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subject: Release of FMT database FMT_021120.CHEMDAT

1. Introduction

In preparation for the actinide solubility calculations that will be performed in support of the Compliance Recertification Application (CRA), I have been reviewing the geochemical parameters used in the Fracture-Matrix Transport (FMT) database. In previous memorandums addressed to you, I recommended parameter values to use in simulations of the speciation and solubility of actinides in Waste Isolation Pilot Plant (WIPP) brines (Giambalvo, 2002a; 2002b; 2002c; 2002d; 2002e). I have completed a new version of the FMT database (FMT_021120.CHEMDAT) that incorporates the parameter values recommended in the previous memos and additional inorganic Am species and organic Th and Np species identified in two of the memos (Giambalvo, 2002a; 2002d). The new database has been archived in Sandia's WIPP Configuration Management System (CMS) (Table 1).

The new database is derived from the version of the FMT database used for compliance baseline performance assessment (PA) calculations (old baseline database)[1]. Differences between the new database and the old baseline database other than those resulting from implementation of the recommendations made in the previous memos are described below. I recommend that the new database be adopted as a new baseline database, and be used to calculate actinide solubilities for the CRA.

This memo includes a comparison of actinide solubilities calculated using the old baseline database and actinide solubilities calculated using the new database.

2. Alkaline metal analogies

In the new database, Pitzer parameters describing Na^+-actinide interactions have been applied to K^+-actinide interactions if K^+-actinide interaction parameters were not otherwise available. In the old baseline database, this analogy between Na^+ and K^+ behavior was made only for inorganic Np species

[1] The old baseline database is identical to the database used in the Performance Assessment Verification Test (PAVT), and is the most recent documented version of the database (Novak, 1997). Two versions are stored in Sandia National Laboratories' Configuration Management System on the WIPP Alpha Cluster in the library WP$NONPA_CMSROOT:[FMT]. FMT_970407.CHEMDAT is compatible with FMT version 2.2. FMT_PH_970407.CHEMDAT is compatible with FMT version 2.4.

WIPP:1.3.5.1.2.1:PA:QA:PKG522917

and for $Th(CO_3)_5^{6-}$. Application of the Na^+/K^+ analogy to Np species was found to be valid by Novak et al. (1997) and Al Mahamid et al. (1998).

In the new database, where either Mg^{2+}-actinide or Ca^{2+}-actinide interaction parameters are available, the parameters have been applied to both sets of interactions. Pitzer parameters describing the interaction of Ca^{2+} with Am-chloride species have been applied to Mg^{2+} interactions with Am-chloride species. Pitzer parameters describing the interaction of Mg^{2+} with Th^{4+} have been applied to Ca^{2+} interactions with Th^{4+}. Pitzer parameters describing the interaction of Mg^{2+} with Np-carbonate species have been applied to Ca^{2+} interactions with Np-carbonate species.

As in the old baseline database, parameters describing the formation of Ca-organic species and interaction of Ca-organic species with other species in solution are assigned by analogy with Mg-organic species.

The Na^+/K^+ and Mg^{2+}/Ca^{2+} analogies have not been thoroughly tested, but theoretical considerations and limited experimental evidence suggest that such analogies should result in better model predictions than would neglecting the effect of interaction with K^+, Mg^{2+}, or Ca^{2+} on the activity of actinide species. At ionic strengths greater than ~1 mol/kg, activity coefficients increase with increasing ionic strength (Pitzer, 1991; Langmuir, 1997). For the actinide species, neglecting interaction with K^+, Mg^{2+}, and Ca^{2+} effectively lowers the ionic strength of the solution being modeled, and results in underprediction of activity coefficients and overprediction of solubilities. This theoretical argument is born out by the work of Al Mahamid et al. (1998), who measured and simulated Np solubility in high Mg brines. Application of Na^+ and K^+ Pitzer parameters to Mg^{2+} interactions with Np species resulted in model predictions of Np solubility within 1 order of magnitude of measured solubility, while neglecting Mg^{2+} interactions resulted in overprediction of Np solubility by 4-5 orders of magnitude (Al Mahamid et al., 1998). The Na^+/K^+ and Mg^{2+}/Ca^{2+} analogies should be at least as good as if not better than the $Na^+/K^+/Mg^{2+}$ analogy because ions of like charge and similar size behave more similarly than ions of unlike charge. All of these analogies should be re-evaluated as more experimental data become available.

3. Actinide analogies

The old baseline database includes Pu(III) and U(IV) species whose parameters are assigned by analogy with Am(III) and Th(IV), respectively. I removed most of the Pu(III) species from the new database (to make room for additional Th, Np, and Am species). Pu(III) species remaining in the new database have been deactivated by setting the normalized chemical potential (μ^0/RT, where R is the ideal gas constant and T is temperature in Kelvin) to 999.999. Setting μ^0/RT to a large positive value prevents formation of a species. Some of the U(IV) species in the old baseline database are modeled with parameters derived from U(IV) experimental data. Others are modeled by analogy with Th(IV). All U(IV) species remain in the new database; those whose parameters are assigned by analogy with Th(IV) have been deactivated.

Because I did not consistently update Pitzer parameters for the deactivated species, the Pu(III) and U(IV) species should not be reactivated without a thorough review of associated Pitzer parameters in the database.

4. Miscellaneous corrections

4.1 ClO_4^-. The ternary interaction parameters (θ and ψ) describing the interaction of ClO_4^- and Na^+ with HOx^-, H_2Cit^-, $HCit^{2-}$, and Cit^{3-} (where Ox is oxalate and Cit is citrate) have been set to zero in the new database. The nonzero values in the old baseline database were referenced to Moore (1996). This reference does not discuss interaction of ClO_4^- with aqueous organic species. Such interaction is irrelevant to geochemical models of WIPP brines, because the brines do not contain ClO_4^-.

4.2 $NpO_2Ac_{(aq)}$. The new database includes newly calculated values for parameters related to formation of $NpO_2Ac_{(aq)}$, because the values calculated by Choppin et al. (2001) and recommended in Giambalvo (2002d) are not consistent with the apparent stability constants (β_{101}) used to calculate them (Figure 1). I recalculated μ^0/RT for $NpO_2Ac_{(aq)}$ and the Pitzer parameter $\lambda(NpO_2Ac_{(aq)} - Cl^-)$ using NONLIN version 2.0[2] and the β_{101} versus NaCl molality data given in Choppin et al. (2001, Tables 20-25). The new value of μ^0/RT (-519.615) is similar to that in the old baseline database (-519.809). The new value of $\lambda(NpO_2Ac_{(aq)} - Cl^-)$ is -0.104. NONLIN input and output files are attached as an appendix.

4.3 Citations. I have updated and corrected reference citations throughout the database.

5. Compatibility with FMT version 2.4

The fictional solid species "H+(solid)" and "OH-/H2O(solid)" have been added to the new database for compatibility with FMT version 2.4[3]. Addition of these species allows simulations to be run in which pH is fixed at a user-specified value. The μ^0/RT value for the fictional solid species "CO_2 (solid)," which allows the user to specify a fixed CO_2 fugacity, has been changed to 0.0 in order to achieve a consistent format (i.e., all fictional solid species have $\mu^0/RT = 0.0$).

6. Model predictions: Comparison to old baseline database

I compared FMT predictions of Am(III), Th(IV), and Np(V) solubilities resulting from the new database to predictions resulting from the old baseline database. Each database was used for 4 solubility calculations: Brine A (Salado Brine) with organic ligands, Brine A without organic ligands, ERDA-6 (Castile brine) with organic ligands, and ERDA-6 without organic ligands. Assumptions concerning mineral equilibria and mineral suppression are the same as those used for the PAVT (Novak, 1997). Brines are forced to be in equilibrium with halite, anhydrite, and brucite. Magnesite, dolomite, calcite, aragonite, gaylussite, and pirssonite are prevented from forming in order to achieve precipitation of the metastable phase hydromagnesite ($Mg_5(CO_3)_4(OH)_2.4H_2O$). FMT input and output files are archived in the CMS (Table 1).

For both brines, with and without organic ligands, the new database results in predictions of Th(IV) and Np(V) solubilities that are essentially the same as those resulting from the old baseline database (Table 2). Predicted solubilities differ by <7%.

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[2] The NONLIN executable is stored in Sandia National Laboratories' Configuration Management System in the library WP$NONPA_CMSROOT:[NL] and class PA96 as NL_NONLIN_PA96.EXE. It runs under the VMS operating system on Sandia's WIPP DEC/COMPAQ Alpha cluster.

[3] FMT version 2.4 is the current baseline version of the software. The executable is stored in the file FMT_QB0204.EXE in Sandia National Laboratories' Configuration Management System on the WIPP DEC/COMPAQ Alpha Cluster in the library WP$NONPA_CMSROOT:[FMT] and class QB0204. It runs under the VMS operating system on the DEC/COMPAQ Alpha cluster.

