SCIENTIFIC NOTEBOOK
NUMBER 278

by

Roberto T. Pabalan

The entries in this electronic scientific notebook #278 document activities conducted during the Period October 1, 1999, through December 31, 1999, under the Radionuclide Transport Key Technical Issue (Project Number 20-1402-871). **Note that no entries were made during the period July 1, 1999 through September 30, 1999. This lack of entry for that period was communicated to Bruce Mabrito via email.**

10/04/99

OBJECTIVE:

The following is a copy of an abstract that was submitted to the MRS'99 Symposium on the Scientific Basis for Nuclear Waste Management XXIII:

Information potentially subject to copyright protection was redacted from this location. The redacted material was an abstract published in an MRS Symposium in 1999.

The entries in this notebook document the calculations that were done for the study. Thermodynamic calculations will be done using Geochemist's Workbench 3.0, whereas sensitivity analyses will be done using the TPA code version 3.2.

Below are thermodynamic data for Tc-bearing aqueous, gaseous, and solid species taken from the LLNL EQ3 database (version 8, release 6, GWB-formatted):

```
TcO4-
   charge= -1.0     ion size=  4.0 A     mole wt.=  161.9976
g
   2 elements in species
    4.0000 O            1.0000 Tc
*  gflag = 1 [reported delG0f used]
*  extrapolation algorithm: 64cri/cob
*  reference-state data source = 84rar
*      delG0f =  -623.800 kj/mol
*      delH0f =  -716.269 kj/mol
*      S0PrTr =   199.000 j/(mol*K)

Tc+++
   charge=  3.0     ion size=  5.0 A     mole wt.=  98.0000 g
   4 species in reaction
 -2.0000 H2O            -1.0000 O2(aq)            1.0000
TcO4-
  4.0000 H+
     500.0000  47.6140  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 83rar
*      delG0f =   105.800 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

TcO++
   charge=  2.0     ion size=  4.5 A     mole wt.=  113.9994
g
   4 species in reaction
 -1.5000 H2O            -0.7500 O2(aq)            1.0000
TcO4-
  3.0000 H+
     500.0000  31.5059  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 83rar
*      delG0f =  -100.600 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

TcO4--
   charge= -2.0     ion size=  4.0 A     mole wt.=  161.9976
g
   4 species in reaction
 -1.0000 H+             -0.2500 O2(aq)            0.5000 H2O
  1.0000 TcO4-
     500.0000  31.8197  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 84rar
*      delG0f =  -564.900 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

TcO4---
   charge= -3.0     ion size=  4.0 A     mole wt.=  161.9976
g
   4 species in reaction
 -2.0000 H+             -0.5000 O2(aq)            1.0000 H2O
  1.0000 TcO4-
     500.0000  63.2889  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 84rar
*      delG0f =  -508.000 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

(TcO(OH)2)2(aq)
   charge=  0.0     ion size=  3.0 A     mole wt.=  296.0282
g
   3 species in reaction
 -4.0000 H+             2.0000 TcO++            4.0000 H2O
     500.0000   0.1271  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 83rar
*      delG0f =  -1149.200 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

H2TcO4(aq)
   charge=  0.0     ion size=  3.0 A     mole wt.=  164.0135
g
   2 species in reaction
  1.0000 TcO4--            2.0000 H+
     500.0000  -9.0049  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 84rar
*      delG0f =  -616.300 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

HTcO4-
   charge= -1.0     ion size=  4.0 A     mole wt.=  163.0055
g
   2 species in reaction
  1.0000 H+             1.0000 TcO4--
     500.0000  -8.7071  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 84rar
*      delG0f =  -614.600 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

TcO(OH)2(aq)
   charge=  0.0     ion size=  3.0 A     mole wt.=  148.0141
g
   3 species in reaction
 -2.0000 H+             1.0000 TcO++            2.0000 H2O
     500.0000   3.3221  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*  gflag = 1 [reported delG0f used]
*  reference-state data source = 83rar
*      delG0f =  -556.000 kj/mol
*      delH0f =     N/A
*      S0PrTr =     N/A

TcOOH+
   charge=  1.0     ion size=  4.0 A     mole wt.=  131.0067
g
   3 species in reaction
 -1.0000 H+             1.0000 H2O            1.0000 TcO++
     500.0000   1.1355  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
```

```
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -331.300 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

HTcO4                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  163.0055 g
   2 species in reaction
 1.0000 H+              1.0000 TcO4-
      6.1472   5.9566   5.7286   5.5327
      5.3565   5.2374  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -589.800 kj/mol
*       delH0f =  -703.945 kj/mol
*       S0PrTr =   126.300 j/(mol*K)

KTcO4                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  201.0959 g
   2 species in reaction
 1.0000 K+              1.0000 TcO4-
     -3.1520  -2.2667  -1.3220  -0.5151
      0.2154   0.7378   1.1244   1.3997
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: Cp integration
*   reference-state data source = 83rar
*       delG0f =  -919.200 kj/mol
*       delH0f = -1021.675 kj/mol
*       S0PrTr =   164.780 j/(mol*K)

NaTcO4                  type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  184.9874 g
   2 species in reaction
 1.0000 Na+             1.0000 TcO4-
     500.0000   1.5208  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -877.000 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

Tc                      type=
   formula=
   mole vol.=   0.000 cc     mole wt.=   98.0000 g
   4 species in reaction
-1.7500 O2(aq)          -0.5000 H2O             1.0000 H+
 1.0000 TcO4-
     102.3170  93.5811  83.2812  73.6302
      63.8614  55.9190  49.2958  43.6058
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: Cp integration
*   reference-state data source = 83rar
*       delG0f =    0.000 kj/mol
*       delH0f =    0.000 kj/mol
*       S0PrTr =   33.500 j/(mol*K)

Tc(OH)2                 type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  132.0147 g
   4 species in reaction
-3.0000 H+              -0.2500 O2(aq)          1.0000 Tc+++
 2.5000 H2O
     500.0000   5.2714  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -461.200 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

Tc(OH)3                 type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  149.0220 g
   3 species in reaction
-3.0000 H+              1.0000 Tc+++            3.0000 H2O
     500.0000  -9.2425  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -658.500 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

Tc2O7                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  307.9958 g
   3 species in reaction
-1.0000 H2O             2.0000 H+               2.0000 TcO4-
     13.5030  13.1077  12.5945  12.1028
     11.5969  11.1884  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -935.600 kj/mol
*       delH0f = -1120.164 kj/mol
*       S0PrTr =   166.000 j/(mol*K)

Tc2S7                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  420.4620 g
   4 species in reaction
-8.0000 H2O             2.0000 TcO4-            7.0000 HS-
 9.0000 H+
    -252.2732 -230.2410 -205.6544 -183.8931
    -163.3409 -148.0562  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -580.600 kj/mol
*       delH0f =  -615.000 kj/mol
*       S0PrTr =   176.000 j/(mol*K)

Tc3O4                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  357.9976 g
   4 species in reaction
-9.0000 H+              -0.2500 O2(aq)          3.0000 Tc+++
 4.5000 H2O
     500.0000  -19.2271  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -863.800 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

Tc4O7                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  503.9958 g
   4 species in reaction
-10.0000 H+             2.0000 Tc+++            2.0000
TcO++
 5.0000 H2O
     500.0000  -26.0149  500.0000  500.0000
```

```
    500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -1324.000 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcO2:2H2O(am)                 type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  166.0294 g
   3 species in reaction
-2.0000 H+              1.0000 TcO++            3.0000 H2O
    500.0000   -4.2319  500.0000  500.0000
    500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -836.300 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcO3                          type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  145.9982 g
   3 species in reaction
-1.0000 H2O             1.0000 TcO4--           2.0000 H+
    500.0000  -23.1483  500.0000  500.0000
    500.0000  500.0000  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   reference-state data source = 83rar
*       delG0f =  -459.849 kj/mol
*       delH0f =  -540.000 kj/mol
*       S0PrTr =    72.400 j/(mol*K)

TcOH                          type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  115.0073 g
   4 species in reaction
-3.0000 H+              -0.5000 O2(aq)          1.0000 Tc+++
 2.0000 H2O
    500.0000   24.9009  500.0000  500.0000
    500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -234.700 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcS2                          type=
   formula=
   mole vol.=   32.000 cc    mole wt.=  162.1320 g
   3 species in reaction
-1.0000 H2O             1.0000 TcO++            2.0000 HS-
    500.0000  -65.9742  500.0000  500.0000
    500.0000  500.0000  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   reference-state data source = 83rar
*       delG0f =  -216.067 kj/mol
*       delH0f =  -224.000 kj/mol
*       S0PrTr =    71.000 j/(mol*K)

TcS3                          type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  194.1980 g
   4 species in reaction
-4.0000 H2O             1.0000 TcO4--           3.0000 HS-
 5.0000 H+
    500.0000 -119.5008  500.0000  500.0000
    500.0000  500.0000  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   reference-state data source = 83rar
*       delG0f =  -262.386 kj/mol
*       delH0f =  -276.000 kj/mol
*       S0PrTr =    84.000 j/(mol*K)

Tc2O7(g)
   mole wt.=  307.9958 g
   3 species in reaction
-1.0000 H2O             2.0000 H+               2.0000 TcO4-
    23.8637   21.3593   18.4240   15.7206
    13.0380   10.9129  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -888.500 kj/mol
*       delH0f =  -988.569 kj/mol
*       S0PrTr =   449.400 j/(mol*K)

TcO2
   mole wt.=  129.999 g
   4 species in reaction
-0.7500 O2(aq)          -0.5000 H2O             1.0000 H+
 1.0000 TcO4-
```

For comparison, below are thermodynamic data from a file downloaded from the LLNL website. This file was modified by Bill Bourcier, as indicated in the following comment lines taken from the file:

```
dataset of thermodynamic data for gwb programs
dataset format: oct94
activity model: debye-huckel
*  THERMODYNAMIC DATABASE: thermo.com.V8.R6.230
*  generated by GEMBOCHS.V2-Jewel.src.R6 03-dec-1996 17:13:36
*  Output package:  gwb
*  Data set:        com
*  altered for UGTA project
*     3/97 deleted Tc data; replaced with Rard new data
*          as listed in his 2/27/97 and 3/4/97 memos
```

TcO4-
charge= -1.0 ion size= 4.0 A mole wt.= 161.9976 g
2 elements in species
4.0000 O 1.0000 Tc
* data from Rard (1997)
* reference-state data
* delG0f = -637.4 kj/mol
* delH0f = -729.4 kj/mol
* S0PrTr = 199.6 j/(mol*K)

Tc+++
charge= 3.0 ion size= 5.0 A mole wt.= 98.0000 g
4 species in reaction
-2.0000 H2O -1.0000 O2(aq) 1.0000 TcO4-
4.0000 H+
500.0000 52.8903 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = 105.8 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

TcO(OH)2(aq)
charge= 0.0 ion size= 3.0 A mole wt.= 147.9980 g
4 species in reaction
0.50000 H2O -0.7500 O2(aq) 1.0000 TcO4-
1.0000 H+
500.0000 35.1932 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -567.5 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

TcOOH+
charge= 1.0 ion size= 4.0 A mole wt.= 131.0067 g
3 species in reaction
1.0000 H+ -1.0000 H2O 1.0000 TcO(OH)2(aq)
500.0000 -2.5020 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -344.6 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

TcO(OH)3-
charge= -1.0 ion size= 4.0 A mole wt.= 165.0060 g
3 species in reaction
-1.0000 H+ 1.0000 H2O 1.0000 TcO(OH)2(aq)
500.0000 10.9106 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -742.4 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

AgTcO4 type=
formula=
mole vol.= 0.000 cc mole wt.= 269.866 g
2 species in reaction
1.0000 Ag+ 1.0000 TcO4-
500.0000 -3.2756 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -579.0 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

CsTcO4 type=
formula=
mole vol.= 0.000 cc mole wt.= 294.903 g
2 species in reaction
1.0000 Cs+ 1.0000 TcO4-
500.0000 -3.6145 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -949.7 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

KTcO4 type=
formula=
mole vol.= 0.000 cc mole wt.= 201.096 g
2 species in reaction
1.0000 K+ 1.0000 TcO4-
500.0000 -2.2840 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -932.9 kj/mol
* delH0f = -1035.1 kj/mol
* S0PrTr = 164.78 j/(mol*K)

NH4TcO4 type=
formula=
mole vol.= 0.000 cc mole wt.= 180.004 g
3 species in reaction
1.0000 NH3(aq) 1.0000 TcO4- 1.0000 H+
500.0000 -10.1417 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -722.0 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

NaTcO4:4H2O type=
formula=
mole vol.= 0.000 cc mole wt.= 256.985 g
3 species in reaction
4.0000 H2O 1.0000 Na+ 1.0000 TcO4-
500.0000 0.8078 500.0000 500.0000
500.0000 500.0000 500.0000 500.0000
* data from Rard (1997)
* reference-state data
* delG0f = -1843.4 kj/mol
* delH0f = kj/mol
* S0PrTr = j/(mol*K)

TcO2 type=
formula=
mole vol.= 0.000 cc mole wt.= 129.999 g
2 species in reaction

```
-1.0000 H2O            1.0000 TcO(OH)2(aq)
    500.0000 -12.5222 500.0000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   data from Rard (1997)
*   reference-state data
*       delG0f =  -401.8  kj/mol
*       delH0f =  -457.8  kj/mol
*       S0PrTr =   50.000 j/(mol*K)

TcO2:1.6H2O            type=
   formula=
   mole vol.=   0.000 cc    mole wt.= 158.798  g
   2 species in reaction
 0.6000 H2O            1.0000 TcO(OH)2(aq)
    500.0000  -8.4945 500.0000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   data from Rard (1997)
*   reference-state data
*       delG0f =  -758.3  kj/mol
*       delH0f =          kj/mol
*       S0PrTr =          j/(mol*K)

Tc2O7                  type=
   formula=
   mole vol.=   0.000 cc    mole wt.= 307.9958 g
   3 species in reaction
-1.0000 H2O            2.0000 H+            2.0000 TcO4-
    500.0000  15.2962 500.0000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   data from Rard (1997)
*   reference-state data
*       delG0f =  -950.3  kj/mol
*       delH0f = -1126.5  kj/mol
*       S0PrTr =  192.000 j/(mol*K)

Tc2O7:H2O              type=
   formula=
   mole vol.=   0.000 cc    mole wt.= 325.9958 g
   2 species in reaction
2.0000 H+            2.0000 TcO4-
    500.0000  14.1019 500.0000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   data from Rard (1997)
*   reference-state data
*       delG0f = -1194.3  kj/mol
*       delH0f = -1414.2  kj/mol
*       S0PrTr =  278.800 j/(mol*K)

TlTcO4                 type=
   formula=
   mole vol.=   0.000 cc    mole wt.= 366.386  g
   2 species in reaction
 1.0000 Tl+           1.0000 TcO4-
    500.0000  -5.3107 500.0000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   data from Rard (1997)
*   reference-state data
*       delG0f =  -700.1  kj/mol
*       delH0f =          kj/mol
*       S0PrTr =          j/(mol*K)

Tc2O7(g)
   mole wt.= 307.9958 g
   3 species in reaction
-1.0000 H2O            2.0000 H+            2.0000 TcO4-
    500.0000  23.2668 500.0000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -904.800 kj/mol
*       delH0f = -1008.100 kj/mol
*       S0PrTr =  436.640 j/(mol*K)

Tc(g)
   mole wt.= 98.0000 g
   4 species in reaction
-0.5000 H2O            1.0000 H+            1.0000 TcO4-
-1.7500 O2(aq)
    500.0000 206.4413 5000.000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   del G from Rard for Tc(g)
*   reference-state data source = Rard, 1997
*       delG0f =  630.700 kj/mol
*       delH0f =  675.000 kj/mol
*       S0PrTr =  181.050 j/(mol*K)

TcO(g)
   mole wt.= 113.9990 g
   4 species in reaction
-0.5000 H2O            1.0000 H+            1.0000 TcO4-
-1.2500 O2(aq)
    500.0000 157.1334 5000.000 500.0000
    500.0000 500.0000 500.0000 500.0000
*   del G from Rard for Tc(g)
*   reference-state data source = Rard, 1997
*       delG0f =  357.5  kj/mol
*       delH0f =  390.000 kj/mol
*       S0PrTr =  244.100 j/(mol*K)

TcO2
   mole wt.= 129.999 g
   4 species in reaction
-0.7500 O2(aq)         -0.5000 H2O          1.0000 H+
 1.0000 TcO4-
```

For additional comparison, the following are values of thermodynamic parameters taken from the NEA website http://www.nea.fr/html/dbtdb/tdbdata/tdbdata.html (password required). The format of the datafile is described below:

Format of the data files

The files are divided into lines ("records"), each with a set of data values separated by blank space(s). A value of 1.0e+33 means the parameter is missing.

The coefficients a through k of the temperature functions mentioned below refer to the following equation:

$$F(T) = a + b\,T + c\,T2 + d/T + e\,T\text{-}2 + f\,\ln(T) + g\,T\ln(T) + h\,T1/2 + i/T1/2 + j\,T3 + k\,T\text{-}3.$$

The information about "format" gives the FORTRAN formatting codes used for output. "a30" means a field of 30 characters (letters, numbers, points etc.). "6x" means 6 blanks, and 2e12.5 means two consecutive fields of numbers in exponent format, each with a total width of 12 characters, 5 of which are decimals. The exponent format used is the one common for most computer programming languages, where 2.98150E+02 means 2.98150×102.

Formation data files

In the formation data files, the below 13 records (lines) are repeated for each chemical formula.

The formulae of certain aqueous species may be written in different ways. For example, (UO2)3O(OH)2(HCO3)+ is the same as (UO2)3(OH)5CO2+. This must be considered when comparisons of different databases are made.

Record 1:

Chemical formula (in basic Hill notation).
ionic strength for all data in the record (normally 0)
temperature for all data in the record, in Kelvin (K)

Format: a30,6x,2e12.5

Record 2:

Gibbs energy of formation, in kJ/mol.
uncertainty in the Gibbs energy of formation.
minimum temperature for the function in Records 3 and 4 (equal to 0.0 if the function is missing).
maximum temperature for the function in Records 3 and 4 (equal to 0.0 if the function is missing).

Format: 4e12.5

Record 3:

temperature function of the Gibbs energy of formation, first part (coefficients a, b, c, d, e, f) (see above)
a = 1.00e+33 means the function is missing.

Format: 6e12.5

Record 4:

temperature function of the Gibbs energy of formation, second part (coefficients g, h, i, j, k) (see above).

Format: 5e12.5

Record 5:

enthalpy of formation, in kJ/mol.
uncertainty in the enthalpy of formation.
minimum temperature for the function in Records 6 and 7 (equal to 0.0 if the function is missing).
maximum temperature for the function in Records 6 and 7 (equal to 0.0 if the function is missing).

Format: 4e12.5

Record 6:

temperature function of the enthalpy of formation, first part (coefficients a, b, c, d, e, f).
a = 1.00e+33 means the function is missing.

Format: 6e12.5

Record 7:

temperature function of the enthalpy of formation, second part (coefficients g, h, i, j, k).

Format: 5e12.5

Record 8:

absolute entropy of the chemical species So, in J/(K·mol).
uncertainty in the entropy.
minimum temperature for the function in Records 9 and 10 (equal to 0.0 if the function is missing).
maximum temperature for the function in Records 9 and 10 (equal to 0.0 if the function is missing).

Format: 4e12.5

Record 9:

temperature function of the entropy, first part (coefficients a, b, c, d, e, f).
a = 1.00e+33 means the function is missing.

Format: 6e12.5

Record 10:

temperature function of the entropy, second part (coefficients g, h, i, j, k).

Format: 5e12.5

Record 11:

heat capacity at constant pressure, in J/(K·mol).
uncertainty in the heat capacity.
minimum temperature for the function in Records 12 and 13 (equal to 0.0 if the function is missing).
maximum temperature for the function in Records 12 and 13 (equal to 0.0 if the function is missing).

Format: 4e12.5

Record 12:

temperature function of the heat capacity, first part (coefficients a, b, c, d, e, f).
a = 1.00e+33 means the function is missing.

Format: 6e12.5

Record 13:

temperature function of the heat capacity, second part (coefficients g, h, i, j, k).

Format: 5e12.5

The parameter values for the Tc-bearing species in the Tcdatafile are:

```
Tc(c)                    1.00000E+33 2.98150E+02
0.00000E+00 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
3.25000E+01 7.00000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
2.49000E+01 1.00000E+00 2.98150E+02 2.43000E+03
```

2.50940E+01 4.31450E-03-2.75460E-07 0.00000E+00-1.31300E+05 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Tc(g) 1.00000E+33 2.98150E+02
6.30709E+02 2.50009E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
6.75000E+02 2.50000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.81052E+02 1.00000E-02 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
2.07950E+01 1.00000E-02 2.98000E+02 6.00000E+02
2.49130E+01-1.91834E-02 2.51283E-05 0.00000E+00-5.61800E+04 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Tc(l) 1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO(g) 1.00000E+33 2.98150E+02
3.57492E+02 5.70007E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
3.90000E+02 5.70000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
2.44109E+02 6.00000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
3.12560E+01 7.50000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO+2 0.00000E+00 2.98150E+02
-1.16801E+02 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO2(c) 1.00000E+33 2.98150E+02
-4.01852E+02 1.17625E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
-4.57800E+02 1.17000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
5.00000E+01 4.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO4- 0.00000E+00 2.98150E+02
-6.37408E+02 7.61601E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00

0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
-7.29400E+02 7.60000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.99600E+02 1.50000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
-1.50000E+01 8.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO4-2 0.00000E+00 2.98150E+02
-5.75761E+02 8.88008E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Tc2O7(c) 1.00000E+33 2.98150E+02
-9.50284E+02 1.55623E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
-1.12650E+03 1.49000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.92000E+02 1.50000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.60400E+02 1.50000E+01 2.98150E+02 3.92700E+02
1.55000E+02 8.60000E-02 0.00000E+00 0.00000E+00-1.80000E+06 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Tc2O7(g) 1.00000E+33 2.98150E+02
-9.04823E+02 1.64616E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
-1.00810E+03 1.60627E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
4.36641E+02 1.20000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.46736E+02 5.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO(OH)+ 0.00000E+00 2.98150E+02
-3.45379E+02 9.00933E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO(OH)2(aq) 1.00000E+33 2.98150E+02
-5.68249E+02 8.84499E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00

1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO2.1.6H2O(s) 1.00000E+33 2.98150E+02
-7.58481E+02 8.37185E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcO(OH)3- 0.00000E+00 2.98150E+02
-7.43172E+02 9.13502E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Tc2O7.H2O(s) 1.00000E+33 2.98150E+02
-1.19430E+03 1.55000E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
-1.41415E+03 1.49054E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
2.78921E+02 7.21383E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcF6(c,cubic) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
2.53520E+02 5.10000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.57840E+02 3.20000E-01 2.68335E+02 3.11136E+02
7.20810E+01 2.87666E-01 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcF6(g) 0.00000E+00 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
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0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
3.59136E+02 4.50000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.20703E+02 2.62900E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00

TcO3F(g) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
3.06879E+02 6.77000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
7.72610E+01 3.08000E-01 2.98150E+02 1.00000E+03
7.61200E+01 5.34274E-02-2.53393E-05 0.00000E+00-1.11446E+06 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcCl6-2 0.00000E+00 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
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0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcOCl5-2 0.00000E+00 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
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TcO2Cl4-3 0.00000E+00 2.98150E+02
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TcO3Cl(g) 1.00000E+33 2.98150E+02
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3.17636E+02 7.97000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
8.03650E+01 3.08000E-01 2.98150E+02 1.00000E+03
8.03360E+01 4.63950E-02-2.20086E-05 0.00000E+00-1.05321E+06 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
TcBr6-2 0.00000E+00 2.98150E+02
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TcS(g) 1.00000E+33 2.98150E+02
4.91923E+02 6.50010E+01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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2.55990E+02 1.00000E+00 0.00000E+00 0.00000E+00
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3.44740E+01 1.00000E+00 0.00000E+00 0.00000E+00
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NH4TcO4(c) 1.00000E+33 2.98150E+02
-7.22000E+02 7.63154E+00 0.00000E+00 0.00000E+00
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(NH4)2TcCl6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
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(NH4)2TcBr6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
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TcC(g) 1.00000E+33 2.98150E+02
7.65600E+02 4.02498E+01 0.00000E+00 0.00000E+00
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8.26500E+02 4.00000E+01 0.00000E+00 0.00000E+00
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TcCO3(OH)2(aq) 0.00000E+00 2.98150E+02
-9.68901E+02 9.01023E+00 0.00000E+00 0.00000E+00
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TcCO3(OH)3- 0.00000E+00 2.98150E+02
-1.15866E+03 9.48600E+00 0.00000E+00 0.00000E+00
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TlTcO4(c) 0.00000E+00 2.98150E+02
-7.00174E+02 7.65263E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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AgTcO4(c) 1.00000E+33 2.98150E+02
-5.78977E+02 7.65365E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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NaTcO4.4H2O(s) 1.00000E+33 2.98150E+02
-1.84341E+03 7.62180E+00 0.00000E+00 0.00000E+00
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KTcO4(c) 1.00000E+33 2.98150E+02
-9.32923E+02 7.60374E+00 0.00000E+00 0.00000E+00

1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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-1.03510E+03 7.60000E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.64780E+02 3.30000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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1.23300E+02 2.50000E-01 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
K2TcCl6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
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0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
K2TcBr6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
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0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Rb2TcCl6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Rb2TcBr6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
CsTcO4(c) 1.00000E+33 2.98150E+02
-9.49698E+02 7.67077E+00 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00

1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Cs2TcCl6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
Cs2TcBr6(c) 1.00000E+33 2.98150E+02
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
1.00000E+33 1.00000E+33 0.00000E+00 0.00000E+00
1.00000E+33 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00
0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00 0.00000E+00

Reaction data files

A value of 1.0e+33 means the parameter is missing. The following 8 records (lines) are repeated for each reaction. Each reaction has an assigned "key species" for which formation data have been derived from the reaction. These reaction data are listed in the formation data files. The temperature dependence function and the data output format codes used are described above.

Record 1:

formula of key species (in basic Hill notation).
ionic strength for all data in the record (normally 0).
temperature for all data in the record in Kelvin (K).

Format: a30,6x,2e12.5

Record 2:

reaction, consisting of the chemical formulae of the species involved, including their stoichiometric coefficient. The reaction string is implicitly equal to zero. This means, that e. g. the reaction

U4+ + 2H2O <=> U(OH)22+ + 2H+

would appear as

*-1.00*U+4 -2.00*H2O(l) +1.00*U(OH)2+2 +2.00*H+*

Note that electrons are not noted explicitly in the case of half cell redox reactions. The number of electrons involved is obtained from the charge imbalance.

Format: A80

Record 3:

continuation of Record 2 if necessary.

Format: A80

Record 4:

equilibrium constant, logarithmic.
uncertainty in the equilibrium constant, logarithmic.

Format: 2e12.5

Record 5:

Gibbs energy of reaction, in kJ/mol.
uncertainty in the Gibbs energy of reaction.

Format: 2e12.5

Record 6:

enthalpy of reaction, in kJ/mol.
uncertainty in the enthalpy of reaction.

Format: 2e12.5

Record 7:

entropy of reaction, in J/(K·mol).
uncertainty in the entropy of reaction.

Format: 2e12.5

Record 8:

heat capacity of reaction at constant pressure, in J/(K·mol).

uncertainty in the heat capacity of reaction.

Format: 2e12.5

Additional information

The temperature function Cp(T) selected in the review for UO2+2 is not compatible with the present notation and is therefore not included in the data file.

The data on UO2(OH)2(aq) are limiting values.

The data on U(OH)5- are limiting values.

The value selected in the review for the formation of U6(OH)15+9 refers to an ionic strength of 3 M and is not listed here.

The parameter values for Tc from the file Tcreac are:

```
Tc(g)                    1.00000E+33 2.98150E+02
 -1.00*Tc(c)  +1.00*Tc(g)

 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 6.75000E+02 2.50000E+01
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO+2                    0.00000E+00 2.98150E+02
 -2.00*H+  +2.00*H2O(l)  -1.00*TcO(OH)2(aq)  +1.00*TcO+2

 4.00000E+00 1.00000E+33
-2.28322E+01 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO2(c)                  0.00000E+00 2.98150E+02
 -4.00*H+  +2.00*H2O(l)  +1.00*Tc(c)  -1.00*TcO2(c)

 1.89886E+01 1.00000E+33
-1.08388E+02 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO4-                    0.00000E+00 2.98150E+02
 +8.00*H+  -4.00*H2O(l)  -1.00*Tc(c)  +1.00*TcO4-

-5.63800E+01 1.00000E+33
 3.21819E+02 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO4-2                   0.00000E+00 2.98150E+02
 -1.00*TcO4-  +1.00*TcO4-2

-1.08000E+01 8.00000E-01
 6.16469E+01 4.56643E+00
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
Tc2O7(g)                 1.00000E+33 2.98150E+02
 -1.00*Tc2O7(c)  +1.00*Tc2O7(g)

 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.18400E+02 6.00000E+00
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO(OH)+                 0.00000E+00 2.98150E+02
 -1.00*H+  +1.00*H2O(l)  +1.00*TcO(OH)+  -
1.00*TcO(OH)2(aq)

 2.50000E+00 3.00000E-01
-1.42701E+01 1.71241E+00
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO(OH)2(aq)             1.00000E+33 2.98150E+02
 +0.60*H2O(l)  +1.00*TcO(OH)2(aq)  -1.00*TcO2.1.6H2O(s)

-8.40000E+00 5.00000E-01
 4.79476E+01 2.85402E+00
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO2.1.6H2O(s)           1.00000E+33 2.98150E+02
 -4.00*H+  +0.40*H2O(l)  +1.00*TcO2.1.6H2O(s)  -
1.00*TcO4-

 3.78290E+01 6.09000E-01
-2.15930E+02 3.47620E+00
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
 1.00000E+33 1.00000E+33
TcO(OH)3-                0.00000E+00 2.98150E+02
 +1.00*H+  -1.00*H2O(l)  -1.00*TcO(OH)2(aq)
+1.00*TcO(OH)3-

-1.09000E+01 4.00000E-01
 6.22177E+01 2.28322E+00
```

1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
Tc2O7.H2O(s) 1.00000E+33 2.98150E+02
-1.00*H2O(g) -1.00*Tc2O7(c) +1.00*Tc2O7.H2O(s)

1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
-4.58200E+01 4.00000E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
TcO2Cl4-3 0.00000E+00 2.98150E+02
+1.00*Cl- +2.00*H+ -1.00*H2O(l) +1.00*TcO2Cl4-3 -
1.00*TcOCl5-2

-2.95000E+00 1.50000E-01
1.68387E+01 8.56206E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
NH4TcO4(c) 1.00000E+33 2.98150E+02
-1.00*NH4+ +1.00*NH4TcO4(c) -1.00*TcO4-

9.10000E-01 7.00000E-02
-5.19432E+00 3.99563E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
TcCO3(OH)2(aq) 0.00000E+00 2.98150E+02
-1.00*CO2(g) +1.00*TcCO3(OH)2(aq) -1.00*TcO(OH)2(aq)

1.10000E+00 3.00000E-01
-6.27885E+00 1.71241E+00
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
TcCO3(OH)3- 0.00000E+00 2.98150E+02
-1.00*CO2(g) +1.00*H+ -1.00*H2O(l) +1.00*TcCO3(OH)3-
-1.00*TcO(OH)2(aq)

-7.20000E+00 6.00000E-01
4.10979E+01 3.42483E+00
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
TlTcO4(c) 0.00000E+00 2.98150E+02
-1.00*TcO4- -1.00*Tl+ +1.00*TlTcO4(c)

5.32000E+00 1.20000E-01
-3.03668E+01 6.84965E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
AgTcO4(c) 1.00000E+33 2.98150E+02
-1.00*Ag+ +1.00*AgTcO4(c) -1.00*TcO4-

3.27000E+00 1.30000E-01
-1.86653E+01 7.42045E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
NaTcO4.4H2O(s) 1.00000E+33 2.98150E+02
-4.00*H2O(l) -1.00*Na+ +1.00*NaTcO4.4H2O(s) -
1.00*TcO4-

-7.90000E-01 4.00000E-02
4.50935E+00 2.28322E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33

CsTcO4(c) 1.00000E+33 2.98150E+02
-1.00*Cs+ +1.00*CsTcO4(c) -1.00*TcO4-

3.65000E+00 1.30000E-01
-2.08344E+01 7.42045E-01
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33
1.00000E+33 1.00000E+33

October 7, 1999

The GWB database thermo.com.V8.R6.full was modified to incorporate the new technetium thermodynamic parameters provided in the NEA TCDATA file (downloaded from the NEA website). The following are the first lines in the new database:

```
dataset of thermodynamic data for gwb programs
dataset format: oct94
activity model: debye-huckel
* THERMODYNAMIC DATABASE: thermo.com.V8.R6.full
* generated by GEMBOCHS.V2-Jewel.src.R6 03-dec-1996 16:55:04
* Output package:  gwb
* Data set:      com
*
* Altered for MRS'99 calculations
* The following 5 aqueous species were commented out:
*        Tc+3, TcO4-3, (TcO(OH)2)2(aq), H2TcO4(aq), HTcO4-
* The following 5 aqueous species were added:
*        TcO+2, TcO4-2, TcCO3(OH)2(aq), TcCO3(OH)3-, TcO(OH)3-
* The following 13 solids were commented out:
*        HTcO4, Tc, Tc(OH)2, Tc(OH)3, NaTcO4, Tc2S7, Tc3O4,
*        Tc4O7, TcO2.2H2O, TcO3, TcOH, TcS2, TcS3

* The following 8 solids were added:
*        AgTcO4, CsTcO4, NaTcO4.4H2O, TcO2, NH4TcO4, TcO2.1.6H2O,
*        Tc2O7.H2O,TlTcO4
* The following 2 gas species were added:
*        Tc(g), TcO(g)
*
```

