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FINAL EC 9/4/24

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date: *August 19, 2024*

to: Sandia National Laboratory WIPP Records Center

from: Paul S. Domski, SNL WIPP Geochemistry *signature on file*

subject: Solubility of Solid Phase $Am(OH)_3$ in Various Solutions at pmH 7 - 14

Introduction

At the Request of the Department of Energy (DOE) (see Appendix A) a series of EQ3/6, Version 8.0a (Wolery, 2013) simulations were performed for the purpose of determining the solubility of americium hydroxide, both the amorphous and the crystalline form, as represented in the WIPP thermodynamic database DATA0.FM6 (Domski, 2023a). The simulations were performed using EQ3NR at pmH values from 7.0 to 14.0 in 0.5 pmH increments. The matrix solutions used for the simulations included pure water, 5.0 molar NaCl, and the equilibrium compositions of GWB and ERDA-6 as documented in Tables 6 and 7 of Domski (2023b). Note, all of the associated files are archived at data/cvs/CVSLIB/WIPP_EXTERNAL/DOE_AWard.

Inputs

The pure water EQ3NR input is listed below. The pmH value was changed for each input file from 7.0 to 14.0 in 0.5 increments. Likewise, a second set of input files were created where the solubility controlling phase was $Am(OH)_3(s)$ representing the crystalline form.

```
|-------------------------------------------------------------------------|
|Aqueous Basis Species/Constraint Species        |Conc., etc. |Units/Constraint|
|  (uspeci(n)/ucospi(n))                         | (covali(n))|(ujf3(jflgi(n)))|
|-------------------------------------------------------------------------|
|H+                                              | 7.0        |pmH             |
|Am+++                                           | 1.00000E-18|Hetero. equil.  |
|->|Am(OH)3(am)                                     | (ucospi(n))             |
|-------------------------------------------------------------------------|
```

The 5.0 M NaCl EQ3NR input is listed below. The pmH value was changed for each input file from 7.0 to 14.0 in 0.5 increments. Likewise, a second set of input files were created where the solubility controlling phase was $Am(OH)_3(s)$ representing the crystalline form.

```
|-------------------------------------------------------------------------|
|Aqueous Basis Species/Constraint Species        |Conc., etc. |Units/Constraint|
|  (uspeci(n)/ucospi(n))                         | (covali(n))|(ujf3(jflgi(n)))|
|-------------------------------------------------------------------------|
|H+                                              | 7.0        |pmH             |
|Na+                                             | 5.00000E+00|Molarity        |
|Cl-                                             | 5.00000E+00|Molarity        |
|Am+++                                           | 1.00000E-18|Hetero. equil.  |
```


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```
|->|Am(OH)3(am)                                          | (ucospi(n))                |
|-----------------------------------------------------------------------------------|
```

The equilibrated GWB composition used as EQ3NR input is listed below. The elemental input values were extracted from the minimum volume case output file, *GWB_01.6o*, from Domski (2023b). Also extracted were the solution density, total dissolved solids, and the ratio of molarity to molality (M:m), all of which were used in the input files for this exercise. The file from Domski (2023b), *GWB_01.6o,* is included with the files for this memo.

The pmH value was changed for each input file from 7.0 to 14.0 in 0.5 increments. The organic concentrations used in the input files are documented in Table 4 of Domski (2023b) Likewise, a second set of input files were created where the solubility controlling phase was $Am(OH)_3(s)$ representing the crystalline form.