The new database results in predicted Am(III) solubilities that are significantly different from solubilities resulting from the old baseline database (Table 2). For ERDA-6 brine without organics and for Brine A with and without organics, the new database results in solubilities more than twice as large as the solubilities resulting from the old baseline database. For ERDA-6 brine with organics, the new database results in a solubility about 6 times smaller than that resulting from the old baseline database. In all cases the solubility controlling solid phase is $AmOHCO_{3(c)}$ when the old database is used and $Am(OH)_{3(s)}$ when the new database is used.

In the three cases in which the new database resulted in higher Am(III) solubility, $Am(OH)_2^+$ is the dominant Am-species in solution. The new database results in larger $Am(OH)_2^+$ concentrations due to corrections to the Am(III) hydrolysis constants. In implementing the Am(III) model in the old baseline database, Novak (1996) took the solubility product constant (K_{sp}) for $Am(OH)_{3(s)}$ and the first and second hydrolysis constants for Am^{3+} from two different sources (Felmy et al. (1989) and Fanghänel et al. (1994), respectively). In order to force compatibility between the K_{sp} and the hydrolysis constants, Novak (1996) adjusted the hydrolysis constants using the constraint that

$$\beta^0_{1n}(\text{Fanghänel})K^0_{sp}(\text{Fanghänel}) = \beta^0_{1n}(\text{adjusted})K^0_{sp}(\text{Felmy}),$$

where β^0_{1n} is the appropriate hydrolysis constant at infinite dilution and the product $\beta^0_{1n}K^0_{sp}$ is the equilibrium constant at infinite dilution for the dissolution reaction

$$Am(OH)_{3(s)} = Am(OH)_n^{3-n} + (3-n)OH^-.$$

As noted in Giambalvo (2002a), one problem with this strategy is that it results in an effective K_{sp} equal to that given by Fanghänel et al. (1994) when $AmOH^+$ or $Am(OH)_2^+$ is the dominant Am-species in solution ($\log K_{sp} = -28.2$), and an effective K_{sp} equal to that given by Felmy et al. (1989) when any other Am-species dominates ($\log K_{sp} = -26.2$). Another problem with this strategy is that when $AmOH^+$ or $Am(OH)_2^+$ is the dominant Am-species in solution it results in effective K_{sp}s for all other Am-bearing solid phases that are two orders of magnitude too low. Thus equilibrium with $AmOHCO_{3(c)}$ maintains artificially low Am solubilities when the old baseline database is used. Implementation of an internally consistent Am(III) model in the new database has corrected this problem.

In the one case in which the new database resulted in lower Am(III) solubility (ERDA-6 with organics), the new database has the same effect on $Am(OH)_2^+$ concentration, but has a stronger effect on $AmEDTA^-$ concentration (Table 3). In this pair of simulations, $AmEDTA^-$ is the dominant Am-species in solution. The predicted $AmEDTA^-$ concentration resulting from the new database is almost 10 times smaller than that resulting from the old database.

7. Conclusion

The new database contains improved models for Am(III), Th(IV), and Np(V) solubility, and should be adopted as the new baseline database for CRA solubility calculations.

8. References

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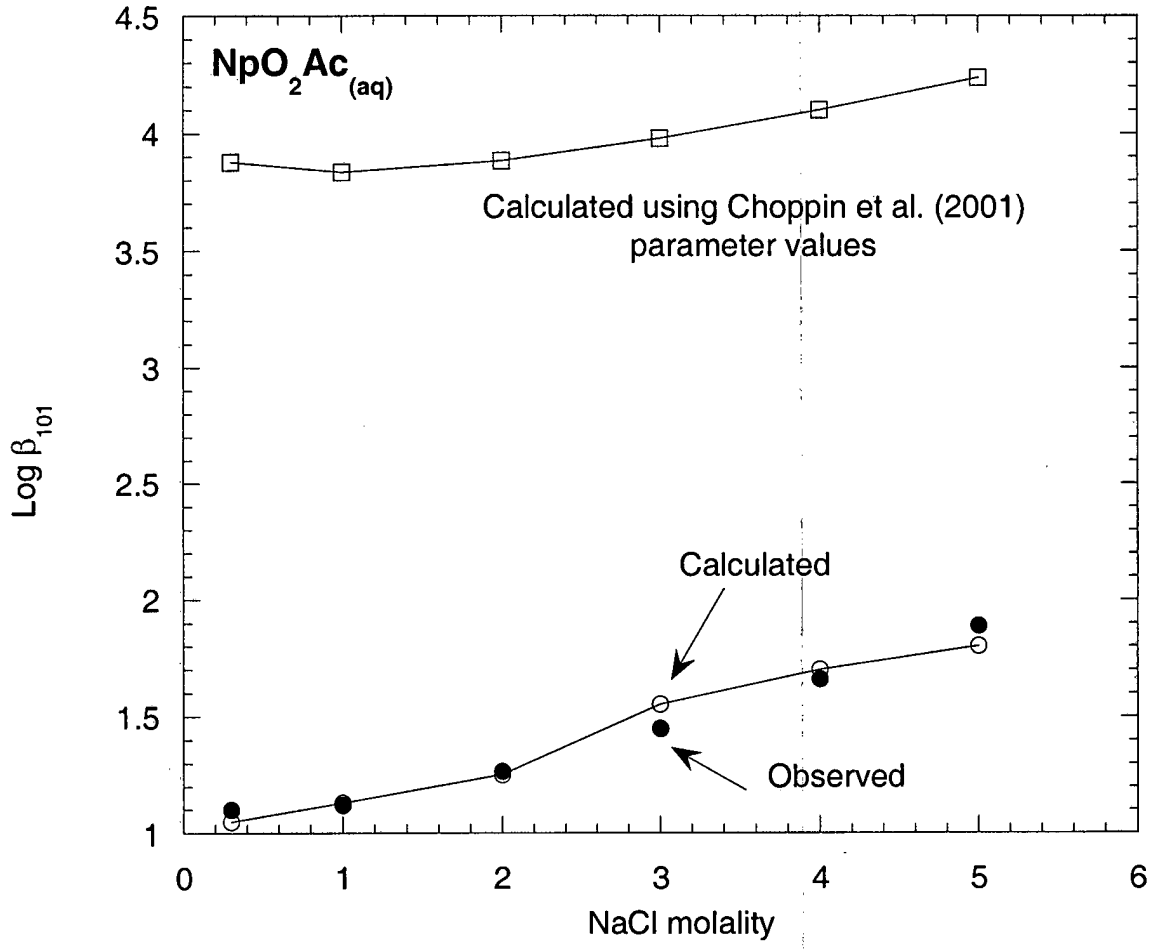

Figure 1. Log β_{101} versus NaCl molality for $NpO_2Ac_{(aq)}$. Filled circles: values from Choppin et al. (2001, Tables 20-25); open squares: model predictions calculated using $\mu^0/RT(NpO_2Ac_{(aq)})$ and $\lambda(NpO_2Ac_{(aq)} - Cl^-)$ values listed in Choppin et al. (2001); open circles: model predictions calculated using $\mu^0/RT(NpO_2Ac_{(aq)})$ and $\lambda(NpO_2Ac_{(aq)} - Cl^-)$ values calculated in this work.

Table 1. Files archived in the CMS FMT library[a], class DB2002

File Name	Purpose
FMT_PH_970407.CHEMDAT	Old baseline database (also in class QB0204)
FMT_021120.CHEMDAT	New database
FMT_021120_BRINEA_NO_ORG.IN[b]	Input for simulations of Brine A without organic ligands.
FMT_021120_BRINEA_ORG.IN[b]	Input for simulations of Brine A with organic ligands.
FMT_021120_ERDA6_NO_ORG.IN[b]	Input for simulations of ERDA-6 without organic ligands.
FMT_021120_ERDA6_ORG.IN[b]	Input for simulations of ERDA-6 with organic ligands.
FMT_021120_BRINEA_NO_ORG.OUT[c]	Output from simulation with new database of Brine A without organic ligands.
FMT_021120_BRINEA_ORG.OUT[c]	Output from simulation with new database of Brine A with organic ligands.
FMT_021120_ERDA6_NO_ORG.OUT[c]	Output from simulation with new database of ERDA-6 without organic ligands.
FMT_021120_ERDA6_ORG.OUT[c]	Output from simulation with new database of ERDA-6 with organic ligands.
FMT_970407_BRINEA_NO_ORG.OUT[c]	Output from simulation with old baseline database of Brine A without organic ligands.
FMT_970407_BRINEA_ORG.OUT[c]	Output from simulation with old baseline database of Brine A with organic ligands.
FMT_970407_ERDA6_NO_ORG.OUT[c]	Output from simulation with old baseline database of ERDA-6 without organic ligands.
FMT_970407_ERDA6_ORG.OUT[c]	Output from simulation with old baseline database of ERDA-6 with organic ligands.