The following are the technetium species in the new database. Technetium species not in the NEA database are commented out. The species TcC(g) and TcS(g), which are in the NEA database, were not included because those species are not expected to be important in YM groundwater systems.

```
TcO4-
   charge= -1.0     ion size=  4.0 A     mole wt.=  161.9976
g
   2 elements in species
    4.0000 O            1.0000 Tc
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =   -637.4   kj/mol
*       delH0f =   -729.4   kj/mol
*       S0PrTr =    199.6   j/(mol*K)

* Not in NEA database
*Tc+++
*   charge=  3.0     ion size=  5.0 A     mole wt.=  98.0000
g
*   4 species in reaction
* -2.0000 H2O            -1.0000 O2(aq)            1.0000
TcO4-
* 4.0000 H+
*     500.0000  47.6140  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =    105.800 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcO++
   charge=  2.0     ion size=  4.5 A     mole wt.=  113.9994
g
   4 species in reaction
 -1.5000 H2O            -0.7500 O2(aq)            1.0000
TcO4-
 3.0000 H+
     500.0000  31.0512  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =   -116.800 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcO4--
   charge= -2.0     ion size=  4.0 A     mole wt.=  161.9976
g
   4 species in reaction
 -1.0000 H+             -0.2500 O2(aq)            0.5000 H2O
  1.0000 TcO4-
     500.0000  32.3010  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =   -575.760 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*TcO4---
```

```
*    charge= -3.0      ion size=  4.0 A      mole wt.=  161.9976
g
*    4 species in reaction
* -2.0000 H+              -0.5000 O2(aq)             1.0000 H2O
*  1.0000 TcO4-
*      500.0000  63.2889  500.0000  500.0000
*      500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 84rar
*       delG0f =  -508.000 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcCO3(OH)2(aq)
    charge=  0.0      ion size=  4.0 A      mole wt.=  192.0238
g
    5 species in reaction
 -0.5000 H2O             -0.7500 O2(aq)             1.0000
TcO4-
  2.0000 H+              1.0000 HCO3-
      500.0000  31.0512  500.0000  500.0000
      500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =  -116.800 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcCO3(OH)3-
    charge= -1.0      ion size=  4.0 A      mole wt.=  209.0311
g
    5 species in reaction
 -0.7500 O2(aq)          1.0000 H+              1.0000 TcO4-
  1.0000 HCO3-           0.5000 H2O
      500.0000   34.4581  500.0000  500.0000
      500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =  -1158.660 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*(TcO(OH)2)2(aq)
*    charge=  0.0      ion size=  3.0 A      mole wt.=  296.0282
g
*    3 species in reaction
* -4.0000 H+              2.0000 TcO++           4.0000 H2O
*      500.0000    0.1271  500.0000  500.0000
*      500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -1149.200 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*H2TcO4(aq)
*    charge=  0.0      ion size=  3.0 A      mole wt.=  164.0135
g
*    2 species in reaction
*  1.0000 TcO4--           2.0000 H+
*      500.0000  -9.0049  500.0000  500.0000
*      500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 84rar
*       delG0f =  -616.300 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*HTcO4-
*    charge= -1.0      ion size=  4.0 A      mole wt.=  163.0055
g
*    2 species in reaction
*  1.0000 H+               1.0000 TcO4--
*      500.0000  -8.7071  500.0000  500.0000
*      500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 84rar
*       delG0f =  -614.600 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcO(OH)2(aq)
    charge=  0.0      ion size=  3.0 A      mole wt.=  147.9980
g
    4 species in reaction
 0.50000 H2O             -0.7500 O2(aq)             1.0000
TcO4-
  1.0000 H+
      500.0000  32.5711  500.0000  500.0000
      500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =  -568.2500 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcOOH+
    charge=  1.0      ion size=  4.0 A      mole wt.=  131.0067
g
    3 species in reaction
  1.0000 H+              -1.0000 H2O                1.0000
TcO(OH)2(aq)
      500.0000  -2.5074  500.0000  500.0000
      500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =  -345.3800 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

TcO(OH)3-
    charge= -1.0      ion size=  4.0 A      mole wt.=  165.0060
g
    3 species in reaction
 -1.0000 H+              1.0000 H2O                 1.0000
TcO(OH)2(aq)
      500.0000  10.9076  500.0000  500.0000
      500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =  -743.17  kj/mol
*       delH0f =          kj/mol
*       S0PrTr =          j/(mol*K)

*Not in NEA database
*HTcO4                    type=
*    formula=
*    mole vol.=   0.000 cc      mole wt.=  163.0055 g
*    2 species in reaction
*  1.0000 H+               1.0000 TcO4-
*       6.1472    5.9566    5.7286    5.5327
*       5.3565    5.2374  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -589.800 kj/mol
*       delH0f =  -703.945 kj/mol
```

```
*       S0PrTr =    126.300 j/(mol*K)

AgTcO4                    type=
   formula=
   mole vol.=   0.000 cc     mole wt.= 269.866  g
   2 species in reaction
  1.0000 Ag+              1.0000 TcO4-
     500.0000   -3.2756  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -579.0   kj/mol
*       delH0f =           kj/mol
*       S0PrTr =           j/(mol*K)

CsTcO4                    type=
   formula=
   mole vol.=   0.000 cc     mole wt.= 294.903  g
   2 species in reaction
  1.0000 Cs+              1.0000 TcO4-
     500.0000   -3.6145  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -949.7   kj/mol
*       delH0f =           kj/mol
*       S0PrTr =           j/(mol*K)

KTcO4                     type=
   formula=
   mole vol.=   0.000 cc     mole wt.= 201.0959 g
   2 species in reaction
  1.0000 K+               1.0000 TcO4-
     500.0000   -2.2868  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -932.923 kj/mol
*       delH0f =  -1035.100 kj/mol
*       S0PrTr =    164.780 j/(mol*K)

NH4TcO4                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.= 180.004  g
   3 species in reaction
  1.0000 NH3(aq)          1.0000 TcO4-            1.0000 H+
     500.0000  -10.1417  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -722.0   kj/mol
*       delH0f =           kj/mol
*       S0PrTr =           j/(mol*K)

*Not in NEA database
*NaTcO4                   type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.= 184.9874 g
*   2 species in reaction
*  1.0000 Na+              1.0000 TcO4-
*     500.0000    1.5208  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =   -877.000 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

NaTcO4:4H2O               type=
   formula=
   mole vol.=   0.000 cc     mole wt.= 256.985  g
   3 species in reaction
  4.0000 H2O              1.0000 Na+              1.0000 TcO4-
     500.0000    0.8078  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -1843.4   kj/mol
*       delH0f =           kj/mol
*       S0PrTr =           j/(mol*K)

*Not in NEA database
*Tc                       type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  98.0000 g
*   4 species in reaction
* -1.7500 O2(aq)          -0.5000 H2O             1.0000 H+
*  1.0000 TcO4-
*     102.3170   93.5811   83.2812   73.6302
*      63.8614   55.9190   49.2958   43.6058
*   gflag = 1 [reported delG0f used]
*   extrapolation algorithm: Cp integration
*   reference-state data source = 83rar
*       delG0f =    0.000 kj/mol
*       delH0f =    0.000 kj/mol
*       S0PrTr =   33.500 j/(mol*K)

*Not in NEA database
*Tc(OH)2                  type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.= 132.0147 g
*   4 species in reaction
* -3.0000 H+              -0.2500 O2(aq)          1.0000
Tc+++
*  2.5000 H2O
*     500.0000    5.2714  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =   -461.200 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

*Not in NEA database
*Tc(OH)3                  type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.= 149.0220 g
*   3 species in reaction
* -3.0000 H+              1.0000 Tc+++            3.0000 H2O
*     500.0000   -9.2425  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =   -658.500 kj/mol
*       delH0f =    N/A
*       S0PrTr =    N/A

TcO2                      type=
   formula=
   mole vol.=   0.000 cc     mole wt.= 129.999  g
   2 species in reaction
 -1.0000 H2O              1.0000 TcO(OH)2(aq)
     500.0000  -12.4010  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -401.852   kj/mol
*       delH0f =   -457.800   kj/mol
*       S0PrTr =    50.000 j/(mol*K)
```

```
TcO2:1.6H2O                    type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  158.798  g
   2 species in reaction
 0.6000 H2O              1.0000 TcO(OH)2(aq)
     500.0000  -8.3954  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =  -758.481   kj/mol
*       delH0f =           kj/mol
*       S0PrTr =           j/(mol*K)

Tc2O7                          type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  307.9958 g
   3 species in reaction
-1.0000 H2O              2.0000 H+              2.0000 TcO4-
     500.0000  15.3029  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =  -950.2840 kj/mol
*       delH0f =  -1126.5000 kj/mol
*       S0PrTr =    192.000 j/(mol*K)

Tc2O7:H2O                      type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  325.9958 g
   2 species in reaction
2.0000 H+              2.0000 TcO4-
     500.0000  14.1058  500.0000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =  -1194.300   kj/mol
*       delH0f =  -1414.150   kj/mol
*       S0PrTr =    278.921 j/(mol*K)

*Not in NEA database
*Tc2S7                         type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  420.4620 g
*   4 species in reaction
* -8.0000 H2O              2.0000 TcO4-              7.0000 HS-
* 9.0000 H+
*     -252.2732 -230.2410 -205.6544 -183.8931
*     -163.3409 -148.0562  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   extrapolation algorithm: constant enthalpy approximatio
*   reference-state data source = 83rar
*       delG0f =  -580.600 kj/mol
*       delH0f =  -615.000 kj/mol
*       S0PrTr =    176.000 j/(mol*K)

*Not in NEA database
*Tc3O4                         type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  357.9976 g
*   4 species in reaction
* -9.0000 H+              -0.2500 O2(aq)            3.0000
Tc+++
* 4.5000 H2O
*     500.0000  -19.2271  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -863.800 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*Tc4O7                         type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  503.9958 g
*   4 species in reaction
*-10.0000 H+              2.0000 Tc+++              2.0000
TcO++
* 5.0000 H2O
*     500.0000  -26.0149  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -1324.000 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*TcO2:2H2O(am)                 type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  166.0294 g
*   3 species in reaction
* -2.0000 H+              1.0000 TcO++              3.0000 H2O
*     500.0000  -4.2319  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -836.300 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*TcO3                          type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  145.9982 g
*   3 species in reaction
* -1.0000 H2O              1.0000 TcO4--             2.0000 H+
*     500.0000  -23.1483  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   reference-state data source = 83rar
*       delG0f =  -459.849 kj/mol
*       delH0f =  -540.000 kj/mol
*       S0PrTr =    72.400 j/(mol*K)

*Not in NEA database
*TcOH                          type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  115.0073 g
*   4 species in reaction
* -3.0000 H+              -0.5000 O2(aq)            1.0000
Tc+++
* 2.0000 H2O
*     500.0000  24.9009  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 1 [reported delG0f used]
*   reference-state data source = 83rar
*       delG0f =  -234.700 kj/mol
*       delH0f =     N/A
*       S0PrTr =     N/A

*Not in NEA database
*TcS2                          type=
*   formula=
*   mole vol.=  32.000 cc     mole wt.=  162.1320 g
*   3 species in reaction
* -1.0000 H2O              1.0000 TcO++              2.0000 HS-
*     500.0000  -65.9742  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
```

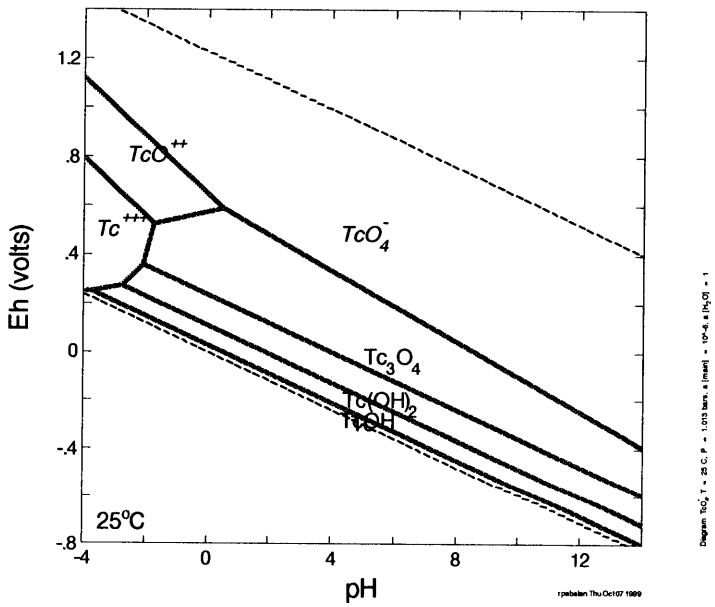
```
*   reference-state data source = 83rar
*       delG0f =   -216.067 kj/mol
*       delH0f =   -224.000 kj/mol
*       S0PrTr =     71.000 j/(mol*K)

*Not in NEA database
*TcS3                    type=
*   formula=
*   mole vol.=   0.000 cc     mole wt.=  194.1980 g
*   4 species in reaction
* -4.0000 H2O              1.0000 TcO4--          3.0000 HS-
* 5.0000 H+
*     500.0000 -119.5008  500.0000  500.0000
*     500.0000  500.0000  500.0000  500.0000
*   gflag = 2 [calculated delG0f(delH0f,S0PrTr) used]
*   reference-state data source = 83rar
*       delG0f =   -262.386 kj/mol
*       delH0f =   -276.000 kj/mol
*       S0PrTr =     84.000 j/(mol*K)

TlTcO4                   type=
   formula=
   mole vol.=   0.000 cc     mole wt.=  366.386  g
   2 species in reaction
 1.0000 Tl+              1.0000 TcO4-
    500.0000   -5.3227  500.0000  500.0000
    500.0000  500.0000  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data
*       delG0f =   -700.174   kj/mol
*       delH0f =             kj/mol
*       S0PrTr =             j/(mol*K)

Tc2O7(g)
   mole wt.=  307.9958 g
   3 species in reaction
 -1.0000 H2O              2.0000 H+              2.0000 TcO4-
     23.8637  23.2673   18.4240   15.7206
     13.0380  10.9129  500.0000  500.0000
*   data from NEA website TCDATA file
*   reference-state data source =
*       delG0f =   -904.823 kj/mol
*       delH0f =  -1008.100 kj/mol
*       S0PrTr =    436.640 j/(mol*K)

Tc(g)
   mole wt.=   98.0000 g
   4 species in reaction
 -0.5000 H2O              1.0000 H+              1.0000 TcO4-
 -1.7500 O2(aq)
     500.0000  206.4413  5000.000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   del G from NEA website TCDATA file
*   reference-state data source =
*       delG0f =    630.700 kj/mol
*       delH0f =    675.000 kj/mol
*       S0PrTr =    181.050 j/(mol*K)

TcO(g)
   mole wt.=  113.9990 g
   4 species in reaction
 -0.5000 H2O              1.0000 H+              1.0000 TcO4-
 -1.2500 O2(aq)
     500.0000  157.1334  5000.000  500.0000
     500.0000  500.0000  500.0000  500.0000
*   del G from NEA website TCDATA file
*   reference-state data source =
*       delG0f =    357.5   kj/mol
*       delH0f =    390.000 kj/mol
*       S0PrTr =    244.100 j/(mol*K)

TcO2
   mole wt.=   129.999 g
   4 species in reaction
 -0.7500 O2(aq)          -0.5000 H2O              1.0000 H+
  1.0000 TcO4-
```

The following results compare technetium Eh-pH diagrams calculated with GWB using three data files: (1) the LLNL V8.R6 datafile, (2) the V8.R6 datafile modified by Bourcier of LLNL, and (3) the V8.R6 datafile modified by R. Pabalan to incorporate the NEA Tc values.

(1) Calculate technetium speciation using LLNL V8.R6 database:

```
Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C,  Pressure is 1.013 bars
Thermo dataset:  D:\Gwb\Gtdata\thermo.dat
Working directory:  d:\

Basis is:
 TcO4-                    Activity = 10^-6 (main species)
 H2O                      Activity = 10^0 (solvent)
 e-  (swapped for O2(aq))       Y-axis variable
 H+                       X-axis variable

X-axis plotted from -4 to 14;  Y-axis from -.8 to 1.4

Act2> go

 Calculating diagram for TcO4-
  Loaded 21 species and minerals:
      TcO4-        Tc+++        TcO++        TcO4--
      TcO4---      (TcO(OH)2)2(aq) H2TcO4(aq)     HTcO4-
      TcO(OH)2(aq)   TcOOH+       HTcO4        Tc
```

Tc(OH)2 Tc(OH)3 Tc2O7 Tc3O4
Tc4O7 TcO2:2H2O(am) TcO3 TcOH
Tc2O7(g)



(2) Calculate using the Bourcier-modified V8.R6 file:

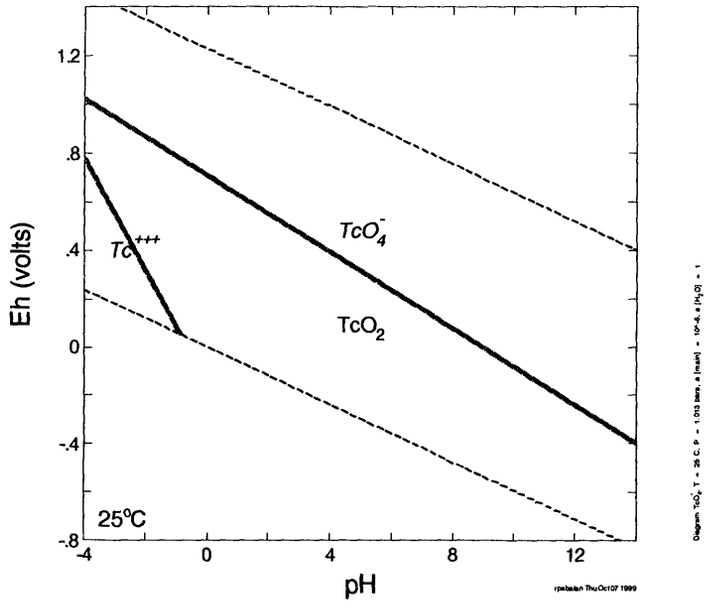
Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Bourcier data_R6.Tc
Working directory: d:

Basis is:
TcO4- Activity = 10^-6 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4

Act2> go

Calculating diagram for TcO4-
Loaded 12 species and minerals:
TcO4- Tc+++ TcO(OH)2(aq) TcOOH+
TcO(OH)3- TcO2 TcO2:1.6H2O Tc2O7
Tc2O7:H2O Tc2O7(g) Tc(g) TcO(g)



(3) Calculate using the datafile with NEA Tc parameters:

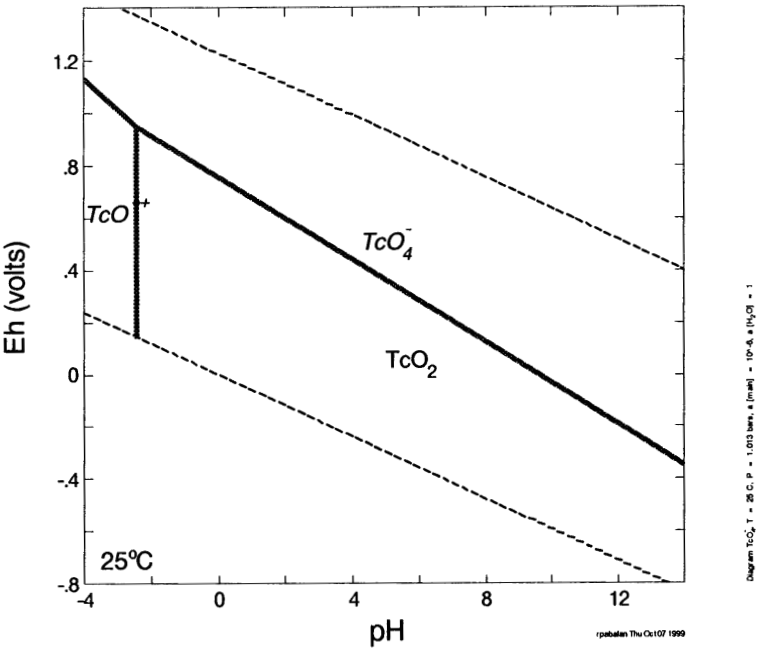
Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:
TcO4- Activity = 10^-6 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4

Act2> go

Calculating diagram for TcO4-
Loaded 13 species and minerals:
TcO4- TcO++ TcO4-- TcO(OH)2(aq)
TcOOH+ TcO(OH)3- TcO2 TcO2:1.6H2O
Tc2O7 Tc2O7:H2O Tc2O7(g) Tc(g)
TcO(g)



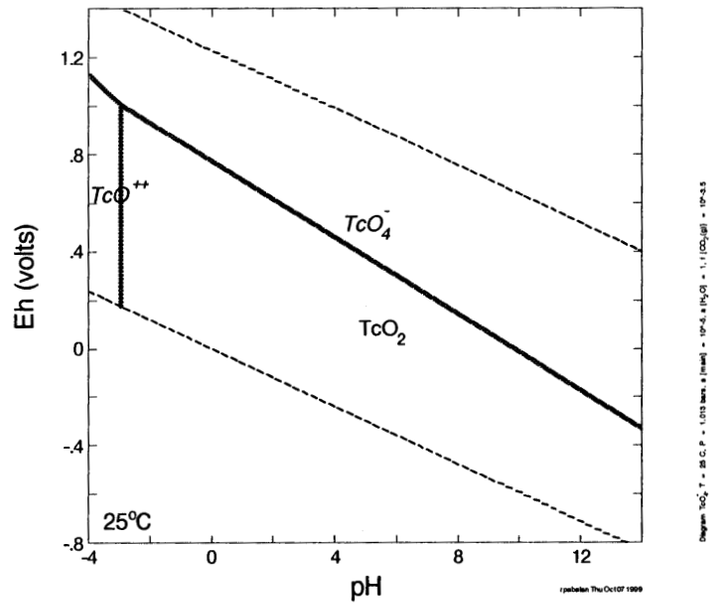
(4) As in (3), but have carbonate in the system. Fix pCO2(g) at $10^{-3.5}$ atm.

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:
TcO4- Activity = 10^-5 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4
Other "show" options: type "show show"
Act2> go

Calculating diagram for TcO4-
Loaded 15 species and minerals:
TcO4- TcO++ TcO4-- TcCO3(OH)2(aq)
TcCO3(OH)3- TcO(OH)2(aq) TcOOH+ TcO(OH)3-
TcO2 TcO2:1.6H2O Tc2O7 Tc2O7:H2O
Tc2O7(g) Tc(g) TcO(g)



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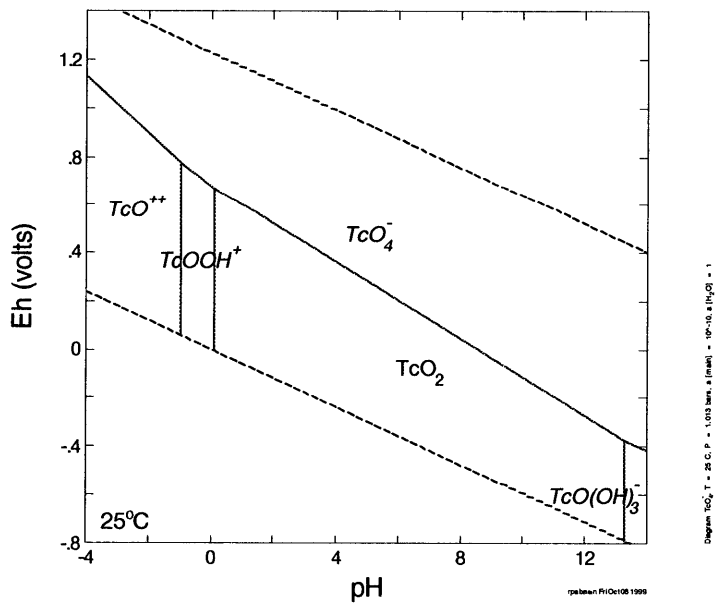
October 8, 1999

(5) As in (3), calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -10

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:
TcO4- Activity = 10^-10 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4

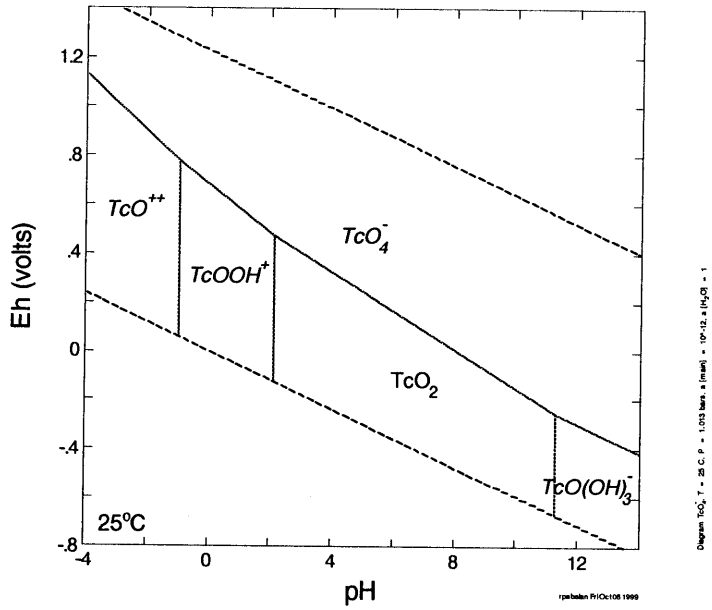


(6) As in (3), calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -12

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:
TcO4- Activity = 10^-12 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



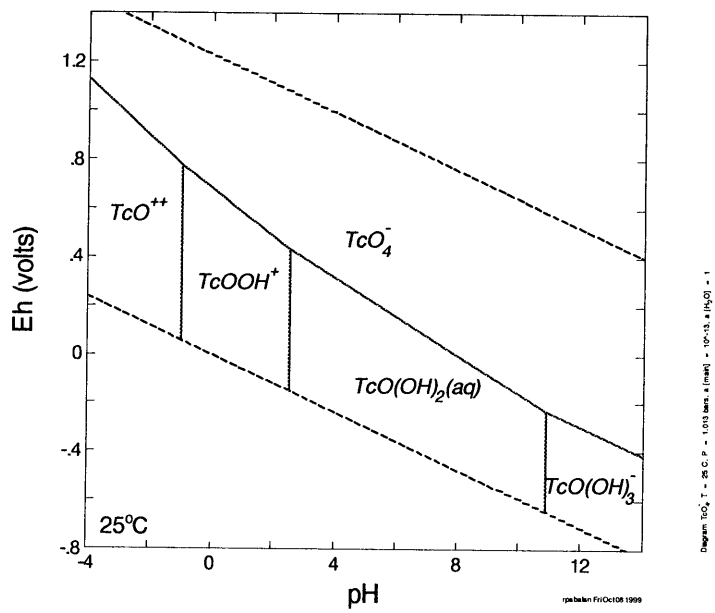
(7) As in (3), `calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -13`

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:

TcO4- Activity = 10^-13 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



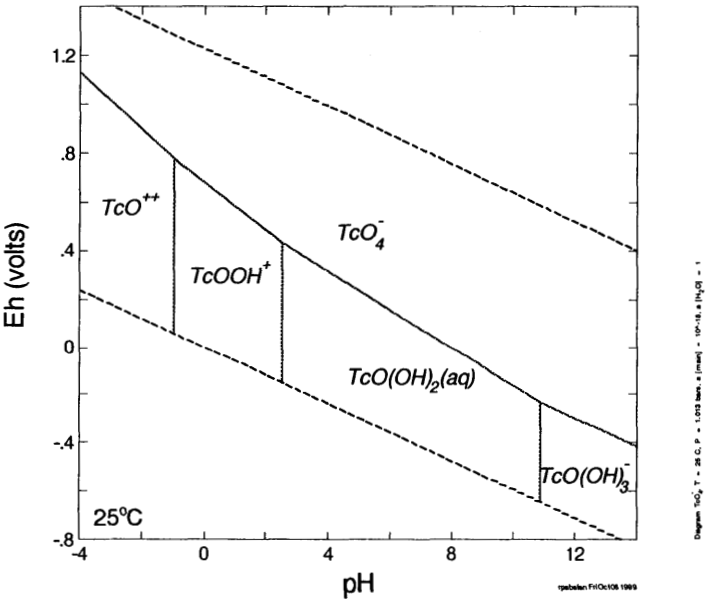
(8) As in (3), `calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -18`

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:

TcO4- Activity = 10^-18 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



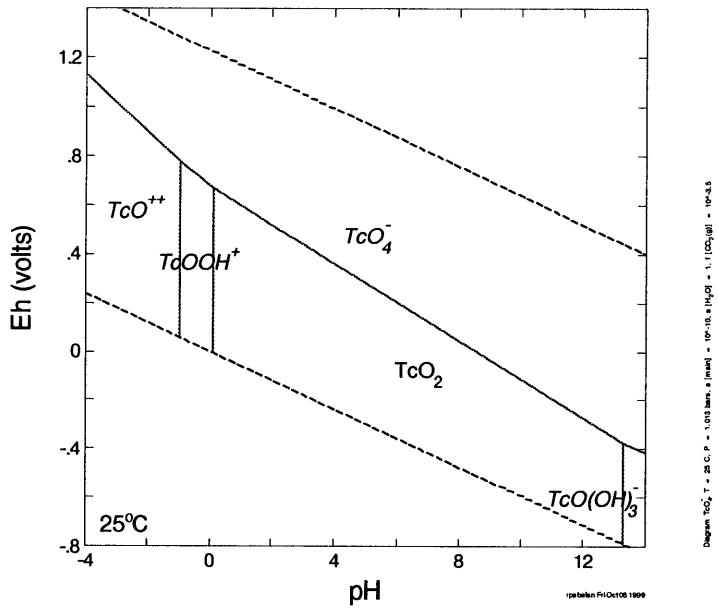
(9) As in (3), `calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -10 and set log fCO2(g) = -3.5`

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:

TcO4- Activity = 10^-10 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



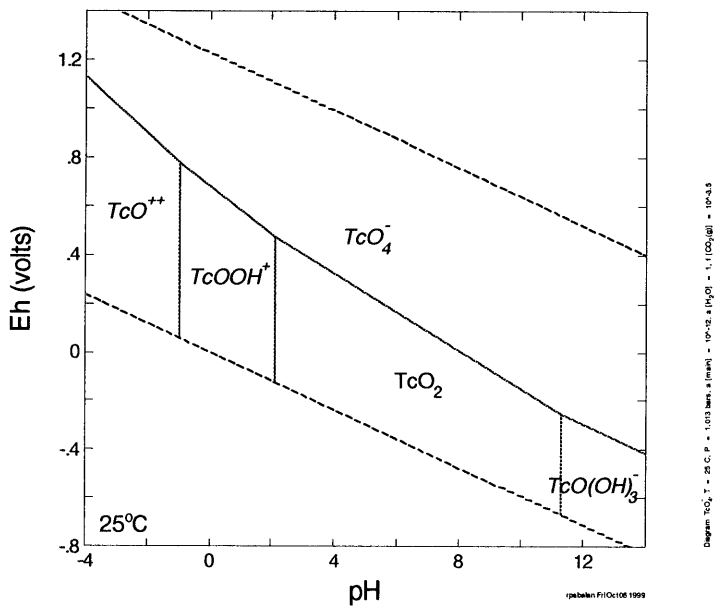
(10) As in (9), calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -12

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:
TcO4- Activity = 10^-12 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable

H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



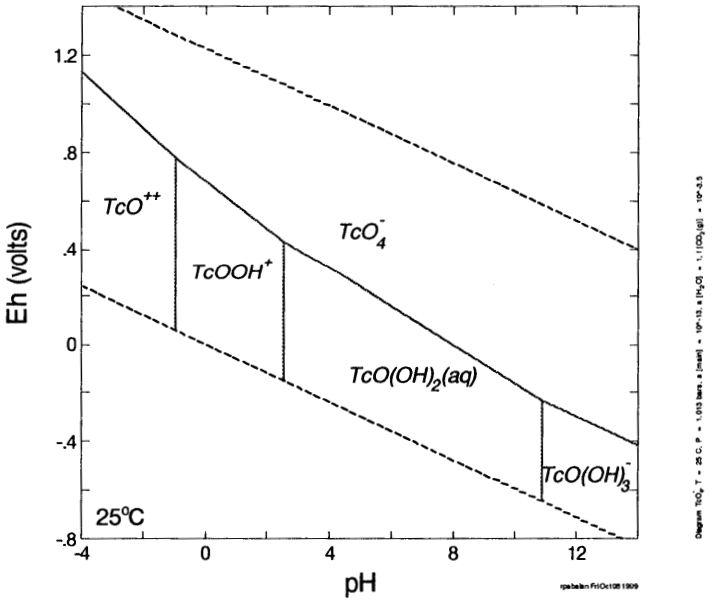
(11) As in (9), calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -13

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:

TcO4- Activity = 10^-13 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



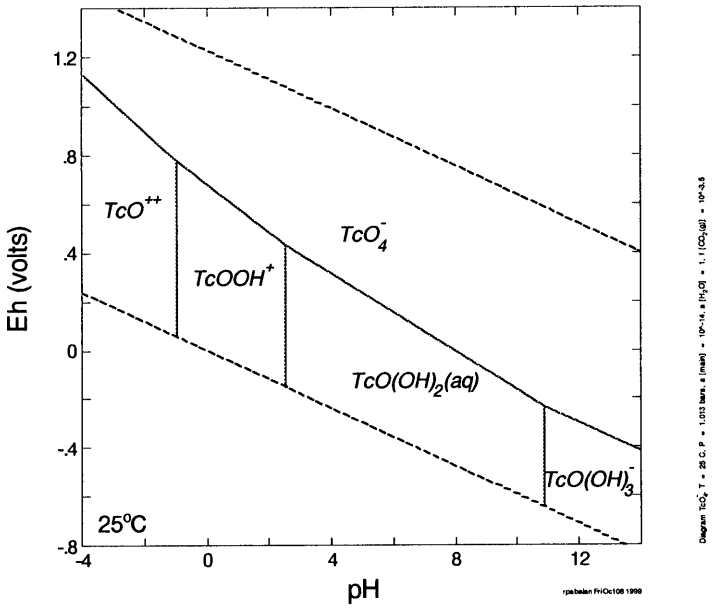
(12) As in (9), calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -14

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:

TcO4- Activity = 10^-14 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



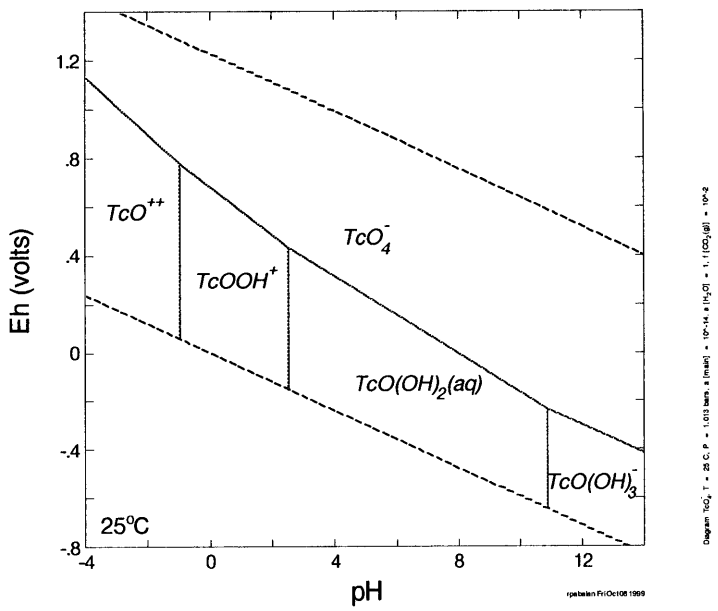
(13) As in (9), calculate using the datafile with NEA Tc parameters, but use log a TcO4- = -14. Also, use log f CO2(g) = -2.00

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

Basis is:
TcO4- *Activity = 10^-14 (main species)*

H2O *Activity = 10^0 (solvent)*
e- *(swapped for O2(aq))* *Y-axis variable*
H+ *X-axis variable*
CO2(g) *(swapped for HCO3-)* *Fugacity = 10^-2*

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4



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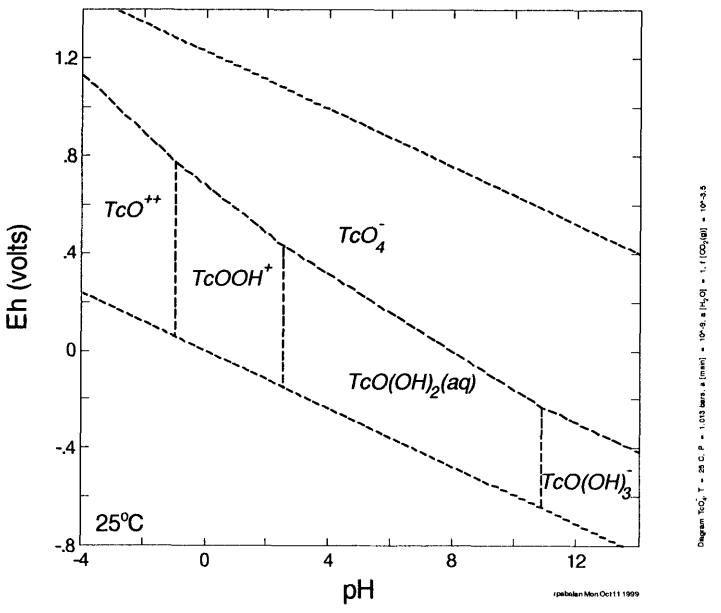
(14) As in (9), but use log a TcO4- = -9 and set log fCO2(g) = -3.5. Also, calculate only aqueous species stability fields.

Diagram speciation of TcO4- on Eh (volts) vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:

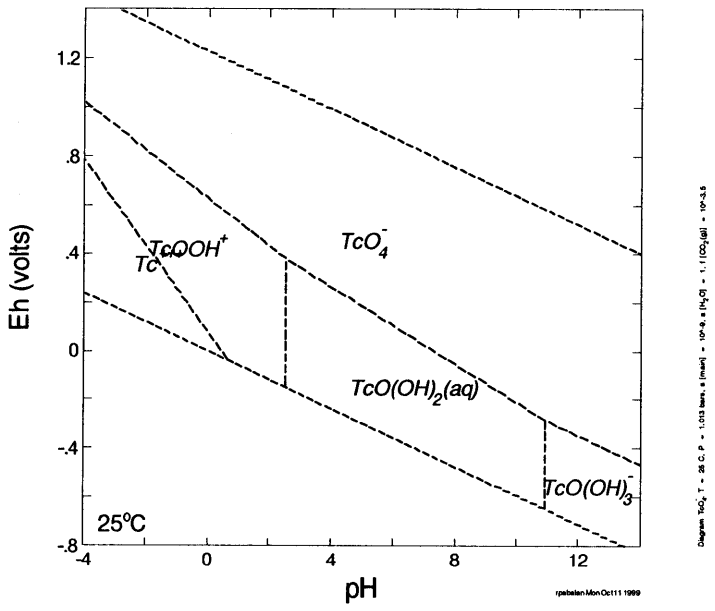
Basis is:
TcO4- Activity = 10^-9 (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Y-axis variable
H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from -4 to 14; Y-axis from -.8 to 1.4
Other "show" options: type "show show"
show options
Act2 option settings:
labels selected
reactions not selected
full_dgm not selected
aqueous_dgm selected
gases selected
overlay not selected
mosaic_bounds selected
water_limits selected
extrapolate not selected

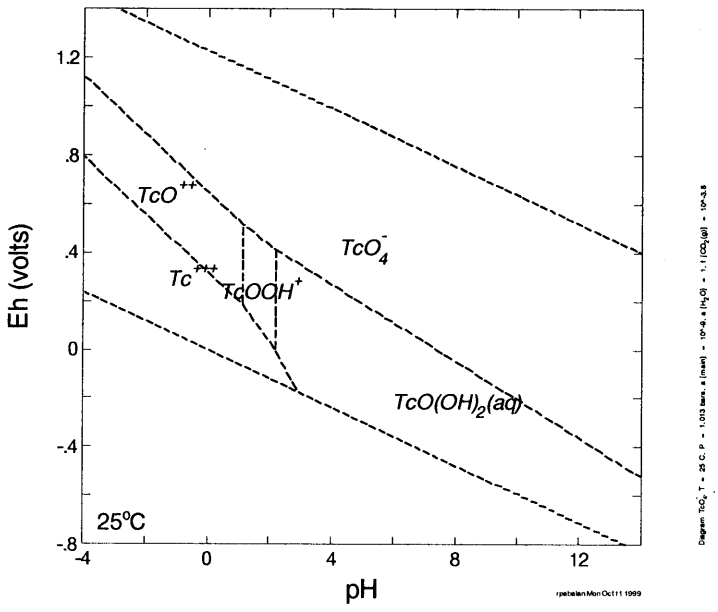
Calculating diagram for TcO4-
Loaded 15 species and minerals:
TcO4- TcO++ TcO4-- TcCO3(OH)2(aq) TcCO3(OH)3- TcO(OH)2(aq) TcOOH+ TcO(OH)3- TcO2 TcO2:1.6H2O Tc2O7 Tc2O7:H2O Tc2O7(g) Tc(g) TcO(g)



(15) As in (14), but use Bourcier database.



