primary analyst performing the tasks under this AP. A memo that states the completion of DATA0.FM1 will be issued to users, and the final database will be submitted to WIPP records to be stored in the CVS under the Sun platform/Solaris OS with all accompanying documentation.

## 4 Special Considerations

No special considerations are needed for this analysis plan.

# 5 Applicable Procedures

All applicable WIPP QA procedures will be followed when conducting this AP.

- Training of personnel will be conducted in accordance with the requirements of NP 2-1, *Qualification and Training*.
- Analyses will be conducted and documented in accordance with the requirements of NP 9-1, *Analyses*.
- All software used will meet the requirements laid out in NP 19-1, *Software Requirements* and NP 9-1, as applicable.
- The analyses will be reviewed following NP 6-1, *Document Review Process*.
- All required records will be submitted to the WIPP Records Center in accordance with NP 17-1, *Records*.

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