[a]WP$NONPA_CMSROOT:[FMT]

[b]In all simulations, input concentrations were read from the IN file, therefore INGUESS files are not archived.

[c]The supplementary FOR088 output files are not archived, because all information contained in these files is also contained in the OUT files.

Table 2. Predicted elemental abundances in solution (mol/kg H_2O) and other output parameters

	ERDA-6				Brine A			
	no organics		with organics		no organics		with organics	
	970407	021120	970407	021120	970407	021120	970407	021120
Hydrogen	1.11E+02	1.11E+02	1.11E+02	1.11E+02	1.11E+02	1.11E+02	1.11E+02	1.11E+02
Oxygen	5.66E+01	5.66E+01	5.66E+01	5.66E+01	5.59E+01	5.59E+01	5.59E+01	5.59E+01
Sodium	6.20E+00	6.20E+00	6.19E+00	6.20E+00	4.68E+00	4.68E+00	4.67E+00	4.69E+00
Potassium	1.11E-01	1.11E-01	1.11E-01	1.11E-01	1.07E+00	1.07E+00	1.07E+00	1.07E+00
Magnesium	4.57E-02	4.57E-02	5.78E-02	5.79E-02	5.09E-01	5.09E-01	5.22E-01	5.20E-01
Calcium	1.34E-02	1.34E-02	1.42E-02	1.44E-02	3.29E-02	3.29E-02	3.33E-02	3.33E-02
Chlorine	5.97E+00	5.97E+00	5.96E+00	5.97E+00	6.68E+00	6.68E+00	6.67E+00	6.68E+00
Sulfur	1.91E-01	1.91E-01	1.91E-01	1.92E-01	6.06E-02	6.06E-02	6.10E-02	6.10E-02
Carbon	7.77E-04	7.77E-04	7.31E-04	7.41E-04	4.13E-04	4.13E-04	4.13E-04	4.13E-04
Boron	7.21E-02	7.21E-02	7.21E-02	7.21E-02	2.77E-02	2.77E-02	2.77E-02	2.77E-02
Bromine	1.26E-02	1.26E-02	1.26E-02	1.26E-02	1.39E-02	1.39E-02	1.39E-02	1.39E-02
Oxalate	0.00E+00	0.00E+00	4.78E-04	4.79E-04	0.00E+00	0.00E+00	4.79E-04	4.79E-04
Acetate	0.00E+00	0.00E+00	1.12E-03	1.12E-03	0.00E+00	0.00E+00	1.12E-03	1.12E-03
EDTA	0.00E+00	0.00E+00	4.28E-06	4.28E-06	0.00E+00	0.00E+00	4.28E-06	4.28E-06
Citrate	0.00E+00	0.00E+00	7.53E-03	7.53E-03	0.00E+00	0.00E+00	7.54E-03	7.55E-03
Th(IV)	4.63E-08	4.59E-08	4.34E-08	4.36E-08	1.37E-08	1.33E-08	1.37E-08	1.34E-08
Am(III)	1.45E-08	6.01E-08	7.54E-07	1.29E-07	1.08E-07	3.88E-07	2.29E-07	4.93E-07
Np(V)	5.28E-07	5.09E-07	6.63E-07	6.20E-07	1.38E-07	1.35E-07	1.49E-07	1.48E-07
pcH (log [mH+])	9.888	9.888	9.856	9.864	9.366	9.366	9.364	9.366
pH (log [aH+])	9.235	9.235	9.205	9.212	8.690	8.690	8.690	8.690
Ionic strength (mol/kg)	6.660	6.660	6.679	6.680	7.409	7.409	7.428	7.422

Continues on following page.

Table 2 (continued). Predicted elemental abundances in solution (mol/kg H_2O) and other output parameters

$Am(OH)_{3(s)}$	none	present	none	present	none	present	none	present
$AmOHCO_{3(c)}$	present	none	present	none	present	none	present	none
$ThO_{2(am)}$	present	present	present	present	present	present	present	present
$KNpO_2CO_3{\cdot}2H_2O_{(s)}$	present	present	present	present	present	present	present	present
Anhydrite	present	present	present	present	present	present	present	present
Halite	present	present	present	present	present	present	present	present
Brucite	present	present	present	present	present	present	present	present
Hydromagnesite $(Mg_5(CO_3)_4(OH)_2{\cdot}4H_2O)$	present	present	present	present	present	present	present	present
Mg-Oxychloride	none	none	none	none	present	present	present	present
Glauberite	present	present	present	present	none	none	none	none

Table 3. Predicted Am speciation (concentrations in mol/kg H_2O)

	ERDA-6				Brine A			
	no organics		with organics		no organics		with organics	
	970407	021120	970407	021120	970407	021120	970407	021120
$Am(CO_3)_2^-$	7.37E-11	6.39E-10	6.89E-11	6.07E-10	2.16E-11	1.58E-10	2.16E-11	1.59E-10
$Am(CO_3)_3^{3-}$	2.03E-09	3.89E-10	1.69E-09	3.39E-10	1.67E-10	3.75E-11	1.68E-10	3.80E-11
$Am(CO_3)_4^{5-}$	NA*	4.42E-12	NA*	3.68E-12	NA*	7.95E-12	NA*	8.27E-12
$Am(OH)_2^+$	1.19E-08	5.79E-08	1.26E-08	6.11E-08	1.05E-07	3.84E-07	1.04E-07	3.83E-07
$Am(OH)_{3(aq)}$	1.59E-10	8.56E-10	1.59E-10	8.57E-10	1.54E-10	9.09E-10	1.54E-10	9.11E-10
$Am(SO_4)_2^-$	NA*	2.53E-13	NA*	2.99E-13	NA*	1.06E-12	NA*	1.07E-12
Am^{3+}	1.23E-11	4.72E-13	1.56E-11	5.59E-13	4.80E-10	2.84E-11	4.92E-10	2.87E-11
$AmAcetate^{2+}$	NA*	NA*	9.96E-12	4.62E-12	NA*	NA*	1.23E-10	1.14E-10
$AmCitrate_{(aq)}$	NA*	NA*	1.60E-08	9.84E-10	NA*	NA*	4.33E-08	5.69E-09
$AmCl^{2+}$	NA*	3.17E-14	NA*	3.78E-14	NA*	9.23E-13	NA*	9.30E-13
$AmCl_2^+$	NA*	1.12E-15	NA*	1.33E-15	NA*	5.44E-14	NA*	5.46E-14
$AmCO_3^+$	2.85E-10	1.16E-10	3.05E-10	1.22E-10	1.13E-09	3.92E-10	1.13E-09	3.92E-10
$AmEDTA^-$	NA*	NA*	7.23E-07	6.52E-08	NA*	NA*	7.75E-08	1.00E-07
$AmOH^{2+}$	2.21E-11	2.04E-10	2.55E-11	2.29E-10	2.43E-10	2.33E-09	2.44E-10	2.34E-09
$AmOxalate^+$	NA*	NA*	2.22E-10	3.28E-12	NA*	NA*	1.46E-09	9.15E-12
$AmSO_4^+$	NA*	2.11E-12	NA*	2.49E-12	NA*	2.96E-11	NA*	2.96E-11

*NA: Not applicable, species not included in simulation.

9. Appendix: NONLIN input and output files for $NpO_2Ac_{(aq)}$ calculations

This appendix contains:

Database files:

- BINARYP.DAT
- COMP.DAT
- LAMBDA.DAT
- TERNARYP.DAT

Input and output files for parameter calculation:

- NPO2AC.COM
- NPO2AC.GMIN
- NPO2AC.IN
- NPO2AC.OUT

Input and output files for parameter verification:

- REV_NPO2AC.COM
- REV_NPO2AC.GMIN
- REV_NPO2AC.IN
- REV_NPO2AC.OUT

Input and output files for checking Choppin et al. (2001) parameter values:

- REV_NPO2AC_CH.COM
- REV_NPO2AC_CH.GMIN
- REV_NPO2AC_CH.IN
- REV_NPO2AC_CH.OUT

Brush, Laurence H

From: Kessel, David S DSK 3/12/03
Sent: Friday, March 07, 2003 8:57 AM
To: Mitchell, Marty J
Cc: Brush, Laurence H; Xiong, Yongliang; Chavez, Mario Joseph
Subject: Release of FMT database FMT_021120.CHEMDAT

Marty,

As you are aware Emily Giambalvo has left the project. In her absence I am delegating comment resolution and signature authority for the subject document to Xiong Yongliang. Please contact me if you have any questions regarding this issue.