(16) As in (14), but use LLNL V8.R6 database.



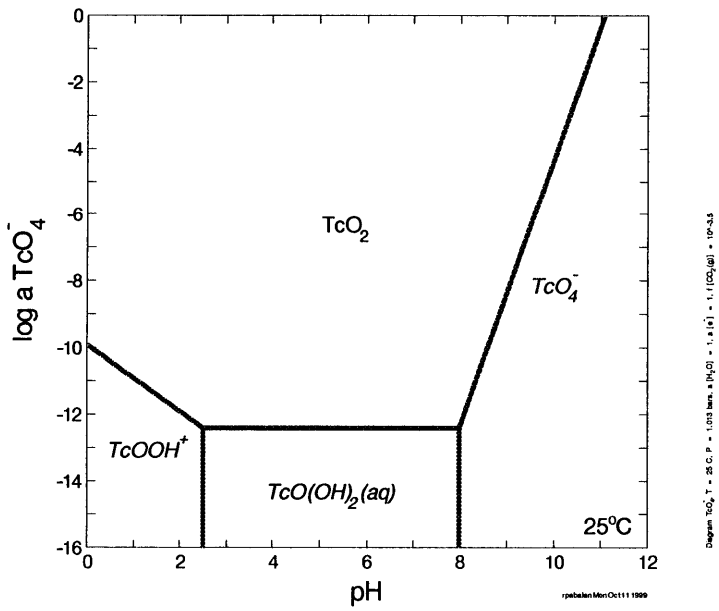
(17) Calculate solubility diagram as a function of pH for fixed Eh = 0.0 v and fCO2 = -3.5 atm. Use NEA Tc data.

Diagram speciation of TcO4- on TcO4- vs pH
Temperature is 25 C, Pressure is 1.013 bars
Thermo dataset: D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory: d:\

Basis is:
TcO4- Y-axis variable (main species)
H2O Activity = 10^0 (solvent)
e- (swapped for O2(aq)) Activity = 10^0
H+ X-axis variable
CO2(g) (swapped for HCO3-) Fugacity = 10^-3.5

X-axis plotted from 0 to 12; Y-axis from -16 to 0

Calculating diagram for TcO4-
Loaded 15 species and minerals:
TcO4- TcO++ TcO4-- TcCO3(OH)2(aq)
TcCO3(OH)3- TcO(OH)2(aq) TcOOH+ TcO(OH)3-
TcO2 TcO2:1.6H2O Tc2O7 Tc2O7:H2O
Tc2O7(g) Tc(g) TcO(g)



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Objective: Calculate solubility of technetium in YM groundwater based on chemical composition derived for WT#17 samples. See lab notebook #278, page 182 for detailed information on data downloaded from the DOE YM web site.

The following WT#17 data are for calcium:

DTN: LAAM831311AQ98.002 s98311_001 Data Report

	Ca (mg/L)	Sample Number	Date	Activity	
1	11.73	WT-005**	01/27/1998	Pumping	
2	11.42	WT-011	06/04/1998	Bailer	1306'
3	11.15	WT-012**	06/04/1998	Bailer	1306'
4	11.64	WT-012**	06/04/1998	Bailer	1306'
5	11.56	WT-013	06/04/1998	Bailer	1350'
6	11.8	WT-013	06/04/1998	Bailer	1350'
7	11.53	WT-013	06/04/1998	Bailer	1350'
8	12.67	WT-037*	07/01/1998	Pumping	
9	14.59	WT-039*	07/01/1998	Pumping	
10	9.18	WT-041*	07/01/1998	Pumping	
11	8.59	WT-043*	07/01/1998	Pumping	
12	8.07	WT-044*	07/01/1998	Pumping	

Average	11.16083333
StDeviation	1.795142224

The following WT#17 data are for sodium:

DTN: LAAM831311AQ98.002 s98311_006 Data Report

	Na (mg/L)	Sample Number	Date	Activity	
1	19.78	WT-005**	01/27/1998	Pumping	
2	21.28	WT-011	06/04/1998	Bailer	1306'
3	20.41	WT-012**	06/04/1998	Bailer	1306'
4	20.87	WT-012**	06/04/1998	Bailer	1306'
5	23.26	WT-013	06/04/1998	Bailer	1350'
6	20.57	WT-013	06/04/1998	Bailer	1350'
7	20.36	WT-013	06/04/1998	Bailer	1350'
8	24.41	WT-037*	07/01/1998	Pumping	
9	21.76	WT-039*	07/01/1998	Pumping	
10	20.32	WT-041*	07/01/1998	Pumping	
11	20.08	WT-043*	07/01/1998	Pumping	
12	20.26	WT-044*	07/01/1998	Pumping	

Average	21.11333333
StDeviation	1.399411132

The following WT#17 data are for potassium:

DTN: LAAM831311AQ98.002 s98311_007 Data Report

	K (mg/L)	Sample Number	Date	Activity	
1	4.51	WT-005**	01/27/1998	Pumping	
2	3.3	WT-011	06/04/1998	Bailer	1306'
3	ND	WT-012**	06/04/1998	Bailer	1306'
4	N/M	WT-012**	06/04/1998	Bailer	1306'
5	5.34	WT-013	06/04/1998	Bailer	1350'
6	N/M	WT-013	06/04/1998	Bailer	1350'
7	ND	WT-013	06/04/1998	Bailer	1350'
8	3.29	WT-037*	07/01/1998	Pumping	
9	2.56	WT-039*	07/01/1998	Pumping	
10	1.44	WT-041*	07/01/1998	Pumping	
11	3.2	WT-043*	07/01/1998	Pumping	
12	2.25	WT-044*	07/01/1998	Pumping	

Average	3.23625
StDeviation	1.237104194

The following WT#17 data are for magnesium:

DTN: LAAM831311AQ98.002 s98311_005 Data Report

	Mg (mg/L)	Sample Number	Date	Activity	
1	0.42	WT-005**	01/27/1998	Pumping	
2	1.01	WT-011	06/04/1998	Bailer	1306'
3	1.41	WT-012**	06/04/1998	Bailer	1306'
4	1.45	WT-012**	06/04/1998	Bailer	1306'
5	1.19	WT-013	06/04/1998	Bailer	1350'
6	1.18	WT-013	06/04/1998	Bailer	1350'
7	1.18	WT-013	06/04/1998	Bailer	1350'
8	1.18	WT-037*	07/01/1998	Pumping	
9	1.94	WT-039*	07/01/1998	Pumping	
10	0.94	WT-041*	07/01/1998	Pumping	
11	0.87	WT-043*	07/01/1998	Pumping	
12	0.83	WT-044*	07/01/1998	Pumping	

Average	1.133333333
StDeviation	0.376595595

The following WT#17 data are for silica:

DTN: LAAM831311AQ98.002 s98311_002 Data Report

	Si (mg/L)	Sample Number	Date	Activity

1	22.15	WT-005**	01/27/1998	Pumping	
2	15.6	WT-011	06/04/1998	Bailer	1306'
3	15.97	WT-012**	06/04/1998	Bailer	1306'
4	15.08	WT-012**	06/04/1998	Bailer	1306'
5	20.44	WT-013	06/04/1998	Bailer	1350'
6	17.14	WT-013	06/04/1998	Bailer	1350'
7	17.23	WT-013	06/04/1998	Bailer	1350'
8	24.71	WT-037*	07/01/1998	Pumping	
9	20.57	WT-039*	07/01/1998	Pumping	
10	24.62	WT-041*	07/01/1998	Pumping	
11	23.69	WT-043*	07/01/1998	Pumping	
12	22.9	WT-044*	07/01/1998	Pumping	

Average	20.00833333
StDeviation	3.647596552

The following WT#17 data are for manganese:

DTN: LAAM831311AQ98.002 s98311_003 Data Report

	Mn (mg/L)	Sample Number	Date	Activity	
1	0.16	WT-005**	01/27/1998	Pumping	
2	0.42	WT-011	06/04/1998	Bailer	1306'
3	0.34	WT-012**	06/04/1998	Bailer	1306'
4	0.37	WT-012**	06/04/1998	Bailer	1306'
5	0.4	WT-013	06/04/1998	Bailer	1350'
6	0.44	WT-013	06/04/1998	Bailer	1350'
7	0.38	WT-013	06/04/1998	Bailer	1350'
8	0.42	WT-037*	07/01/1998	Pumping	
9	0.73	WT-039*	07/01/1998	Pumping	
10	0.4	WT-041*	07/01/1998	Pumping	
11	0.41	WT-043*	07/01/1998	Pumping	
12	0.41	WT-044*	07/01/1998	Pumping	

Average	0.406666667
StDeviation	0.125577454

The following WT#17 data are for iron:

DTN: LAAM831311AQ98.002 s98311_004 Data Report

	Fe (mg/L)	Sample Number	Date	Activity	
1	0.6	WT-005**	01/27/1998	Pumping	
2	1.16	WT-011	06/04/1998	Bailer	1306'
3	0.78	WT-012**	06/04/1998	Bailer	1306'
4	0.85	WT-012**	06/04/1998	Bailer	1306'
5	0.05	WT-013	06/04/1998	Bailer	1350'

6	1.82	WT-013	06/04/1998	Bailer	1350'
7	1.73	WT-013	06/04/1998	Bailer	1350'
8	2.13	WT-037*	07/01/1998	Pumping	
9	0.03	WT-039*	07/01/1998	Pumping	
10	0.01	WT-041*	07/01/1998	Pumping	
11	0.04	WT-043*	07/01/1998	Pumping	
12	0.16	WT-044*	07/01/1998	Pumping	

Average	0.78
StDeviation	0.775101166

The following WT#17 data are for chloride:

DTN: LAAM831311AQ98.001 s98310_001 Data Report

	Cl (mg/L)	Sample Number	Date	Method	Condition	
1	6.33	WT-005	01/27/1998	Pumping	Filtered/Acidified	
2	6.76	WT-006	01/27/1998	Pumping	Unfiltered	
3	6.97	WT-007	01/28/1998	Pumping	Unfiltered	
4	15.61	WT-011	06/04/1998	Bailer	Unfiltered,	1306*
5	15.77	WT-013	06/04/1998	Bailer	Unfiltered,	1350*
6	15.8	WT-036	07/01/1998	Pumping	Unfiltered	
7	16.36	WT-037	07/01/1998	Pumping	Filtered	
8	8.77	WT-038	07/01/1998	Pumping	Unfiltered	
9	9.39	WT-039	07/01/1998	Pumping	Filtered	
10	5.83	WT-040	07/01/1998	Pumping	Unfiltered	
11	5.78	WT-041	07/01/1998	Pumping	Filtered	
12	5.89	WT-042	07/01/1998	Pumping	Unfiltered	
13	6.03	WT-043	07/01/1998	Pumping	Filtered	
14	6.09	WT-044	07/01/1998	Pumping	Filtered	
15	6.57	WT-045	07/01/1998	Pumping	Unfiltered	

Average	9.196666667
StDeviation	4.302077018

The following WT#17 data are for bromide:

DTN: LAAM831311AQ98.001 s98310_002 Data Report

	Br (mg/L)	Sample Number	Date	Method	Condition	
1	nm	WT-005	01/27/1998	Pumping	Filtered/Acidified	
2	0.19	WT-006	01/27/1998	Pumping	Unfiltered	
3	0.2	WT-007	01/28/1998	Pumping	Unfiltered	
4	2.78	WT-011	06/04/1998	Bailer	Unfiltered,	1306*
5	2.91	WT-013	06/04/1998	Bailer	Unfiltered,	1350*

6	2.88	WT-036	07/01/1998	Pumping	Unfiltered
7	2.94	WT-037	07/01/1998	Pumping	Filtered
8	1.16	WT-038	07/01/1998	Pumping	Unfiltered
9	1.28	WT-039	07/01/1998	Pumping	Filtered
10	0.16	WT-040	07/01/1998	Pumping	Unfiltered
11	0.15	WT-041	07/01/1998	Pumping	Filtered
12	0.18	WT-042	07/01/1998	Pumping	Unfiltered
13	0.19	WT-043	07/01/1998	Pumping	Filtered
14	0.22	WT-044	07/01/1998	Pumping	Filtered
15	0.26	WT-045	07/01/1998	Pumping	Unfiltered

Average	1.107142857
StDeviation	1.217389569

The following WT#17 data are for fluoride:

DTN: LAAM831311AQ98.001 s98310_003 Data Report

	F (mg/L)	Sample Number	Date	Method	Condition
1	nm	WT-005	01/27/1998	Pumping	Filtered/Acidified
2	nm	WT-006	01/27/1998	Pumping	Unfiltered
3	nm	WT-007	01/28/1998	Pumping	Unfiltered
4	2.51	WT-011	06/04/1998	Bailer	Unfiltered, 1306*
5	2.51	WT-013	06/04/1998	Bailer	Unfiltered, 1350*
6	2.89	WT-036	07/01/1998	Pumping	Unfiltered
7	3.32	WT-037	07/01/1998	Pumping	Filtered
8	4.01	WT-038	07/01/1998	Pumping	Unfiltered
9	4.5	WT-039	07/01/1998	Pumping	Filtered
10	3.42	WT-040	07/01/1998	Pumping	Unfiltered
11	3.41	WT-041	07/01/1998	Pumping	Filtered
12	3.22	WT-042	07/01/1998	Pumping	Unfiltered
13	3.41	WT-043	07/01/1998	Pumping	Filtered
14	3.44	WT-044	07/01/1998	Pumping	Filtered
15	3.41	WT-045	07/01/1998	Pumping	Unfiltered

Average	3.3375
StDeviation	0.557643174

The following WT#17 data are for nitrate:

DTN: LAAM831311AQ98.001 s98310_004 Data Report

	NO3 (mg/L)	Sample Number	Date	Method	Condition

1	nm	WT-005	01/27/1998	Pumping	Filtered/Acidified	
2	nm	WT-006	01/27/1998	Pumping	Unfiltered	
3	nm	WT-007	01/28/1998	Pumping	Unfiltered	
4	0.07	WT-011	06/04/1998	Bailer	Unfiltered,	1306*
5	nd	WT-013	06/04/1998	Bailer	Unfiltered,	1350*
6	nm	WT-036	07/01/1998	Pumping	Unfiltered	
7	nm	WT-037	07/01/1998	Pumping	Filtered	
8	nm	WT-038	07/01/1998	Pumping	Unfiltered	
9	0.49	WT-039	07/01/1998	Pumping	Filtered	
10	4.92	WT-040	07/01/1998	Pumping	Unfiltered	
11	4.96	WT-041	07/01/1998	Pumping	Filtered	
12	4.93	WT-042	07/01/1998	Pumping	Unfiltered	
13	5.03	WT-043	07/01/1998	Pumping	Filtered	
14	4.99	WT-044	07/01/1998	Pumping	Filtered	
15	4.99	WT-045	07/01/1998	Pumping	Unfiltered	

Average	3.7975
StDeviation	2.17423057

The following WT#17 data are for phosphate:

Phosphate data all less than 0.1 ppm

The following WT#17 data are for sulfate:

DTN: LAAM831311AQ98.001 s98310_006 Data Report

	SO4 (mg/L)	Sample Number	Date	Method	Condition	
1	18.79	WT-005	01/27/1998	Pumping	Filtered/Acidified	
2	20.30	WT-006	01/27/1998	Pumping	Unfiltered	
3	21.88	WT-007	01/28/1998	Pumping	Unfiltered	
4	10.58	WT-011	06/04/1998	Bailer	Unfiltered,	1306*
5	14.44	WT-013	06/04/1998	Bailer	Unfiltered,	1350*
6	0.37	WT-036	07/01/1998	Pumping	Unfiltered	
7	0.62	WT-037	07/01/1998	Pumping	Filtered	
8	10.86	WT-038	07/01/1998	Pumping	Unfiltered	
9	11.72	WT-039	07/01/1998	Pumping	Filtered	

10	19.99	WT-040	07/01/1998	Pumping	Unfiltered
11	19.82	WT-041	07/01/1998	Pumping	Filtered
12	19.45	WT-042	07/01/1998	Pumping	Unfiltered
13	19.43	WT-043	07/01/1998	Pumping	Filtered
14	19.19	WT-044	07/01/1998	Pumping	Filtered
15	19.75	WT-045	07/01/1998	Pumping	Unfiltered

Average	15.146
StDeviation	6.998553728

The following are pH values of WT#17 water taken from borehole measurements:

	pH	Date measured	Temp (C)	Depth (feet)			
1	nm	05/06/1998	nm	1100	Measured	going	down
2	nm	05/06/1998	nm	1200	Measured	going	down
3	nm	05/06/1998	nm	1227	Measured	going	down
4	nm	05/06/1998	nm	1257	Measured	going	down
5	nm	05/06/1998	nm	1267	Measured	going	down
6	nm	05/06/1998	nm	1277	Measured	going	down
7	nm	05/06/1998	nm	1280	Measured	going	down
8	nm	05/06/1998	nm	1287	Measured	going	down
9	nm	05/06/1998	nm	1288	Measured	going	down
10	nm	05/06/1998	nm	1289	Measured	going	down
11	7	05/06/1998	27.8	1290	Measured	going	down
12	nm	05/06/1998	nm	1291	Measured	going	down
13	nm	05/06/1998	nm	1294	Measured	going	down
14	7.07	05/06/1998	31.0	1300	Measured	going	down
15	7.09	05/06/1998	31.0	1310	Measured	going	down
16	7.06	05/06/1998	31.1	1320	Measured	going	down
17	7.06	05/06/1998	31.2	1330	Measured	going	down
18	7.04	05/06/1998	31.3	1340	Measured	going	down
19	7.02	05/06/1998	31.3	1350	Measured	going	down
20	7.03	05/06/1998	31.3	1360	Measured	going	down
21	7.03	05/06/1998	31.4	1370	Measured	going	down
22	nm	07/15/1998	nm	1100	Measured	going	down
23	nm	07/15/1998	nm	1200	Measured	going	down

24	nm	07/15/1998	nm	1227	Measured	going	down
25	nm	07/15/1998	nm	1257	Measured	going	down
26	nm	07/15/1998	nm	1267	Measured	going	down
27	nm	07/15/1998	nm	1277	Measured	going	down
28	nm	07/15/1998	nm	1280	Measured	going	down
29	nm	07/15/1998	nm	1287	Measured	going	down
30	nm	07/15/1998	nm	1288	Measured	going	down
31	nm	07/15/1998	nm	1289	Measured	going	down
32		7.22 07/15/1998	31.1	1290	Measured	going	down
33	nm	07/15/1998	nm	1291	Measured	going	down
34	nm	07/15/1998	nm	1294	Measured	going	down
35		7.2 07/15/1998	31.2	1300	Measured	going	down
36		7.21 07/15/1998	31.3	1310	Measured	going	down
37		7.22 07/15/1998	31.3	1320	Measured	going	down
38		7.22 07/15/1998	31.3	1330	Measured	going	down
39		7.23 07/15/1998	31.4	1340	Measured	going	down
40		7.22 07/15/1998	31.4	1350	Measured	going	down
41		7.23 07/15/1998	31.4	1360	Measured	going	down
42		7.23 07/15/1998	31.4	1370	Measured	going	down
43	nm	07/15/1998	nm	1100	Measured	coming	up
44	nm	07/15/1998	nm	1200	Measured	coming	up
45	nm	07/15/1998	nm	1227	Measured	coming	up
46	nm	07/15/1998	nm	1257	Measured	coming	up
47	nm	07/15/1998	nm	1267	Measured	coming	up
48	nm	07/15/1998	nm	1277	Measured	coming	up
49	nm	07/15/1998	nm	1280	Measured	coming	up
50	nm	07/15/1998	nm	1287	Measured	coming	up
51	nm	07/15/1998	nm	1288	Measured	coming	up
52	nm	07/15/1998	nm	1289	Measured	coming	up
53	nm	07/15/1998	nm	1290	Measured	coming	up
54	nm	07/15/1998	nm	1291	Measured	coming	up
55	nm	07/15/1998	nm	1294	Measured	coming	up
56	nm	07/15/1998	nm	1300	Measured	coming	up
57		7.19 07/15/1998	31.3	1310	Measured	coming	up

58	nm	07/15/1998	nm	1320	Measured	coming	up
59	nm	07/15/1998	nm	1330	Measured	coming	up
60	7.21	07/15/1998	31.4	1340	Measured	coming	up
61	nm	07/15/1998	nm	1350	Measured	coming	up
62	nm	07/15/1998	nm	1360	Measured	coming	up
63	7.23	07/15/1998	31.4	1370	Measured	coming	up

Average	7.143333
StDev	0.090019

The following is a summary of chemistry data for WT#17. All data in the previous tables were used to calculate averages and standard deviations.

	Average (ppm)	Std Deviation
Sodium	21.113	1.399
Silicon	20.008	3.648
Calcium	11.161	1.795
Potassium	3.236	1.237
Magnesium	1.133	0.377
Iron	0.780	0.775
Manganese	0.407	0.126
Sulfate	15.146	6.999
Chloride	9.197	4.302
Nitrate	3.798	2.174
Fluoride	3.338	0.558
Bromide	1.107	1.217
Phosphate	<0.1	
pH	7.14	0.09

(1) GWB calculations using a subset of WT#17 composition.

```
Temperature is 25 C
    Thermo dataset:  D:\Gwb\Gtdata\LLNL
database\Pabalan.dat
    Working directory:  d:\
    Options: Debye-Huckel

    Basis is:
     H2O                1 kg solvent
     Na+               total mg/kg = 21.113
     SiO2(aq)            total mg/kg = 20.008
     Ca++              total mg/kg = 11.161
     K+               total mg/kg = 3.236
     SO4--              total mg/kg = 15.146
     Cl-              total mg/kg = 9.197
     HCO3-              charge balance
     H+               pH = 7.14

    No reactants specified.

    Other "show" options: type "show show"
React> go

Solving for initial system.

 Loaded:  34 aqueous species,
        68 minerals,
         3 gases,
         0 surface species,
         9 elements,
         6 oxides.

N-R converged in  16 its., resmax =   5.15e-013, Xi =  0.0000
 Charge balance: HCO3- molality adjusted from .001 to
.001131
```

3 supersaturated phases, most = Quartz
Swapping Quartz in for SiO2(aq)
Swapping CO2(aq) in for H+

The following is the React output file

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 7.140
Ionic strength = 0.001972
Activity of water = 0.999991
Solvent mass = 1.000000 kg
Solution mass = 1.000152 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 152 mg/kg sol'n

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
HCO3-	0.0009771	59.61	0.9520	-3.0314
Na+	0.0009164	21.06	0.9520	-3.0593
SiO2(aq)	0.0003324	19.97	1.0000	-3.4783
Ca++	0.0002721	10.90	0.8252	-3.6487
Cl-	0.0002594	9.194	0.9513	-3.6078
SO4--	0.0001532	14.71	0.8208	-3.9005
CO2(aq)	0.0001490	6.558	1.0000	-3.8267
K+	8.269e-005	3.232	0.9513	-4.1042
CaSO4(aq)	3.646e-006	0.4963	1.0000	-5.4382
CaHCO3+	2.443e-006	0.2469	0.9520	-5.6335
NaHCO3(aq)	1.157e-006	0.09719	1.0000	-5.9366
NaSO4-	7.613e-007	0.09062	0.9520	-6.1398
CO3--	7.327e-007	0.04396	0.8220	-6.2202
HSiO3-	5.378e-007	0.04145	0.9520	-6.2908
CaCO3(aq)	2.872e-007	0.02874	1.0000	-6.5419
OH-	1.467e-007	0.002495	0.9516	-6.8551
KSO4-	7.874e-008	0.01064	0.9520	-7.1252
H+	7.586e-008	7.644e-005	0.9550	-7.1400
NaCl(aq)	3.597e-008	0.002102	1.0000	-7.4441
NaHSiO3(aq)	1.988e-008	0.001989	1.0000	-7.7016
CaCl+	1.173e-008	0.0008858	0.9520	-7.9521

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Quartz	0.5210s/sat	Cristobalite(bet	-0.4730
Tridymite	0.3495s/sat	SiO2(am)	-0.7647
Chalcedony	0.2498s/sat	Calcite	-1.3889
Cristobalite(alp	-0.0295	Aragonite	-1.5333
Ice	-0.1387	Monohydrocalcite	-2.2226
Coesite	-0.2890		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
H2O(g)	0.02598	-1.585
CO2(g)	0.004387	-2.358
HCl(g)	8.846e-018	-17.053

Original basis	total moles	In fluid moles	Sorbed mg/kg	Sorbed moles	mg/kg
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
H+	0.0001474	0.0001474	0.1485		
H2O	55.51	55.51	9.999e+005		
HCO3-	0.001131	0.001131	68.98		
K+	8.277e-005	8.277e-005	3.236		
Na+	0.0009184	0.0009184	21.11		
SO4--	0.0001577	0.0001577	15.14		
SiO2(aq)	0.0003330	0.0003330	20.00		

Elemental composition	total moles	In fluid moles	Sorbed mg/kg	Sorbed moles	mg/kg
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001131	0.001131	13.58		
Chlorine	0.0002594	0.0002594	9.196		
Hydrogen	111.0	111.0	1.119e+005		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.236		
Silicon	0.0003330	0.0003330	9.351		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		

N-R converged in 7 its., resmax = 4.54e-014, Xi = 0.0000

No reaction path specified.

```
Step #   0          Xi = 0.0000
Temperature =  25.0 C   Pressure =  1.013 bars
pH =  7.141
Ionic strength     =   0.001972
Activity of water  =   0.999991
Solvent mass       =   1.000000 kg
Solution mass      =   1.000138 kg
Solution density   =   1.013   g/cm3
Chlorinity         =   0.000259 molal
Dissolved solids   =        138 mg/kg sol'n

 Minerals in system     moles     log moles     grams
volume (cm3)
-----------------------------------------------------------
 Quartz          0.0002327    -3.633      0.01398
0.005279
                                  ____________
____________
      (total)                     0.01398      0.005279

 Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
-----------------------------------------------------------
 HCO3-          0.0009775      59.63     0.9520     -
3.0313
 Na+            0.0009164      21.06     0.9520     -
3.0593
 Ca++           0.0002721      10.90     0.8252     -
3.6487
 Cl-            0.0002594      9.194     0.9513     -3.6078
 SO4--          0.0001532      14.71     0.8208     -
3.9005
 CO2(aq)        0.0001486      6.541     1.0000     -
3.8279
 SiO2(aq)       0.0001002      6.017     1.0000     -
3.9993
 K+             8.269e-005     3.232     0.9513     -
4.1042
 CaSO4(aq)      3.646e-006     0.4963    1.0000     -
5.4382
 CaHCO3+        2.444e-006     0.2470    0.9520     -
5.6333
 NaHCO3(aq)     1.158e-006     0.09723   1.0000
-5.9365
 NaSO4-         7.613e-007     0.09062   0.9520     -
6.1398
 CO3--          7.351e-007     0.04411   0.8220     -
6.2188
 CaCO3(aq)      2.881e-007     0.02883   1.0000     -
6.5404
 HSiO3-         1.625e-007     0.01253   0.9520     -
6.8105
 OH-            1.471e-007     0.002502  0.9516     -
6.8538
 KSO4-          7.874e-008     0.01064   0.9520     -
7.1252
 H+             7.563e-008     7.622e-005  0.9550     -
7.1413
 NaCl(aq)       3.597e-008     0.002102  1.0000     -
7.4440
 CaCl+          1.173e-008     0.0008858 0.9520     -
7.9521
  (only species > 1e-8 molal listed)

 Mineral saturation states
            log Q/K                    log Q/K
-----------------------------------------------------
 Quartz          0.0000 sat   Cristobalite(bet  -0.9940
 Ice            -0.1387       SiO2(am)          -1.2857
 Tridymite      -0.1715       Calcite           -1.3874
 Chalcedony     -0.2712       Aragonite         -1.5318
 Cristobalite(alp -0.5505     Monohydrocalcite  -2.2211
 Coesite        -0.8100
  (only minerals with log Q/K > -3 listed)

 Gases          fugacity     log fug.
-----------------------------------
 H2O(g)          0.02598     -1.585
 CO2(g)          0.004376    -2.359
 HCl(g)          8.820e-018  -17.055

                          In fluid              Sorbed
 Original basis  total moles    moles      mg/kg      moles
mg/kg
-----------------------------------------------------------
 Ca++          0.0002785   0.0002785     11.16
 Cl-           0.0002594   0.0002594     9.196
 H+            0.0001474   0.0001474     0.1485
 H2O             55.51       55.51  9.999e+005
 HCO3-          0.001131    0.001131     68.98
 K+            8.277e-005  8.277e-005     3.236
 Na+           0.0009184   0.0009184     21.11
 SO4--         0.0001577   0.0001577     15.14
 SiO2(aq)      0.0003330   0.0001003     6.027

 Elemental composition          In fluid              Sorbed
            total moles    moles      mg/kg      moles
mg/kg
-----------------------------------------------------------
 Calcium       0.0002785   0.0002785     11.16
 Carbon         0.001131    0.001131     13.58
 Chlorine      0.0002594   0.0002594     9.196
 Hydrogen        111.0       111.0  1.119e+005
 Oxygen          55.51       55.51  8.880e+005
 Potassium     8.277e-005  8.277e-005     3.236
 Silicon       0.0003330   0.0001003     2.817
 Sodium        0.0009184   0.0009184     21.11
 Sulfur        0.0001577   0.0001577     5.055
```

Note that charge balance was attained by adjusting HCO3- molality from .001 to .001131. Also, the calculated log fCO2(g) = -2.358.

(2) As is (1), but charge balance on H+ and set log fCO2(g) = -2.00.

React> show

```
Temperature is 25 C
```

```
Thermo dataset:  D:\Gwb\Gtdata\LLNL database\Pabalan.dat
Working directory:  d:\
Options: Debye-Huckel

Basis is:
H2O                    1 kg solvent
Na+                    total mg/kg = 21.113
SiO2(aq)               total mg/kg = 20.008
Ca++                   total mg/kg = 11.161
K+                     total mg/kg = 3.236
SO4--                  total mg/kg = 15.146
Cl-                    total mg/kg = 9.197
CO2(g)  (swapped for HCO3-)    fugacity = .01
H+                     charge balance

No reactants specified.

Other "show" options: type "show show"
React> go
```

```
Solving for initial system.

Loaded:  34 aqueous species,
         68 minerals,
          3 gases,
          0 surface species,
          9 elements,
          6 oxides.

N-R converged in  23 its., resmax =     7e-013, Xi =  0.0000
Charge balance: H+ molality adjusted from .001 to -.0009833

Removing CO2(g) from basis vector
Swapping HCO3- in for CO2(g)
3 supersaturated phases, most = Quartz
Swapping Quartz in for SiO2(aq)
Swapping CO2(aq) in for H+
N-R converged in  7 its., resmax =  2.18e-014, Xi =  0.0000

No reaction path specified.
```

The following is the React output file:

```
Step #    0          Xi = 0.0000
Temperature =  25.0 C    Pressure =  1.013 bars
pH =  6.783
Ionic strength     =   0.001972
Activity of water  =   0.999991
Solvent mass       =   1.000000 kg
Solution mass      =   1.000160 kg
Solution density   =   1.013   g/cm3
Chlorinity         =   0.000259 molal
Dissolved solids   =        160 mg/kg sol'n

No minerals in system.

Aqueous species      molality   mg/kg sol'n   act. coef.   log act.
-----------------------------------------------------------------------
HCO3-          0.0009787      59.71     0.9520     -3.0307
Na+            0.0009164      21.06     0.9520     -3.0593
CO2(aq)        0.0003397      14.95     1.0000     -3.4689
SiO2(aq)       0.0003328      19.99     1.0000     -3.4779
Ca++           0.0002722      10.91     0.8252     -3.6485
Cl-            0.0002594      9.194     0.9513     -3.6078
SO4--          0.0001532      14.71     0.8208     -3.9006
K+             8.269e-005     3.232     0.9513     -4.1042
CaSO4(aq)      3.648e-006     0.4966    1.0000     -5.4379
CaHCO3+        2.448e-006     0.2475    0.9520     -5.6325
NaHCO3(aq)     1.159e-006     0.09735   1.0000     -5.9359
NaSO4-         7.613e-007     0.09062   0.9520     -6.1398
CO3--          3.225e-007     0.01935   0.8219     -6.5766
HSiO3-         2.365e-007     0.01823   0.9520     -6.6475
H+             1.726e-007     0.0001740 0.9550     -6.7829
CaCO3(aq)      1.265e-007     0.01266   1.0000     -6.8980
KSO4-          7.874e-008     0.01064   0.9520     -7.1252
OH-            6.446e-008     0.001096  0.9516     -7.2122
NaCl(aq)       3.597e-008     0.002102  1.0000     -7.4441
CaCl+          1.174e-008     0.0008863 0.9520     -7.9518
 (only species > 1e-8 molal listed)

Mineral saturation states
                    log Q/K                    log Q/K
-----------------------------------------------------------------
Quartz          0.5214s/sat  Cristobalite(bet  -0.4726
Tridymite       0.3499s/sat  SiO2(am)          -0.7643
Chalcedony      0.2502s/sat  Calcite           -1.7450
Cristobalite(alp  -0.0291    Aragonite         -1.8894
Ice             -0.1387      Monohydrocalcite  -2.5787
Coesite         -0.2886
 (only minerals with log Q/K > -3 listed)

Gases            fugacity     log fug.
-----------------------------------------
H2O(g)           0.02598      -1.585
CO2(g)           0.01000      -2.000
HCl(g)           2.013e-017   -16.696

                          In fluid          Sorbed
Original basis  total moles    moles      mg/kg      moles      mg/kg
-----------------------------------------------------------------------
Ca++          0.0002785   0.0002785     11.16
Cl-           0.0002594   0.0002594     9.196
H+            0.0003391   0.0003391     0.3417
H2O           55.51       55.51         9.998e+005
HCO3-         0.001322    0.001322      80.68
K+            8.277e-005  8.277e-005    3.235
Na+           0.0009184   0.0009184     21.11
SO4--         0.0001577   0.0001577     15.14
SiO2(aq)      0.0003330   0.0003330     20.00
```

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001322	0.001322	15.88		
Chlorine	0.0002594	0.0002594	9.196		
Hydrogen	111.0	111.0	1.119e+005		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.235		
Silicon	0.0003330	0.0003330	9.351		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 6.783
Ionic strength = 0.001972
Activity of water = 0.999991
Solvent mass = 1.000000 kg
Solution mass = 1.000146 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 146 mg/kg sol'n

Minerals in system	moles	log moles	grams	volume (cm3)
Quartz	0.0002328	-3.633	0.01399	0.005281
(total)			0.01399	0.005281

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
HCO3-	0.0009789	59.72	0.9520	-3.0306
Na+	0.0009164	21.06	0.9520	-3.0593
CO2(aq)	0.0003395	14.94	1.0000	-3.4691
Ca++	0.0002722	10.91	0.8252	-3.6485
Cl-	0.0002594	9.194	0.9513	-3.6078
SO4--	0.0001532	14.71	0.8208	-3.9006
SiO2(aq)	0.0001002	6.017	1.0000	-3.9993
K+	8.269e-005	3.232	0.9513	-4.1042
CaSO4(aq)	3.648e-006	0.4966	1.0000	-5.4379
CaHCO3+	2.449e-006	0.2475	0.9520	-5.6324
NaHCO3(aq)	1.159e-006	0.09737	1.0000	-5.9358
NaSO4-	7.613e-007	0.09062	0.9520	-6.1398
CO3--	3.228e-007	0.01937	0.8219	-6.5763
H+	1.725e-007	0.0001738	0.9550	-6.7832
CaCO3(aq)	1.266e-007	0.01267	1.0000	-6.8977
KSO4-	7.874e-008	0.01064	0.9520	-7.1252
HSiO3-	7.124e-008	0.005491	0.9520	-7.1686
OH-	6.451e-008	0.001097	0.9516	-7.2119
NaCl(aq)	3.597e-008	0.002102	1.0000	-7.4440
CaCl+	1.174e-008	0.0008863	0.9520	-7.9518

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Quartz	0.0000 sat	Cristobalite(bet	-0.9940
Ice	-0.1387	SiO2(am)	-1.2857
Tridymite	-0.1715	Calcite	-1.7447
Chalcedony	-0.2712	Aragonite	-1.8891
Cristobalite(alp	-0.5505	Monohydrocalcite	-2.5784
Coesite	-0.8100		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
H2O(g)	0.02598	-1.585
CO2(g)	0.009995	-2.000
HCl(g)	2.012e-017	-16.696

Original basis	In fluid total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
H+	0.0003391	0.0003391	0.3417		
H2O	55.51	55.51	9.999e+005		
HCO3-	0.001322	0.001322	80.68		
K+	8.277e-005	8.277e-005	3.236		
Na+	0.0009184	0.0009184	21.11		
SO4--	0.0001577	0.0001577	15.14		
SiO2(aq)	0.0003330	0.0001002	6.022		

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001322	0.001322	15.88		
Chlorine	0.0002594	0.0002594	9.196		
Hydrogen	111.0	111.0	1.119e+005		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.236		
Silicon	0.0003330	0.0001002	2.815		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		

Note that with the resulting pH, based on a log fCO2(g) of –2.0, is 6.783, compared to the measured value of 7.14 ± 0.09.