```
|-----------------------------------------------------------------------------------|
|Aqueous Basis Species/Constraint Species          |Conc., etc. |Units/Constraint|
| (uspeci(n)/ucospi(n))                            | (covali(n))|(ujf3(jflgi(n)))|
|-----------------------------------------------------------------------------------|
|Na+                                               | 5.04000E+00|Molality        |
|K+                                                | 6.28000E-01|Molality        |
|Mg++                                              | 9.11000E-01|Molality        |
|Ca++                                              | 1.48000E-02|Molality        |
|Cl-                                               | 5.70000E+00|Molality        |
|HCO3-                                             | 2.66000E-04|Molality        |
|SO4--                                             | 2.40000E-01|Molality        |
|B(OH)4-                                           | 2.13000E-01|Molality        |
|Br-                                               | 3.58000E-02|Molality        |
|H+                                                | 7.00000E+00|pmH             |
|NpO2+                                             | 1.00000E-18|Hetero. equil.  |
|->|KNpO2CO3                                          | (ucospi(n))                |
|Th++++                                            | 0.00000E+00|Hetero. equil.  |
|->|ThO2(am)                                          | (ucospi(n))                |
|Am+++                                             | 1.00000E-18|Hetero. equil.  |
|->|Am(OH)3(am)                                       | (ucospi(n))                |
|Acetate-                                          | 1.72000E-02|Molarity        |
|Citrate---                                        | 1.79000E-03|Molarity        |
|EDTA----                                          | 4.22000E-05|Molarity        |
|Oxalate--                                         | 5.76000E-03|Molality        |
|Pb++                                              | 1.30000E-02|Molality        |
|Fe++                                              | 6.15000E-05|Molality        |
|-----------------------------------------------------------------------------------|
```

The equilibrated ERDA-6 composition used as EQ3NR input is listed below. The elemental input values were extracted from the minimum volume case output file, *ERDA_01.6o*, from Domski (2023b). Also extracted were the solution density, total dissolved solids, and the ratio of molarity to molality (M:m), all of which were used in the input files for this exercise. The file from Domski (2023b), *ERDA_01.6o,* is included with the files for this memo.

The pmH value was changed for each input file from 7.0 to 14.0 in 0.5 increments. The organic concentrations used in the input files are documented in Table 4 of Domski (2023b) Likewise, a second set of input files were created where the solubility controlling phase was $Am(OH)_3(s)$ representing the crystalline form.