Dave Kessel
Manager Performance Assessment Department 6821

```
011000 000170   .0765   .2664    .000 .00127   Na+ - Cl-       HMW84
001000 000170   .1775   .2945    .000  .0008   H+ - Cl-        HMW84
093080 000170   .1415   .281     .000  .0000   NpO2+ Cl-       Neck et al. 1995
093081 000170   .1415   .281     .000  .0000   NpO2+F Cl-
095000 000170   .6117 5.403     .000 -.0284   Am+++ - Cl-     FRF90
095001 000170   .6117 5.403     .000 -.0284   Am+++F - Cl-
090000 000170 1.092    13.7    -160.  -.112   Th++++ - Cl-    Roy92
090001 000170 1.092    13.7    -160.  -.112   Th++++F - Cl-
012000 000170 .35235 1.6815     .0    .00519  Mg++ - Cl-      HMW84
012001 000170 .35235 1.6815     .0    .00519  Mg++F - Cl-
011000 000990 -.2176   1.74     .000   .122   Na+ - Ox=       SAND99/Mizera
011000 000991 -.2176   1.74     .000   .122   Na+ - Ox=F
011000 000890  .1426    .22     .000-.00629   Na+ - Ac-       NBC96
011000 000891  .1426    .22     .000-.00629   Na+ - Ac-F
011000 000860 0.0887   5.22     .0    .047    Na+ - Cit=-    SAND99/Mizera
011000 000861 0.0887   5.22     .0    .047    Na+ - Cit=-F
011000 000870  1.016   11.6     .0    .001    Na+ - EDTA==   SAND99/Mizera
011000 000871  1.016   11.6     .0    .001    Na+ - EDTA==F
011000 012861   .000   0.29     .0    .000     Na+ - MgCit-F  Fix Beta(1) followin
011000 012860   .1742  0.29     .0  -.06923    Na+ - MgCit-
011000 012870   .2134  1.74     .0    .00869   Na+ - MgEDTA=
011000 012871   .000   1.74     .0    .000     Na+ - MgEDTA=F Fix Beta(1) followin
095890 000170 0.3088   1.74     .000 -.132     AmAc++ - Cl-
095891 000170 0.000    1.74     .000 -.000     AmAc++F - Cl-  Fix Beta(1) followin
095892 000170 1.014    1.74     .000 -.265     AmAc++moore - Cl-   SAND99/Moore et
012890 000170-.0833    0.29     .000  .0987   MgAc+ - Cl-
012891 000170 0.0      0.29     .000  .000    MgAc+F - Cl-   Fix Beta(1) followin

000000

cation anion   beta0 beta1   beta2  cphi
```

References:

HMW84 = Harvie, Moller, and Weare, 1984. Geochimica et Cosmochimica Acta 48:723-
SAND99 = Choppin et al., 2001. SAND99-0943.
Moore et al 99 = Moore et al., 1999. J. Soln. Chem. 28:521-531.
Mizera = Mizera et al., 1999. in Actinide Speciation in High Ionic Strength Medi
Reed et al., p. 113-124.
FRF90 = Felmy, Rai, and Fulton, 1990. Radiochimica Acta 50:193-204.
Roy92 = Roy et al., 1992. J. Phys. Chem. 96:11065-11072.
NBC96 = Novak, Borkowski, and Choppin, 1996. Radiochimica Acta 74:111-116.
Neck et al. 1995 = Neck, Fanghanel, Rudolph, and Kim, 1995. RCA 69:39-47.

```
001080H2O            0       -95.6635  1 2.0 8 1.0
001000H+             1        000.000  1 1.0
011000Na+            1       -105.651 11 1.0
012000Mg++           2       -183.468 12 1.0
012001Mg++F          2       -183.468 12 1.072 1.0
000170Cl-           -1       -52.9550 17 1.0

093080NpO2+          1       -369.105 93 1.0 8 2.0
093081NpO2+F         1       -369.105 93 1.0 8 2.072 1.0
093890NpO2Ac(aq)     0       -519.615 93 1.0 8 2.089 1.0
093891NpO2Ac(aq)F    0        999.999 93 1.0 8 2.089 1.073 1.0
093892NpO2Ac(aq)Ch   0       -526.061 93 1.0 8 2.089 1.0

090000Th++++         4       -284.227 90 1.0
090001Th++++F        4       -284.227 90 1.072 1.0
095000Am+++          3       -241.694 95 1.0
095001Am+++F         3       -241.694 95 1.072 1.0

095890AmAc++         2       -395.356 95 1.089 1.0
095891AmAc++F        2        999.999 95 1.089 1.073 1.0
095892AmAc++moore    2       -395.239 95 1.089 1.0

090870ThEDTA         0       -285.419 90 1.087 1.0
090871ThEDTAF        0        999.999 90 1.087 1.073 1.0

000890Ac-           -1       -147.347 89 1.0
000891Ac-F          -1       -147.347 89 1.071 1.0

000990Ox=           -2         13.017 99 1.0
000991Ox=F          -2         13.017 99 1.071 1.0

000870EDTA==        -4          53.05 87 1.0
000871EDTA==F       -4          53.05 87 1.071 1.0

000860Cit=-         -3         33.410 86 1.0
000861Cit=-F        -3         33.410 86 1.071 1.0

012890MgAc+          1       -333.378 89 1.012 1.0
012891MgAc+F         1        999.999 89 1.012 1.073 1.0

012990MgOx           0       -179.185 99 1.012 1.0
012991MgOxF          0        999.99  99 1.012 1.073 1.0

012870MgEDTA=       -2       -153.734 87 1.012 1.0
012871MgEDTA=F      -2        999.999 87 1.012 1.073 1.0

012860MgCit-        -1       -162.261 86 1.012 1.0
012861MgCit-F       -1        999.999 86 1.012 1.073 1.0

193890NpO2Ac/Np/Ac   0          0.000 73 1.071-1.072-1.0
195890AmAc/Am/Ac(s)  0          0.000 73 1.071-1.072-1.0
190870ThEDTA/Th/EDT  0          0.000 73 1.071-1.072-1.0
112890MgAc/Mg/Ac(s)  0          0.000 73 1.071-1.072-1.0
112990MgOx/Mg/Ox(s)  0          0.000 73 1.071-1.072-1.0
112870MgEDTA/Mg/EDT  0          0.000 73 1.071-1.072-1.0
112860MgCit/Mg/Cit   0          0.000 73 1.071-1.072-1.0

    -1


Reference keys in order are:
H+ HMW84
Na+ HMW84
Mg++ HMW84
```

Cl- HMW84
NpO2+ ERG_Np_memo
NpO2Ac(aq)Ch SAND99
Th++++ FRM91
Am+++ FRF90
AmAc++moore SAND99/Moore et al. 99
Ac- NBC96
Ox= SAND99/Mizera
EDTA== SAND99/Mizera
Cit=- SAND99/Mizera

References:
HMW84 = Harvie, Moller, and Weare, 1984. Geochimica et Cosmochimica Acta 48:723-
ERG_Np_memo = Giambalvo memo to Brush, 26 July 2002, ERMS# 522990
SAND99 = Choppin et al., 2001. SAND99-0943.
Moore et al 99 = Moore et al., 1999. J. Soln. Chem. 28:521-531.
Mizera = Mizera et al., 1999. in Actinide Speciation in High Ionic Strength Medi
Reed et al., p. 113-124.
FRF90 = Felmy, Rai, and Fulton, 1990. Radiochimica Acta 50:193-204.
NBC96 = Novak, Borkowski, and Choppin, 1996. Radiochimica Acta 74:111-116.
FRM91 = Felmy, Rai, and Mason, 1991. Radiochimica Acta 55:177-185.

```
012990 000170 0.0189
012991 000170  0.000
090870 000170 0.1111
090871 000170 0.0000
093890 000170 -.1040
093891 000170 0.0000
000000

MgOx Cl lambda
MgOxF Cl lambda
ThEDTA Cl lambda
ThEDTAF Cl lambda
NpO2Ac(aq) Cl lambda
NpO2Ac(aq)F Cl lambda

Neutral/ion interaction parameters
```

```
011000 001000   .036 000170   -.004
011000 012000   .07  000170   -.012
011000 012001   .07  000170   -.012
012000 001000   .10  000170   -.011
012001 001000   .10  000170   -.011
000170 000890  -.090 011000  .01029
000170 000891  -.090 011000  .01029
090000 001000   .600 000170  .37
090001 001000   .600 000170  .37
090000 011000   .420 000170  .21
090001 011000   .420 000170  .21
000000
```