(3) According to Lauren Browning, quartz is kinetically impaired from precipitating at 25 °C, thus one should suppress its precipitation during the GWB calculation. Cristobalite(alpha) was supposed to be the expected silica mineral to precipitate from YM waters. A GWB run was done with quartz suppressed, which resulted in tridymite being precipitated. Another run was done with tridymite suppressed, which resulted in chalcedony being precipitated. L. Browning suggested suppressing all minerals, and then unsuppressing cristobalite(alpha). Another GWB run was made doing that, and the results indicated that no mineral was saturated or supersaturated. The React output of the last run is:

```
Step #   0          Xi = 0.0000
Temperature =  25.0 C    Pressure =  1.013 bars
pH = 7.140
Ionic strength     =   0.001972
Activity of water  =   0.999991
Solvent mass       =   1.000000 kg
Solution mass      =   1.000152 kg
Solution density   =   1.013   g/cm3
Chlorinity         =   0.000259 molal
Dissolved solids   =        152 mg/kg sol'n

No minerals in system.

Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
-------------------------------------------------------------
HCO3-          0.0009771      59.61     0.9520     -
3.0314
Na+            0.0009164      21.06     0.9520     -
3.0593
SiO2(aq)       0.0003324      19.97     1.0000     -
3.4783
Ca++           0.0002721      10.90     0.8252     -
3.6487
Cl-            0.0002594      9.194     0.9513     -3.6078
SO4--          0.0001532      14.71     0.8208     -
3.9005
CO2(aq)        0.0001490      6.558     1.0000     -
3.8267
K+             8.269e-005     3.232     0.9513     -
4.1042
CaSO4(aq)      3.646e-006     0.4963    1.0000     -
5.4382
CaHCO3+        2.443e-006     0.2469    0.9520     -
5.6335
NaHCO3(aq)     1.157e-006     0.09719   1.0000
-5.9366
NaSO4-         7.613e-007     0.09062   0.9520     -
6.1398
CO3--          7.327e-007     0.04396   0.8220     -
6.2202
HSiO3-         5.378e-007     0.04145   0.9520     -
6.2908
CaCO3(aq)      2.872e-007     0.02874   1.0000     -
6.5419
OH-            1.467e-007     0.002495  0.9516     -
6.8551
KSO4-          7.874e-008     0.01064   0.9520     -
7.1252
H+             7.586e-008     7.644e-005  0.9550   -
7.1400
NaCl(aq)       3.597e-008     0.002102  1.0000     -
7.4441
NaHSiO3(aq)    1.988e-008     0.001989  1.0000
-7.7016
CaCl+          1.173e-008     0.0008858 0.9520     -
7.9521
 (only species > 1e-8 molal listed)

Mineral saturation states
                 log Q/K                    log Q/K
-------------------------------------------------------------
Quartz        0.5210s/sat  Cristobalite(bet  -0.4730
Tridymite     0.3495s/sat  SiO2(am)          -0.7647
Chalcedony    0.2498s/sat  Calcite           -1.3889
Cristobalite(alp  -0.0295  Aragonite         -1.5333
Ice           -0.1387      Monohydrocalcite  -2.2226
Coesite       -0.2890
 (only minerals with log Q/K > -3 listed)

Gases          fugacity     log fug.
-------------------------------------------
H2O(g)          0.02598     -1.585
CO2(g)          0.004387    -2.358
HCl(g)          8.846e-018  -17.053

                          In fluid              Sorbed
Original basis  total moles    moles      mg/kg      moles
mg/kg
-------------------------------------------------------------
Ca++          0.0002785   0.0002785     11.16
Cl-           0.0002594   0.0002594     9.196
H+            0.0001474   0.0001474     0.1485
H2O             55.51       55.51  9.999e+005
HCO3-         0.001131    0.001131      68.98
K+            8.277e-005  8.277e-005    3.236
Na+           0.0009184   0.0009184     21.11
SO4--         0.0001577   0.0001577     15.14
SiO2(aq)      0.0003330   0.0003330     20.00

Elemental composition          In fluid              Sorbed
          total moles    moles      mg/kg      moles
mg/kg
-------------------------------------------------------------
Calcium       0.0002785   0.0002785     11.16
Carbon        0.001131    0.001131      13.58
Chlorine      0.0002594   0.0002594     9.196
Hydrogen        111.0       111.0  1.119e+005
Oxygen          55.51       55.51  8.880e+005
Potassium     8.277e-005  8.277e-005    3.236
Silicon       0.0003330   0.0003330     9.351
Sodium        0.0009184   0.0009184     21.11
Sulfur        0.0001577   0.0001577     5.055
```

(4) Calculate speciation with GWB using all cations and anions (except phosphate) in the WT#17 datafile. Set fO2(g) = 0.2 atm.

```
Temperature is 25 C
    Thermo dataset:  D:\Gwb\Gtdata\LLNL
database\Pabalan.dat
    Working directory:  d:\
    Options: Debye-Huckel

    Basis is:
     H2O                    1 kg solvent
     Na+                    total mg/kg = 21.113
     SiO2(aq)               total mg/kg = 20.008
     Ca++                   total mg/kg = 11.161
     K+                     total mg/kg = 3.236
     SO4--                  total mg/kg = 15.146
     Cl-                    total mg/kg = 9.197
     HCO3-                  charge balance
     H+                     pH = 7.14
     F-                     total mg/kg = 3.338
     Mg++                   total mg/kg = 1.133
     Fe++                   total mg/kg = .78
     Mn++                   total mg/kg = .407
     Br-                    total mg/kg = 1.107
     NO3-  (swapped for NH3(aq))    total mg/kg = 3.798
     O2(g)  (swapped for O2(aq))    fugacity = .2

    No reactants specified.

    Other "show" options: type "show show"
React> go

Solving for initial system.

    -- Could not open React_output.txt file.
      (Do you have write permission in this directory?)
React> go

Solving for initial system.

 Loaded:  422 aqueous species,
        184 minerals,
        27 gases,
         0 surface species,
        15 elements,
        12 oxides.

N-R converged in  29 its., resmax =   4.66e-013, Xi =  0.0000
 Charge balance: HCO3- molality adjusted from .001 to
.0009682

 Removing O2(g) from basis vector
 Swapping O2(aq) in for O2(g)
 17 supersaturated phases, most = Birnessite
 Swapping Birnessite in for H+
N-R converged in  42 its., resmax =   1.01e-013, Xi =  0.0000
 12 supersaturated phases, most = Hematite
 Swapping Hematite in for Fe++
 Swapping CO2(aq) in for Mn++
N-R converged in  10 its., resmax =   1.15e-015, Xi =  0.0000
 5 supersaturated phases, most = Pyrolusite
 Swapping Pyrolusite in for O2(aq)
N-R converged in  18 its., resmax =   8.75e-012, Xi =  0.0000
 Birnessite is undersaturated
 Swapping N2(aq) in for Birnessite
N-R converged in 149 its., resmax =   2.82e-014, Xi =
0.0000
 3 supersaturated phases, most = Quartz
 Swapping Quartz in for SiO2(aq)
 Swapping O2(aq) in for N2(aq)
N-R converged in   9 its., resmax =   4.64e-015, Xi =  0.0000

No reaction path specified.
```

The following is the React output file for run (3):

```
    Step #    0          Xi = 0.0000
    Temperature =  25.0 C   Pressure =  1.013 bars
    pH =  7.140          log fO2 =   -0.699
    Eh =   0.7963 volts     pe =  13.4617
    Ionic strength     =   0.002130
    Activity of water  =   0.999991
    Solvent mass       =   1.000000 kg
    Solution mass      =   1.000162 kg
    Solution density   =   1.013   g/cm3
    Chlorinity         =   0.000259 molal
    Dissolved solids   =        162 mg/kg sol'n

 No minerals in system.

 Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
 ---------------------------------------------------------------------
  Na+            0.0009166      21.07     0.9502      -
3.0600
  HCO3-          0.0008364      51.03     0.9502      -
3.0997
  SiO2(aq)       0.0003324      19.97     1.0000      -
3.4783
  Ca++           0.0002723      10.91     0.8195      -
3.6514
  Cl-            0.0002594      9.193     0.9495     -3.6086
  O2(aq)         0.0002528      8.087     1.0000      -
3.5973
  F-             0.0001753      3.330     0.9499     -3.7785
  SO4--          0.0001519      14.59     0.8149      -
3.9073
  CO2(aq)        0.0001273      5.604     1.0000      -
3.8950
  K+             8.269e-005     3.232     0.9495      -
4.1050
  NO3-           6.118e-005     3.793     0.9495      -
4.2358
  Mg++           4.493e-005     1.092     0.8240      -
4.4316
  Br-            1.385e-005     1.107     0.9495      -
4.8809
  Fe(OH)3(aq)    1.197e-005     1.279     1.0000      -
4.9219
  Mn++           7.075e-006     0.3886    0.8195      -
5.2367
  CaSO4(aq)      3.568e-006     0.4857    1.0000      -
5.4475
  CaHCO3+        2.079e-006     0.2101    0.9502      -
5.7044
  Fe(OH)2+       1.951e-006     0.1753    0.9502      -
5.7319
  MgSO4(aq)      1.183e-006     0.1423    1.0000      -
5.9272
```

NaHCO3(aq)	9.871e-007	0.08291	1.0000	-6.0056
NaSO4-	7.497e-007	0.08924	0.9502	-6.1473
CO3--	6.306e-007	0.03784	0.8161	-6.2885
HSiO3-	5.387e-007	0.04152	0.9502	-6.2908
MgHCO3+	3.362e-007	0.02868	0.9502	-6.4956
CaCO3(aq)	2.439e-007	0.02441	1.0000	-6.6128
CaF+	1.879e-007	0.01110	0.9502	-6.7482
MnSO4(aq)	1.618e-007	0.02442	1.0000	-6.7911
OH-	1.470e-007	0.002499	0.9499	-6.8551
MgF+	1.460e-007	0.006323	0.9502	-6.8577
MnCO3(aq)	9.880e-008	0.01136	1.0000	-7.0052
KSO4-	7.752e-008	0.01048	0.9502	-7.1327
H+	7.598e-008	7.656e-005	0.9535	-7.1400
CaNO3+	6.839e-008	0.006980	0.9502	-7.1872
Fe(OH)4-	4.368e-008	0.005410	0.9502	-7.3819
MnHCO3+	3.693e-008	0.004281	0.9502	-7.4548
NaCl(aq)	3.584e-008	0.002094	1.0000	-7.4456
MnF+	2.735e-008	0.002022	0.9502	-7.5852
NaHSiO3(aq)	1.985e-008	0.001986	1.0000	-7.7023
MgCO3(aq)	1.815e-008	0.001530	1.0000	-7.7412
HF(aq)	1.777e-008	0.0003554	1.0000	-7.7504
NaF(aq)	1.458e-008	0.0006122	1.0000	-7.8361
CaCl+	1.166e-008	0.0008804	0.9502	-7.9555

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Birnessite	49.8570s/sat	Cristobalite(alp	-0.0295
Todorokite	43.2328s/sat	Ice	-0.1387
Hematite	14.0477s/sat	Coesite	-0.2890
Bixbyite	9.0913s/sat	Cristobalite(bet	-0.4730
Pyrolusite	8.6813s/sat	SiO2(am)	-0.7647
MnO2(gamma)	7.1635s/sat	Rhodochrosite	-1.0036
Hausmannite	7.0093s/sat	Fluorite	-1.1714
Goethite	6.5436s/sat	Calcite	-1.4598
Manganite	4.2275s/sat	Aragonite	-1.6042
Ferrite-Ca	3.2632s/sat	Mn(OH)3	-2.2783
Magnetite	3.1715s/sat	Monohydrocalcite	-2.2935
Ferrite-Mg	2.9496s/sat	Dolomite-ord	-2.5159
Andradite	2.2722s/sat	Dolomite	-2.5159
Fe(OH)3	1.4225s/sat	Sellaite	-2.6044
Quartz	0.5210s/sat	Magnesite	-2.6849
Tridymite	0.3495s/sat	Jarosite	-2.7346
Chalcedony	0.2498s/sat		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.003749	-2.426
HF(g)	9.308e-013	-12.031
HCl(g)	8.829e-018	-17.054
N2(g)	4.882e-019	-18.311
NO2(g)	1.433e-019	-18.844
Br2(g)	1.437e-020	-19.843
HBr(g)	1.252e-021	-20.902
NO(g)	2.147e-025	-24.668
Cl2(g)	5.056e-027	-26.296
SiF4(g)	1.098e-032	-31.959
H2(g)	6.267e-042	-41.203
CO(g)	7.337e-048	-47.134
SO2(g)	7.631e-055	-54.117
NH3(g)	8.356e-069	-68.078
Na(g)	1.304e-076	-75.885
K(g)	2.253e-078	-77.647
F2(g)	4.412e-080	-79.355
Mg(g)	2.498e-131	-130.602
H2S(g)	4.637e-142	-141.334
CH4(g)	4.611e-145	-144.336
Ca(g)	2.236e-153	-152.651
C(g)	3.848e-189	-188.415
Si(g)	1.473e-220	-219.832
S2(g)	1.221e-226	-225.913
C2H4(g)	1.068e-236	-235.971

Original basis	total moles	In fluid moles	Sorbed mg/kg	Sorbed moles	Sorbed mg/kg
Br-	1.385e-005	1.385e-005	1.107		
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
F-	0.0001757	0.0001757	3.337		
Fe++	1.397e-005	1.397e-005	0.7799		
H+	3.845e-005	3.845e-005	0.03875		
H2O	55.51	55.51	9.998e+005		
HCO3-	0.0009682	0.0009682	59.07		
K+	8.277e-005	8.277e-005	3.235		
Mg++	4.662e-005	4.662e-005	1.133		
Mn++	7.408e-006	7.408e-006	0.4069		
NH3(aq)	6.125e-005	6.125e-005	1.043		
Na+	0.0009184	0.0009184	21.11		
O2(aq)	0.0003788	0.0003788	12.12		
SO4--	0.0001577	0.0001577	15.14		
SiO2(aq)	0.0003330	0.0003330	20.00		

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.0009682	0.0009682	11.63		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.337		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.397e-005	0.7799		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.235		
Silicon	0.0003330	0.0003330	9.351		
Sodium	0.0009184	0.0009184	21.11		

Sulfur	0.0001577	0.0001577	5.055

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 7.080 log fO2 = -0.705
Eh = 0.7998 volts pe = 13.5197
Ionic strength = 0.002107
Activity of water = 0.999991
Solvent mass = 1.000001 kg
Solution mass = 1.000146 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 145 mg/kg sol'n

Minerals in system	moles	log moles	grams	volume (cm3)
Hematite	6.983e-006	-5.156	0.001115	0.0002114
Pyrolusite	7.408e-006	-5.130	0.0006441	
Quartz	0.0002327	-3.633	0.01398	0.005279
(total)			0.01574	0.005491*

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
Na+	0.0009166	21.07	0.9505	-3.0599
HCO3-	0.0008207	50.07	0.9505	-3.1078
Ca++	0.0002724	10.91	0.8203	-3.6508
Cl-	0.0002594	9.194	0.9498	-3.6085
O2(aq)	0.0002491	7.969	1.0000	-3.6037
F-	0.0001753	3.330	0.9501	-3.7784
SO4--	0.0001521	14.61	0.8157	-3.9064
CO2(aq)	0.0001434	6.310	1.0000	-3.8435
SiO2(aq)	0.0001002	6.017	1.0000	-3.9993
K+	8.269e-005	3.232	0.9498	-4.1049
NO3-	6.118e-005	3.793	0.9498	-4.2357
Mg++	4.493e-005	1.092	0.8247	-4.4311
Br-	1.385e-005	1.107	0.9498	-4.8808
CaSO4(aq)	3.580e-006	0.4873	1.0000	-5.4461
CaHCO3+	2.042e-006	0.2064	0.9505	-5.7120
MgSO4(aq)	1.186e-006	0.1428	1.0000	-5.9259
NaHCO3(aq)	9.691e-007	0.08140	1.0000	-6.0136
NaSO4-	7.512e-007	0.08942	0.9505	-6.1463
CO3--	5.389e-007	0.03234	0.8169	-6.3563
MgHCO3+	3.302e-007	0.02817	0.9505	-6.5033
CaCO3(aq)	2.089e-007	0.02091	1.0000	-6.6800
CaF+	1.882e-007	0.01112	0.9505	-6.7475
MgF+	1.462e-007	0.006330	0.9505	-6.8571
HSiO3-	1.414e-007	0.01090	0.9505	-6.8715
OH-	1.281e-007	0.002178	0.9501	-6.9148
H+	8.714e-008	8.782e-005	0.9537	-7.0803
KSO4-	7.768e-008	0.01050	0.9505	-7.1317
CaNO3+	6.847e-008	0.006989	0.9505	-7.1866
NaCl(aq)	3.586e-008	0.002096	1.0000	-7.4453
HF(aq)	2.039e-008	0.0004079	1.0000	-7.6906
MgCO3(aq)	1.554e-008	0.001310	1.0000	-7.8085
NaF(aq)	1.459e-008	0.0006127	1.0000	-7.8358
CaCl+	1.167e-008	0.0008815	0.9505	-7.9549

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Pyrolusite	0.0000 sat	Fluorite	-1.1705
Quartz	0.0000 sat	SiO2(am)	-1.2857
Hematite	0.0000 sat	MnO2(gamma)	-1.5178
Ice	-0.1387	Calcite	-1.5270
Tridymite	-0.1715	Aragonite	-1.6714
Chalcedony	-0.2712	Monohydrocalcite	-2.3607
Goethite	-0.4802	Sellaite	-2.6036
Cristobalite(alp	-0.5505	Dolomite-ord	-2.6505
Coesite	-0.8100	Dolomite	-2.6505
Cristobalite(bet	-0.9940	Magnesite	-2.7522

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.1971	-0.705
H2O(g)	0.02598	-1.585
CO2(g)	0.004221	-2.375
HF(g)	1.068e-012	-11.971
HCl(g)	1.013e-017	-16.994
N2(g)	6.670e-019	-18.176
NO2(g)	1.650e-019	-18.782
Br2(g)	1.878e-020	-19.726
HBr(g)	1.437e-021	-20.843
NO(g)	2.492e-025	-24.604
Cl2(g)	6.609e-027	-26.180
SiF4(g)	5.741e-033	-32.241
H2(g)	6.313e-042	-41.200
CO(g)	8.322e-048	-47.080
SO2(g)	1.014e-054	-53.994
NH3(g)	9.876e-069	-68.005
Na(g)	1.141e-076	-75.943
K(g)	1.972e-078	-77.705
F2(g)	5.769e-080	-79.239
Mg(g)	1.914e-131	-130.718
H2S(g)	6.299e-142	-141.201
CH4(g)	5.348e-145	-144.272
Ca(g)	1.714e-153	-152.766
C(g)	4.397e-189	-188.357
Si(g)	4.503e-221	-220.347

```
S2(g)          2.220e-226   -225.654
C2H4(g)          1.416e-236   -235.849

                          In fluid              Sorbed
 Original basis  total moles    moles      mg/kg      moles
mg/kg
-----------------------------------------------------------------
 Br-          1.385e-005  1.385e-005      1.107
 Ca++           0.0002785   0.0002785      11.16
 Cl-          0.0002594   0.0002594      9.196
 F-           0.0001757   0.0001757      3.338
 Fe++           1.397e-005  1.349e-012  7.531e-008
 H+           3.845e-005  8.120e-005    0.08183
 H2O              55.51       55.51  9.999e+005
 HCO3-          0.0009682   0.0009682      59.07
 K+           8.277e-005  8.277e-005      3.236
 Mg++           4.662e-005  4.662e-005      1.133
 Mn++           7.408e-006  2.040e-014  1.121e-009
 NH3(aq)        6.125e-005  6.125e-005      1.043
 Na+          0.0009184   0.0009184      21.11
 O2(aq)         0.0003788   0.0003716      11.89
 SO4--          0.0001577   0.0001577      15.14
```

```
 SiO2(aq)       0.0003330   0.0001003      6.026

 Elemental composition          In fluid              Sorbed
          total moles    moles      mg/kg      moles
mg/kg
-----------------------------------------------------------------
 Bromine        1.385e-005  1.385e-005      1.107
 Calcium        0.0002785   0.0002785      11.16
 Carbon         0.0009682   0.0009682      11.63
 Chlorine       0.0002594   0.0002594      9.196
 Fluorine       0.0001757   0.0001757      3.338
 Hydrogen           111.0       111.0  1.119e+005
 Iron           1.397e-005  1.349e-012  7.531e-008
 Magnesium      4.662e-005  4.662e-005      1.133
 Manganese      7.408e-006  2.040e-014  1.121e-009
 Nitrogen       6.125e-005  6.125e-005      0.8578
 Oxygen             55.51       55.51  8.880e+005
 Potassium      8.277e-005  8.277e-005      3.236
 Silicon        0.0003330   0.0001003      2.817
 Sodium         0.0009184   0.0009184      21.11
 Sulfur         0.0001577   0.0001577      5.055
```

(5) As in (3), but set Eh to 0 volt

```
React> show
    Temperature is 25 C
    Thermo dataset:  D:\Gwb\Gtdata\LLNL
database\Pabalan.dat
    Working directory:  d:\
    Options: Debye-Huckel

    Basis is:
    H2O                    1 kg solvent
    Na+                    total mg/kg = 21.113
    SiO2(aq)               total mg/kg = 20.008
    Ca++                   total mg/kg = 11.161
    K+                     total mg/kg = 3.236
    SO4--                  total mg/kg = 15.146
    Cl-                    total mg/kg = 9.197
    HCO3-                  charge balance
    H+                     pH = 7.14
    F-                     total mg/kg = 3.338
    Mg++                   total mg/kg = 1.133
    Fe++                   total mg/kg = .78
    Mn++                   total mg/kg = .407
    Br-                    total mg/kg = 1.107
    NO3-  (swapped for NH3(aq))    total mg/kg = 3.798
    e-  (swapped for O2(aq))       Eh = 0

    No reactants specified.
```

```
React> go

Solving for initial system.

 Loaded:  422 aqueous species,
        184 minerals,
         27 gases,
          0 surface species,
         15 elements,
         12 oxides.

N-R converged in  41 its., resmax =   3.7e-011, Xi =  0.0000
 Charge balance: HCO3- molality adjusted from .001 to
.001068

 Removing e- from basis vector
 Swapping N2(aq) in for e-
 7 supersaturated phases, most = Hematite
 Swapping Hematite in for NO3-
 Swapping CO2(aq) in for H+
N-R converged in  11 its., resmax =  1.47e-013, Xi =  0.0000
 3 supersaturated phases, most = Quartz
 Swapping Quartz in for SiO2(aq)
N-R converged in   7 its., resmax =  3.06e-013, Xi =  0.0000

No reaction path specified.
```

```
    Step #    0          Xi = 0.0000
    Temperature =  25.0 C    Pressure =  1.013 bars
    pH =  7.140            log fO2 =  -54.546
    Eh =  0.0000 volts     pe =  0.0000
    Ionic strength     =   0.002160
    Activity of water  =   0.999991
    Solvent mass       =   1.000000 kg
    Solution mass      =   1.000156 kg
    Solution density   =   1.013   g/cm3
    Chlorinity         =  0.000259 molal
    Dissolved solids   =        156 mg/kg sol'n

No minerals in system.
```

```
 Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
-----------------------------------------------------------------
  HCO3-          0.0009189      56.06     0.9499      -
3.0591
  Na+            0.0009165      21.07     0.9499      -
3.0602
  SiO2(aq)       0.0003324      19.97     1.0000      -
3.4783
  Ca++           0.0002722      10.91     0.8185      -
3.6521
  Cl-            0.0002594      9.193     0.9492     -3.6087
```

F-	0.0001753	3.329	0.9496	-3.7788
SO4--	0.0001518	14.58	0.8138	-3.9083
CO2(aq)	0.0001398	6.154	1.0000	-3.8544
K+	8.269e-005	3.232	0.9492	-4.1052
Mg++	4.490e-005	1.091	0.8229	-4.4324
N2(aq)	3.063e-005	0.8578	1.0000	-4.5139
Br-	1.385e-005	1.107	0.9492	-4.8811
Fe++	9.679e-006	0.5405	0.8185	-5.1012
Mn++	7.066e-006	0.3881	0.8185	-5.2378
FeHCO3+	3.820e-006	0.4464	0.9499	-5.4402
CaSO4(aq)	3.554e-006	0.4838	1.0000	-5.4493
CaHCO3+	2.279e-006	0.2304	0.9499	-5.6645
MgSO4(aq)	1.178e-006	0.1417	1.0000	-5.9290
NaHCO3(aq)	1.084e-006	0.09101	1.0000	-5.9652
NaSO4-	7.479e-007	0.08903	0.9499	-6.1485
CO3--	6.934e-007	0.04161	0.8150	-6.2479
HSiO3-	5.389e-007	0.04154	0.9499	-6.2908
MgHCO3+	3.686e-007	0.03144	0.9499	-6.4558
CaCO3(aq)	2.674e-007	0.02676	1.0000	-6.5729
FeCO3(aq)	2.404e-007	0.02785	1.0000	-6.6190
CaF+	1.875e-007	0.01108	0.9499	-6.7492
MnSO4(aq)	1.610e-007	0.02431	1.0000	-6.7932
FeSO4(aq)	1.551e-007	0.02356	1.0000	-6.8094
OH-	1.470e-007	0.002500	0.9496	-6.8551
MgF+	1.457e-007	0.006310	0.9499	-6.8588
MnCO3(aq)	1.082e-007	0.01244	1.0000	-6.9657
KSO4-	7.735e-008	0.01045	0.9499	-7.1339
H+	7.600e-008	7.659e-005	0.9532	-7.1400
MnHCO3+	4.046e-008	0.004691	0.9499	-7.4153
FeOH+	3.640e-008	0.002652	0.9499	-7.4612
NaCl(aq)	3.581e-008	0.002093	1.0000	-7.4459
FeF+	3.180e-008	0.002379	0.9499	-7.5199
MnF+	2.727e-008	0.002016	0.9499	-7.5866
MgCO3(aq)	1.989e-008	0.001677	1.0000	-7.7014
NaHSiO3(aq)	1.984e-008	0.001985	1.0000	-7.7025
HF(aq)	1.776e-008	0.0003552	1.0000	-7.7507
NaF(aq)	1.457e-008	0.0006117	1.0000	-7.8366
CaCl+	1.164e-008	0.0008788	0.9499	-7.9565

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Hematite	6.5069s/sat	Siderite	-0.8282
Magnetite	5.3219s/sat	Rhodochrosite	-0.9641
Cronstedtite-7A	5.2346s/sat	Fluorite	-1.1727
Goethite	2.7732s/sat	Calcite	-1.4199
Quartz	0.5210s/sat	Aragonite	-1.5643
Tridymite	0.3495s/sat	Ferrosilite	-1.7466
Chalcedony	0.2498s/sat	Greenalite	-2.0902
Cristobalite(alp	-0.0295	Monohydrocalcite	-2.2536
Ice	-0.1387	Fe(OH)3	-2.3479
Coesite	-0.2890	Dolomite	-2.4362
Minnesotaite	-0.3572	Dolomite-ord	-2.4362
Cristobalite(bet	-0.4730	Sellaite	-2.6056
SiO2(am)	-0.7647	Magnesite	-2.6451

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
N2(g)	0.04704	-1.327
H2O(g)	0.02598	-1.585
CO2(g)	0.004117	-2.385
HF(g)	9.303e-013	-12.031
H2(g)	5.252e-015	-14.280
HCl(g)	8.826e-018	-17.054
NH3(g)	6.294e-020	-19.201
CO(g)	6.753e-021	-20.171
HBr(g)	1.251e-021	-20.903
SO2(g)	6.381e-028	-27.195
SiF4(g)	1.096e-032	-31.960
H2S(g)	2.283e-034	-33.641
CH4(g)	2.499e-037	-36.602
NO(g)	7.953e-044	-43.099
Br2(g)	1.713e-047	-46.766
Cl2(g)	6.028e-054	-53.220
O2(g)	2.847e-055	-54.546
Na(g)	3.774e-063	-62.423
K(g)	6.521e-065	-64.186
NO2(g)	6.331e-065	-64.199
S2(g)	4.215e-065	-64.375
C2H4(g)	4.467e-075	-74.350
Mg(g)	2.090e-104	-103.680
F2(g)	5.258e-107	-106.279
Ca(g)	1.871e-126	-125.728
C(g)	2.969e-135	-134.527
Si(g)	1.034e-166	-165.985

		In fluid	Sorbed	
Original basis	total moles	moles	mg/kg	moles mg/kg
Br-	1.385e-005	1.385e-005	1.107	
Ca++	0.0002785	0.0002785	11.16	
Cl-	0.0002594	0.0002594	9.196	
F-	0.0001757	0.0001757	3.337	
Fe++	1.397e-005	1.397e-005	0.7799	
H+	-0.9999	-0.9999	-1008.	
H2O	56.01	56.01	1.009e+006	
HCO3-	0.001068	0.001068	65.13	
K+	8.277e-005	8.277e-005	3.235	
Mg++	4.662e-005	4.662e-005	1.133	

Mn++	7.408e-006	7.408e-006	0.4069
NH3(aq)	6.125e-005	6.125e-005	1.043
Na+	0.0009184	0.0009184	21.11
O2(aq)	-0.2500	-0.2500	-7997.
SO4--	0.0001577	0.0001577	15.14
SiO2(aq)	0.0003330	0.0003330	20.00

Elemental composition	In fluid total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001068	0.001068	12.82		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.337		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.397e-005	0.7799		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.235		
Silicon	0.0003330	0.0003330	9.351		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 7.123 log fO2 = -66.955
Eh = -0.1825 volts pe = -3.0853
Ionic strength = 0.002154
Activity of water = 0.999991
Solvent mass = 1.000000 kg
Solution mass = 1.000142 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 142 mg/kg sol'n

Minerals in system	moles	log moles	grams	volume (cm3)
Hematite	1.659e-006	-5.780	0.0002649	5.022e-005
Quartz	0.0002327	-3.633	0.01398	0.005279
(total)			0.01425	0.005329

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
Na+	0.0009165	21.07	0.9500	-3.0602
HCO3-	0.0009149	55.82	0.9500	-3.0609
Ca++	0.0002722	10.91	0.8187	-3.6520
Cl-	0.0002594	9.194	0.9493	-3.6087
F-	0.0001753	3.330	0.9496	-3.7787
SO4--	0.0001518	14.58	0.8140	-3.9080
CO2(aq)	0.0001449	6.374	1.0000	-3.8391
SiO2(aq)	0.0001002	6.017	1.0000	-3.9993
K+	8.269e-005	3.232	0.9493	-4.1052
Mg++	4.490e-005	1.091	0.8232	-4.4323
N2(aq)	3.007e-005	0.8424	1.0000	-4.5218
Br-	1.385e-005	1.107	0.9493	-4.8810
Fe++	7.395e-006	0.4130	0.8187	-5.2179
Mn++	7.071e-006	0.3884	0.8187	-5.2374
CaSO4(aq)	3.557e-006	0.4842	1.0000	-5.4489
FeHCO3+	2.907e-006	0.3397	0.9500	-5.5588
CaHCO3+	2.270e-006	0.2295	0.9500	-5.6662
MgSO4(aq)	1.179e-006	0.1419	1.0000	-5.9286
NH4+	1.096e-006	0.01977	0.9489	-5.9829
NaHCO3(aq)	1.079e-006	0.09063	1.0000	-5.9670
NaSO4-	7.483e-007	0.08908	0.9500	-6.1482
CO3--	6.636e-007	0.03982	0.8152	-6.2668
MgHCO3+	3.671e-007	0.03132	0.9500	-6.4575
CaCO3(aq)	2.560e-007	0.02562	1.0000	-6.5917
CaF+	1.876e-007	0.01108	0.9500	-6.7490
FeCO3(aq)	1.759e-007	0.02038	1.0000	-6.7547
MnSO4(aq)	1.612e-007	0.02434	1.0000	-6.7926
HSiO3-	1.561e-007	0.01203	0.9500	-6.8289
MgF+	1.458e-007	0.006312	0.9500	-6.8586
OH-	1.413e-007	0.002403	0.9496	-6.8722
FeSO4(aq)	1.186e-007	0.01801	1.0000	-6.9260
MnCO3(aq)	1.037e-007	0.01192	1.0000	-6.9842
H+	7.905e-008	7.966e-005	0.9533	-7.1229
KSO4-	7.739e-008	0.01046	0.9500	-7.1336
MnHCO3+	4.032e-008	0.004675	0.9500	-7.4167
NaCl(aq)	3.582e-008	0.002093	1.0000	-7.4459
MnF+	2.730e-008	0.002018	0.9500	-7.5861
FeOH+	2.674e-008	0.001948	0.9500	-7.5950
FeF+	2.430e-008	0.001819	0.9500	-7.6366
MgCO3(aq)	1.905e-008	0.001606	1.0000	-7.7202
HF(aq)	1.847e-008	0.0003695	1.0000	-7.7335
NaF(aq)	1.457e-008	0.0006118	1.0000	-7.8365
CaCl+	1.164e-008	0.0008791	0.9500	-7.9563

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Hematite	0.0000 sat	SiO2(am)	-1.2857
Quartz	0.0000 sat	Magnetite	-1.3360
Ice	-0.1387	Calcite	-1.4387
Tridymite	-0.1715	Aragonite	-1.5831
Pyrite	-0.1929	Cronstedtite-7A	-2.0953
Chalcedony	-0.2712	Monohydrocalcite	-2.2724
Goethite	-0.4802	Ferrosilite	-2.4186
Cristobalite(alp	-0.5505	Dolomite	-2.4738
Coesite	-0.8100	Dolomite-ord	-2.4738
Siderite	-0.9639	Sellaite	-2.6054
Rhodochrosite	-0.9826	Magnesite	-2.6639
Cristobalite(bet	-0.9940	Minnesotaite	-2.8942
Fluorite	-1.1724		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
N2(g)	0.04620	-1.335
H2O(g)	0.02598	-1.585
CO2(g)	0.004264	-2.370
H2(g)	8.419e-009	-8.075
H2S(g)	1.632e-009	-8.787
NH3(g)	1.266e-010	-9.898
CH4(g)	1.709e-012	-11.767
HF(g)	9.679e-013	-12.014
CO(g)	1.121e-014	-13.950
HCl(g)	9.182e-018	-17.037
HBr(g)	1.302e-021	-20.885
SO2(g)	1.107e-021	-20.956
S2(g)	8.378e-028	-27.077
SiF4(g)	3.868e-033	-32.413
C2H4(g)	8.128e-038	-37.090
NO(g)	4.917e-050	-49.308
Br2(g)	1.157e-053	-52.937
Na(g)	4.594e-060	-59.338
Cl2(g)	4.070e-060	-59.390
K(g)	7.937e-062	-61.100
O2(g)	1.108e-067	-66.955
NO2(g)	2.442e-077	-76.612
Mg(g)	3.097e-098	-97.509
F2(g)	3.550e-113	-112.450
Ca(g)	2.772e-120	-119.557
C(g)	7.900e-123	-122.102
Si(g)	8.008e-155	-154.096

Original basis	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Br-	1.385e-005	1.385e-005	1.107		
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
F-	0.0001757	0.0001757	3.338		
Fe++	1.397e-005	1.065e-005	0.5946		
H+	0.0001379	0.0001445	0.1456		
H2O	55.51	55.51	9.999e+005		
HCO3-	0.001068	0.001068	65.13		
K+	8.277e-005	8.277e-005	3.236		
Mg++	4.662e-005	4.662e-005	1.133		
Mn++	7.408e-006	7.408e-006	0.4069		
NH3(aq)	6.125e-005	6.125e-005	1.043		
Na+	0.0009184	0.0009184	21.11		
O2(aq)	4.594e-005	4.511e-005	1.443		
SO4--	0.0001577	0.0001577	15.14		
SiO2(aq)	0.0003330	0.0001003	6.027		

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001068	0.001068	12.82		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.338		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.065e-005	0.5946		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.236		
Silicon	0.0003330	0.0001003	2.817		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		

(6) As in (4), but calculate solubility of technetium by specifying TcO4- concentration to be 1.e-12 molal. (Also, set print options for species to be long, so values less than 10e-8 print out also).

```
Temperature is 25 C
   Thermo dataset: D:\Gwb\Gtdata\LLNL
database\Pabalan.dat
   Working directory: d:\
   Options: Debye-Huckel

   Basis is:
    H2O                    1 kg solvent
    NO3- (swapped for NH3(aq))    total mg/kg = 3.798
    e- (swapped for O2(aq))       Eh = 0
    Na+                    total mg/kg = 21.113
    SiO2(aq)               total mg/kg = 20.008
    Ca++                   total mg/kg = 11.161
    K+                     total mg/kg = 3.236
    SO4--                  total mg/kg = 15.146
    Cl-                    total mg/kg = 9.197
    HCO3-                  charge balance
    H+                     pH = 7.14
    F-                     total mg/kg = 3.338
    Mg++                   total mg/kg = 1.133
    Fe++                   total mg/kg = .78
    Mn++                   total mg/kg = .407
    Br-                    total mg/kg = 1.107
    TcO4-                  total molality = 1e-12

   No reactants specified.

React> go

Solving for initial system.

Loaded:  430 aqueous species,
         191 minerals,
          30 gases,
```

0 surface species,
16 elements,
13 oxides.