```
|-----------------------------------------------------------------------------------|
|Aqueous Basis Species/Constraint Species          |Conc., etc. |Units/Constraint|
```

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(uspeci(n)/ucospi(n))	(covali(n))	(ujf3(jflgi(n)))
Na+	5.79000E+00	Molality
K+	1.09000E-01	Molality
Mg++	4.39000E-01	Molality
Ca++	1.46000E-02	Molality
Cl-	5.71000E+00	Molality
HCO3-	2.80000E-04	Molality
SO4--	1.97000E-01	Molality
B(OH)4-	7.06000E-02	Molality
Br-	1.23000E-02	Molality
H+	7.00000E+00	pmH
NpO2+	1.00000E-18	Hetero. equil.
->KNpO2CO3		(ucospi(n))
Th++++	0.00000E+00	Hetero. equil.
->ThO2(am)		(ucospi(n))
Am+++	1.00000E-18	Hetero. equil.
->Am(OH)3(am)		(ucospi(n))
Acetate-	1.72000E-02	Molarity
Citrate---	1.79000E-03	Molarity
EDTA----	4.22000E-05	Molarity
Oxalate--	5.76000E-03	Molarity
Pb++	6.09000E-03	Molality
Fe++	6.46000E-05	Molality

Results

The results of the EQ3NR simulations are tabulated in the spreadsheet *An(III)_Solubility.xlsx* (included with this memo) and graphically displayed in Figure 1.

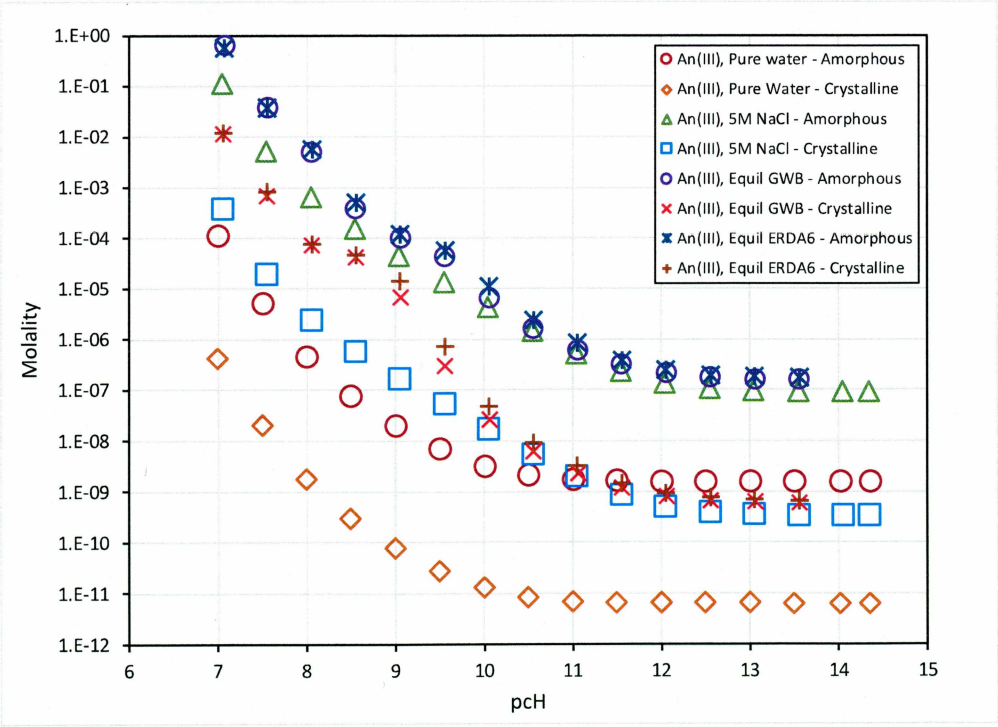

Figure 1. An(III) solubility as a function of pcH for the amorphous and crystalline $Am(OH)_3$ solid phase in four solutions: 1) Pure water; 2) 5.0 M NaCl solution; 3) equilibrated GWB brine; and 4) equilibrated ERDA-6 brine.

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Summary

EQ3/6 calculations were preformed at the request of the DOE Appendix A) to examine the solubility of solid phase $Am(OH)_3$ in both the amorphous and crystalline form, in four different solutions at pmH values ranging from 7 to 14. The solutions included pure water, 5M NaCl, and the equilibrated GWB and ERDA-6 compositions from the RPPCR (Domski, 2023b). The model results fall within the expected ranges of the experimental data shown in the plot in Appendix A.