Na+	H+	theta	Cl-	psi	HMW84
Na+	Mg++	theta	Cl-	psi	HMW84
Na+	Mg++F	theta	Cl-	psi	
Mg++	H+	theta	Cl-	psi	HMW84
Mg++F	H+	theta	Cl-	psi	
Cl-	Ac-	theta	Na+	psi	NBC96
Cl-	Ac-F	theta	Na+	psi	
Th++++	H+	theta	Cl-	psi	Roy92
Th++++F	H+	theta	Cl-	psi	
Th++++	Na+	theta	Cl-	psi	RFSMMN
Th++++F	Na+	theta	Cl-	psi	

References:

RFSMMN = Rai et al., 1997. Radiochimica Acta 79:239-247.
HMW84 = Harvie, Moller, and Weare, 1984. Geochimica et Cosmochimica Acta 48:723-
Roy92 = Roy et al., 1992. J. Phys. Chem. 96:11065-11072.
NBC96 = Novak, Borkowski, and Choppin, 1996. Radiochimica Acta 74:111-116.

```
$ set def u1:[ergiamb.nonlin.npo2ac]
$!
$ define GMIN npo2ac.gmin
$ define INPUT npo2ac.in
$ define COMP comp.dat
$ define BINARYP binaryp.dat
$ define LAMBDA lambda.dat
$ define TERNARYP ternaryp.dat
$ define OUTPUT npo2ac.out
$!
run u1:[ergiamb.nonlin.cms_files]nl_nonlin_pa96.exe
$!
$ deassign GMIN
$ deassign INPUT
$ deassign COMP
$ deassign BINARYP
$ deassign LAMBDA
$ deassign TERNARYP
$ deassign OUTPUT
end
```

```
    0     0     0     0
0
001080      0.00000000       1   0 H2O
011000      0.00000000       2   1 Na+
001000      0.00000000       3   2 H+
093081      0.00000000       4   3 NpO2+F
093891      0.00000000       5   1 NpO2Ac(aq)F
000170      0.00000000       6   1 Cl-
000891      0.00000000       7   2 Ac-F
000000
193890      0.00000000       8    NpO2Ac/Np/Ac
-1
-1
```

```
000005 -300.0          u0 NpO2Ac(aq)F
100101  0.1            lambda NpO2Ac(aq)F Cl-
-1
     6   -1
0   0  0.3000  1.000E-07  0.01000000  0.00112    0.30000  0.01000
0   0  1.0000  1.000E-07  0.01000000  0.00135    1.00000  0.01000
0   0  2.0000  1.000E-07  0.01000000  0.00178    2.00000  0.01000
0   0  3.0000  1.000E-07  0.01000000  0.00355    3.00000  0.01000
0   0  4.0000  1.000E-07  0.01000000  0.00501    4.00000  0.01000
0   0  5.0000  1.000E-07  0.01000000  0.00631    5.00000  0.01000
-1

0   0  Na+       H+          NpO2+      NpO2Ac(aq)    Cl-       Ac-
```

NONLIN V2.0

NONLIN was developed by A.R. Felmy

INPUT file name is U1:[ERGIAMB.NONLIN.NPO2AC]NPO2AC.IN;1
GMIN file name is U1:[ERGIAMB.NONLIN.NPO2AC]NPO2AC.GMIN;1
OUTPUT file name is U1:[ERGIAMB.NONLIN.NPO2AC]NPO2AC.OUT;1

AQUEOUS SPECIES

ID	NAME	MOLES	Z	u0rt
1080	H2O	0.000000000000	0.	-95.663
11000	Na+	0.000000000000	1.	-105.651
1000	H+	0.000000000000	1.	0.000
93081	NpO2+F	0.000000000000	1.	-369.105
93891	NpO2Ac(aq)F	0.000000000000	0.	999.999
170	Cl-	0.000000000000	-1.	-52.955
891	Ac-F	0.000000000000	-1.	-147.347

SOLID PHASES

ID	NAME	MOLES	Z	u0rt
193890	NpO2Ac/Np/Ac	0.000000000000	0.	0.000

CONSTRAINT EQUATIONS

	0	1	8	11	93	72	89	73	17
H2O	0.0	2.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0
Na+	1.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0
H+	1.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
NpO2+F	1.0	0.0	2.0	0.0	1.0	1.0	0.0	0.0	0.0
NpO2Ac(aq)F	0.0	0.0	2.0	0.0	1.0	0.0	1.0	1.0	0.0
Cl-	-1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0
Ac-F	-1.0	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0
NpO2Ac/Np/Ac	0.0	0.0	0.0	0.0	0.0	-1.0	0.0	1.0	0.0

Closed input files GMIN and COMP

COMP file name is U1:[ERGIAMB.NONLIN.NPO2AC]COMP.DAT;1
BINARYP file name is U1:[ERGIAMB.NONLIN.NPO2AC]BINARYP.DAT;1
TERNARYP file name is U1:[ERGIAMB.NONLIN.NPO2AC]TERNARYP.DAT;1
LAMBDA file name is U1:[ERGIAMB.NONLIN.NPO2AC]LAMBDA.DAT;1

non-ideal electrolyte parameters

single electrolyte parameters

Na+	Cl-	0.07650	0.26640	0.00000	0.00127
Na+	Ac-F	0.14260	0.22000	0.00000	-0.00629
H+	Cl-	0.17750	0.29450	0.00000	0.00080
H+	Ac-F	0.00000	0.00000	0.00000	0.00000
NpO2+F	Cl-	0.14150	0.28100	0.00000	0.00000
NpO2+F	Ac-F	0.00000	0.00000	0.00000	0.00000

ternary electrolyte parameters

			Cl-	Ac-F
Na+	H+	0.03600	-0.00400	0.00000
Na+	NpO2+F	0.00000	0.00000	0.00000
H+	NpO2+F	0.00000	0.00000	0.00000

			Na+	H+	NpO2+F
Cl-	Ac-F	-0.09000	0.01029	0.00000	0.00000

neutral ion parameters

	NpO2Ac(aq)F
Na+	0.0000

```
H+                0.0000
NpO2+F            0.0000
Cl-               0.0000
Ac-F              0.0000
NpO2Ac(aq)F       0.0000

        higher order lambdas

                        Cl-          Ac-F
NpO2Ac(aq)F Na+          0.00000      0.00000
NpO2Ac(aq)F H+           0.00000      0.00000
NpO2Ac(aq)F NpO2+F       0.00000      0.00000

 TOTAL NUMBER OF SPECIES =  8
 NUMBER OF COMPONENTS    = 10
 INDEPENDENT CONSTRAINTS =  7

          SOLUBILITY DATA

          input molalities


   Na+          H+           NpO2+F       NpO2Ac(aq)F  Cl-           Ac-F
   3.000E-01    1.000E-07    1.000E-02    1.120E-03    3.000E-01    1.000E-02
   1.000E+00    1.000E-07    1.000E-02    1.350E-03    1.000E+00    1.000E-02
   2.000E+00    1.000E-07    1.000E-02    1.780E-03    2.000E+00    1.000E-02
   3.000E+00    1.000E-07    1.000E-02    3.550E-03    3.000E+00    1.000E-02
   4.000E+00    1.000E-07    1.000E-02    5.010E-03    4.000E+00    1.000E-02
   5.000E+00    1.000E-07    1.000E-02    6.310E-03    5.000E+00    1.000E-02

 -3.0000E+02  1.0000E-01
           L2 NORM OF THE RESIDUALS  5.4049534E+02



 -5.1961E+02 -1.0401E-01
           L2 NORM OF THE RESIDUALS  3.4260887E-01



 -5.1961E+02 -1.0401E-01
           L2 NORM OF THE RESIDUALS  3.4260887E-01



          ADJUSTED PARAMETERS
  u0rt(                          NpO2Ac(aq)F )   -5.1961477E+02
lambda(            NpO2Ac(aq)F Cl-           )   -1.0401487E-01

 DATA SET( 1)

  input     calc       diff
 0.000000000000000E+000  0.117052858444651
 0.000000000000000E+000 -2.027867567011299E-002
 0.000000000000000E+000  2.970997461974888E-002
 0.000000000000000E+000 -0.232574063358413
 0.000000000000000E+000 -9.301552937407108E-002
 0.000000000000000E+000  0.199105435338026

 AVERAGE DEVIATION   =  1.1529E-01

 STANDARD DEVIATION =  1.3987E-01

FINAL MOLALITIES
```