N-R converged in 42 its., resmax = 3.7e-011, Xi = 0.0000
Charge balance: HCO3- molality adjusted from .001 to .001068

Removing e- from basis vector
Swapping N2(aq) in for e-
8 supersaturated phases, most = Hematite
Swapping Hematite in for NO3-
N-R converged in 33 its., resmax = 2.34e-014, Xi = 0.0000
4 supersaturated phases, most = Quartz
Swapping Quartz in for SiO2(aq)
Swapping CO2(aq) in for H+
Swapping TcO(OH)2(aq) in for TcO4-
N-R converged in 9 its., resmax = 7.47e-014, Xi = 0.0000
1 supersaturated phase, TcO2
Swapping TcO2 in for TcO(OH)2(aq)
N-R converged in 3 its., resmax = 2.67e-015, Xi = 0.0000

No reaction path specified.

The following is the React output file.

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 7.140 log fO2 = -54.546
Eh = 0.0000 volts pe = 0.0000
Ionic strength = 0.002160
Activity of water = 0.999991
Solvent mass = 1.000000 kg
Solution mass = 1.000156 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 156 mg/kg sol'n

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
HCO3-	0.0009189	56.06	0.9499	-3.0591
Na+	0.0009165	21.07	0.9499	-3.0602
SiO2(aq)	0.0003324	19.97	1.0000	-3.4783
Ca++	0.0002722	10.91	0.8185	-3.6521
Cl-	0.0002594	9.193	0.9492	-3.6087
F-	0.0001753	3.329	0.9496	-3.7788
SO4--	0.0001518	14.58	0.8138	-3.9083
CO2(aq)	0.0001398	6.154	1.0000	-3.8544
K+	8.269e-005	3.232	0.9492	-4.1052
Mg++	4.490e-005	1.091	0.8229	-4.4324
N2(aq)	3.063e-005	0.8578	1.0000	-4.5139
Br-	1.385e-005	1.107	0.9492	-4.8811
Fe++	9.679e-006	0.5405	0.8185	-5.1012
Mn++	7.066e-006	0.3881	0.8185	-5.2378
FeHCO3+	3.820e-006	0.4464	0.9499	-5.4402
CaSO4(aq)	3.554e-006	0.4838	1.0000	-5.4493
CaHCO3+	2.279e-006	0.2304	0.9499	-5.6645
MgSO4(aq)	1.178e-006	0.1417	1.0000	-5.9290
NaHCO3(aq)	1.084e-006	0.09101	1.0000	-5.9652
NaSO4-	7.479e-007	0.08903	0.9499	-6.1485
CO3--	6.934e-007	0.04161	0.8150	-6.2479
HSiO3-	5.389e-007	0.04154	0.9499	-6.2908
MgHCO3+	3.686e-007	0.03144	0.9499	-6.4558
CaCO3(aq)	2.674e-007	0.02676	1.0000	-6.5729
FeCO3(aq)	2.404e-007	0.02785	1.0000	-6.6190
CaF+	1.875e-007	0.01108	0.9499	-6.7492
MnSO4(aq)	1.610e-007	0.02431	1.0000	-6.7932
FeSO4(aq)	1.551e-007	0.02356	1.0000	-6.8094
OH-	1.470e-007	0.002500	0.9496	-6.8551
MgF+	1.457e-007	0.006310	0.9499	-6.8588
MnCO3(aq)	1.082e-007	0.01244	1.0000	-6.9657
KSO4-	7.735e-008	0.01045	0.9499	-7.1339
H+	7.600e-008	7.659e-005	0.9532	-7.1400
MnHCO3+	4.046e-008	0.004691	0.9499	-7.4153
FeOH+	3.640e-008	0.002652	0.9499	-7.4612
NaCl(aq)	3.581e-008	0.002093	1.0000	-7.4459
FeF+	3.180e-008	0.002379	0.9499	-7.5199
MnF+	2.727e-008	0.002016	0.9499	-7.5866
MgCO3(aq)	1.989e-008	0.001677	1.0000	-7.7014
NaHSiO3(aq)	1.984e-008	0.001985	1.0000	-7.7025
HF(aq)	1.776e-008	0.0003552	1.0000	-7.7507
NaF(aq)	1.457e-008	0.0006117	1.0000	-7.8366

CaCl+	1.164e-008	0.0008788	0.9499	-7.9565
MgCl+	7.019e-009	0.0004194	0.9499	-8.1761
MnCl+	3.000e-009	0.0002711	0.9499	-8.5453
MnOH+	2.160e-009	0.0001554	0.9499	-8.6878
Fe(OH)3(aq)	2.031e-009	0.0002170	1.0000	-8.6923
NaCO3-	1.693e-009	0.0001405	0.9499	-8.7937
FeCl+	1.419e-009	0.0001295	0.9499	-8.8704
HSO4-	8.977e-010	8.713e-005	0.9499	-9.0692
KCl(aq)	6.187e-010	4.612e-005	1.0000	-9.2085
NaBr(aq)	5.034e-010	5.179e-005	1.0000	-9.2981
CaOH+	4.573e-010	2.610e-005	0.9499	-9.3621
Fe(OH)2+	3.312e-010	2.976e-005	0.9499	-9.5023
NaOH(aq)	1.928e-011	7.708e-007	1.0000	-10.7150
KBr(aq)	1.890e-011	2.249e-006	1.0000	-10.7235
Fe(OH)4-	7.414e-012	9.183e-007	0.9499	-11.1523
HCl(aq)	3.813e-012	1.390e-007	1.0000	-11.4187
Fe(OH)2(aq)	3.792e-012	3.407e-007	1.0000	-11.4212
KOH(aq)	3.757e-012	2.107e-007	1.0000	-11.4252
CaCl2(aq)	3.067e-012	3.404e-007	1.0000	-11.5132
TcO(OH)2(aq)	9.992e-013	1.479e-007	1.0000	-12.0004
H2SiO4--	8.535e-013	8.030e-008	0.8138	-12.1583
HF2-	7.511e-013	2.929e-008	0.9499	-12.1466
Mn2(OH)3+	1.166e-013	1.876e-008	0.9499	-12.9556
FeOH++	8.445e-014	6.152e-009	0.8150	-13.1622
Mn(OH)2(aq)	6.953e-014	6.184e-009	1.0000	-13.1578
H6(H2SiO4)4--	6.551e-014	2.505e-008	0.8138	-13.2732
Mn2OH+++	2.009e-014	2.549e-009	0.6329	-13.8956
KHSO4(aq)	4.572e-015	6.225e-010	1.0000	-14.3399
FeCO3+	2.411e-015	2.793e-010	0.9499	-14.6401
Fe(OH)3-	2.193e-015	2.344e-010	0.9499	-14.6812
FeCl2(aq)	1.688e-015	2.139e-010	1.0000	-14.7727
H2F2(aq)	8.521e-016	3.409e-011	1.0000	-15.0695
NH4+	5.240e-016	9.451e-012	0.9488	-15.3035
TcO4-	4.468e-016	7.237e-011	0.9499	-15.3722
TcO(OH)3-	1.796e-016	2.963e-011	0.9499	-15.7680
TcCO3(OH)2(aq)	1.667e-016	3.200e-011	1.0000	-15.7781
MnCl3-	4.225e-017	6.814e-012	0.9499	-16.3964
TcOOH+	2.451e-017	3.211e-012	0.9499	-16.6330
TcCO3(OH)3-	1.191e-017	2.489e-012	0.9499	-16.9464
Formate	7.585e-018	3.414e-013	0.9496	-17.1425
FeF2+	5.039e-018	4.728e-013	0.9499	-17.3200
H2(aq)	4.124e-018	8.313e-015	1.0000	-17.3847
NH3(aq)	3.940e-018	6.709e-014	1.0000	-17.4045
FeF++	2.160e-018	1.616e-013	0.8150	-17.7545
Fe+++	1.190e-018	6.647e-014	0.6487	-18.1122
Mn(OH)3-	9.477e-019	1.004e-013	0.9499	-18.0456
H2SO4(aq)	6.178e-020	6.058e-015	1.0000	-19.2092
Ca(For)+	4.547e-020	3.869e-015	0.9499	-19.3645
FeSO4+	8.500e-021	1.291e-015	0.9499	-20.0929
Mg(For)+	7.542e-021	5.228e-016	0.9499	-20.1448
Na(For)(aq)	7.034e-021	4.783e-016	1.0000	-20.1528
Fe(For)+	4.037e-021	4.071e-016	0.9499	-20.4163
Formic_acid(aq)	2.955e-021	1.360e-016	1.0000	-20.5295
Mn(For)+	2.197e-021	2.196e-016	0.9499	-20.6805
H4(H2SiO4)4----	1.162e-021	4.419e-016	0.4382	-21.2932
K(For)(aq)	6.059e-022	5.096e-017	1.0000	-21.2176
FeCl4--	4.502e-022	8.897e-017	0.8138	-21.4361
SO3--	1.897e-022	1.519e-017	0.8150	-21.8107
HSO3-	1.892e-022	1.534e-017	0.9499	-21.7453
FeCl++	3.606e-023	3.292e-018	0.8150	-22.5318
Fe(OH)4--	3.534e-023	4.378e-018	0.8138	-22.5412
Fe(SO4)2-	2.029e-023	5.030e-018	0.9499	-22.7151
CO(aq)	6.648e-024	1.862e-019	1.0000	-23.1773
FeCl2+	6.646e-024	8.423e-019	0.9499	-23.1997
Fe2(OH)2++++	2.859e-025	4.165e-020	0.4459	-24.8945
TcO++	2.130e-025	2.428e-020	0.8150	-24.7605
Oxalate	1.693e-025	1.490e-020	0.8138	-24.8608
Mn(OH)4--	1.293e-025	1.590e-020	0.8138	-24.9778
TcO4--	8.266e-027	1.339e-021	0.8138	-26.1722
H2SO3(aq)	1.326e-027	1.088e-022	1.0000	-26.8775

SO2(aq) 9.438e-028 6.046e-023 1.0000 -27.0251
NH4SO4- 3.233e-028 3.688e-023 0.9499 -27.5127
H-Oxalate 1.897e-028 1.689e-023 0.9499 -27.7442
SiF6-- 4.504e-029 6.398e-024 0.8138 -28.4359
Mg4(OH)4++++ 2.698e-029 4.458e-024 0.4459 -28.9197
Mn+++ 2.392e-031 1.314e-026 0.6329 -30.8199
Fe3(OH)4(5+) 2.934e-032 6.911e-027 0.2856 -32.0767
FeCl4- 4.843e-034 9.571e-029 0.9499 -33.3372
Oxalic_acid(aq) 2.433e-034 2.190e-029 1.0000 -33.6139
HS- 3.509e-035 1.160e-030 0.9496 -34.4773
H2S(aq) 2.346e-035 7.995e-031 1.0000 -34.6296
Ca(For)2(aq) 2.307e-036 3.001e-031 1.0000 -35.6370
Fe(For)2(aq) 4.275e-037 6.235e-032 1.0000 -36.3691
Mg(For)2(aq) 3.826e-037 4.374e-032 1.0000 -36.4172
Mn(For)2(aq) 1.849e-037 2.680e-032 1.0000 -36.7330
Na(For)2- 2.798e-038 3.162e-033 0.9499 -37.5754
Urea(aq) 4.205e-039 2.525e-034 1.0000 -38.3762
K(For)2- 2.304e-039 2.974e-034 0.9499 -38.6599
Formaldehyde(aq) 4.703e-040 1.412e-035 1.0000 -39.3277
Methane(aq) 3.528e-040 5.659e-036 1.0000 -39.4525
S-- 6.543e-041 2.098e-036 0.8161 -40.2724
S2O3-- 2.195e-041 2.461e-036 0.8138 -40.7481
Methanol(aq) 1.154e-042 3.697e-038 1.0000 -41.9379
HBrO(aq) 5.360e-044 5.194e-039 1.0000 -43.2708
BrO- 3.181e-045 3.050e-040 0.9499 -44.5197
S2O6-- 1.973e-045 3.158e-040 0.8138 -44.7945
Acetate 7.988e-046 4.716e-041 0.9503 -45.1197
HCN(aq) 2.013e-046 5.439e-042 1.0000 -45.6962
HS2O3- 1.406e-047 1.591e-042 0.9499 -46.8742
HClO(aq) 9.934e-048 5.210e-043 1.0000 -47.0029
Br2(aq) 8.304e-048 1.327e-042 1.0000 -47.0807
ClO- 3.892e-048 2.002e-043 0.9499 -47.4321
Acetic_acid(aq) 3.144e-048 1.888e-043 1.0000 -47.5025
CN- 1.700e-048 4.423e-044 0.9492 -47.7921
CaCH3COO+ 1.518e-048 1.505e-043 0.9499 -47.8409
S2O5-- 6.047e-049 8.713e-044 0.8138 -48.3080
MgCH3COO+ 5.614e-049 4.679e-044 0.9499 -48.2730
Glycolate 5.569e-049 4.178e-044 0.9499 -48.2766
NaCH3COO(aq) 5.208e-049 4.272e-044 1.0000 -48.2833
FeCH3COO+ 1.235e-049 1.418e-044 0.9499 -48.9308
MnCH3COO+ 7.614e-050 8.677e-045 0.9499 -49.1407
KCH3COO(aq) 3.245e-050 3.184e-045 1.0000 -49.4888
NO2- 2.868e-050 1.319e-045 0.9492 -49.5650
HO2- 1.375e-050 4.539e-046 0.9499 -49.8839
Ca(Glyc)+ 5.541e-051 6.378e-046 0.9499 -50.2787
Br3- 2.797e-051 6.704e-046 0.9499 -50.5756
Fe(Glyc)+ 6.631e-052 8.678e-047 0.9499 -51.2007
Na(Glyc)(aq) 5.165e-052 5.062e-047 1.0000 -51.2870
Mg(Glyc)+ 4.396e-052 4.367e-047 0.9499 -51.3793
Glycolic_acid(aq 2.612e-052 1.986e-047 1.0000 -51.5830
Mn(Glyc)+ 1.230e-052 1.598e-047 0.9499 -51.9326
K(Glyc)(aq) 4.449e-053 5.077e-048 1.0000 -52.3518
HNO2(aq) 3.278e-054 1.541e-049 1.0000 -53.4844
Malonate 9.338e-055 9.527e-050 0.8138 -54.1193
S2O4-- 5.027e-055 6.440e-050 0.8161 -54.3870
H-Malonate 2.874e-056 2.962e-051 0.9499 -55.5638
Methanamine(aq) 1.033e-057 3.207e-053 1.0000 -56.9860
HSO5- 9.238e-058 1.044e-052 0.9499 -57.0567
O2(aq) 3.598e-058 1.151e-053 1.0000 -57.4439
SCN- 8.884e-060 5.159e-055 0.9496 -59.0739
Glycine(aq) 1.555e-060 1.167e-055 1.0000 -59.8082
Malonic_acid(aq) 1.405e-060 1.461e-055 1.0000 -59.8525
NH4CH3COO(aq) 6.158e-061 4.746e-056 1.0000 -60.2106
Acetamide(aq) 6.134e-061 3.622e-056 1.0000 -60.2123
N3- 1.069e-063 4.492e-059 0.9499 -62.9932
NO3- 9.342e-064 5.792e-059 0.9492 -63.0522
Fe(Gly)+ 6.195e-064 8.046e-059 0.9499 -63.2303
Mg(Gly)+ 3.923e-064 3.859e-059 0.9499 -63.4287
Mn(Gly)+ 9.951e-065 1.283e-059 0.9499 -64.0244
S2-- 4.788e-065 3.070e-060 0.8138 -64.4093

FeNO2++ 3.644e-065 3.711e-060 0.8150 -64.5273
Ca(Gly)+ 1.879e-065 2.144e-060 0.9499 -64.7485
Acetaldehyde(aq) 6.988e-066 3.078e-061 1.0000 -65.1557
HN3(aq) 3.689e-066 1.587e-061 1.0000 -65.4331
CaNO3+ 1.042e-066 1.064e-061 0.9499 -66.0043
MnO4-- 3.574e-067 4.250e-062 0.8138 -66.5363
MnNO3+ 8.557e-069 1.001e-063 0.9499 -68.0900
S3O6-- 3.870e-072 7.438e-067 0.8138 -71.5018
HNO3(aq) 3.201e-072 2.017e-067 1.0000 -71.4947
Ethanol(aq) 6.196e-073 2.854e-068 1.0000 -72.2079
S2O8-- 5.904e-074 1.134e-068 0.8138 -73.3184
Ethane(aq) 1.420e-074 4.268e-070 1.0000 -73.8479
MnO4- 1.269e-076 1.509e-071 0.9496 -75.9190
Ethylene(aq) 2.120e-077 5.947e-073 1.0000 -76.6736
Propanoate 4.000e-078 2.922e-073 0.9499 -77.4203
Lactate 3.423e-079 3.048e-074 0.9499 -78.4880
Propanoic_acid(a 2.133e-080 1.580e-075 1.0000 -79.6711
FeNO3++ 8.403e-081 9.902e-076 0.8150 -80.1644
Ca(Prop)+ 4.195e-081 4.746e-076 0.9499 -80.3995
Na(Prop)(aq) 3.648e-081 3.504e-076 1.0000 -80.4379
Ca(Lac)+ 2.005e-081 2.588e-076 0.9499 -80.7203
Fe(Prop)+ 1.262e-081 1.627e-076 0.9499 -80.9213
Mg(Prop)+ 7.623e-082 7.421e-077 0.9499 -81.1402
Mn(Prop)+ 4.332e-082 5.545e-077 0.9499 -81.3856
Fe(Lac)+ 3.555e-082 5.150e-077 0.9499 -81.4715
Na(Lac)(aq) 3.174e-082 3.557e-077 1.0000 -81.4984
K(Prop)(aq) 3.143e-082 3.525e-077 1.0000 -81.5027
Mg(Lac)+ 2.964e-082 3.360e-077 0.9499 -81.5505
Lactic_acid(aq) 1.718e-082 1.547e-077 1.0000 -81.7651
S4O6-- 6.119e-083 1.372e-077 0.8138 -82.3029
Mn(Lac)+ 5.329e-083 7.673e-078 0.9499 -82.2957
K(Lac)(aq) 2.734e-083 3.504e-078 1.0000 -82.5632
ClO2- 7.272e-085 4.904e-080 0.9499 -84.1606
Succinate 1.646e-085 1.910e-080 0.8138 -84.8732
Ethyne(aq) 6.720e-087 1.749e-082 1.0000 -86.1726
H-Succinate 4.352e-087 5.094e-082 0.9499 -86.3837
HClO2(aq) 7.399e-089 5.064e-084 1.0000 -88.1308
S3-- 2.918e-089 2.807e-084 0.8138 -88.6244
Ethanamine(aq) 7.623e-090 3.436e-085 1.0000 -89.1179
Succinic_acid(aq 4.829e-090 5.702e-085 1.0000 -89.3162
Alanine(aq) 1.309e-091 1.166e-086 1.0000 -90.8831
Ca(CH3COO)2(aq) 1.744e-092 2.757e-087 1.0000 -91.7586
Mg(CH3COO)2(aq) 2.342e-093 3.334e-088 1.0000 -92.6304
Acetone(aq) 2.104e-093 1.222e-088 1.0000 -92.6770
Fe(CH3COO)2(aq) 1.394e-093 2.425e-088 1.0000 -92.8557
Mn(CH3COO)2(aq) 3.823e-094 6.615e-089 1.0000 -93.4175
Serine(aq) 1.791e-094 1.882e-089 1.0000 -93.7468
Na(CH3COO)2- 1.731e-094 2.441e-089 0.9499 -93.7841
Fe(Ala)+ 5.503e-095 7.920e-090 0.9499 -94.2817
K(CH3COO)2- 7.956e-096 1.250e-090 0.9499 -95.1216
Mn(Ala)+ 8.252e-097 1.180e-091 0.9499 -96.1057
Ca(Ala)+ 3.181e-097 4.076e-092 0.9499 -96.5198
Mg(Ala)+ 2.768e-097 3.110e-092 0.9499 -96.5802
Propanal(aq) 1.240e-097 7.203e-093 1.0000 -96.9065
Ca(Glyc)2(aq) 4.841e-098 9.205e-093 1.0000 -97.3151
Fe(Glyc)2(aq) 1.560e-098 3.213e-093 1.0000 -97.8068
Mg(Glyc)2(aq) 2.063e-099 3.598e-094 1.0000 -98.6854
Mn(Glyc)2(aq) 4.355e-100 8.927e-095 1.0000 -99.3610
Na(Glyc)2- 2.337e-100 4.044e-095 0.9499 -99.6537
K(Glyc)2- 1.924e-101 3.638e-096 0.9499 -100.7382
Aspartic_acid(aq 8.356e-102 1.112e-096 1.0000 -101.0780
1-Propanol(aq) 3.491e-105 2.098e-100 1.0000 -104.4571
NH4(CH3COO)2- 3.631e-106 4.942e-101 0.9499 -105.4623
Propane(aq) 3.422e-107 1.509e-102 1.0000 -106.4658
1-Propene(aq) 1.766e-107 7.429e-103 1.0000 -106.7531
ClO3- 9.707e-108 8.099e-103 0.9496 -107.0354
BrO3- 6.780e-109 8.671e-104 0.9496 -108.1912
Butanoate 6.800e-111 5.922e-106 0.9499 -110.1898
2-Hydroxybutanoa 7.493e-112 7.724e-107 0.9499 -111.1477
Butanoic_acid(aq 3.011e-113 2.652e-108 1.0000 -112.5213

S4-- 1.072e-113 1.374e-108 0.8138 -113.0594
Na(But)(aq) 5.895e-114 6.489e-109 1.0000 -113.2295
Ca(But)+ 4.912e-114 6.246e-109 0.9499 -113.3311
Fe(But)+ 2.185e-114 3.124e-109 0.9499 -113.6828
Asparagine(aq) 2.100e-114 2.773e-109 1.0000 -113.6779
Mg(But)+ 8.527e-115 9.498e-110 0.9499 -114.0915
Mn(But)+ 6.241e-115 8.864e-110 0.9499 -114.2270
K(But)(aq) 5.078e-115 6.407e-110 1.0000 -114.2943
1-Propyne(aq) 3.618e-115 1.449e-110 1.0000 -114.4416
2-Hydroxybutanoi 3.342e-115 3.478e-110 1.0000 -114.4761
Glutarate 2.193e-117 2.853e-112 0.8138 -116.7485
H-Glutarate 3.554e-119 4.659e-114 0.9499 -118.4716
1-Propanamine(aq 1.423e-121 8.413e-117 1.0000 -120.8467
Ethylacetate(aq) 5.605e-122 4.938e-117 1.0000 -121.2514
Glutaric_acid(aq 5.344e-122 7.059e-117 1.0000 -121.2722
S5O6-- 1.274e-122 3.265e-117 0.8138 -121.9843
Diglycine(aq) 6.269e-123 8.282e-118 1.0000 -122.2028
Fe(Gly)2(aq) 3.493e-123 7.124e-118 1.0000 -122.4568
Mg(Gly)2(aq) 1.363e-123 2.350e-118 1.0000 -122.8655
a-Aminobutyric_a 4.754e-124 4.902e-119 1.0000 -123.3229
Mn(Gly)2(aq) 3.156e-124 6.407e-119 1.0000 -123.5009
Diketopiperazine 4.756e-125 5.426e-120 1.0000 -124.3228
Ca(Gly)2(aq) 3.674e-127 6.912e-122 1.0000 -126.4349
Threonine(aq) 3.203e-127 3.815e-122 1.0000 -126.4945
Mn(NO3)2(aq) 1.811e-131 3.239e-126 1.0000 -130.7422
Butanal(aq) 8.830e-132 6.366e-127 1.0000 -131.0540
Glutamic_acid(aq 1.424e-132 2.095e-127 1.0000 -131.8465
ClO4- 6.559e-135 6.522e-130 0.9496 -134.2057
S5-- 2.372e-138 3.803e-133 0.8138 -137.7143
1-Butanol(aq) 1.214e-138 8.999e-134 1.0000 -137.9157
Mn(CH3COO)3- 6.641e-139 1.541e-133 0.9499 -138.2000
n-Butane(aq) 7.255e-140 4.216e-135 1.0000 -139.1394
1-Butene(aq) 1.903e-140 1.067e-135 1.0000 -139.7206
Pentanoate 1.391e-143 1.407e-138 0.9499 -142.8789
2-Hydroxypentano 2.418e-144 2.831e-139 0.9499 -143.6389
Pentanoic_acid(a 6.703e-146 6.845e-141 1.0000 -145.1737
Na(Pent)(aq) 1.227e-146 1.523e-141 1.0000 -145.9112
Ca(Pent)+ 5.876e-147 8.295e-142 0.9499 -146.2533
Glutamine(aq) 4.085e-147 5.969e-142 1.0000 -146.3888
Fe(Pent)+ 4.048e-147 6.353e-142 0.9499 -146.4151
K(Pent)(aq) 1.057e-147 1.482e-142 1.0000 -146.9760
Mg(Pent)+ 9.979e-148 1.251e-142 0.9499 -147.0232
Mn(Pent)+ 9.636e-148 1.504e-142 0.9499 -147.0384
2-Hydroxypentano 6.500e-148 7.678e-143 1.0000 -147.1871
1-Butyne(aq) 5.945e-148 3.215e-143 1.0000 -147.2258
Adipate 1.387e-151 1.999e-146 0.8138 -150.9475
H-Adipate 2.210e-153 3.207e-148 0.9499 -152.6779
BrO4- 1.411e-153 2.030e-148 0.9499 -152.8729
Alanylglycine(aq 2.379e-154 3.476e-149 1.0000 -153.6236
1-Butanamine(aq) 1.534e-154 1.122e-149 1.0000 -153.8142
Adipic_acid(aq) 3.934e-156 5.749e-151 1.0000 -155.4051
Methionine(aq) 2.988e-156 4.457e-151 1.0000 -155.5247
Valine(aq) 1.364e-156 1.597e-151 1.0000 -155.8653
Fe(Prop)2(aq) 4.798e-158 9.689e-153 1.0000 -157.3190
Ca(Prop)2(aq) 2.706e-158 5.039e-153 1.0000 -157.5676
Mn(Prop)2(aq) 8.851e-159 1.780e-153 1.0000 -158.0530
Na(Prop)2- 7.894e-159 1.335e-153 0.9499 -158.1250
Ca(Lac)2(aq) 7.106e-159 1.550e-153 1.0000 -158.1484
Mg(Prop)2(aq) 5.405e-159 9.211e-154 1.0000 -158.2672
Fe(Lac)2(aq) 4.676e-159 1.094e-153 1.0000 -158.3301
Mg(Lac)2(aq) 9.591e-160 1.941e-154 1.0000 -159.0181
K(Prop)2- 6.486e-160 1.201e-154 0.9499 -159.2103
Mn(Lac)2(aq) 1.934e-160 4.507e-155 1.0000 -159.7135
Na(Lac)2- 9.039e-161 1.818e-155 0.9499 -160.0662
K(Lac)2- 7.439e-162 1.616e-156 0.9499 -161.1508
Phenol(aq) 3.477e-163 3.272e-158 1.0000 -162.4588
Pentanal(aq) 3.101e-164 2.670e-159 1.0000 -163.5086
Benzene(aq) 1.358e-168 1.061e-163 1.0000 -167.8671
1-Pentanol(aq) 4.025e-170 3.547e-165 1.0000 -169.3953
Benzoate 1.660e-172 2.010e-167 0.9513 -171.8015

n-Pentane(aq) 1.311e-172 9.455e-168 1.0000 -171.8825
1-Pentene(aq) 3.075e-173 2.156e-168 1.0000 -172.5122
Benzoic_acid(aq) 1.814e-175 2.215e-170 1.0000 -174.7414
Hexanoate 2.325e-176 2.677e-171 0.9499 -175.6558
2-Hydroxyhexanoa 3.357e-177 4.402e-172 0.9499 -176.4964
Hexanoic_acid(aq 1.159e-178 1.346e-173 1.0000 -177.9359
o-Phthalate 5.926e-180 9.723e-175 0.8138 -179.3168
2-Hydroxyhexanoi 1.265e-180 1.671e-175 1.0000 -179.8980
1-Pentyne(aq) 1.010e-180 6.882e-176 1.0000 -179.9955
Ca(o-Phthalate)(2.825e-181 5.768e-176 1.0000 -180.5489
H(o-Phthalate)- 9.409e-182 1.553e-176 0.9499 -181.0488
Na(o-Phthalate)- 2.215e-182 4.144e-177 0.9499 -181.6770
Pimelate 3.509e-183 5.549e-178 0.8138 -182.5443
H-Pimelate 5.786e-185 9.207e-180 0.9499 -184.2600
Fe(Ala)2(aq) 2.140e-185 4.964e-180 1.0000 -184.6696
o-Phthalic_acid(5.770e-186 9.585e-181 1.0000 -185.2388
Pimelic_acid(aq) 1.219e-187 1.952e-182 1.0000 -186.9139
Mn(Ala)2(aq) 3.951e-188 9.131e-183 1.0000 -187.4032
1-Pentanamine(aq 3.108e-188 2.709e-183 1.0000 -187.5075
Isoleucine(aq) 4.509e-190 5.913e-185 1.0000 -189.3459
Mg(Ala)2(aq) 3.815e-190 7.648e-185 1.0000 -189.4185
Leucine(aq) 3.217e-190 4.219e-185 1.0000 -189.4926
Ca(Ala)2(aq) 1.103e-190 2.384e-185 1.0000 -189.9576
Hexanal(aq) 4.927e-197 4.934e-192 1.0000 -196.3074
Toluene(aq) 1.585e-198 1.460e-193 1.0000 -197.7999
p-Toluate 3.445e-202 4.654e-197 0.9499 -201.4852
m-Toluate 1.973e-202 2.666e-197 0.9499 -201.7271
1-Hexanol(aq) 1.630e-203 1.665e-198 1.0000 -202.7879
o-Toluate 2.036e-204 2.751e-199 0.9499 -203.7135
p-Toluic_acid(aq 5.540e-205 7.542e-200 1.0000 -204.2565
m-Toluic_acid(aq 2.464e-205 3.355e-200 1.0000 -204.6083
n-Hexane(aq) 1.700e-205 1.465e-200 1.0000 -204.7695
1-Hexene(aq) 7.706e-206 6.484e-201 1.0000 -205.1132
o-Toluic_acid(aq 1.131e-207 1.539e-202 1.0000 -206.9466
Heptanoate 5.223e-209 6.745e-204 0.9499 -208.3044
2-Hydroxyheptano 6.532e-210 9.481e-205 0.9499 -209.2073
Heptanoic_acid(a 2.808e-211 3.655e-206 1.0000 -210.5516
2-Hydroxyheptano 2.461e-213 3.597e-208 1.0000 -212.6089
1-Hexyne(aq) 1.379e-213 1.133e-208 1.0000 -212.8603
Suberate 3.059e-217 5.266e-212 0.8138 -216.6040
H-Suberate 4.793e-219 8.300e-214 0.9499 -218.3417
1-Hexanamine(aq) 5.946e-221 6.015e-216 1.0000 -220.2258
Suberic_acid(aq) 1.062e-221 1.850e-216 1.0000 -220.9737
Fe(But)2(aq) 1.375e-223 3.162e-218 1.0000 -222.8618
Ca(But)2(aq) 3.888e-224 8.330e-219 1.0000 -223.4103
Na(But)2- 2.013e-224 3.969e-219 0.9499 -223.7185
Mn(But)2(aq) 1.837e-224 4.209e-219 1.0000 -223.7359
Mg(But)2(aq) 6.911e-225 1.372e-219 1.0000 -224.1604
K(But)2- 1.654e-225 3.528e-220 0.9499 -224.8037
Heptanal(aq) 1.050e-230 1.199e-225 1.0000 -229.9786
1-Heptanol(aq) 2.790e-237 3.242e-232 1.0000 -236.5544
n-Heptane(aq) 3.308e-238 3.314e-233 1.0000 -237.4805
1-Heptene(aq) 1.425e-238 1.399e-233 1.0000 -237.8462
Octanoate 1.671e-241 2.393e-236 0.9499 -240.7992
2-Hydroxyoctanoa 1.271e-242 2.023e-237 0.9499 -241.9183
Octanoic_acid(aq 9.064e-244 1.307e-238 1.0000 -243.0427
2-Hydroxyoctanoi 4.787e-246 7.668e-241 1.0000 -245.3199
1-Heptyne(aq) 1.730e-246 1.664e-241 1.0000 -245.7618
Tyrosine(aq) 4.105e-250 7.436e-245 1.0000 -249.3867
Phenylalanine(aq 1.508e-250 2.491e-245 1.0000 -249.8216
Azelate 5.363e-252 9.986e-247 0.8138 -251.3601
Leucylglycine(aq 1.874e-252 3.527e-247 1.0000 -251.7272
1-Heptanamine(aq 1.157e-253 1.332e-248 1.0000 -252.9368
H-Azelate 8.265e-254 1.547e-248 0.9499 -253.1050
Azelaic_acid(aq) 1.895e-256 3.566e-251 1.0000 -255.7225
Octanal(aq) 8.873e-263 1.137e-257 1.0000 -262.0519
n-Propylbenzene(8.266e-264 9.934e-259 1.0000 -263.0827
1-Octanol(aq) 9.008e-270 1.173e-264 1.0000 -269.0454
n-Octane(aq) 5.815e-271 6.641e-266 1.0000 -270.2355
1-Octene(aq) 1.913e-271 2.146e-266 1.0000 -270.7184

Nonanoate	3.538e-274	5.562e-269	0.9499	-273.4735
2-Hydroxynonanoa	2.557e-275	4.429e-270	0.9499	-274.6146
Nonanoic_acid(aq	1.301e-276	2.059e-271	1.0000	-275.8856
2-Hydroxynonanoi	9.632e-279	1.678e-273	1.0000	-278.0163
1-Octyne(aq)	2.941e-279	3.241e-274	1.0000	-278.5315
Sebacate	4.965e-285	9.941e-280	0.8138	-284.3936
1-Octanamine(aq)	2.250e-286	2.908e-281	1.0000	-285.6478
H-Sebacate	8.186e-287	1.647e-281	0.9499	-286.1092
Fe(Pent)2(aq)	4.724e-289	1.219e-283	1.0000	-288.3257
Sebacic_acid(aq)	1.974e-289	3.991e-284	1.0000	-288.7047
Na(Pent)2-	8.516e-290	1.918e-284	0.9499	-289.0921
Ca(Pent)2(aq)	5.973e-290	1.447e-284	1.0000	-289.2238
Mn(Pent)2(aq)	4.475e-290	1.151e-284	1.0000	-289.3492
Mg(Pent)2(aq)	1.037e-290	2.349e-285	1.0000	-289.9842
K(Pent)2-	7.008e-291	1.691e-285	0.9499	-290.1767
Nonanal(aq)	1.318e-295	1.874e-290	1.0000	-294.8801
Decanoate	4.439e-307	7.601e-302	0.9499	-300.0000
2-Hydroxydecanoa	5.059e-308	9.472e-303	0.9499	-300.0000
Tryptophan(aq)	8.945e-309	1.827e-303	1.0000	-300.0000
Decanoic_acid(aq	2.531e-309	4.360e-304	1.0000	-300.0000
2-Hydroxydecanoi	1.906e-311	3.587e-306	1.0000	-300.0000
Ethylbenzene(aq)	0.0000	0.0000	1.0000	-300.0000
Dodecanoic_acid(	0.0000	0.0000	1.0000	-300.0000
Dodecanoate	0.0000	0.0000	0.9499	-300.0000
n-Pentylbenzene(	0.0000	0.0000	1.0000	-300.0000
Decanal(aq)	0.0000	0.0000	1.0000	-300.0000
n-Octylbenzene(a	0.0000	0.0000	1.0000	-300.0000
n-Hexylbenzene(a	0.0000	0.0000	1.0000	-300.0000
2-Hexanone(aq)	0.0000	0.0000	1.0000	-300.0000
n-Heptylbenzene(	0.0000	0.0000	1.0000	-300.0000
n-Butylbenzene(a	0.0000	0.0000	1.0000	-300.0000
Undecanoic_acid(	0.0000	0.0000	1.0000	-300.0000
Undecanoate	0.0000	0.0000	0.9499	-300.0000
2-Heptanone(aq)	0.0000	0.0000	1.0000	-300.0000
2-Butanone(aq)	0.0000	0.0000	1.0000	-300.0000
2-Pentanone(aq)	0.0000	0.0000	1.0000	-300.0000
2-Octanone(aq)	0.0000	0.0000	1.0000	-300.0000