References

Domski, P.S. 2023a. "An Update to the WIPP EQ3/6 Database DATA0.FM1 with the Creation of DATA0.FM6. Carlsbad, NM: Sandia National Laboratories. ERMS 579370.

Domski, P.S. 2023b. "Prediction of Baseline Actinide Solubilities for the Replacement Panels Planned Change Request (RPPCR) Performance Assessment". Carlsbad, NM: Sandia National Laboratories. ERMS 579503.

Wolery T. J. 2013. EQ3/6 - Software for Geochemical Modeling, Version 8.0a. LLNL-CODE-2013-683958. Lawrence Livermore National Laboratory, Livermore, CA.

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Appendix A.

From: Anderson Ward <Anderson.Ward@cbfo.doe.gov>
Sent: Tuesday, August 13, 2024 8:31 AM
To: Domski, Paul Stanley <psdomsk@sandia.gov>; Miller, Chammi Sumudumali <csmille@sandia.gov>
Subject: [EXTERNAL] An Solubility Plot

Hi Paul,

I trust all is well.

I'm putting together a presentation of the EPA Stakeholders meeting ***and*** a response to the EPA's question on An solubility and need your help.

We have collected a large amount of published solubility data and need to compare our current An+3 model to these data. Please generate the total concentration (molal), as a function of pH in the range 6 to 14, for the amorphous and crystalline phases using the current model. My goal is to overlay them on the plot below, which is derived mostly from the reference *Hala and Miyamoto J. Phys. Chem. Ref. Data, Vol. 36, No. 4, 2007*. The points I want to make are: (i) at pH > 7 solubility is mostly independent of pH and (ii) at pH > 7, the data shows no real difference in solubility of +3 and +4.

It would be great if I could get the results by Thursday August 15, 2024.

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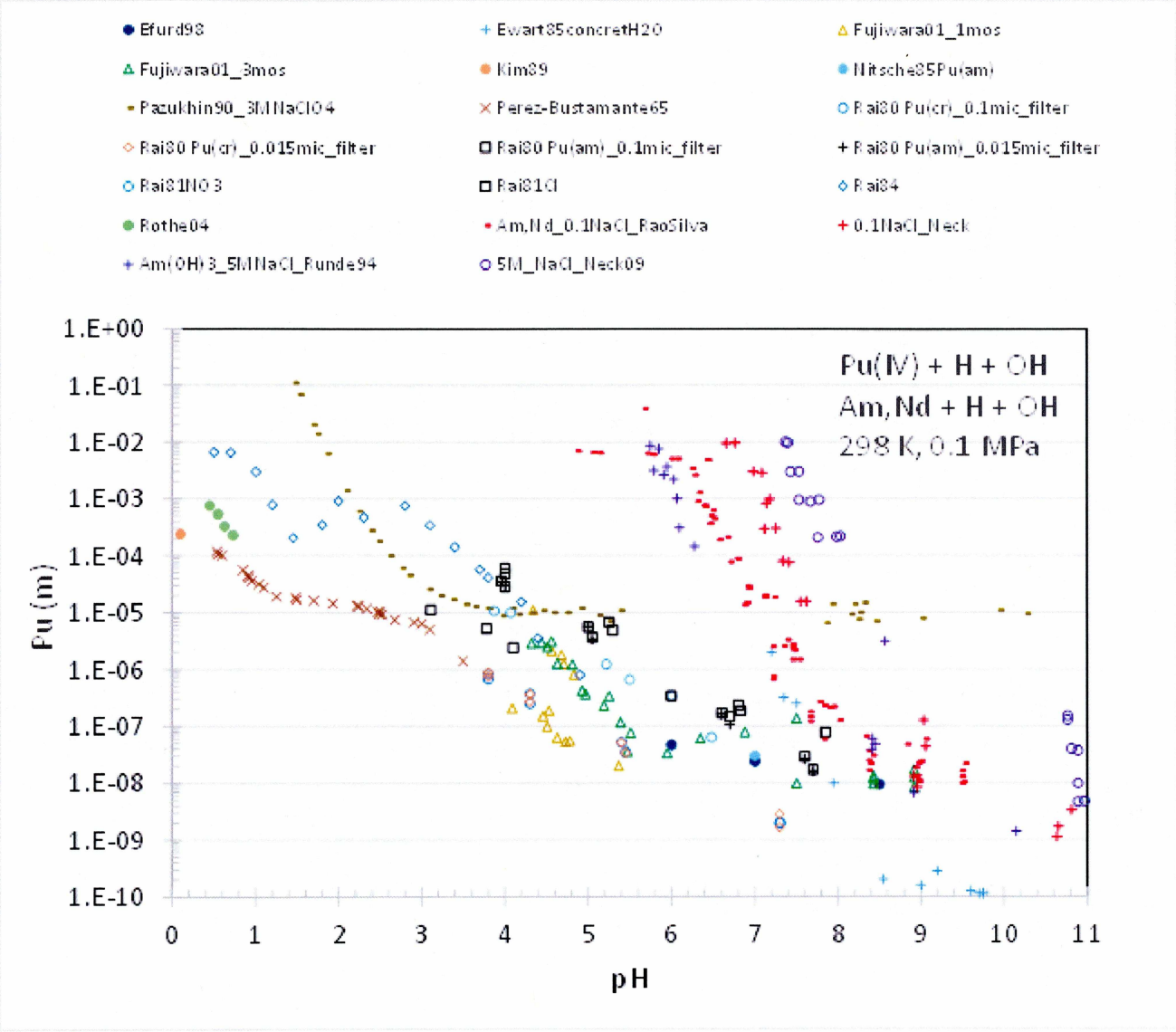

Andy

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