Na+	H+	NpO2+F	NpO2Ac(aq)F	Cl-	Ac-F
3.000E-01	1.000E-07	1.000E-02	1.120E-03	3.000E-01	1.000E-02
1.000E+00	1.000E-07	1.000E-02	1.350E-03	1.000E+00	1.000E-02
2.000E+00	1.000E-07	1.000E-02	1.780E-03	2.000E+00	1.000E-02
3.000E+00	1.000E-07	1.000E-02	3.550E-03	3.000E+00	1.000E-02
4.000E+00	1.000E-07	1.000E-02	5.010E-03	4.000E+00	1.000E-02
5.000E+00	1.000E-07	1.000E-02	6.310E-03	5.000E+00	1.000E-02

FINAL LOG ACTIVITIES

Na+	H+	NpO2+F	NpO2Ac(aq)F	Cl-	Ac-F
-6.743E-01	-7.114E+00	-2.135E+00	-2.978E+00	-6.750E-01	-2.166E+00
-1.834E-01	-7.061E+00	-2.126E+00	-2.960E+00	-1.842E-01	-2.217E+00
1.256E-01	-6.937E+00	-2.061E+00	-2.930E+00	1.248E-01	-2.229E+00
3.306E-01	-6.798E+00	-1.978E+00	-2.721E+00	3.296E-01	-2.217E+00
4.957E-01	-6.652E+00	-1.885E+00	-2.662E+00	4.946E-01	-2.190E+00
6.405E-01	-6.502E+00	-1.786E+00	-2.652E+00	6.393E-01	-2.153E+00

Calculated Log Activity Coefficients

Na+	H+	NpO2+F	NpO2Ac(aq)F	Cl-	Ac-F
-0.1515	-0.1145	-0.1349	-0.0271	-0.1521	-0.1657
-0.1834	-0.0610	-0.1256	-0.0903	-0.1842	-0.2168
-0.1754	0.0626	-0.0615	-0.1807	-0.1762	-0.2295
-0.1466	0.2019	0.0217	-0.2710	-0.1475	-0.2171
-0.1064	0.3481	0.1148	-0.3614	-0.1074	-0.1904
-0.0585	0.4979	0.2144	-0.4517	-0.0597	-0.1532

SOLID PHASE NOW IN EQUILIBRIUM

DATA SET(1)

input	calc	diff
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	

AVERAGE DEVIATION = 0.0000E+00

STANDARD DEVIATION = 0.0000E+00

FINAL MOLALITIES

Na+	H+	NpO2+F	NpO2Ac(aq)F	Cl-	Ac-F
3.000E-01	1.000E-07	1.000E-02	1.120E-03	3.000E-01	1.000E-02
1.000E+00	1.000E-07	1.000E-02	1.350E-03	1.000E+00	1.000E-02
2.000E+00	1.000E-07	1.000E-02	1.780E-03	2.000E+00	1.000E-02
3.000E+00	1.000E-07	1.000E-02	3.550E-03	3.000E+00	1.000E-02
4.000E+00	1.000E-07	1.000E-02	5.010E-03	4.000E+00	1.000E-02
5.000E+00	1.000E-07	1.000E-02	6.310E-03	5.000E+00	1.000E-02

FINAL LOG ACTIVITIES

Na+	H+	NpO2+F	NpO2Ac(aq)F	Cl-	Ac-F
-6.743E-01	-7.114E+00	-2.135E+00	-2.978E+00	-6.750E-01	-2.166E+00
-1.834E-01	-7.061E+00	-2.126E+00	-2.960E+00	-1.842E-01	-2.217E+00
1.256E-01	-6.937E+00	-2.061E+00	-2.930E+00	1.248E-01	-2.229E+00
3.306E-01	-6.798E+00	-1.978E+00	-2.721E+00	3.296E-01	-2.217E+00
4.957E-01	-6.652E+00	-1.885E+00	-2.662E+00	4.946E-01	-2.190E+00
6.405E-01	-6.502E+00	-1.786E+00	-2.652E+00	6.393E-01	-2.153E+00

Calculated Log Activity Coefficients

Na+	H+	NpO2+F	NpO2Ac(aq)F	Cl-	Ac-F
-0.1515	-0.1145	-0.1349	-0.0271	-0.1521	-0.1657
-0.1834	-0.0610	-0.1256	-0.0903	-0.1842	-0.2168
-0.1754	0.0626	-0.0615	-0.1807	-0.1762	-0.2295
-0.1466	0.2019	0.0217	-0.2710	-0.1475	-0.2171
-0.1064	0.3481	0.1148	-0.3614	-0.1074	-0.1904
-0.0585	0.4979	0.2144	-0.4517	-0.0597	-0.1532

FINAL L2 NORM OF THE RESIDUALS 3.4260887E-01

EXIT PARAMETER 3

FINAL APPROXIMATE SOLUTION

0 0 5 -5.1961477E+02
10 1 1 -1.0401487E-01

```
$ set def u1:[ergiamb.nonlin.npo2ac]
$!
$ define GMIN rev_npo2ac.gmin
$ define INPUT rev_npo2ac.in
$ define COMP comp.dat
$ define BINARYP binaryp.dat
$ define LAMBDA lambda.dat
$ define TERNARYP ternaryp.dat
$ define OUTPUT rev_npo2ac.out
$!
run u1:[ergiamb.nonlin.cms_files]nl_nonlin_pa96.exe
$!
$ deassign GMIN
$ deassign INPUT
$ deassign COMP
$ deassign BINARYP
$ deassign LAMBDA
$ deassign TERNARYP
$ deassign OUTPUT
end
```

```
    0    0    0    0
0
001080      0.00000000       1  0 H2O
011000      0.00000000       2  1 Na+
001000      0.00000000       3  2 H+
093080      0.00000000       4  3 NpO2+
093890      0.00000000       5  1 NpO2Ac(aq)
000170      0.00000000       6  1 Cl-
000890      0.00000000       7  2 Ac-
000000
-1
-1
```

```
-1
     6   -1
0   0   0.3000   1.000E-07   0.01000000   0.00112    0.30000   0.01000
0   0   1.0000   1.000E-07   0.01000000   0.00135    1.00000   0.01000
0   0   2.0000   1.000E-07   0.01000000   0.00178    2.00000   0.01000
0   0   3.0000   1.000E-07   0.01000000   0.00355    3.00000   0.01000
0   0   4.0000   1.000E-07   0.01000000   0.00501    4.00000   0.01000
0   0   5.0000   1.000E-07   0.01000000   0.00631    5.00000   0.01000
-1

0   0   Na+      H+          NpO2+        NpO2Ac(aq)   Cl-       Ac-
```

NONLIN V2.0

NONLIN was developed by A.R. Felmy

INPUT file name is U1:[ERGIAMB.NONLIN.NPO2AC]REV_NPO2AC.IN;1
GMIN file name is U1:[ERGIAMB.NONLIN.NPO2AC]REV_NPO2AC.GMIN;1
OUTPUT file name is U1:[ERGIAMB.NONLIN.NPO2AC]REV_NPO2AC.OUT;1

AQUEOUS SPECIES

ID	NAME	MOLES	Z	u0rt
1080	H2O	0.000000000000	0.	-95.663
11000	Na+	0.000000000000	1.	-105.651
1000	H+	0.000000000000	1.	0.000
93080	NpO2+	0.000000000000	1.	-369.105
93890	NpO2Ac(aq)	0.000000000000	0.	-519.615
170	Cl-	0.000000000000	-1.	-52.955
890	Ac-	0.000000000000	-1.	-147.347

SOLID PHASES

ID	NAME	MOLES	Z	u0rt

CONSTRAINT EQUATIONS

	0	1	8	11	93	89	17
H2O	0.0	2.0	1.0	0.0	0.0	0.0	0.0
Na+	1.0	0.0	0.0	1.0	0.0	0.0	0.0
H+	1.0	1.0	0.0	0.0	0.0	0.0	0.0
NpO2+	1.0	0.0	2.0	0.0	1.0	0.0	0.0
NpO2Ac(aq)	0.0	0.0	2.0	0.0	1.0	1.0	0.0
Cl-	-1.0	0.0	0.0	0.0	0.0	0.0	1.0
Ac-	-1.0	0.0	0.0	0.0	0.0	1.0	0.0

Closed input files GMIN and COMP

COMP file name is U1:[ERGIAMB.NONLIN.NPO2AC]COMP.DAT;1
BINARYP file name is U1:[ERGIAMB.NONLIN.NPO2AC]BINARYP.DAT;1
TERNARYP file name is U1:[ERGIAMB.NONLIN.NPO2AC]TERNARYP.DAT;1
LAMBDA file name is U1:[ERGIAMB.NONLIN.NPO2AC]LAMBDA.DAT;1

non-ideal electrolyte parameters

single electrolyte parameters

Na+	Cl-	0.07650	0.26640	0.00000	0.00127
Na+	Ac-	0.14260	0.22000	0.00000	-0.00629
H+	Cl-	0.17750	0.29450	0.00000	0.00080
H+	Ac-	0.00000	0.00000	0.00000	0.00000
NpO2+	Cl-	0.14150	0.28100	0.00000	0.00000
NpO2+	Ac-	0.00000	0.00000	0.00000	0.00000

ternary electrolyte parameters

			Cl-	Ac-	
Na+	H+	0.03600	-0.00400	0.00000	
Na+	NpO2+	0.00000	0.00000	0.00000	
H+	NpO2+	0.00000	0.00000	0.00000	