Mineral saturation states

	log Q/K		log Q/K
Hematite	6.5069s/sat	Siderite	-0.8282
Magnetite	5.3219s/sat	Rhodochrosite	-0.9641
Cronstedtite-7A	5.2346s/sat	Fluorite	-1.1727
Goethite	2.7732s/sat	Calcite	-1.4199
Quartz	0.5210s/sat	Aragonite	-1.5643
TcO2	0.4006s/sat	Ferrosilite	-1.7466
Tridymite	0.3495s/sat	Greenalite	-2.0902
Chalcedony	0.2498s/sat	Monohydrocalcite	-2.2536
Cristobalite(alp	-0.0295	Fe(OH)3	-2.3479
Ice	-0.1387	Dolomite	-2.4362
Coesite	-0.2890	Dolomite-ord	-2.4362
Minnesotaite	-0.3572	Sellaite	-2.6056
Cristobalite(bet	-0.4730	Magnesite	-2.6451
SiO2(am)	-0.7647		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
N2(g)	0.04704	-1.327
H2O(g)	0.02598	-1.585
CO2(g)	0.004117	-2.385
HF(g)	9.303e-013	-12.031
H2(g)	5.252e-015	-14.280
HCl(g)	8.826e-018	-17.054
NH3(g)	6.294e-020	-19.201
CO(g)	6.753e-021	-20.171
HBr(g)	1.251e-021	-20.903
SO2(g)	6.381e-028	-27.195
SiF4(g)	1.096e-032	-31.960
H2S(g)	2.283e-034	-33.641
CH4(g)	2.499e-037	-36.602
NO(g)	7.953e-044	-43.099
Br2(g)	1.713e-047	-46.766
Cl2(g)	6.028e-054	-53.220
O2(g)	2.847e-055	-54.546
Na(g)	3.774e-063	-62.423
K(g)	6.521e-065	-64.186
NO2(g)	6.331e-065	-64.199
S2(g)	4.215e-065	-64.375
Tc2O7(g)	5.109e-069	-68.292
C2H4(g)	4.467e-075	-74.350
Mg(g)	2.090e-104	-103.680
F2(g)	5.258e-107	-106.279
TcO(g)	1.443e-108	-107.841
Ca(g)	1.871e-126	-125.728
Tc(g)	3.744e-129	-128.427
C(g)	2.969e-135	-134.527
Si(g)	1.034e-166	-165.985

Original basis	total moles	In fluid moles	mg/kg	Sorbed moles	mg/kg
Br-	1.385e-005	1.385e-005	1.107		
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
F-	0.0001757	0.0001757	3.337		
Fe++	1.397e-005	1.397e-005	0.7799		
H+	-0.9999	-0.9999	-1008.		
H2O	56.01	56.01	1.009e+006		
HCO3-	0.001068	0.001068	65.13		
K+	8.277e-005	8.277e-005	3.235		

Mg++	4.662e-005	4.662e-005	1.133
Mn++	7.408e-006	7.408e-006	0.4069
NH3(aq)	6.125e-005	6.125e-005	1.043
Na+	0.0009184	0.0009184	21.11
O2(aq)	-0.2500	-0.2500	-7997.
SO4--	0.0001577	0.0001577	15.14
SiO2(aq)	0.0003330	0.0003330	20.00
TcO4-	1.000e-012	1.000e-012	1.620e-007

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001068	0.001068	12.82		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.337		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.397e-005	0.7799		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.235		
Silicon	0.0003330	0.0003330	9.351		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		
Technetium	1.000e-012	1.000e-012	9.798e-008		

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 7.123 log fO2 = -66.955
Eh = -0.1825 volts pe = -3.0853
Ionic strength = 0.002154
Activity of water = 0.999991
Solvent mass = 1.000000 kg
Solution mass = 1.000142 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 142 mg/kg sol'n

Minerals in system	moles	log moles	grams	volume (cm3)
Hematite	1.659e-006	-5.780	0.0002649	5.022e-005
Quartz	0.0002327	-3.633	0.01398	0.005279
TcO2	6.027e-013	-12.220	7.835e-011	
(total)			0.01425	0.005329*

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
Na+	0.0009165	21.07	0.9500	-3.0602
HCO3-	0.0009149	55.82	0.9500	-3.0609
Ca++	0.0002722	10.91	0.8187	-3.6520
Cl-	0.0002594	9.194	0.9493	-3.6087
F-	0.0001753	3.330	0.9496	-3.7787
SO4--	0.0001518	14.58	0.8140	-3.9080
CO2(aq)	0.0001449	6.374	1.0000	-3.8391
SiO2(aq)	0.0001002	6.017	1.0000	-3.9993
K+	8.269e-005	3.232	0.9493	-4.1052
Mg++	4.490e-005	1.091	0.8232	-4.4323
N2(aq)	3.007e-005	0.8424	1.0000	-4.5218
Br-	1.385e-005	1.107	0.9493	-4.8810
Fe++	7.395e-006	0.4130	0.8187	-5.2179
Mn++	7.071e-006	0.3884	0.8187	-5.2374
CaSO4(aq)	3.557e-006	0.4842	1.0000	-5.4489
FeHCO3+	2.907e-006	0.3397	0.9500	-5.5588
CaHCO3+	2.270e-006	0.2295	0.9500	-5.6662
MgSO4(aq)	1.179e-006	0.1419	1.0000	-5.9286
NH4+	1.096e-006	0.01977	0.9489	-5.9829
NaHCO3(aq)	1.079e-006	0.09063	1.0000	-5.9670
NaSO4-	7.483e-007	0.08908	0.9500	-6.1482
CO3--	6.636e-007	0.03982	0.8152	-6.2668
MgHCO3+	3.671e-007	0.03132	0.9500	-6.4575
CaCO3(aq)	2.560e-007	0.02562	1.0000	-6.5917
CaF+	1.876e-007	0.01108	0.9500	-6.7490
FeCO3(aq)	1.759e-007	0.02038	1.0000	-6.7547
MnSO4(aq)	1.612e-007	0.02434	1.0000	-6.7926
HSiO3-	1.561e-007	0.01203	0.9500	-6.8289
MgF+	1.458e-007	0.006312	0.9500	-6.8586
OH-	1.413e-007	0.002403	0.9496	-6.8722
FeSO4(aq)	1.186e-007	0.01801	1.0000	-6.9260
MnCO3(aq)	1.037e-007	0.01192	1.0000	-6.9842
H+	7.905e-008	7.966e-005	0.9533	-7.1229
KSO4-	7.739e-008	0.01046	0.9500	-7.1336
MnHCO3+	4.032e-008	0.004675	0.9500	-7.4167
NaCl(aq)	3.582e-008	0.002093	1.0000	-7.4459
MnF+	2.730e-008	0.002018	0.9500	-7.5861
FeOH+	2.674e-008	0.001948	0.9500	-7.5950
FeF+	2.430e-008	0.001819	0.9500	-7.6366
MgCO3(aq)	1.905e-008	0.001606	1.0000	-7.7202

HF(aq)	1.847e-008	0.0003695	1.0000	-7.7335
NaF(aq)	1.457e-008	0.0006118	1.0000	-7.8365
CaCl+	1.164e-008	0.0008791	0.9500	-7.9563
NH3(aq)	7.924e-009	0.0001349	1.0000	-8.1011
MgCl+	7.021e-009	0.0004195	0.9500	-8.1759
NaHSiO3(aq)	5.747e-009	0.0005751	1.0000	-8.2406
MnCl+	3.002e-009	0.0002713	0.9500	-8.5448
MnOH+	2.078e-009	0.0001495	0.9500	-8.7045
NaCO3-	1.621e-009	0.0001345	0.9500	-8.8126
FeCl+	1.084e-009	9.898e-005	0.9500	-8.9871
HSO4-	9.343e-010	9.068e-005	0.9500	-9.0518
KCl(aq)	6.188e-010	4.612e-005	1.0000	-9.2085
NaBr(aq)	5.035e-010	5.180e-005	1.0000	-9.2980
CaOH+	4.397e-010	2.510e-005	0.9500	-9.3791
HS-	2.410e-010	7.971e-006	0.9496	-9.6404
H2S(aq)	1.677e-010	5.714e-006	1.0000	-9.7755
KBr(aq)	1.891e-011	2.250e-006	1.0000	-10.7234
NaOH(aq)	1.853e-011	7.411e-007	1.0000	-10.7321
Formate	1.211e-011	5.449e-007	0.9496	-10.9394
H2(aq)	6.611e-012	1.332e-008	1.0000	-11.1797
HCl(aq)	3.967e-012	1.446e-007	1.0000	-11.4016
KOH(aq)	3.612e-012	2.026e-007	1.0000	-11.4423
CaCl2(aq)	3.069e-012	3.406e-007	1.0000	-11.5130
Fe(OH)2(aq)	2.678e-012	2.406e-007	1.0000	-11.5722
Fe(OH)3(aq)	1.133e-012	1.211e-007	1.0000	-11.9457
HF2-	7.814e-013	3.048e-008	0.9500	-12.1294
TcO(OH)2(aq)	3.972e-013	5.877e-008	1.0000	-12.4010
H2SiO4--	2.376e-013	2.235e-008	0.8140	-12.7136
Fe(OH)2+	1.922e-013	1.727e-008	0.9500	-12.7386
Mn2(OH)3+	1.038e-013	1.669e-008	0.9500	-13.0062
Ca(For)+	7.260e-014	6.177e-009	0.9500	-13.1613
Mn(OH)2(aq)	6.432e-014	5.720e-009	1.0000	-13.1917
Mn2OH+++	1.934e-014	2.453e-009	0.6333	-13.9120
Mg(For)+	1.204e-014	8.346e-010	0.9500	-13.9416
Na(For)(aq)	1.123e-014	7.635e-010	1.0000	-13.9497
Fe(For)+	4.924e-015	4.966e-010	0.9500	-14.3300
Formic_acid(aq)	4.905e-015	2.257e-010	1.0000	-14.3093
KHSO4(aq)	4.759e-015	6.479e-010	1.0000	-14.3225
Fe(OH)4-	3.976e-015	4.925e-010	0.9500	-14.4228
Mn(For)+	3.509e-015	3.507e-010	0.9500	-14.4771
Methane(aq)	2.412e-015	3.870e-011	1.0000	-14.6175
Fe(OH)3-	1.489e-015	1.591e-010	0.9500	-14.8493
FeCl2(aq)	1.290e-015	1.635e-010	1.0000	-14.8894
K(For)(aq)	9.671e-016	8.134e-011	1.0000	-15.0145
H2F2(aq)	9.222e-016	3.690e-011	1.0000	-15.0352
H6(H2SiO4)4--	4.988e-016	1.907e-010	0.8140	-15.3915
S--	4.320e-016	1.385e-011	0.8164	-15.4526
HSO3-	3.157e-016	2.559e-011	0.9500	-15.5231
SO3--	3.042e-016	2.435e-011	0.8152	-15.6056
S2O3--	1.569e-016	1.759e-011	0.8140	-15.8938
TcO(OH)3-	6.864e-017	1.132e-011	0.9500	-16.1857
TcCO3(OH)2(aq)	6.863e-017	1.318e-011	1.0000	-16.1635
FeOH++	5.097e-017	3.713e-012	0.8152	-16.3814
MnCl3-	4.230e-017	6.822e-012	0.9500	-16.3959
CO(aq)	1.104e-017	3.091e-013	1.0000	-16.9571
TcOOH+	1.013e-017	1.328e-012	0.9500	-17.0165
TcCO3(OH)3-	4.714e-018	9.852e-013	0.9500	-17.3489
FeCO3+	1.449e-018	1.679e-013	0.9500	-17.8612
Mn(OH)3-	8.427e-019	8.928e-014	0.9500	-18.0966
NH4SO4-	6.765e-019	7.718e-014	0.9500	-18.1920
Oxalate	2.691e-019	2.368e-014	0.8140	-18.6595
H2SO4(aq)	6.688e-020	6.559e-015	1.0000	-19.1747
Urea(aq)	1.762e-020	1.058e-015	1.0000	-19.7541
Acetate	5.438e-021	3.211e-016	0.9503	-20.2866
FeF2+	3.165e-021	2.970e-016	0.9500	-20.5219
H2SO3(aq)	2.301e-021	1.888e-016	1.0000	-20.6381
SO2(aq)	1.638e-021	1.049e-016	1.0000	-20.7857
S2--	1.410e-021	9.039e-017	0.8140	-20.9403
FeF++	1.356e-021	1.015e-016	0.8152	-20.9565
Fe+++	7.471e-022	4.172e-017	0.6491	-21.3143

FeCl4-- 3.441e-022 6.800e-017 0.8140 -21.5527
H-Oxalate 3.137e-022 2.792e-017 0.9500 -21.5258
HS2O3- 1.046e-022 1.183e-017 0.9500 -22.0028
Fe(OH)4-- 2.306e-023 2.857e-018 0.8140 -22.7264
Acetic_acid(aq) 2.227e-023 1.337e-018 1.0000 -22.6523
CaCH3COO+ 1.034e-023 1.025e-018 0.9500 -23.0077
H4(H2SiO4)4---- 8.167e-024 3.106e-018 0.4387 -23.4457
Ca(For)2(aq) 5.879e-024 7.649e-019 1.0000 -23.2307
FeSO4+ 5.340e-024 8.110e-019 0.9500 -23.2948
Methanol(aq) 4.922e-024 1.577e-019 1.0000 -23.3078
MgCH3COO+ 3.823e-024 3.186e-019 0.9500 -23.4398
NaCH3COO(aq) 3.547e-024 2.909e-019 1.0000 -23.4502
Mg(For)2(aq) 9.751e-025 1.115e-019 1.0000 -24.0110
Fe(For)2(aq) 8.323e-025 1.214e-019 1.0000 -24.0797
FeCH3COO+ 6.424e-025 7.380e-020 0.9500 -24.2145
MnCH3COO+ 5.188e-025 5.913e-020 0.9500 -24.3073
Mn(For)2(aq) 4.715e-025 6.835e-020 1.0000 -24.3265
KCH3COO(aq) 2.209e-025 2.168e-020 1.0000 -24.6557
SCN- 1.271e-025 7.383e-021 0.9496 -24.9182
Mn(OH)4-- 1.105e-025 1.359e-020 0.8140 -25.0459
TcO++ 9.160e-026 1.044e-020 0.8152 -25.1269
TcO4- 8.413e-026 1.363e-020 0.9500 -25.0973
Na(For)2- 7.129e-026 8.057e-021 0.9500 -25.1692
FeCl++ 2.264e-026 2.067e-021 0.8152 -25.7338
Fe(SO4)2- 1.275e-026 3.162e-021 0.9500 -25.9167
NH4CH3COO(aq) 8.770e-027 6.759e-022 1.0000 -26.0570
Acetamide(aq) 8.736e-027 5.160e-022 1.0000 -26.0587
K(For)2- 5.868e-027 7.577e-022 0.9500 -26.2538
FeCl2+ 4.174e-027 5.290e-022 0.9500 -26.4017
S3-- 3.830e-027 3.684e-022 0.8140 -26.5062
Formaldehyde(aq) 1.252e-027 3.757e-023 1.0000 -26.9026
Oxalic_acid(aq) 4.184e-028 3.767e-023 1.0000 -27.3784
Mg4(OH)4++++ 2.305e-029 3.808e-024 0.4464 -28.9876
SiF6-- 1.589e-029 2.258e-024 0.8140 -28.8881
Methanamine(aq) 8.860e-030 2.751e-025 1.0000 -29.0526
Malonate 6.329e-030 6.457e-025 0.8140 -29.2881
Glycolate 2.365e-030 1.775e-025 0.9500 -29.6484
HCN(aq) 6.721e-031 1.816e-026 1.0000 -30.1726
Ethane(aq) 4.141e-031 1.245e-026 1.0000 -30.3829
H-Malonate 2.027e-031 2.089e-026 0.9500 -30.7154
Fe2(OH)2++++ 1.041e-031 1.516e-026 0.4464 -31.3329
Ca(Glyc)+ 2.354e-032 2.710e-027 0.9500 -31.6504
Glycine(aq) 1.382e-032 1.037e-027 1.0000 -31.8595
S4-- 6.272e-033 8.043e-028 0.8140 -32.2920
CN- 5.458e-033 1.420e-028 0.9493 -32.2856
Na(Glyc)(aq) 2.194e-033 2.151e-028 1.0000 -32.6588
Fe(Glyc)+ 2.153e-033 2.817e-028 0.9500 -32.6893
TcO4-- 1.894e-033 3.068e-028 0.8140 -32.8120
Mg(Glyc)+ 1.868e-033 1.855e-028 0.9500 -32.7510
Glycolic_acid(aq 1.154e-033 8.777e-029 1.0000 -32.9377
Mn(Glyc)+ 5.227e-034 6.793e-029 0.9500 -33.3040
Mn+++ 1.966e-034 1.080e-029 0.6333 -33.9048
K(Glyc)(aq) 1.890e-034 2.157e-029 1.0000 -33.7236
Propanoate 1.162e-034 8.486e-030 0.9500 -33.9572
Acetaldehyde(aq) 7.933e-035 3.494e-030 1.0000 -34.1006
Ethanol(aq) 1.127e-035 5.193e-031 1.0000 -34.9479
Malonic_acid(aq) 1.030e-035 1.072e-030 1.0000 -34.9870
Fe(Gly)+ 4.044e-036 5.253e-031 0.9500 -35.4155
Mg(Gly)+ 3.352e-036 3.297e-031 0.9500 -35.4970
S2O4-- 2.242e-036 2.872e-031 0.8164 -35.7375
S2O5-- 1.682e-036 2.424e-031 0.8140 -35.8635
Mn(Gly)+ 8.507e-037 1.097e-031 0.9500 -36.0925
Propanoic_acid(a 6.443e-037 4.772e-032 1.0000 -36.1909
FeCl4- 3.042e-037 6.011e-032 0.9500 -36.5391
Ca(Gly)+ 1.605e-037 1.832e-032 0.9500 -36.8168
Ca(Prop)+ 1.219e-037 1.379e-032 0.9500 -36.9363
Na(Prop)(aq) 1.060e-037 1.018e-032 1.0000 -36.9748
Fe(Prop)+ 2.801e-038 3.611e-033 0.9500 -37.5749
Mg(Prop)+ 2.214e-038 2.156e-033 0.9500 -37.6770
Mn(Prop)+ 1.259e-038 1.612e-033 0.9500 -37.9221

K(Prop)(aq) 9.129e-039 1.024e-033 1.0000 -38.0396
S5-- 6.190e-039 9.923e-034 0.8140 -38.2977
S2O6-- 3.424e-039 5.482e-034 0.8140 -38.5549
S4O6-- 2.111e-039 4.734e-034 0.8140 -38.7649
Ethylene(aq) 3.858e-040 1.082e-035 1.0000 -39.4136
Lactate 6.201e-042 5.523e-037 0.9500 -41.2298
Fe3(OH)4(5+) 6.194e-042 1.459e-036 0.2861 -41.7515
Succinate 4.758e-042 5.522e-037 0.8140 -41.4120
Ca(CH3COO)2(aq) 8.086e-043 1.279e-037 1.0000 -42.0923
Ethanamine(aq) 2.790e-043 1.258e-038 1.0000 -42.5545
H-Succinate 1.309e-043 1.532e-038 0.9500 -42.9053
Mg(CH3COO)2(aq) 1.086e-043 1.546e-038 1.0000 -42.9642
Acetone(aq) 1.019e-043 5.917e-039 1.0000 -42.9919
Fe(CH3COO)2(aq) 4.940e-044 8.590e-039 1.0000 -43.3063
Ca(Lac)+ 3.633e-044 4.692e-039 0.9500 -43.4620
Mn(CH3COO)2(aq) 1.774e-044 3.069e-039 1.0000 -43.7510
Na(CH3COO)2- 8.023e-045 1.132e-039 0.9500 -44.1179
Na(Lac)(aq) 5.752e-045 6.445e-040 1.0000 -44.2402
Mg(Lac)+ 5.372e-045 6.089e-040 0.9500 -44.2922
Alanine(aq) 4.961e-045 4.419e-040 1.0000 -44.3044
Fe(Lac)+ 4.922e-045 7.132e-040 0.9500 -44.3301
Propane(aq) 4.258e-045 1.877e-040 1.0000 -44.3708
Lactic_acid(aq) 3.238e-045 2.916e-040 1.0000 -44.4898
Mn(Lac)+ 9.664e-046 1.392e-040 0.9500 -45.0371
K(Lac)(aq) 4.955e-046 6.350e-041 1.0000 -45.3050
K(CH3COO)2- 3.688e-046 5.797e-041 0.9500 -45.4555
Succinic_acid(aq 1.511e-046 1.784e-041 1.0000 -45.8207
NH4(CH3COO)2- 3.521e-047 4.792e-042 0.9500 -46.4756
S3O6-- 2.995e-047 5.756e-042 0.8140 -46.6130
Propanal(aq) 6.007e-048 3.488e-043 1.0000 -47.2214
Fe(Ala)+ 1.533e-048 2.206e-043 0.9500 -47.8369
Butanoate 8.424e-049 7.336e-044 0.9500 -48.0967
1-Propanol(aq) 2.710e-049 1.628e-044 1.0000 -48.5670
HBrO(aq) 3.479e-050 3.371e-045 1.0000 -49.4586
Mn(Ala)+ 3.010e-050 4.304e-045 0.9500 -49.5438
Ca(Ala)+ 1.159e-050 1.486e-045 0.9500 -49.9580
Mg(Ala)+ 1.009e-050 1.134e-045 0.9500 -50.0185
Butanoic_acid(aq 3.880e-051 3.418e-046 1.0000 -50.4111
BrO- 1.985e-051 1.903e-046 0.9500 -50.7246
1-Propene(aq) 1.371e-051 5.767e-047 1.0000 -50.8631
Na(But)(aq) 7.305e-052 8.040e-047 1.0000 -51.1364
Ca(But)+ 6.087e-052 7.741e-047 0.9500 -51.2378
Fe(But)+ 2.069e-052 2.958e-047 0.9500 -51.7064
Mg(But)+ 1.057e-052 1.177e-047 0.9500 -51.9983
Mn(But)+ 7.739e-053 1.099e-047 0.9500 -52.1336
K(But)(aq) 6.292e-053 7.939e-048 1.0000 -52.2012
Methionine(aq) 9.188e-054 1.371e-048 1.0000 -53.0368
HClO(aq) 6.447e-054 3.382e-049 1.0000 -53.1906
Br2(aq) 5.607e-054 8.959e-049 1.0000 -53.2513
Serine(aq) 4.236e-054 4.451e-049 1.0000 -53.3730
ClO- 2.428e-054 1.249e-049 0.9500 -53.6370
Aspartic_acid(aq 3.281e-055 4.366e-050 1.0000 -54.4840
Glutarate 2.705e-055 3.518e-050 0.8140 -54.6573
Ethyne(aq) 7.629e-056 1.986e-051 1.0000 -55.1175
2-Hydroxybutanoa 5.791e-056 5.970e-051 0.9500 -55.2595
1-Propanamine(aq 2.222e-056 1.313e-051 1.0000 -55.6532
HO2- 8.249e-057 2.722e-052 0.9500 -56.1059
H-Glutarate 4.561e-057 5.979e-052 0.9500 -56.3632
Br3- 1.889e-057 4.527e-052 0.9500 -56.7461
Asparagine(aq) 1.658e-058 2.190e-053 1.0000 -57.7805
a-Aminobutyric_a 7.687e-059 7.926e-054 1.0000 -58.1142
n-Butane(aq) 3.851e-059 2.238e-054 1.0000 -58.4144
2-Hydroxybutanoi 2.687e-059 2.797e-054 1.0000 -58.5708
NO2- 1.346e-059 6.193e-055 0.9493 -58.8935
Ethylacetate(aq) 7.224e-060 6.364e-055 1.0000 -59.1412
Glutaric_acid(aq 7.134e-060 9.423e-055 1.0000 -59.1467
S5O6-- 1.960e-060 5.023e-055 0.8140 -59.7972
N3- 1.266e-060 5.320e-056 0.9500 -59.9198
Ca(Glyc)2(aq) 8.738e-061 1.661e-055 1.0000 -60.0586
Fe(Glyc)2(aq) 2.152e-061 4.430e-056 1.0000 -60.6672

Mg(Glyc)2(aq)	3.724e-062	6.494e-057	1.0000	-61.4290
Mn(Glyc)2(aq)	7.865e-063	1.612e-057	1.0000	-62.1043
Pentanoate	7.352e-063	7.434e-058	0.9500	-62.1559
HN3(aq)	4.544e-063	1.955e-058	1.0000	-62.3425
Na(Glyc)2-	4.216e-063	7.296e-058	0.9500	-62.3974
Butanal(aq)	1.824e-063	1.315e-058	1.0000	-62.7389
HNO2(aq)	1.600e-063	7.523e-059	1.0000	-62.7957
HSO5-	5.998e-064	6.781e-059	0.9500	-63.2443
1-Butanol(aq)	4.021e-064	2.980e-059	1.0000	-63.3957
K(Glyc)2-	3.471e-064	6.565e-059	0.9500	-63.4819
Mn(CH3COO)3-	2.098e-064	4.868e-059	0.9500	-63.7005
Pentanoic_acid(a	3.685e-065	3.763e-060	1.0000	-64.4335
1-Propyne(aq)	1.752e-065	7.018e-061	1.0000	-64.7565
Na(Pent)(aq)	6.485e-066	8.048e-061	1.0000	-65.1881
1-Butene(aq)	6.301e-066	3.535e-061	1.0000	-65.2006
Ca(Pent)+	3.106e-066	4.385e-061	0.9500	-65.5300
Fe(Pent)+	1.635e-066	2.566e-061	0.9500	-65.8088
K(Pent)(aq)	5.586e-067	7.832e-062	1.0000	-66.2529
Mg(Pent)+	5.275e-067	6.616e-062	0.9500	-66.3000
Mn(Pent)+	5.097e-067	7.953e-062	0.9500	-66.3150
Diglycine(aq)	4.950e-067	6.539e-062	1.0000	-66.3054
Glutamic_acid(aq	2.385e-067	3.508e-062	1.0000	-66.6225
Fe(Gly)2(aq)	1.948e-067	3.973e-062	1.0000	-66.7104
Mg(Gly)2(aq)	9.949e-068	1.715e-062	1.0000	-67.0022
Threonine(aq)	3.231e-068	3.848e-063	1.0000	-67.4907
Mn(Gly)2(aq)	2.305e-068	4.679e-063	1.0000	-67.6374
Diketopiperazine	3.755e-069	4.284e-064	1.0000	-68.4254
2-Hydroxypentano	7.972e-070	9.336e-065	0.9500	-69.1207
O2(aq)	1.400e-070	4.481e-066	1.0000	-69.8537
1-Butanamine(aq)	1.021e-070	7.470e-066	1.0000	-69.9908
Adipate	7.297e-071	1.052e-065	0.8140	-70.2262
Fe(Prop)2(aq)	3.093e-071	6.247e-066	1.0000	-70.5096
Ca(Gly)2(aq)	2.681e-071	5.046e-066	1.0000	-70.5716
Ca(Prop)2(aq)	2.284e-071	4.252e-066	1.0000	-70.6414
Mn(Prop)2(aq)	7.474e-072	1.503e-066	1.0000	-71.1265
Na(Prop)2-	6.660e-072	1.126e-066	0.9500	-71.1988
Mg(Prop)2(aq)	4.561e-072	7.773e-067	1.0000	-71.3410
Glutamine(aq)	1.376e-072	2.011e-067	1.0000	-71.8614
H-Adipate	1.210e-072	1.756e-067	0.9500	-71.9395
Valine(aq)	9.407e-073	1.102e-067	1.0000	-72.0266
K(Prop)2-	5.472e-073	1.013e-067	0.9500	-72.2841
n-Pentane(aq)	2.968e-073	2.141e-068	1.0000	-72.5276
2-Hydroxypentano	2.229e-073	2.633e-068	1.0000	-72.6518
Adipic_acid(aq)	2.240e-075	3.274e-070	1.0000	-74.6497
Benzene(aq)	1.987e-075	1.552e-070	1.0000	-74.7018
Phenol(aq)	3.174e-076	2.987e-071	1.0000	-75.4984
1-Pentanol(aq)	5.685e-077	5.011e-072	1.0000	-76.2452
Hexanoate	5.243e-077	6.036e-072	0.9500	-76.3027
Pentanal(aq)	2.732e-077	2.353e-072	1.0000	-76.5635
FeNO2++	1.074e-077	1.094e-072	0.8152	-77.0578
NO3-	2.736e-079	1.696e-074	0.9493	-78.5855
Hexanoic_acid(aq	2.718e-079	3.157e-074	1.0000	-78.5657
Benzoate	2.419e-079	2.929e-074	0.9514	-78.6381
1-Butyne(aq)	1.228e-079	6.642e-075	1.0000	-78.9107
MnO4--	1.189e-079	1.414e-074	0.8140	-79.0142
Alanylglycine(aq	8.013e-080	1.171e-074	1.0000	-79.0962
1-Pentene(aq)	4.343e-080	3.046e-075	1.0000	-79.3622
S2O8--	3.988e-080	7.662e-075	0.8140	-79.4886
CaNO3+	3.053e-082	3.116e-077	0.9500	-81.5375
Benzoic_acid(aq)	2.749e-082	3.357e-077	1.0000	-81.5608
Pimelate	7.876e-084	1.246e-078	0.8140	-83.1930
2-Hydroxyhexanoa	4.722e-084	6.191e-079	0.9500	-83.3482
MnNO3+	2.508e-084	2.933e-079	0.9500	-83.6229
Ca(Lac)2(aq)	2.334e-084	5.093e-079	1.0000	-83.6319
Fe(Lac)2(aq)	1.174e-084	2.745e-079	1.0000	-83.9305
Mg(Lac)2(aq)	3.150e-085	6.376e-080	1.0000	-84.5017
H-Pimelate	1.351e-085	2.150e-080	0.9500	-84.8916
1-Pentanamine(aq	8.830e-086	7.695e-081	1.0000	-85.0541
Mn(Lac)2(aq)	6.356e-086	1.481e-080	1.0000	-85.1968
Na(Lac)2-	2.968e-086	5.968e-081	0.9500	-85.5499

Toluene(aq) 9.894e-087 9.115e-082 1.0000 -86.0046
o-Phthalate 8.594e-087 1.410e-081 0.8140 -86.1552
K(Lac)2- 2.442e-087 5.305e-082 0.9500 -86.6345
2-Hydroxyhexanoi 1.851e-087 2.446e-082 1.0000 -86.7327
n-Hexane(aq) 1.642e-087 1.415e-082 1.0000 -86.7846
Isoleucine(aq) 1.327e-087 1.740e-082 1.0000 -86.8772
HNO3(aq) 9.752e-088 6.144e-083 1.0000 -87.0109
Leucine(aq) 9.466e-088 1.241e-082 1.0000 -87.0238
Ca(o-Phthalate)(4.100e-088 8.371e-083 1.0000 -87.3872
Pimelic_acid(aq) 2.962e-088 4.743e-083 1.0000 -87.5285
H(o-Phthalate)- 1.420e-088 2.344e-083 0.9500 -87.8700
Na(o-Phthalate)- 3.213e-089 6.011e-084 0.9500 -88.5153
p-Toluate 2.141e-090 2.892e-085 0.9500 -89.6917
m-Toluate 1.226e-090 1.657e-085 0.9500 -89.9337
Heptanoate 5.023e-091 6.487e-086 0.9500 -90.3214
Hexanal(aq) 1.852e-091 1.855e-086 1.0000 -90.7323
1-Hexanol(aq) 9.820e-092 1.003e-086 1.0000 -91.0079
MnO4- 3.469e-092 4.125e-087 0.9496 -91.4823
Fe(Ala)2(aq) 2.172e-092 5.038e-087 1.0000 -91.6632
o-Toluate 1.265e-092 1.710e-087 0.9500 -91.9201
o-Phthalic_acid(9.058e-093 1.505e-087 1.0000 -92.0430
p-Toluic_acid(aq 3.582e-093 4.876e-088 1.0000 -92.4459
Heptanoic_acid(a 2.809e-093 3.657e-088 1.0000 -92.5514
m-Toluic_acid(aq 1.593e-093 2.169e-088 1.0000 -92.7977
1-Pentyne(aq) 8.905e-094 6.065e-089 1.0000 -93.0504
1-Hexene(aq) 4.643e-094 3.907e-089 1.0000 -93.3332
Mn(Ala)2(aq) 5.252e-095 1.214e-089 1.0000 -94.2797
o-Toluic_acid(aq 7.311e-096 9.952e-091 1.0000 -95.1360
Mg(Ala)2(aq) 5.068e-097 1.016e-091 1.0000 -96.2952
ClO2- 2.830e-097 1.909e-092 0.9500 -96.5704
Ca(Ala)2(aq) 1.465e-097 3.167e-092 1.0000 -96.8343
2-Hydroxyheptano 3.919e-098 5.688e-093 0.9500 -97.4291
Suberate 2.928e-099 5.042e-094 0.8140 -98.6227
Fe(But)2(aq) 1.613e-099 3.710e-094 1.0000 -98.7924
FeNO3++ 1.545e-099 1.820e-094 0.8152 -98.8998
1-Hexanamine(aq) 7.205e-100 7.290e-095 1.0000 -99.1424
Ca(But)2(aq) 5.971e-100 1.279e-094 1.0000 -99.2240
Na(But)2- 3.090e-100 6.093e-095 0.9500 -99.5323
Mn(But)2(aq) 2.823e-100 6.467e-095 1.0000 -99.5493
Mg(But)2(aq) 1.061e-100 2.106e-095 1.0000 -99.9742
H-Suberate 4.775e-101 8.268e-096 0.9500 -100.3433
HClO2(aq) 2.996e-101 2.051e-096 1.0000 -100.5235
K(But)2- 2.539e-101 5.415e-096 0.9500 -100.6176
2-Hydroxyheptano 1.536e-101 2.245e-096 1.0000 -100.8137
n-Heptane(aq) 1.363e-101 1.365e-096 1.0000 -100.8656
Suberic_acid(aq) 1.101e-103 1.918e-098 1.0000 -102.9582
Octanoate 6.857e-105 9.818e-100 0.9500 -104.1862
Heptanal(aq) 1.684e-106 1.923e-101 1.0000 -105.7735
1-Heptanol(aq) 7.172e-107 8.333e-102 1.0000 -106.1443
Octanoic_acid(aq 3.868e-107 5.578e-102 1.0000 -106.4125
1-Hexyne(aq) 5.186e-108 4.259e-103 1.0000 -107.2852
1-Heptene(aq) 3.663e-108 3.596e-103 1.0000 -107.4361
2-Hydroxyoctanoa 3.252e-112 5.177e-107 0.9500 -111.5101
1-Heptanamine(aq 5.979e-114 6.888e-109 1.0000 -113.2234
n-Propylbenzene(9.389e-115 1.128e-109 1.0000 -114.0274
Azelate 2.190e-115 4.078e-110 0.8140 -114.7488
2-Hydroxyoctanoi 1.275e-115 2.042e-110 1.0000 -114.8946
n-Octane(aq) 1.022e-115 1.167e-110 1.0000 -114.9905
Phenylalanine(aq 5.218e-117 8.619e-112 1.0000 -116.2825
H-Azelate 3.512e-117 6.575e-112 0.9500 -116.4767
Nonanoate 6.192e-119 9.734e-114 0.9500 -118.2305
Azelaic_acid(aq) 8.376e-120 1.576e-114 1.0000 -119.0770
Octanal(aq) 6.070e-120 7.781e-115 1.0000 -119.2168
1-Octanol(aq) 9.877e-121 1.286e-115 1.0000 -120.0054
Nonanoic_acid(aq 2.369e-121 3.748e-116 1.0000 -120.6254
Leucylglycine(aq 4.900e-122 9.221e-117 1.0000 -121.3098
1-Heptyne(aq) 2.775e-122 2.668e-117 1.0000 -121.5567
1-Octene(aq) 2.097e-122 2.353e-117 1.0000 -121.6783
Tyrosine(aq) 8.860e-123 1.605e-117 1.0000 -122.0525
2-Hydroxynonanoa 2.792e-126 4.836e-121 0.9500 -125.5764

ClO3-	2.357e-126	1.967e-121	0.9496	-125.6501
BrO3-	1.646e-127	2.105e-122	0.9496	-126.8059
Fe(Pent)2(aq)	1.009e-127	2.603e-122	1.0000	-126.9963
1-Octanamine(aq)	4.962e-128	6.412e-123	1.0000	-127.3044
Na(Pent)2-	2.379e-128	5.357e-123	0.9500	-127.6459
Ca(Pent)2(aq)	1.669e-128	4.044e-123	1.0000	-127.7775
Mn(Pent)2(aq)	1.251e-128	3.217e-123	1.0000	-127.9027
Mg(Pent)2(aq)	2.898e-129	6.564e-124	1.0000	-128.5379
K(Pent)2-	1.958e-129	4.724e-124	0.9500	-128.7305
2-Hydroxynonanoi	1.094e-129	1.906e-124	1.0000	-128.9610
Sebacate	8.651e-130	1.732e-124	0.8140	-129.1523
H-Sebacate	1.484e-131	2.986e-126	0.9500	-130.8509
Decanoate	3.314e-133	5.674e-128	0.9500	-132.5020
Nonanal(aq)	3.846e-134	5.470e-129	1.0000	-133.4150
Sebacic_acid(aq)	3.722e-134	7.527e-129	1.0000	-133.4292
Decanoic_acid(aq	1.966e-135	3.386e-130	1.0000	-134.7064
1-Octyne(aq)	2.012e-136	2.217e-131	1.0000	-135.6964
2-Hydroxydecanoa	2.356e-140	4.412e-135	0.9500	-139.6501
2-Hydroxydecanoi	9.233e-144	1.738e-138	1.0000	-143.0347
Undecanoate	2.750e-147	5.095e-142	0.9500	-146.5829
Tryptophan(aq)	2.750e-147	5.616e-142	1.0000	-146.5607
Decanal(aq)	8.075e-148	1.262e-142	1.0000	-147.0929
Undecanoic_acid(	1.660e-149	3.091e-144	1.0000	-148.7800
n-Hexylbenzene(a	4.769e-157	7.738e-152	1.0000	-156.3216
ClO4-	9.935e-160	9.879e-155	0.9496	-159.0253
Dodecanoate	2.321e-161	4.625e-156	0.9500	-160.6566
Mn(NO3)2(aq)	1.554e-162	2.781e-157	1.0000	-161.8085
Dodecanoic_acid(	1.377e-163	2.758e-158	1.0000	-162.8610
2-Butanone(aq)	2.496e-171	5.398e-166	1.0000	-170.6028
BrO4-	2.137e-178	3.075e-173	0.9500	-177.6925
2-Pentanone(aq)	3.286e-214	8.490e-209	1.0000	-213.4833
2-Hexanone(aq)	3.816e-256	1.147e-250	1.0000	-255.4184
2-Heptanone(aq)	2.181e-298	7.471e-293	1.0000	-297.6613
Ethylbenzene(aq)	2.867e-301	9.129e-296	1.0000	-300.0000
n-Pentylbenzene(	0.0000	0.0000	1.0000	-300.0000
n-Octylbenzene(a	0.0000	0.0000	1.0000	-300.0000
n-Heptylbenzene(	0.0000	0.0000	1.0000	-300.0000
n-Butylbenzene(a	0.0000	0.0000	1.0000	-300.0000
2-Octanone(aq)	0.0000	0.0000	1.0000	-300.0000

Mineral saturation states

	log Q/K		log Q/K
Hematite	0.0000 sat	Fluorite	-1.1724
Quartz	0.0000 sat	SiO2(am)	-1.2857
TcO2	0.0000 sat	Magnetite	-1.3360
Ice	-0.1387	Calcite	-1.4387
Tridymite	-0.1715	Aragonite	-1.5831
Pyrite	-0.1929	Cronstedtite-7A	-2.0953
Chalcedony	-0.2712	Monohydrocalcite	-2.2724
Goethite	-0.4802	Ferrosilite	-2.4186
Cristobalite(alp	-0.5505	Dolomite	-2.4738
Coesite	-0.8100	Dolomite-ord	-2.4738
Siderite	-0.9639	Sellaite	-2.6054
Rhodochrosite	-0.9826	Magnesite	-2.6639
Cristobalite(bet	-0.9940	Minnesotaite	-2.8942

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
N2(g)	0.04620	-1.335
H2O(g)	0.02598	-1.585
CO2(g)	0.004264	-2.370
H2(g)	8.419e-009	-8.075
H2S(g)	1.632e-009	-8.787
NH3(g)	1.266e-010	-9.898
CH4(g)	1.709e-012	-11.767
HF(g)	9.679e-013	-12.014
CO(g)	1.121e-014	-13.950
HCl(g)	9.182e-018	-17.037
HBr(g)	1.302e-021	-20.885
SO2(g)	1.107e-021	-20.956
S2(g)	8.378e-028	-27.077
SiF4(g)	3.868e-033	-32.413
C2H4(g)	8.128e-038	-37.090
NO(g)	4.917e-050	-49.308
Br2(g)	1.157e-053	-52.937
Na(g)	4.594e-060	-59.338
Cl2(g)	4.070e-060	-59.390
K(g)	7.937e-062	-61.100
O2(g)	1.108e-067	-66.955
NO2(g)	2.442e-077	-76.612
Tc2O7(g)	1.960e-088	-87.708
Mg(g)	3.097e-098	-97.509
TcO(g)	9.195e-103	-102.036
F2(g)	3.550e-113	-112.450
Tc(g)	3.824e-117	-116.417
Ca(g)	2.772e-120	-119.557
C(g)	7.900e-123	-122.102
Si(g)	8.008e-155	-154.096

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
Br-	1.385e-005	1.385e-005	1.107		
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
F-	0.0001757	0.0001757	3.338		
Fe++	1.397e-005	1.065e-005	0.5946		
H+	0.0001379	0.0001445	0.1456		

H2O	55.51	55.51	9.999e+005
HCO3-	0.001068	0.001068	65.13
K+	8.277e-005	8.277e-005	3.236
Mg++	4.662e-005	4.662e-005	1.133
Mn++	7.408e-006	7.408e-006	0.4069
NH3(aq)	6.125e-005	6.125e-005	1.043
Na+	0.0009184	0.0009184	21.11
O2(aq)	4.594e-005	4.511e-005	1.443
SO4--	0.0001577	0.0001577	15.14
SiO2(aq)	0.0003330	0.0001003	6.027
TcO4-	1.000e-012	3.973e-013	6.436e-008

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001068	0.001068	12.82		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.338		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.065e-005	0.5946		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.236		
Silicon	0.0003330	0.0001003	2.817		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		
Technetium	1.000e-012	3.973e-013	3.893e-008		

Note that the calculate concentration of technetium, in equilibrium with TcO2(s), is 3.973e-13 moles per liter.

Two other GWB runs were made to check if at high Eh (0.0 volt) a technetium phase would precipitate. One run had the same conditions as in (6) except Eh was set to 0.0 v, and the second run had Eh=0.0 v and TcO4- = 1.e-5 molal. The results indicated no technetium phase was saturated, consistent with the Eh-pH diagrams shown in previous pages of this notebook, where TcO4- aqueous species was predominant at high Eh.

Another GWB calculation was made, this time without NO3- in the input file. Eh was set to 0.0 v, and TcO4- was set to 1.e-12 molal. The resulting solubility is the same—3.973E-12 molal.

(7) Calculate solubility of technetium in pure water under reducing conditions (Eh = 0 v). Use NEA Tc data.

```
Temperature is 25 C
    Thermo dataset: D:\Gwb\Gtdata\LLNL
database\Pabalan.dat
    Working directory: d:\
    Options: Debye-Huckel

    Basis is:
     H2O                    1 kg solvent
     TcO4-                  total molality = 1e-12
     e-  (swapped for O2(aq))      Eh = 0
     H+                     charge balance

    No reactants specified.

    Other "show" options: type "show show"
React> go

Solving for initial system.

Loaded:  12 aqueous species,
         5 minerals,
         6 gases,
         0 surface species,
         3 elements,
         1 oxides.

N-R converged in  13 its., resmax =  1.01e-013, Xi =  0.0000
 Charge balance: H+ molality adjusted from .001 to 4e-12

 Removing e- from basis vector
 Swapping TcO(OH)2(aq) in for e-
 1 supersaturated phase, TcO2
 Swapping TcO2 in for TcO(OH)2(aq)
N-R converged in  3 its., resmax =  8.92e-016, Xi =  0.0000

No reaction path specified.
```

The following is the React output file:

```
Step #    0          Xi = 0.0000
Temperature =  25.0 C   Pressure =  1.013 bars
pH =  6.998          log fO2 =  -55.115
Eh =  0.0000 volts     pe =  0.0000

Ionic strength     =   0.000000
Activity of water  =   1.000000
Solvent mass       =   1.000000 kg
Solution mass      =   1.000006 kg
```

```
Solution density   =   1.013   g/cm3
Chlorinity         =   0.000000 molal
Dissolved solids   =         6 mg/kg sol'n

No minerals in system.

Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
-----------------------------------------------------------
H+            1.006e-007    0.0001014    0.9996     -
6.9975
OH-           1.006e-007    0.001711     0.9996     -
6.9976
(only species > 1e-8 molal listed)

Mineral saturation states
              log Q/K                    log Q/K
-----------------------------------------------------------
TcO2          0.4009s/sat  Ice           -0.1387
(only minerals with log Q/K > -3 listed)

Gases            fugacity     log fug.
-----------------------------------------
H2O(g)            0.02598     -1.585
H2(g)           1.012e-014    -13.995
O2(g)           7.667e-056    -55.115
Tc2O7(g)         7.147e-070   -69.146
TcO(g)          2.782e-108    -107.556
Tc(g)           1.391e-128    -127.857

                          In fluid              Sorbed
Original basis  total moles    moles      mg/kg       moles
mg/kg
-----------------------------------------------------------
H+              -1.000      -1.000     -1008.
H2O              56.01       56.01  1.009e+006
O2(aq)          -0.2500     -0.2500     -8000.
TcO4-         1.000e-012  1.000e-012  1.620e-007

Elemental composition            In fluid              Sorbed
              total moles    moles      mg/kg       moles
mg/kg
-----------------------------------------------------------
Hydrogen          111.0       111.0  1.119e+005
Oxygen            55.51       55.51  8.881e+005
Technetium    1.000e-012  1.000e-012  9.800e-008

Step #    0          Xi = 0.0000
Temperature =  25.0 C    Pressure =  1.013 bars
pH =  6.998            log fO2 =  -54.482
Eh =   0.0094 volts      pe =   0.1583
Ionic strength     =    0.000000
```

```
Activity of water  =   1.000000
Solvent mass       =   1.000000 kg
Solution mass      =   1.000006 kg
Solution density   =   1.013   g/cm3
Chlorinity         =   0.000000 molal
Dissolved solids   =         6 mg/kg sol'n

Minerals in system     moles      log moles     grams
volume (cm3)
-----------------------------------------------------------
TcO2          6.026e-013   -12.220    7.834e-011
                                    ____________
____________
(total)                    7.834e-011        0.0000*

Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
-----------------------------------------------------------
H+            1.006e-007    0.0001014    0.9996     -
6.9975
OH-           1.006e-007    0.001711     0.9996     -
6.9976
(only species > 1e-8 molal listed)

Mineral saturation states
              log Q/K                    log Q/K
-----------------------------------------------------------
TcO2          0.0000 sat   Ice           -0.1387
(only minerals with log Q/K > -3 listed)

Gases            fugacity     log fug.
-----------------------------------------
H2O(g)            0.02598     -1.585
H2(g)           4.882e-015    -14.311
O2(g)           3.296e-055    -54.482
Tc2O7(g)         1.006e-069   -68.998
TcO(g)          5.331e-109    -108.273
Tc(g)           1.286e-129    -128.891

                          In fluid              Sorbed
Original basis  total moles    moles      mg/kg       moles
mg/kg
-----------------------------------------------------------
H+            1.000e-012  3.975e-013  4.006e-010
H2O              55.51       55.51  1.000e+006
O2(aq)       -7.499e-013 -2.979e-013 -9.534e-009
TcO4-         1.000e-012  3.974e-013  6.438e-008

Elemental composition            In fluid              Sorbed
              total moles    moles      mg/kg       moles
mg/kg
-----------------------------------------------------------
Hydrogen          111.0       111.0  1.119e+005
Oxygen            55.51       55.51  8.881e+005
Technetium    1.000e-012  3.974e-013  3.894e-008
```

Note that the solubility of technetium in pure water is the same as in the WT#17 water.

October 15, 1999

The calculated solubility of technetium is a few orders of magnitude lower than reported by Meyer et al. (1991; Radiochimica Acta 55, p. 11) and Eriksen et al. (1992; Radiochimica Acta 58/59). For example, Meyer et al. reported values in the range 10^{-8} and 10^{-9} moles/liter. The solids used in those experiments are not crystalline phases, rather are amorphous and hydrated oxides, with the approximate stoichiometry of TcO2.nH2O.

For the purposes of this study, the solubility of technetium under reducing conditions was assumed to be controlled by the TcO2·1.6H2O phase. The Gibbs free energy of formation for this phase is given in the NEA TCDATA file. A GWB calculation was done to determine the solubility of TcO2·1.6H2O in the WT#17 well water. The calculation is similar to that of (7) done on October 12, 1999, except the precipitation of TcO2 was suppressed. The following give the input and output files for that run:

```
Temperature is 25 C
   Thermo dataset: D:\Gwb\Gtdata\LLNL
database\Pabalan.dat
   Working directory: d:\
   Options: Debye-Huckel

   Basis is:
    H2O                    1 kg solvent
    NO3- (swapped for NH3(aq))    total mg/kg = 3.798
    e- (swapped for O2(aq))       Eh = 0
    Na+                    total mg/kg = 21.113
    SiO2(aq)               total mg/kg = 20.008
    Ca++                   total mg/kg = 11.161
    K+                     total mg/kg = 3.236
    SO4--                  total mg/kg = 15.146
    Cl-                    total mg/kg = 9.197
    HCO3-                  charge balance
    H+                     pH = 7.14
    F-                     total mg/kg = 3.338
    Mg++                   total mg/kg = 1.133
    Fe++                   total mg/kg = .78
    Mn++                   total mg/kg = .407
    Br-                    total mg/kg = 1.107
    TcO4-                  total molality = 1e-8

   No reactants specified.

Suppressed species:
      TcO2
```

```
Solving for initial system.
 ...suppressed loading of:  TcO2
 ...suppressed loading of:  TcO2

 Loaded:  430 aqueous species,
       191 minerals,
       30 gases,
        0 surface species,
       16 elements,
       12 oxides.

N-R converged in  42 its., resmax =   3.7e-011, Xi =  0.0000
 Charge balance: HCO3- molality adjusted from .001 to
.001068

 Removing e- from basis vector
 Swapping N2(aq) in for e-
 8 supersaturated phases, most = Hematite
 Swapping Hematite in for NO3-
N-R converged in  34 its., resmax =   4.02e-013, Xi =  0.0000
 4 supersaturated phases, most = Quartz
 Swapping Quartz in for SiO2(aq)
 Swapping CO2(aq) in for H+
 Swapping TcO(OH)2(aq) in for TcO4-
N-R converged in  9 its., resmax =   6.87e-014, Xi =  0.0000
 1 supersaturated phase, TcO2:1.6H2O
 Swapping TcO2:1.6H2O in for TcO(OH)2(aq)
N-R converged in  3 its., resmax =   9.87e-014, Xi =  0.0000

No reaction path specified.
```

```
      Step #   0          Xi = 0.0000
      Temperature =  25.0 C   Pressure =  1.013 bars
      pH =  7.140          log fO2 =  -54.546
      Eh =  0.0000 volts     pe =  0.0000
      Ionic strength     =   0.002160
      Activity of water  =   0.999991
      Solvent mass       =   1.000000 kg
      Solution mass      =   1.000156 kg
      Solution density   =   1.013   g/cm3
      Chlorinity         =   0.000259 molal
      Dissolved solids   =        156 mg/kg sol'n

 No minerals in system.

 Aqueous species      molality    mg/kg sol'n   act. coef.
log act.
 ---------------------------------------------------------------
```

```
  HCO3-          0.0009189      56.06     0.9499      -
3.0591
  Na+            0.0009165      21.07     0.9499      -
3.0602
  SiO2(aq)       0.0003324      19.97     1.0000      -
3.4783
  Ca++           0.0002722      10.91     0.8185      -
3.6521
  Cl-            0.0002594      9.193     0.9492     -3.6087
  F-             0.0001753      3.329     0.9496     -3.7788
  SO4--          0.0001518      14.58     0.8138      -
3.9083
  CO2(aq)        0.0001398      6.154     1.0000      -
3.8544
  K+             8.269e-005     3.232     0.9492      -
4.1052
```

Mg++	4.490e-005	1.091	0.8229	-4.4324
N2(aq)	3.063e-005	0.8578	1.0000	-4.5139
Br-	1.385e-005	1.107	0.9492	-4.8811
Fe++	9.679e-006	0.5405	0.8185	-5.1012
Mn++	7.066e-006	0.3881	0.8185	-5.2378
FeHCO3+	3.820e-006	0.4464	0.9499	-5.4402
CaSO4(aq)	3.554e-006	0.4838	1.0000	-5.4493
CaHCO3+	2.279e-006	0.2304	0.9499	-5.6645
MgSO4(aq)	1.178e-006	0.1417	1.0000	-5.9290
NaHCO3(aq)	1.084e-006	0.09101	1.0000	-5.9652
NaSO4-	7.479e-007	0.08903	0.9499	-6.1485
CO3--	6.934e-007	0.04161	0.8150	-6.2479
HSiO3-	5.389e-007	0.04154	0.9499	-6.2908
MgHCO3+	3.686e-007	0.03144	0.9499	-6.4558
CaCO3(aq)	2.674e-007	0.02676	1.0000	-6.5729
FeCO3(aq)	2.404e-007	0.02785	1.0000	-6.6190
CaF+	1.875e-007	0.01108	0.9499	-6.7492
MnSO4(aq)	1.610e-007	0.02431	1.0000	-6.7932
FeSO4(aq)	1.551e-007	0.02356	1.0000	-6.8094
OH-	1.470e-007	0.002500	0.9496	-6.8551
MgF+	1.457e-007	0.006310	0.9499	-6.8588
MnCO3(aq)	1.082e-007	0.01244	1.0000	-6.9657
KSO4-	7.735e-008	0.01045	0.9499	-7.1339
H+	7.600e-008	7.659e-005	0.9532	-7.1400
MnHCO3+	4.046e-008	0.004691	0.9499	-7.4153
FeOH+	3.640e-008	0.002652	0.9499	-7.4612
NaCl(aq)	3.581e-008	0.002093	1.0000	-7.4459
FeF+	3.180e-008	0.002379	0.9499	-7.5199
MnF+	2.727e-008	0.002016	0.9499	-7.5866
MgCO3(aq)	1.989e-008	0.001677	1.0000	-7.7014
NaHSiO3(aq)	1.984e-008	0.001985	1.0000	-7.7025
HF(aq)	1.776e-008	0.0003552	1.0000	-7.7507
NaF(aq)	1.457e-008	0.0006117	1.0000	-7.8366
CaCl+	1.164e-008	0.0008788	0.9499	-7.9565

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Hematite	6.5069s/sat	SiO2(am)	-0.7647
Magnetite	5.3219s/sat	Siderite	-0.8282
Cronstedtite-7A	5.2346s/sat	Rhodochrosite	-0.9641
TcO2	4.4006s/sat	Fluorite	-1.1727
Goethite	2.7732s/sat	Calcite	-1.4199
Quartz	0.5210s/sat	Aragonite	-1.5643
TcO2:1.6H2O	0.3950s/sat	Ferrosilite	-1.7466
Tridymite	0.3495s/sat	Greenalite	-2.0902
Chalcedony	0.2498s/sat	Monohydrocalcite	-2.2536
Cristobalite(alp	-0.0295	Fe(OH)3	-2.3479
Ice	-0.1387	Dolomite	-2.4362
Coesite	-0.2890	Dolomite-ord	-2.4362
Minnesotaite	-0.3572	Sellaite	-2.6056
Cristobalite(bet	-0.4730	Magnesite	-2.6451

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
N2(g)	0.04704	-1.327
H2O(g)	0.02598	-1.585
CO2(g)	0.004117	-2.385
HF(g)	9.303e-013	-12.031
H2(g)	5.252e-015	-14.280
HCl(g)	8.826e-018	-17.054
NH3(g)	6.294e-020	-19.201
CO(g)	6.753e-021	-20.171
HBr(g)	1.251e-021	-20.903
SO2(g)	6.381e-028	-27.195
SiF4(g)	1.096e-032	-31.960
H2S(g)	2.283e-034	-33.641
CH4(g)	2.499e-037	-36.602
NO(g)	7.953e-044	-43.099
Br2(g)	1.713e-047	-46.766
Cl2(g)	6.028e-054	-53.220
O2(g)	2.847e-055	-54.546
Tc2O7(g)	5.109e-061	-60.292
Na(g)	3.774e-063	-62.423
K(g)	6.521e-065	-64.186
NO2(g)	6.331e-065	-64.199
S2(g)	4.215e-065	-64.375
C2H4(g)	4.467e-075	-74.350
Mg(g)	2.090e-104	-103.680
TcO(g)	1.443e-104	-103.841
F2(g)	5.258e-107	-106.279
Tc(g)	3.744e-125	-124.427
Ca(g)	1.871e-126	-125.728
C(g)	2.969e-135	-134.527
Si(g)	1.034e-166	-165.985

Original basis	total moles	In fluid moles	Sorbed mg/kg	Sorbed moles mg/kg
Br-	1.385e-005	1.385e-005	1.107	
Ca++	0.0002785	0.0002785	11.16	
Cl-	0.0002594	0.0002594	9.196	
F-	0.0001757	0.0001757	3.337	
Fe++	1.397e-005	1.397e-005	0.7799	
H+	-0.9999	-0.9999	-1008.	
H2O	56.01	56.01	1.009e+006	
HCO3-	0.001068	0.001068	65.13	
K+	8.277e-005	8.277e-005	3.235	
Mg++	4.662e-005	4.662e-005	1.133	
Mn++	7.408e-006	7.408e-006	0.4069	
NH3(aq)	6.125e-005	6.125e-005	1.043	
Na+	0.0009184	0.0009184	21.11	
O2(aq)	-0.2500	-0.2500	-7997.	

SO4--	0.0001577	0.0001577	15.14
SiO2(aq)	0.0003330	0.0003330	20.00
TcO4-	1.000e-008	1.000e-008	0.001620

Elemental composition	In fluid total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001068	0.001068	12.82		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.337		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.397e-005	0.7799		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.235		
Silicon	0.0003330	0.0003330	9.351		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		
Technetium	1.000e-008	1.000e-008	0.0009798		

Step # 0 Xi = 0.0000
Temperature = 25.0 C Pressure = 1.013 bars
pH = 7.123 log fO2 = -66.955
Eh = -0.1825 volts pe = -3.0853
Ionic strength = 0.002154
Activity of water = 0.999991
Solvent mass = 1.000000 kg
Solution mass = 1.000142 kg
Solution density = 1.013 g/cm3
Chlorinity = 0.000259 molal
Dissolved solids = 142 mg/kg sol'n

Minerals in system	moles	log moles	grams	volume (cm3)
Hematite	1.659e-006	-5.780	0.0002649	5.022e-005
Quartz	0.0002327	-3.633	0.01398	0.005279
TcO2:1.6H2O	5.975e-009	-8.224	9.488e-007	
(total)			0.01425	0.005329*

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
Na+	0.0009165	21.07	0.9500	-3.0602
HCO3-	0.0009149	55.82	0.9500	-3.0609
Ca++	0.0002722	10.91	0.8187	-3.6520
Cl-	0.0002594	9.194	0.9493	-3.6087
F-	0.0001753	3.330	0.9496	-3.7787
SO4--	0.0001518	14.58	0.8140	-3.9080
CO2(aq)	0.0001449	6.374	1.0000	-3.8391
SiO2(aq)	0.0001002	6.017	1.0000	-3.9993
K+	8.269e-005	3.232	0.9493	-4.1052
Mg++	4.490e-005	1.091	0.8232	-4.4323
N2(aq)	3.007e-005	0.8424	1.0000	-4.5218
Br-	1.385e-005	1.107	0.9493	-4.8810
Fe++	7.395e-006	0.4130	0.8187	-5.2179
Mn++	7.071e-006	0.3884	0.8187	-5.2374
CaSO4(aq)	3.557e-006	0.4842	1.0000	-5.4489
FeHCO3+	2.907e-006	0.3397	0.9500	-5.5588
CaHCO3+	2.270e-006	0.2295	0.9500	-5.6662
MgSO4(aq)	1.179e-006	0.1419	1.0000	-5.9286
NH4+	1.096e-006	0.01977	0.9489	-5.9829
NaHCO3(aq)	1.079e-006	0.09063	1.0000	-5.9670
NaSO4-	7.483e-007	0.08908	0.9500	-6.1482
CO3--	6.636e-007	0.03982	0.8152	-6.2668
MgHCO3+	3.671e-007	0.03132	0.9500	-6.4575
CaCO3(aq)	2.560e-007	0.02562	1.0000	-6.5917
CaF+	1.876e-007	0.01108	0.9500	-6.7490
FeCO3(aq)	1.759e-007	0.02038	1.0000	-6.7547
MnSO4(aq)	1.612e-007	0.02434	1.0000	-6.7926
HSiO3-	1.561e-007	0.01203	0.9500	-6.8289
MgF+	1.458e-007	0.006312	0.9500	-6.8586
OH-	1.413e-007	0.002403	0.9496	-6.8722
FeSO4(aq)	1.186e-007	0.01801	1.0000	-6.9260
MnCO3(aq)	1.037e-007	0.01192	1.0000	-6.9842
H+	7.905e-008	7.966e-005	0.9533	-7.1229
KSO4-	7.739e-008	0.01046	0.9500	-7.1336
MnHCO3+	4.032e-008	0.004675	0.9500	-7.4167
NaCl(aq)	3.582e-008	0.002093	1.0000	-7.4459
MnF+	2.730e-008	0.002018	0.9500	-7.5861
FeOH+	2.674e-008	0.001948	0.9500	-7.5950
FeF+	2.430e-008	0.001819	0.9500	-7.6366
MgCO3(aq)	1.905e-008	0.001606	1.0000	-7.7202
HF(aq)	1.847e-008	0.0003695	1.0000	-7.7335
NaF(aq)	1.457e-008	0.0006118	1.0000	-7.8365
CaCl+	1.164e-008	0.0008791	0.9500	-7.9563

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
TcO2	4.0056s/sat	Fluorite	-1.1724
TcO2:1.6H2O	0.0000 sat	SiO2(am)	-1.2857
Hematite	0.0000 sat	Magnetite	-1.3360
Quartz	0.0000 sat	Calcite	-1.4387
Ice	-0.1387	Aragonite	-1.5831
Tridymite	-0.1715	Cronstedtite-7A	-2.0953
Pyrite	-0.1929	Monohydrocalcite	-2.2724
Chalcedony	-0.2712	Ferrosilite	-2.4186
Goethite	-0.4802	Dolomite	-2.4738
Cristobalite(alp	-0.5505	Dolomite-ord	-2.4738
Coesite	-0.8100	Sellaite	-2.6054
Siderite	-0.9639	Magnesite	-2.6639
Rhodochrosite	-0.9826	Minnesotaite	-2.8942
Cristobalite(bet	-0.9940		

(only minerals with log Q/K > -3 listed)

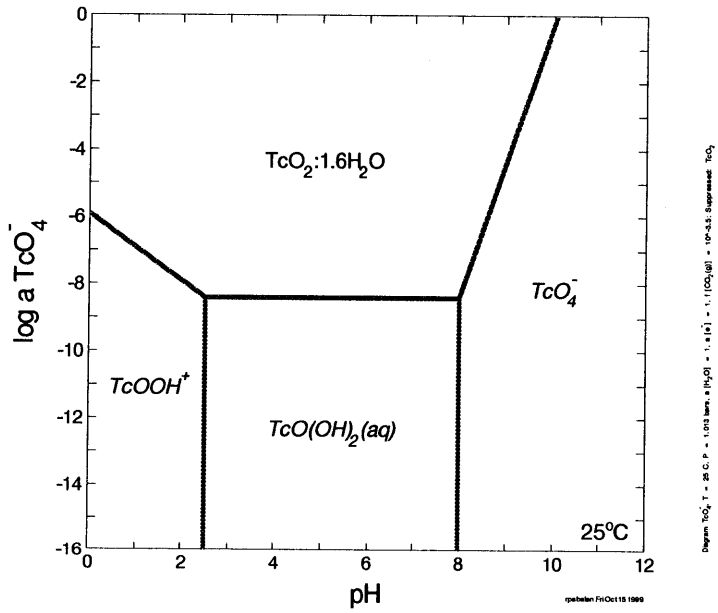
Gases	fugacity	log fug.
N2(g)	0.04620	-1.335
H2O(g)	0.02598	-1.585
CO2(g)	0.004264	-2.370
H2(g)	8.419e-009	-8.075
H2S(g)	1.632e-009	-8.787
NH3(g)	1.266e-010	-9.898
CH4(g)	1.709e-012	-11.767
HF(g)	9.679e-013	-12.014
CO(g)	1.121e-014	-13.950
HCl(g)	9.182e-018	-17.037
HBr(g)	1.302e-021	-20.885
SO2(g)	1.107e-021	-20.956
S2(g)	8.378e-028	-27.077
SiF4(g)	3.868e-033	-32.413
C2H4(g)	8.128e-038	-37.090
NO(g)	4.917e-050	-49.308
Br2(g)	1.157e-053	-52.937
Na(g)	4.594e-060	-59.338
Cl2(g)	4.070e-060	-59.390
K(g)	7.937e-062	-61.100
O2(g)	1.108e-067	-66.955
NO2(g)	2.442e-077	-76.612
Tc2O7(g)	2.012e-080	-79.696
Mg(g)	3.097e-098	-97.509
TcO(g)	9.315e-099	-98.031
Tc(g)	3.874e-113	-112.412
F2(g)	3.550e-113	-112.450
Ca(g)	2.772e-120	-119.557
C(g)	7.900e-123	-122.102
Si(g)	8.008e-155	-154.096

Original basis	total moles	In fluid moles	mg/kg	Sorbed moles	mg/kg
Br-	1.385e-005	1.385e-005	1.107		
Ca++	0.0002785	0.0002785	11.16		
Cl-	0.0002594	0.0002594	9.196		
F-	0.0001757	0.0001757	3.338		
Fe++	1.397e-005	1.065e-005	0.5946		
H+	0.0001379	0.0001445	0.1456		
H2O	55.51	55.51	9.999e+005		
HCO3-	0.001068	0.001068	65.13		
K+	8.277e-005	8.277e-005	3.236		
Mg++	4.662e-005	4.662e-005	1.133		
Mn++	7.408e-006	7.408e-006	0.4069		
NH3(aq)	6.125e-005	6.125e-005	1.043		
Na+	0.0009184	0.0009184	21.11		
O2(aq)	4.593e-005	4.511e-005	1.443		
SO4--	0.0001577	0.0001577	15.14		
SiO2(aq)	0.0003330	0.0001003	6.027		
TcO4-	1.000e-008	4.025e-009	0.0006520		

Elemental composition	total moles	In fluid moles	mg/kg	Sorbed moles	mg/kg
Bromine	1.385e-005	1.385e-005	1.107		
Calcium	0.0002785	0.0002785	11.16		
Carbon	0.001068	0.001068	12.82		
Chlorine	0.0002594	0.0002594	9.196		
Fluorine	0.0001757	0.0001757	3.338		
Hydrogen	111.0	111.0	1.119e+005		
Iron	1.397e-005	1.065e-005	0.5946		
Magnesium	4.662e-005	4.662e-005	1.133		
Manganese	7.408e-006	7.408e-006	0.4069		
Nitrogen	6.125e-005	6.125e-005	0.8578		
Oxygen	55.51	55.51	8.880e+005		
Potassium	8.277e-005	8.277e-005	3.236		
Silicon	0.0003330	0.0001003	2.817		
Sodium	0.0009184	0.0009184	21.11		
Sulfur	0.0001577	0.0001577	5.055		
Technetium	1.000e-008	4.025e-009	0.0003944		

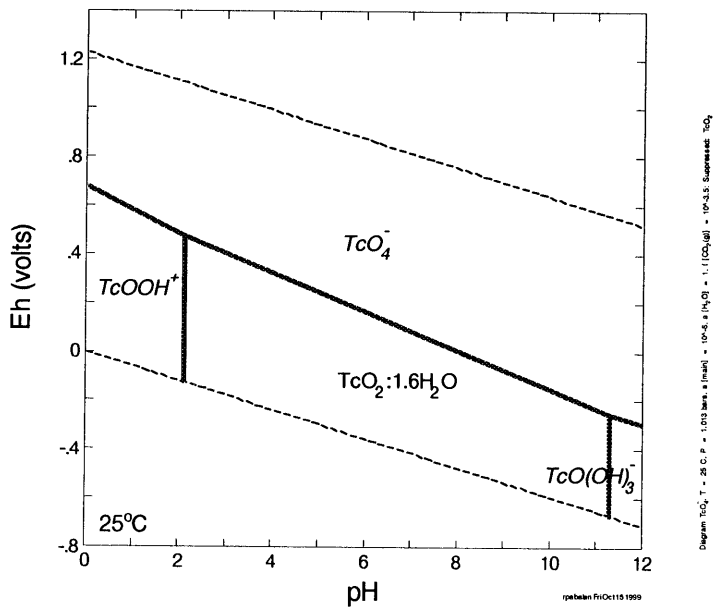
Note that the calculated technetium solubility (for TcO2.1.6H2O) is 4.0E-9 molal, which is consistent with published solubility values and much higher than 4.0E-13 molal, the solubility calculated previously for TcO2(solid). The value of 4.0e-9 molal will be used in subsequent sensitivity analyses with the TPA 3.2 code. This value is equal to 4.0E-7 kg/m^3, the concentration unit used in the TPA 3.2 input file.

The following is a solubility diagram as a function of pH for fixed Eh=0.0 v and fCO2 = $10^{-3.5}$ atm. For comparison, see page 269. To calculate this figure, TcO2 was suppressed.



Compared to the figure on page 269, the stability field of the technetium solid is much smaller.

The following is the calculated Eh-pH diagram for technetium at a TcO4- activity of 1.e-8 molal, pCO2=$10^{-3.5}$ atm (with TcO2 solid suppressed):



Entries into Scientific Notebook No. 278 for the period October 1, 1999, to December 31, 1999, have been made by

Roberto T. Pabalan / Date

No original entry into this Scientific Notebook has been removed.

The entries in this electronic scientific notebook #278 document activities conducted during the Period January 1, 2001, through September 13, 2001, under the Radionuclide Transport Key Technical Issue (Project Number 20-1402-871). No entries were made from January 1, 2001 through July 14, 2001.

July 15, 2001

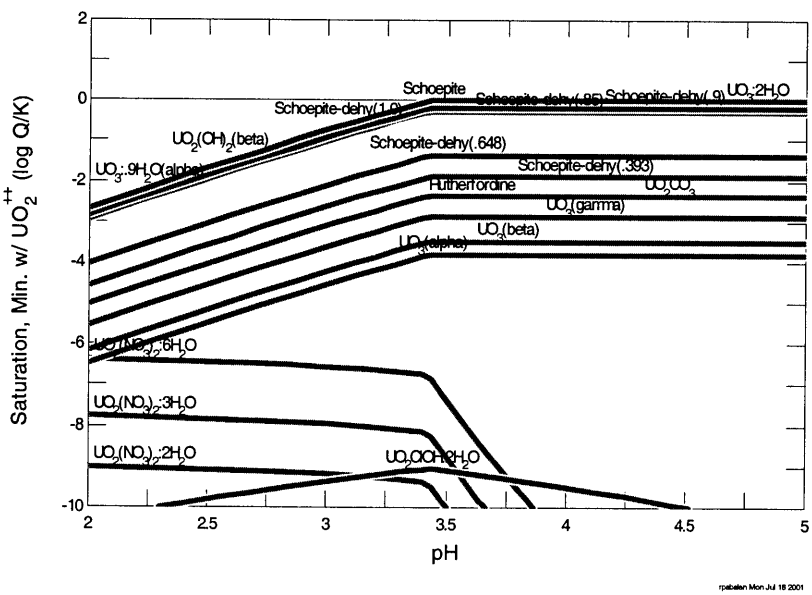
Speciationcalculations were conducted using Geochemist's Workbench (GWB) to determine the uranium mineral saturation as a function of pH of solutions at different uranium concentrations. The mineral saturations are plotted below and the GWB results are also listed below.