			Na+	H+	NpO2+
Cl-	Ac-	-0.09000	0.01029	0.00000	0.00000

neutral ion parameters

	NpO2Ac(aq)
Na+	0.0000
H+	0.0000
NpO2+	0.0000

Cl-	-0.1040
Ac-	0.0000
NpO2Ac(aq)	0.0000

higher order lambdas

		Cl-	Ac-
NpO2Ac(aq)	Na+	0.00000	0.00000
NpO2Ac(aq)	H+	0.00000	0.00000
NpO2Ac(aq)	NpO2+	0.00000	0.00000

TOTAL NUMBER OF SPECIES = 7
NUMBER OF COMPONENTS = 7
INDEPENDENT CONSTRAINTS = 6

SOLUBILITY DATA

input molalities

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
3.000E-01	1.000E-07	1.000E-02	1.120E-03	3.000E-01	1.000E-02
1.000E+00	1.000E-07	1.000E-02	1.350E-03	1.000E+00	1.000E-02
2.000E+00	1.000E-07	1.000E-02	1.780E-03	2.000E+00	1.000E-02
3.000E+00	1.000E-07	1.000E-02	3.550E-03	3.000E+00	1.000E-02
4.000E+00	1.000E-07	1.000E-02	5.010E-03	4.000E+00	1.000E-02
5.000E+00	1.000E-07	1.000E-02	6.310E-03	5.000E+00	1.000E-02

ADJUSTED PARAMETERS

DATA SET(1)

input	calc	diff
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	

AVERAGE DEVIATION = 0.0000E+00

STANDARD DEVIATION = 0.0000E+00

FINAL MOLALITIES

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
3.000E-01	1.000E-07	9.888E-03	1.232E-03	3.000E-01	9.888E-03
1.000E+00	1.000E-07	1.002E-02	1.329E-03	1.000E+00	1.002E-02
2.000E+00	1.000E-07	9.960E-03	1.820E-03	2.000E+00	9.960E-03
3.000E+00	1.000E-07	1.047E-02	3.082E-03	3.000E+00	1.047E-02
4.000E+00	1.000E-07	1.023E-02	4.779E-03	4.000E+00	1.023E-02
5.000E+00	1.000E-07	9.443E-03	6.867E-03	5.000E+00	9.443E-03

FINAL LOG ACTIVITIES

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
-6.743E-01	-7.114E+00	-2.140E+00	-2.937E+00	-6.750E-01	-2.171E+00
-1.834E-01	-7.061E+00	-2.125E+00	-2.967E+00	-1.842E-01	-2.216E+00
1.256E-01	-6.937E+00	-2.063E+00	-2.921E+00	1.248E-01	-2.231E+00
3.306E-01	-6.798E+00	-1.958E+00	-2.782E+00	3.297E-01	-2.197E+00
4.957E-01	-6.652E+00	-1.875E+00	-2.682E+00	4.946E-01	-2.180E+00
6.404E-01	-6.502E+00	-1.810E+00	-2.615E+00	6.392E-01	-2.178E+00

Calculated Log Activity Coefficients

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
-0.1514	-0.1145	-0.1348	-0.0271	-0.1521	-0.1657
-0.1834	-0.0610	-0.1256	-0.0903	-0.1842	-0.2168
-0.1754	0.0626	-0.0615	-0.1807	-0.1762	-0.2294
-0.1465	0.2018	0.0217	-0.2710	-0.1475	-0.2171
-0.1063	0.3481	0.1148	-0.3613	-0.1074	-0.1904
-0.0586	0.4979	0.2144	-0.4517	-0.0598	-0.1532

SOLID PHASE NOW IN EQUILIBRIUM

DATA SET(1)

input	calc	diff
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	

AVERAGE DEVIATION = 0.0000E+00

STANDARD DEVIATION = 0.0000E+00

FINAL MOLALITIES

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
3.000E-01	3.964E+01	9.911E+00	1.112E-02	4.985E+01	7.000E-15
1.000E+00	3.948E+01	9.869E+00	1.135E-02	5.034E+01	7.000E-15
2.000E+00	3.923E+01	9.809E+00	1.178E-02	5.104E+01	7.000E-15
3.000E+00	3.899E+01	9.748E+00	1.355E-02	5.174E+01	7.000E-15
4.000E+00	3.875E+01	9.687E+00	1.501E-02	5.243E+01	7.000E-15
5.000E+00	3.850E+01	9.625E+00	1.631E-02	5.313E+01	7.000E-15

FINAL LOG ACTIVITIES

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
1.829E+00	1.587E+01	1.079E+01	-8.767E+00	1.503E+01	-1.995E+01
2.442E+00	1.589E+01	1.089E+01	-8.811E+00	1.499E+01	-1.969E+01
2.874E+00	1.593E+01	1.102E+01	-8.871E+00	1.493E+01	-1.932E+01
3.184E+00	1.596E+01	1.114E+01	-8.884E+00	1.488E+01	-1.894E+01
3.444E+00	1.598E+01	1.127E+01	-8.913E+00	1.482E+01	-1.856E+01
3.679E+00	1.600E+01	1.140E+01	-8.950E+00	1.478E+01	-1.818E+01

Calculated Log Activity Coefficients

Na+	H+	NpO2+	NpO2Ac(aq)	Cl-	Ac-
2.3515	14.2684	9.7971	-6.8127	13.3330	-5.7956
2.4421	14.2979	9.8908	-6.8662	13.2859	-5.5353
2.5735	14.3356	10.0236	-6.9418	13.2219	-5.1611
2.7069	14.3685	10.1551	-7.0163	13.1615	-4.7843
2.8423	14.3964	10.2854	-7.0898	13.1048	-4.4050
2.9796	14.4194	10.4142	-7.1621	13.0517	-4.0235

FINAL L2 NORM OF THE RESIDUALS 0.0000000E+00

EXIT PARAMETER 0

FINAL APPROXIMATE SOLUTION

```
$ set def u1:[ergiamb.nonlin.npo2ac]
$!
$ define GMIN rev_npo2ac_ch.gmin
$ define INPUT rev_npo2ac_ch.in
$ define COMP comp.dat
$ define BINARYP binaryp.dat
$ define LAMBDA lambda.dat
$ define TERNARYP ternaryp.dat
$ define OUTPUT rev_npo2ac_ch.out
$!
run u1:[ergiamb.nonlin.cms_files]nl_nonlin_pa96.exe
$!
$ deassign GMIN
$ deassign INPUT
$ deassign COMP
$ deassign BINARYP
$ deassign LAMBDA
$ deassign TERNARYP
$ deassign OUTPUT
end
```

```
    0     0     0     0
0
001080       0.00000000        1   0 H2O
011000       0.00000000        2   1 Na+
001000       0.00000000        3   2 H+
093080       0.00000000        4   3 NpO2+
093892       0.00000000        5   1 NpO2Ac(aq)Ch
000170       0.00000000        6   1 Cl-
000890       0.00000000        7   2 Ac-
000000
-1
-1
```

```
-1
     6   -1
0   0   0.3000   1.000E-07   0.01000000   0.00112    0.30000   0.01000
0   0   1.0000   1.000E-07   0.01000000   0.00135    1.00000   0.01000
0   0   2.0000   1.000E-07   0.01000000   0.00178    2.00000   0.01000
0   0   3.0000   1.000E-07   0.01000000   0.00355    3.00000   0.01000
0   0   4.0000   1.000E-07   0.01000000   0.00501    4.00000   0.01000
0   0   5.0000   1.000E-07   0.01000000   0.00631    5.00000   0.01000
-1

0   0   Na+      H+          NpO2+        NpO2Ac(aq)   Cl-       Ac-
```

; NONLIN V2.0

NONLIN was developed by A.R. Felmy

INPUT file name is U1:[ERGIAMB.NONLIN.NPO2AC]REV_NPO2AC_CH.IN;1
GMIN file name is U1:[ERGIAMB.NONLIN.NPO2AC]REV_NPO2AC_CH.GMIN;1
OUTPUT file name is U1:[ERGIAMB.NONLIN.NPO2AC]REV_NPO2AC_CH.OUT;1

AQUEOUS SPECIES

ID	NAME	MOLES	Z	u0rt
1080	H2O	0.000000000000	0.	-95.663
11000	Na+	0.000000000000	1.	-105.651
1000	H+	0.000000000000	1.	0.000
93080	NpO2+	0.000000000000	1.	-369.105
93892	NpO2Ac(aq)Ch	0.000000000000	0.	-526.061
170	Cl-	0.000000000000	-1.	-52.955
890	Ac-	0.000000000000	-1.	-147.347

SOLID PHASES

ID	NAME	MOLES	Z	u0rt

CONSTRAINT EQUATIONS

	0	1	8	11	93	89	17
H2O	0.0	2.0	1.0	0.0	0.0	0.0	0.0
Na+	1.0	0.0	0.0	1.0	0.0	0.0	0.0
H+	1.0	1.0	0.0	0.0	0.0	0.0	0.0
NpO2+	1.0	0.0	2.0	0.0	1.0	0.0	0.0
NpO2Ac(aq)Ch	0.0	0.0	2.0	0.0	1.0	1.0	0.0
Cl-	-1.0	0.0	0.0	0.0	0.0	0.0	1.0
Ac-	-1.0	0.0	0.0	0.0	0.0	1.0	0.0

Closed input files GMIN and COMP

COMP file name is U1:[ERGIAMB.NONLIN.NPO2AC]COMP.DAT;1
BINARYP file name is U1:[ERGIAMB.NONLIN.NPO2AC]BINARYP.DAT;1
TERNARYP file name is U1:[ERGIAMB.NONLIN.NPO2AC]TERNARYP.DAT;1
LAMBDA file name is U1:[ERGIAMB.NONLIN.NPO2AC]LAMBDA.DAT;1

non-ideal electrolyte parameters

single electrolyte parameters

Na+	Cl-	0.07650	0.26640	0.00000	0.00127
Na+	Ac-	0.14260	0.22000	0.00000	-0.00629
H+	Cl-	0.17750	0.29450	0.00000	0.00080
H+	Ac-	0.00000	0.00000	0.00000	0.00000
NpO2+	Cl-	0.14150	0.28100	0.00000	0.00000
NpO2+	Ac-	0.00000	0.00000	0.00000	0.00000

ternary electrolyte parameters

			Cl-	Ac-	
Na+	H+	0.03600	-0.00400	0.00000	
Na+	NpO2+	0.00000	0.00000	0.00000	
H+	NpO2+	0.00000	0.00000	0.00000	
			Na+	H+	NpO2+
Cl-	Ac-	-0.09000	0.01029	0.00000	0.00000

neutral ion parameters

	NpO2Ac(aq)Ch
Na+	0.0000
H+	0.0000
NpO2+	0.0000

Cl-	0.0000
Ac-	0.0000
NpO2Ac(aq)Ch	0.0000

higher order lambdas

	Cl-	Ac-
NpO2Ac(aq)ChNa+	0.00000	0.00000
NpO2Ac(aq)ChH+	0.00000	0.00000
NpO2Ac(aq)ChNpO2+	0.00000	0.00000

TOTAL NUMBER OF SPECIES = 7
NUMBER OF COMPONENTS = 7
INDEPENDENT CONSTRAINTS = 6

SOLUBILITY DATA

input molalities

Na+	H+	NpO2+	NpO2Ac(aq)Ch	Cl-	Ac-
3.000E-01	1.000E-07	1.000E-02	1.120E-03	3.000E-01	1.000E-02
1.000E+00	1.000E-07	1.000E-02	1.350E-03	1.000E+00	1.000E-02
2.000E+00	1.000E-07	1.000E-02	1.780E-03	2.000E+00	1.000E-02
3.000E+00	1.000E-07	1.000E-02	3.550E-03	3.000E+00	1.000E-02
4.000E+00	1.000E-07	1.000E-02	5.010E-03	4.000E+00	1.000E-02
5.000E+00	1.000E-07	1.000E-02	6.310E-03	5.000E+00	1.000E-02

ADJUSTED PARAMETERS

DATA SET(1)

input	calc	diff
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	

AVERAGE DEVIATION = 0.0000E+00

STANDARD DEVIATION = 0.0000E+00

FINAL MOLALITIES

Na+	H+	NpO2+	NpO2Ac(aq)Ch	Cl-	Ac-
3.000E-01	1.000E-07	1.149E-03	9.971E-03	3.000E-01	1.149E-03
1.000E+00	1.000E-07	1.219E-03	1.013E-02	1.000E+00	1.219E-03
2.000E+00	1.000E-07	1.177E-03	1.060E-02	2.000E+00	1.177E-03
3.000E+00	1.000E-07	1.141E-03	1.241E-02	3.000E+00	1.141E-03
4.000E+00	1.000E-07	1.054E-03	1.396E-02	4.000E+00	1.054E-03
5.000E+00	1.000E-07	9.453E-04	1.536E-02	5.000E+00	9.453E-04

FINAL LOG ACTIVITIES

Na+	H+	NpO2+	NpO2Ac(aq)Ch	Cl-	Ac-
-6.736E-01	-7.112E+00	-3.072E+00	-2.001E+00	-6.737E-01	-3.103E+00
-1.838E-01	-7.060E+00	-3.038E+00	-1.994E+00	-1.838E-01	-3.129E+00
1.250E-01	-6.937E+00	-2.990E+00	-1.975E+00	1.249E-01	-3.158E+00
3.298E-01	-6.797E+00	-2.920E+00	-1.906E+00	3.297E-01	-3.159E+00
4.948E-01	-6.651E+00	-2.862E+00	-1.855E+00	4.948E-01	-3.167E+00
6.395E-01	-6.502E+00	-2.810E+00	-1.813E+00	6.395E-01	-3.177E+00

Calculated Log Activity Coefficients

Na+	H+	NpO2+	NpO2Ac(aq)	ChCl-	Ac-
-0.1508	-0.1118	-0.1322	0.0000	-0.1508	-0.1632
-0.1838	-0.0597	-0.1243	0.0000	-0.1838	-0.2155
-0.1761	0.0634	-0.0606	0.0000	-0.1761	-0.2286
-0.1473	0.2025	0.0224	0.0000	-0.1474	-0.2164
-0.1072	0.3486	0.1154	0.0000	-0.1073	-0.1898
-0.0594	0.4983	0.2149	0.0000	-0.0595	-0.1526

SOLID PHASE NOW IN EQUILIBRIUM

DATA SET(1)

input	calc	diff
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	
0.000000000000000E+000	0.000000000000000E+000	

AVERAGE DEVIATION = 0.0000E+00

STANDARD DEVIATION = 0.0000E+00

FINAL MOLALITIES

Na+	H+	NpO2+	NpO2Ac(aq)	ChCl-	Ac-
3.000E-01	3.964E+01	9.910E+00	1.112E-02	4.985E+01	7.000E-15
1.000E+00	3.947E+01	9.868E+00	1.135E-02	5.034E+01	7.000E-15
2.000E+00	3.923E+01	9.808E+00	1.178E-02	5.104E+01	7.000E-15
3.000E+00	3.899E+01	9.748E+00	1.355E-02	5.174E+01	7.000E-15
4.000E+00	3.875E+01	9.687E+00	1.501E-02	5.243E+01	7.000E-15
5.000E+00	3.850E+01	9.625E+00	1.631E-02	5.312E+01	7.000E-15

FINAL LOG ACTIVITIES

Na+	H+	NpO2+	NpO2Ac(aq)	ChCl-	Ac-
1.829E+00	1.587E+01	1.079E+01	-1.762E+00	1.503E+01	-1.995E+01
2.442E+00	1.589E+01	1.088E+01	-1.754E+00	1.499E+01	-1.969E+01
2.875E+00	1.593E+01	1.101E+01	-1.739E+00	1.493E+01	-1.932E+01
3.184E+00	1.596E+01	1.114E+01	-1.680E+00	1.488E+01	-1.894E+01
3.444E+00	1.598E+01	1.127E+01	-1.637E+00	1.483E+01	-1.856E+01
3.679E+00	1.600E+01	1.140E+01	-1.603E+00	1.478E+01	-1.818E+01

Calculated Log Activity Coefficients

Na+	H+	NpO2+	NpO2Ac(aq)	ChCl-	Ac-
2.3516	14.2676	9.7966	0.1918	13.3338	-5.7954
2.4422	14.2970	9.8903	0.1908	13.2867	-5.5351
2.5735	14.3348	10.0230	0.1894	13.2227	-5.1609
2.7069	14.3675	10.1545	0.1879	13.1625	-4.7841
2.8423	14.3953	10.2846	0.1864	13.1059	-4.4048
2.9796	14.4182	10.4134	0.1849	13.0528	-4.0233

FINAL L2 NORM OF THE RESIDUALS 0.0000000E+00

EXIT PARAMETER 0

FINAL APPROXIMATE SOLUTION