July 16, 2001

React> show
Temperature is 25 C
Thermo dataset: thermo.dat
Working directory: d:\
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
NO3- (swapped for NH3(aq)) total molarity = .1 (charge balance)
UO2++ total molarity = .05
CO2(g) (swapped for HCO3-) fugacity = .000316227766
H+ pH = 2
O2(g) (swapped for O2(aq)) fugacity = .2
Cl- total molality = 1e-8

Reactants:
Slide pH to 6



React> show

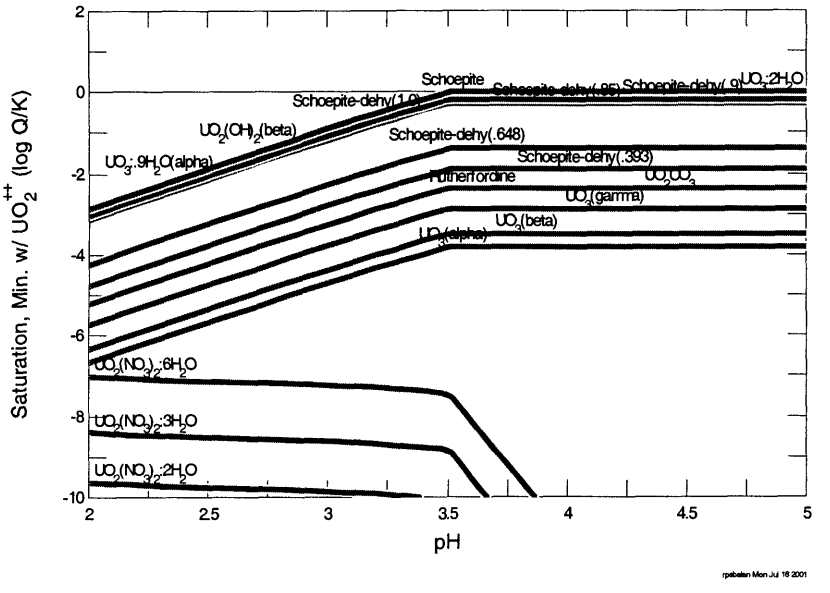
Temperature is 25 C
Thermo dataset: D:\Gwb\Gtdata\DTurner\thermo.dat
Working directory: d:\
Options: Debye-Huckel

Basis is:

H2O	1 kg solvent	
CO2(g) (swapped for HCO3-)	fugacity = .000316227766	
NO3- (swapped for NH3(aq))	total molarity = .05 (charge balance)	
O2(g) (swapped for O2(aq))	fugacity = .2	
UO2++	total molarity = .025	
H+	pH = 2	

Reactants:
Slide pH to 6

Other "show" options: type "show show"
React>

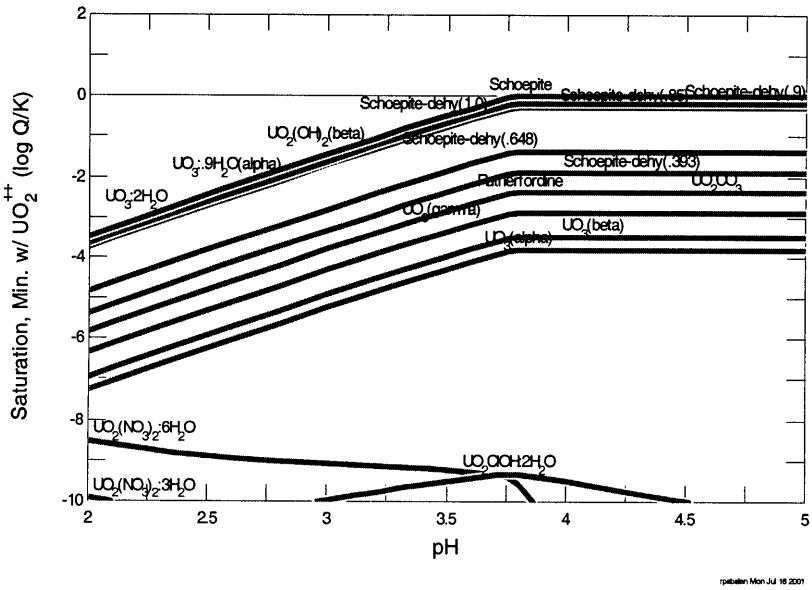


```
React> show
    Temperature is 25 C
    Thermo dataset:  D:\Gwb\Gtdata\DTurner\thermo.dat
    Working directory:  d:\
    Options: Debye-Huckel

    Basis is:
     H2O                        1 kg solvent
     NO3-  (swapped for NH3(aq))     total molarity = .0084 (charge balance)
     UO2++                        total molarity = .0042
     CO2(g)  (swapped for HCO3-)     fugacity = .000316227766
     H+                         pH = 2
     O2(g)  (swapped for O2(aq))     fugacity = .2
     Cl-                        total molality = 1e-8

    Reactants:
     Slide pH to 6

    Other "show" options: type "show show"
React>
```



```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  2.663                log fO2 =   -0.699
Eh =   1.0612 volts        pe =  17.9389
Ionic strength       =     0.146706
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.019764 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =        19381 mg/kg sol'n
```

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
NO3-	0.09751	5929.	0.7309	-1.1471
UO2++	0.04283	1.134e+004	0.3206	-1.8623
H+	0.002649	2.618	0.8207	-2.6628
UO2NO3+	0.002485	809.1	0.7512	-2.7289
(UO2)2OH+++	0.002026	1107.	0.0840	-3.7691
(UO2)2(OH)2++	0.0002886	162.5	0.3206	-4.0337
O2(aq)	0.0002528	7.932	1.0000	-3.5973
UO2OH+	5.217e-005	14.68	0.7512	-4.4069
CO2(aq)	1.074e-005	0.4636	1.0000	-4.9689
HNO3(aq)	7.720e-006	0.4770	1.0000	-5.1124
N2(aq)	4.307e-007	0.01183	1.0000	-6.3659
(UO2)3(OH)4++	4.258e-007	0.3666	0.3206	-6.8649
(UO2)3(OH)5+	1.840e-008	0.01615	0.7512	-7.8594

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Ice	-0.1387	Schoepite-dehy(.	-1.5535
UO3:2H2O	-1.3701	Schoepite-dehy(.	-1.6338
Schoepite	-1.3701	Schoepite-dehy(1	-1.6399
UO2(OH)2(beta)	-1.4825	Schoepite-dehy(.	-2.7431
UO3:.9H2O(alpha)	-1.5535		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
N2(g)	0.0006615	-3.179
CO2(g)	0.0003162	-3.500
NO2(g)	5.274e-012	-11.278
NO(g)	7.905e-018	-17.102
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	3.076e-061	-60.512
UO3(g)	3.275e-068	-67.485
UO2(g)	4.562e-121	-120.341
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489

```
   UO(g)                 2.523e-205    -204.598
   C2H4(g)               7.603e-239    -238.119
   U(g)                  3.273e-290    -289.485

                                        In fluid                  Sorbed
  Original basis  total moles     moles       mg/kg        moles       mg/kg
 ---------------------------------------------------------------------------
---
   H+                  -0.09999     -0.09999      -98.83
   H2O                    55.41        55.41  9.789e+005
   HCO3-             1.075e-005   1.075e-005      0.6431
   NH3(aq)               0.1000       0.1000       1670.
   O2(aq)                0.2003       0.2003       6284.
   UO2++                0.05000      0.05000  1.324e+004

  Elemental composition                 In fluid                  Sorbed
                   total moles     moles       mg/kg        moles       mg/kg
 ---------------------------------------------------------------------------
---
   Carbon            1.075e-005   1.075e-005      0.1266
   Hydrogen               111.0        111.0  1.097e+005
   Nitrogen              0.1000       0.1000       1374.
   Oxygen                 55.91        55.91  8.772e+005
   Uranium               0.05000      0.05000  1.167e+004
```

React> show
Temperature is 25 C
Thermo dataset: D:\Gwb\Gtdata\DTurner\the
Working directory: d:\
Options: Debye-Huckel

Basis is:
H2O 1 kg solv
CO2(g) (swapped for HCO3-) fugacity
NO3- (swapped for NH3(aq)) total mol
O2(g) (swapped for O2(aq)) fugacity
UO2++ total mol
H+ charge ba

No reactants specified.

Other "show" options: type "show show"
React>

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  3.426                log fO2 =   -0.699
Eh =   1.0160 volts        pe =  17.1754
Ionic strength       =     0.012414
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.001677 kg
Solution density     =     1.013    g/cm3
```

```
        Chlorinity          =    0.000000 molal
        Dissolved solids    =        1674 mg/kg sol'n

 No minerals in system.

 Aqueous species        molality    mg/kg sol'n    act. coef.     log act.
-------------------------------------------------------------------------
  NO3-                   0.008363         517.7      0.8896       -2.1285
  UO2++                  0.003674         990.4      0.6380       -2.6300
  H+                    0.0004131        0.4157      0.9071       -3.4262
  O2(aq)                0.0002528         8.075      1.0000       -3.5973
  (UO2)2(OH)2++         0.0001422         81.51      0.6380       -4.0422
  (UO2)2OH+++          7.796e-005         43.36      0.3691       -4.5410
  UO2OH+               4.346e-005         12.45      0.8929       -4.4111
  UO2NO3+              3.725e-005         12.35      0.8929       -4.4780
  CO2(aq)              1.074e-005        0.4720      1.0000       -4.9689
  (UO2)3(OH)4++        1.205e-006         1.056      0.6380       -6.1142
  (UO2)3(OH)5+         5.059e-007        0.4521      0.8929       -6.3451
  HNO3(aq)             1.389e-007      0.008739      1.0000       -6.8572
  UO2CO3(aq)           1.760e-008      0.005798      1.0000       -7.7546
  HCO3-                1.452e-008     0.0008842      0.8929       -7.8874
   (only species > 1e-8 molal listed)

 Mineral saturation states
                     log Q/K                            log Q/K
-------------------------------------------------------------------
  Ice                -0.1387        Schoepite-dehy(.   -0.8746
  UO3:2H2O           -0.6109        Schoepite-dehy(1   -0.8807
  Schoepite          -0.6109        Schoepite-dehy(.   -1.9839
  UO2(OH)2(beta)     -0.7233        Schoepite-dehy(.   -2.5019
  Schoepite-dehy(.   -0.7943        UO2CO3             -2.9645
  UO3:.9H2O(alpha)   -0.7943        Rutherfordine      -2.9848
   (only minerals with log Q/K > -3 listed)

 Gases                 fugacity      log fug.
-----------------------------------------------
  O2(g)                    0.2000       -0.699
  H2O(g)                  0.02598       -1.585
  CO2(g)                0.0003162       -3.500
  N2(g)                2.142e-007       -6.669
  NO2(g)               9.491e-014      -13.023
  NO(g)                1.422e-019      -18.847
  H2(g)                6.267e-042      -41.203
  CO(g)                6.189e-049      -48.208
  NH3(g)               5.535e-063      -62.257
  UO3(g)               1.881e-067      -66.726
  UO2(g)               2.620e-120     -119.582
  CH4(g)               3.890e-146     -145.410
  C(g)                 3.246e-190     -189.489
  UO(g)                1.450e-204     -203.839
  C2H4(g)              7.603e-239     -238.119
  U(g)                 1.880e-289     -288.726

                                     In fluid                  Sorbed
 Original basis  total moles     moles        mg/kg        moles        mg/kg
-------------------------------------------------------------------------------
---
```

```
   H+                -0.008389    -0.008389      -8.441
   H2O                   55.50        55.50  9.982e+005
   HCO3-            1.077e-005   1.077e-005      0.6563
   NH3(aq)            0.008400     0.008400       142.8
   O2(aq)              0.01705      0.01705       544.8
   UO2++              0.004200     0.004200       1132.

  Elemental composition                In fluid                  Sorbed
                  total moles     moles       mg/kg        moles       mg/kg
 ---------------------------------------------------------------------------
---
   Carbon           1.077e-005   1.077e-005      0.1292
   Hydrogen             111.0        111.0  1.117e+005
   Nitrogen           0.008400     0.008400       117.5
   Oxygen               55.54        55.54  8.872e+005
   Uranium            0.004200     0.004200       998.0
```

React> show
Temperature is 25 C
Thermo dataset: D:\Gwb\Gtdata\DTurner\thermo.dat
Working directory: d:\
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
CO2(g) (swapped for HCO3-) fugacity = .000316227766
NO3- (swapped for NH3(aq)) total molarity = .05
O2(g) (swapped for O2(aq)) fugacity = .2
UO2++ total molarity = .025
H+ charge balance

No reactants specified.

Other "show" options: type "show show"
React>

```
          Step #      0              Xi = 0.0000
          Temperature =  25.0 C      Pressure =  1.013 bars
          pH =  2.899                log fO2 =   -0.699
          Eh =   1.0472 volts        pe =  17.7024
          Ionic strength       =     0.073913
          Activity of water    =     1.000000
          Solvent mass         =     1.000000 kg
          Solution mass        =     1.009892 kg
          Solution density     =     1.013    g/cm3
          Chlorinity           =     0.000000 molal
          Dissolved solids     =         9795 mg/kg sol'n

  No minerals in system.

  Aqueous species        molality    mg/kg sol'n    act. coef.     log act.
 ---------------------------------------------------------------------------
   NO3-                   0.04918         3020.       0.7823       -1.4148
```

UO2++	0.02181	5832.	0.4040	-2.0549
H+	0.001495	1.492	0.8433	-2.8993
(UO2)2OH+++	0.0008791	484.9	0.1375	-3.9177
UO2NO3+	0.0008132	267.4	0.7955	-3.1892
(UO2)2(OH)2++	0.0002804	159.4	0.4040	-3.9458
O2(aq)	0.0002528	8.009	1.0000	-3.5973
UO2OH+	5.451e-005	15.49	0.7955	-4.3629
CO2(aq)	1.074e-005	0.4681	1.0000	-4.9689
HNO3(aq)	2.418e-006	0.1509	1.0000	-5.6166
(UO2)3(OH)4++	7.888e-007	0.6859	0.4040	-6.4966
(UO2)3(OH)5+	6.996e-008	0.06201	0.7955	-7.2545
N2(aq)	4.224e-008	0.001172	1.0000	-7.3742

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Ice	-0.1387	UO3:.9H2O(alpha)	-1.2730
Schoepite	-1.0896	Schoepite-dehy(.	-1.3533
UO3:2H2O	-1.0896	Schoepite-dehy(1	-1.3594
UO2(OH)2(beta)	-1.2020	Schoepite-dehy(.	-2.4626
Schoepite-dehy(.	-1.2730	Schoepite-dehy(.	-2.9806

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	6.489e-005	-4.188
NO2(g)	1.652e-012	-11.782
NO(g)	2.476e-018	-17.606
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	9.634e-062	-61.016
UO3(g)	6.247e-068	-67.204
UO2(g)	8.701e-121	-120.060
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	4.813e-205	-204.318
C2H4(g)	7.603e-239	-238.119
U(g)	6.244e-290	-289.205

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-0.04999	-0.04999	-49.89		
H2O	55.46	55.46	9.893e+005		
HCO3-	1.075e-005	1.075e-005	0.6497		
NH3(aq)	0.05000	0.05000	843.2		
O2(aq)	0.1003	0.1003	3177.		
UO2++	0.02500	0.02500	6685.		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg

```
 ---------------------------------------------------------------------------
---
   Carbon          1.075e-005   1.075e-005      0.1279
   Hydrogen             111.0        111.0  1.108e+005
   Nitrogen           0.05000      0.05000       693.5
   Oxygen               55.71        55.71  8.826e+005
   Uranium            0.02500      0.02500       5892.
```

July 16, 2001

Preparation of Uranyl-Loaded Clinoptilolite Samples for EXAFS Analysis

Objective: Clinoptilolite with sorbed uranium(VI) will be prepared for EXAFS analysis at Argonne National Laboratory. Two types of samples will be prepared, one with ion-exchanged uranium and another with surface complexed uranium.

Procedure:

a) Prepared 0.0.025 m uranium nitrate solution

Weighed 6.2750 g $^{238}UO_2(NO_3)_2 \cdot 6H_2O$ reagent (Mallinkrodt Lot # 8640KCAP) in weighing boat. Transferred the solids into a 500-mL polypropylene bottle.

Tared 500 g of deionized water into the bottle. The bottle was capped, then shaken to dissolve the uranium solid.

The pH of the 0.025 m uranium solution was measured to be 2.93. The value calculated using GWB is 2.90. The measured pH of standard solutions are 3.98 (pH 4 standard) and 5.03 (pH 5 standard). No recalibration of the pH electroded was considered necessary because the measured values of the pH standards are close to their stated values.

b) Reacted 0.025 m uranium solution with clinoptilolite

1030 hrs: Weighed and transferred 1.0017 g of clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe). This batch of clinoptilolite has undergone purification by various methods discussed in Pabalan (1994). The same batch was used to prepare homoionic clinoptilolite for use in a SwRI IR&D project on ion exchange being conducted by P. Bertetti and R. Pabalan. Label the bottle CDV*SC*1.

c) Prepared 1×10^{-4} m uranium nitrate solution

Weighed 1.2555 g $^{238}UO_2(NO_3)_2 \cdot 6H_2O$ reagent (Mallinkrodt Lot # 8640KCAP) in weighing boat. Transferred the solids into a 500-mL polypropylene bottle. (Weighing boat had a tared weight of 0.0025 g after the transfer).

Tared 500 g of deionized water into the bottle. The bottle was capped, then shaken to dissolve the uranium solid.

The pH of the 5×10^{-3} m uranium solution was measured to be 3.39. The value calculated using GWB is 3.378 (SEE INSERT BELOW). The measured pH of standard solutions are 3.98 (pH 4 standard) and 5.01 (pH 5 standard). No recalibration of the pH electroded was considered necessary because the measured values of the pH standards are close to their stated values.

Transferred 20.04 g of the 5×10^{-3} molal uranium solution to a 1-L polypropylene bottle and diluted to 1000 g with deionized water to make a 1-L 1×10^{-4} molal uranium solution. The pH of the 1×10^{-4} m uranium solution was measured to be 4.48. The value calculated using GWB is 4.459 (SEE INSERT BELOW). The measured pH of standard solutions are 4.00 (pH 4 standard) and 5.01 (pH 5 standard).

d) Transferred 29.97 g of the 1×10^{-4} m uranium solution for later analysis of uranium concentration

e) Reacted $1x10^{-4}$ m uranium solution with clinoptilolite

1500 hrs: Weighed and transferred 0.009712 g of clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe). Weighing boat had residual weighed of 0.0009 g. Label the bottle CDV*IE*1.

f) Adjusted pH of CDV*SC*1 (clinoptilolite/$1x10^{-4}$ m uranium solution mixture) using 1 M NaHCO3 solution prepared by Alka Jain on 4/10/01 as documented in lab notebook 420 (p. 63):

Initial pH after adding clinoptilolite = 4.95
pH after adding 100 microliter of NaHCO3 solution = 5.79
pH after adding another 100 microliter of NaHCO3 solution = 6.35.

g) The mixtures were placed on the gyratory shaker.

GWB RESULT FOR $5X10^{-3}$ MOLAL URANYL NITRATE SOLUTION

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  3.378                log fO2 =   -0.699
Eh =   1.0189 volts        pe =  17.2241
Ionic strength       =     0.014792
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.001993 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =         1989 mg/kg sol'n

No minerals in system.

Aqueous species        molality    mg/kg sol'n    act. coef.     log act.
-----------------------------------------------------------------------------
NO3-                    0.009949        615.6       0.8812       -2.0572
UO2++                   0.004386        1182.       0.6163       -2.5682
H+                     0.0004651       0.4678       0.9013       -3.3776
O2(aq)                 0.0002528        8.072       1.0000       -3.5973
(UO2)2(OH)2++          0.0001565        89.64       0.6163       -4.0158
(UO2)2OH+++           9.998e-005        55.58       0.3421       -4.4660
UO2NO3+               5.107e-005        16.92       0.8851       -4.3449
UO2OH+                4.520e-005        12.95       0.8851       -4.3979
CO2(aq)               1.074e-005       0.4718       1.0000       -4.9689
(UO2)3(OH)4++         1.222e-006        1.071       0.6163       -6.1233
(UO2)3(OH)5+          4.468e-007       0.3992       0.8851       -6.4029
HNO3(aq)              1.831e-007      0.01152       1.0000       -6.7372
UO2CO3(aq)            1.622e-008     0.005341       1.0000       -7.7900
HCO3-                 1.309e-008    0.0007972       0.8851       -7.9360
 (only species > 1e-8 molal listed)

Mineral saturation states
                     log Q/K                          log Q/K
-----------------------------------------------------------------
Ice                 -0.1387        Schoepite-dehy(.  -0.9100
UO3:2H2O            -0.6463        Schoepite-dehy(1  -0.9161
Schoepite           -0.6463        Schoepite-dehy(.  -2.0193
UO2(OH)2(beta)      -0.7587        Schoepite-dehy(.  -2.5373
Schoepite-dehy(.    -0.8297        UO2CO3            -2.9999
UO3:.9H2O(alpha)    -0.8297
 (only minerals with log Q/K > -3 listed)

Gases                 fugacity      log fug.
-----------------------------------------------
```

O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	3.722e-007	-6.429
NO2(g)	1.251e-013	-12.903
NO(g)	1.875e-019	-18.727
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	7.296e-063	-62.137
UO3(g)	1.734e-067	-66.761
UO2(g)	2.415e-120	-119.617
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	1.336e-204	-203.874
C2H4(g)	7.603e-239	-238.119
U(g)	1.733e-289	-288.761

Original basis	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
H+	-0.009989	-0.009989	-10.05		
H2O	55.50	55.50	9.978e+005		
HCO3-	1.077e-005	1.077e-005	0.6560		
NH3(aq)	0.01000	0.01000	170.0		
O2(aq)	0.02025	0.02025	646.8		
UO2++	0.005000	0.005000	1347.		

Elemental composition	total moles	In fluid moles	In fluid mg/kg	Sorbed moles	Sorbed mg/kg
Carbon	1.077e-005	1.077e-005	0.1291		
Hydrogen	111.0	111.0	1.117e+005		
Nitrogen	0.01000	0.01000	139.8		
Oxygen	55.55	55.55	8.870e+005		
Uranium	0.005000	0.005000	1188.		

GWB RESULT FOR 1.X10^{-4} MOLAL URANYL NITRATE SOLUTION

```
Step #     0              Xi = 0.0000
Temperature =  25.0 C     Pressure =  1.013 bars
pH =  4.459               log fO2 =   -0.699
Eh =   0.9549 volts       pe =  16.1426
Ionic strength      =     0.000277
Activity of water   =     1.000000
Solvent mass        =     1.000000 kg
Solution mass       =     1.000054 kg
Solution density    =     1.013    g/cm3
Chlorinity          =     0.000000 molal
Dissolved solids    =           54 mg/kg sol'n
```

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
O2(aq)	0.0002528	8.088	1.0000	-3.5973
NO3-	0.0002000	12.40	0.9809	-3.7074
UO2++	6.735e-005	18.18	0.9264	-4.2049
H+	3.541e-005	0.03568	0.9815	-4.4590
UO2OH+	1.135e-005	3.259	0.9810	-4.9532
CO2(aq)	1.074e-005	0.4727	1.0000	-4.9689
(UO2)2(OH)2++	8.070e-006	4.633	0.9264	-5.1263
(UO2)3(OH)5+	1.266e-006	1.134	0.9810	-5.9057
(UO2)2OH+++	2.610e-007	0.1454	0.8424	-6.6579
(UO2)3(OH)4++	2.117e-007	0.1859	0.9264	-6.7075
HCO3-	1.425e-007	0.008693	0.9810	-6.8546
UO2CO3(aq)	5.447e-008	0.01798	1.0000	-7.2638
(UO2)4(OH)7+	2.826e-008	0.03389	0.9810	-7.5572

UO2NO3+	2.380e-008	0.007902	0.9810	-7.6317

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
UO3:2H2O	-0.1201	Schoepite-dehy(1	-0.3899
Schoepite	-0.1201	Schoepite-dehy(.	-1.4931
Ice	-0.1387	Schoepite-dehy(.	-2.0111
UO2(OH)2(beta)	-0.2325	UO2CO3	-2.4737
Schoepite-dehy(.	-0.3035	Rutherfordine	-2.4940
UO3:.9H2O(alpha)	-0.3035	UO3(gamma)	-2.9941
Schoepite-dehy(.	-0.3838		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	1.281e-012	-11.893
NO2(g)	2.321e-016	-15.634
NO(g)	3.478e-022	-21.459
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	1.353e-065	-64.869
UO3(g)	5.823e-067	-66.235
UO2(g)	8.112e-120	-119.091
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	4.487e-204	-203.348
C2H4(g)	7.603e-239	-238.119
U(g)	5.821e-289	-288.235

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-0.0001891	-0.0001891	-0.1905		
H2O	55.51	55.51	9.999e+005		
HCO3-	1.094e-005	1.094e-005	0.6676		
NH3(aq)	0.0002000	0.0002000	3.406		
O2(aq)	0.0006528	0.0006528	20.89		
UO2++	0.0001000	0.0001000	27.00		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	1.094e-005	1.094e-005	0.1314		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	0.0002000	0.0002000	2.801		
Oxygen	55.51	55.51	8.881e+005		
Uranium	0.0001000	0.0001000	23.80		

July 18, 2001

The pH of CDV*SC*1 was adjusted by Alka Jain to 6.37 by adding 75 microliters of 1 M NaHCO3 solution.

The mixtures were placed back on the gyratory shaker.

July 20, 2001

The pH of CDV*SC*1 was measured by Alka Jain to be 6.64. No pH adjustment was made.

The mixtures were placed back on the gyratory shaker.

July 24, 2001

1030 hrs. – The pH of CDV*SC*1 and CDV*IE*1 were measured. The measured pH of standards solutions are 4.03 and 7.01 (for pH4 and pH7 standards, respectively). The measured pH of CDV*IE*1 is 3.09, whereas the measured pH of CDV*SC*1 is 6.71. No pH adjustment was made.

The mixtures were placed back on the gyratory shaker.

July 26, 2001

1015 hrs. – The pH of CDV*SC*1 and CDV*IE*1 were measured. The measured pH of standards solutions are 4.00 and 7.00 (for pH4 and pH7 standards, respectively). The measured pH of CDV*IE*1 is 3.09, whereas the measured pH of CDV*SC*1 is 6.78.

1025 hrs. – The pH of CDV*SC*1 was adjusted by adding microliters of 1-M HNO3 solution (prepared on 04/13/01, see labnotebook 309, page 254).

+ 25 microliter 1-M HNO3: measured pH=6.64
+ additional 25 microliter 1-M HNO3: measured pH=6.46
+ additional 20 microliter 1-M HNO3: measured pH=6.41

The mixtures were placed back on the gyratory shaker. The cap on the CDV*SC*1 bottle was loosened to allow CO2(g) in.

1545 hrs. – The pH of CDV*SC*1 was measured to be 6.65. The pH of the standard solutions were measured to be 4.03 and 7.02 (pH4 and pH7 standards, respectively). The pH of CDV*SC*1 was adjusted by adding 25 microliters of 1-M HNO3 solution (prepared on 04/13/01, see labnotebook 309, page 254). The pH after adjustment was 6.32.

The mixture was placed back on the gyratory shaker. The cap on the CDV*SC*1 bottle was loosened to allow CO2(g) in.

July 27, 2001

0915 hrs. – The pH of CDV*SC*1 and CDV*IE*1 were measured. The measured pH of standards solutions are 4.01 and 7.02 (for pH4 and pH7 standards, respectively). The measured pH of CDV*IE*1 is 3.10, whereas the measured pH of CDV*SC*1 is 6.78. The pH of CDV*SC*1 was adjusted by adding 50 microliters of 1-M HNO3 solution (prepared on 04/13/01, see labnotebook 309, page 254). The pH after adjustment was 5.80.

The mixture was placed back on the gyratory shaker. The cap on the CDV*SC*1 bottle was loosened to allow CO2(g) in.

1050 hrs. – The pH of CDV*SC*1 was measured to be 6.30. The mixture was placed back on the gyratory shaker. The cap on the CDV*SC*1 bottle was loosened to allow CO2(g) in.

1240 hrs. – The pH of CDV*SC*1 was measured to be 6.05. The mixture was placed back on the gyratory shaker. The cap on the CDV*SC*1 bottle was loosened to allow CO2(g) in.

July 30, 2001

0845 hrs. – The pH of CDV*SC*1 and CDV*IE*1 were measured. The measured pH of standards solutions are 3.97 and 6.97 (for pH4 and pH7 standards, respectively). The measured pH of CDV*IE*1 is 3.07, whereas the measured pH of CDV*SC*1 is 6.30. The mixtures was placed back on the gyratory shaker. The cap on the CDV*SC*1 bottle was loosened to allow CO2(g) in.

1520 hrs. – Two 50-mL aliquots of the CDV*SC*1 solution was taken and transferred into 50-mL PP bottles (labeled CDV*SC*1*F*1 and CDV*SC*1*F*2). Two 50-mL aliquots of the CDV*IE*1 solution was taken and transferred into 50-mL PP bottles (labeled CDV*IE*1*F*1 and CDV*IE*1*F*2).

About 500 mL of the CDV*SC*1 solution were decanted into a 500 mL polypropylene bottle. The pH of the decanted solution was measured to be 6.39.

The remaining CDV*SC*1 mixture was filtered (Fisher brand P5 qualitative glass fiber filter paper, with medium porosity and medium flow rate, Cat. No. 09-801F, 19 cm diameter). The filtrant was collected into an Erlenmeyer flask, and later transferred back into the original 1-L polypropylene bottle. No colloidal or particulate material was observed in the filtrant. The wet clinoptilolite paste was scooped from the filter paper using a Teflon-lined spatula and transferred into a plastic vial. After transferring most of the clinoptilolite, the vial was capped and labeled for later shipment to Argonne National Laboratory.

The CDV*IE*1 mixture was shaken to suspend the clinoptilolite, and the mixture filtered through the Fisher filter paper. The filtrant was collected into a 500-mL polypropylene bottle. The wet clinoptilolite paste was scooped from the filter paper using a spatula and transferred into a plastic vial. The vial was later capped and labeled for later shipment to ANL.

July 31, 2001

The following table shows the estimated activity of U-238 in the uranyl-loaded samples. According to Lynne Soderholm (Argonne National Laboratory), each individual sample must have a specific activity less than 2 nCi/g for unregulated transportation and for unregulated receipt by ANL staff.

U specific activity (Ci/g)	Sample	U mol/L	solution ppm	U grams in 1 liter	% sorbed	U g on solid	U activity on solid (Ci/g)
3.40E-07	SC*1	1.00E-04	2.38E+01	2.38E-05	100	2.38E-05	8.09E-12
	IE*1	0.025	5.95E+03	5.95E-03	100	5.95E-03	2.02E-09
	IE*1	0.025	5.95E+03	5.95E-03	25	1.49E-03	5.06E-10

The first two results are for conservative assumptions of 100% sorption of U-238 by the 1 g of clinoptilolite reacted with the U solution. The third result is based on an assumption, also conservative, that 25 % of the U ion exchanged with the clinoptilolite at low pH.

The two uranyl-loaded samples were sent to L. Soderholm (ANL) via Fedex.

Entries into Scientific Notebook No. 278 for the period January 1, 2001 through September 13, 2001, have been made by

R T Pabalan 9/14/01

Roberto T. Pabalan / Date

No original entry into this Scientific Notebook has been removed.

R T Pabalan 9/14/01

Roberto T. Pabalan / Date

R. Palalan SN
update copy. 3/26/2002

The entries in this electronic scientific notebook #278 document activities conducted during the Period September 14, 2001, through March 17, 2002, under the Radionuclide Transport Key Technical Issue (Project Number 20-1402-871). 278E

October 23, 2001

The following is an email from Richard Reeder dated October 19, 2001 regarding the EXAFS analysis that he did on the U-loaded clinoptilolite samples I prepared:

Hi Bobby,

Sorry it's taken me so long to get to the clinoptilolite samples that you prepared for us to run at the BESSRC beamline at the APS. Results are similar to but also different from previous samples run at NSLS. You had not indicated which sample was prepared at which pH, and so I figured it would be a nice test for me to determine this by the splitting in the equatorial shell. In the attachment I show three fits: two for CDV*SC*1 and one for CDV*IE*1. Just looking at the FT magnitude for IE (clinopie), you will see obvious splitting of the peak from the equatorial shells. So I initially assumed this was run at pH 6, and the other at pH 3. The splitting in this sample was confirmed by the fitting:

Shell	N	R(A)	D-W(A**2)	
U-Oax	2*	1.78	0.002	* fixed N
U-Oeq1	3.2	2.36	0.005	
U-Oeq2	2.0	2.49	0.002	
MS	3	3.60	0.005	

In the FT mag(s) for SC there appears to be a single peak for the equatorial shells but with a very distinct shoulder on the high-R side. I fitted this first with a single eq. shell and then with a split shell:

Shell	N	R(A)	D-W(A**2)
U-Oax	2*	1.80	0.002
U-Oeq	5.3	2.41	0.008
MS	4	3.62	0.005

Shell	N	R(A)	D-W(A**2)
U-Oax	2*	1.80	0.002
U-Oeq1	3.1	2.36	0.003
U-Oeq2	2.1	2.49	0.002
MS	4	3.62	0.005

Comparing these latter fits (single vs split shell), it's clear from the fit plots that the single-shell fit misses a feature on the high-R side of the eq. peak. (In all fits, I seem to be missing some very

weak contribution between the eq. peak and the multiple-scattering peak. I still working on this using various possible shells (including Si); nothing so far gives a contribution with reasonable fit values.) By allowing for two equatorial oxygen shells the fit is visibly improved, with a slight but I think statistically significant improvement in chi**2.

Now, if you compare the fit results for IE and the split-shell SC, they are virtually identical, despite the difference in appearance of the FT mags. In other samples, I've seen that the appearance of the equatorial peak is very sensitive to splitting, because the two components are interfering destructively. In fact the amplitude of the equatorial peak is also very sensitive to the axial oxygen contribution. And I notice a slight difference in U-Oax between the two samples, but possibly within error.

So what's similar to the previous samples? Recall that I got a best fit earlier to the pH 3 sample with a weak additional oxygen at about 2.2 A in the equatorial shell. We didn't attach much significance to that at the time.

The differences are more striking, however, particularly in the distances. In the earlier fits of the pH 6.4 sample, the eq. shells were at 2.28 and 2.45 A. With the most recent samples, the shells are at 2.36 and 2.49 A. The degree of splitting is smaller and the actual U-O distances are longer. Errors on these distances are 0.02-0.03 A, so these differences are real. Of course, this assumes that the equatorial components are best fitted with two shells. I tried fitting with three shells, and let it go. In all cases two of the three shells become identical (i.e., two shells seems reasonable).

The data quality from the APS is very good; I'm very happy about that. But we noticed immediately that the uranium loading on the samples was very high. We had to detune by 50% and add 8 layers of Al foil over the detector to reduce counts to an acceptable level! The high U produced a fluorescence peak that was greatly above the Rb and Sr peaks, so much so that we could ignore them and run at the L3 -edge. The samples were wet when we opened them, and the mounting went fine. I think I mentioned that they sat for better than a week in Lynne's lab before we ran them. I'm wondering what could be responsible for the differences. High loading? Extra time? Sample prep or Na exchange? I recall reading in some pub that at high uranyl loading, different or multiple sites were occuppied. And on some sorbents, splitting is always observed. Could the high loading have caused sorption in the exchange sites in the cavities? Any thoughts about this? I'm going to have my postdoc fit the spectra independently just to see if he finds something different, but I don't expect anything much different.

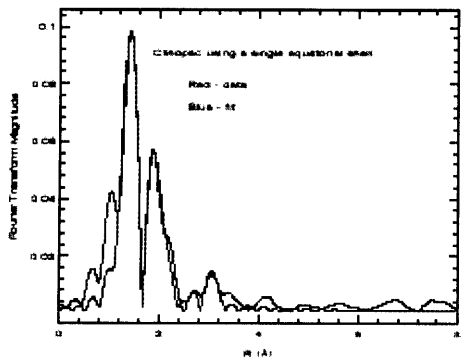
Best,

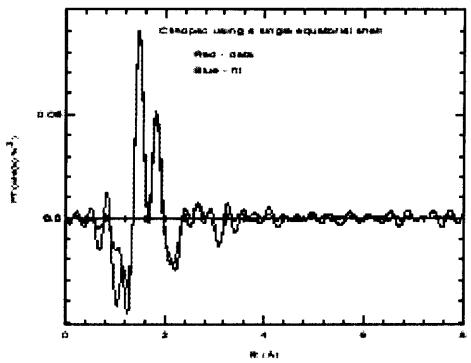
Rich

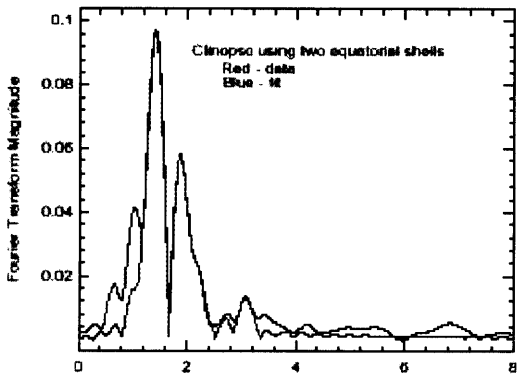
(See attached file: Clinop_APS.pdf)

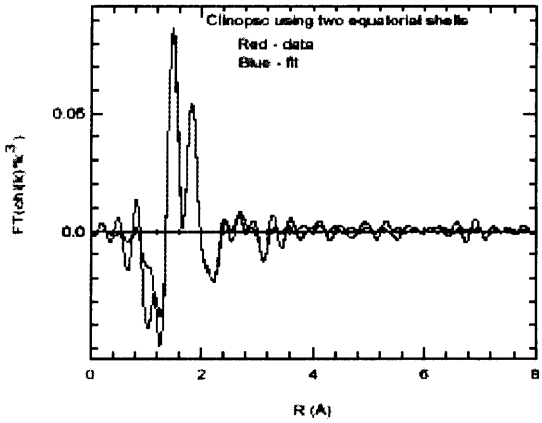
Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240









Entries into Scientific Notebook No. 278 for the period September 14, 2001 through March 17, 2002, have been made by

Roberto T. Pabalan / Date

No original entry into this Scientific Notebook has been removed.

Roberto T. Pabalan / Date

The entries in this electronic scientific notebook #278 document activities conducted during the Period March 26, 2002, through September 5, 2002, under the Radionuclide Transport Key Technical Issue (Project Number 20-1402-871).

July 9, 2002

1. Calculate pH of 0.025 m uranyl nitrate solution (for comparison with previous result on page 323 of ESN#278.

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
CO2(g) (swapped for HCO3-) fugacity = .000316227766
NO3- (swapped for NH3(aq)) total molality = .05
UO2++ total molality = .025
H+ charge balance

No reactants specified.

```
Output file:

          Step #     0              Xi = 0.0000
          Temperature =  25.0 C     Pressure =  1.013 bars
          pH =  2.899               log fO2 =   -0.699
          Eh =   1.0472 volts       pe =  17.7024
          Ionic strength      =     0.073913
          Activity of water   =     1.000000
          Solvent mass        =     1.000000 kg
          Solution mass       =     1.009892 kg
          Solution density    =     1.013    g/cm3
          Chlorinity          =     0.000000 molal
          Dissolved solids    =         9795 mg/kg sol'n
          Rock mass           =     0.000000 kg

 No minerals in system.

 Aqueous species       molality    mg/kg sol'n    act. coef.     log act.
-------------------------------------------------------------------------
  NO3-                   0.04918         3020.       0.7823       -1.4148
  UO2++                  0.02181         5832.       0.4040       -2.0549
  H+                    0.001496         1.493       0.8433       -2.8992
  (UO2)2OH+++          0.0008790         484.8       0.1375       -3.9178
  UO2NO3+              0.0008132         267.4       0.7955       -3.1892
  (UO2)2(OH)2++        0.0002803         159.3       0.4040       -3.9460
  O2(aq)               0.0002528         8.009       1.0000       -3.5973
  UO2OH+              5.450e-005         15.49       0.7955       -4.3630
  CO2(aq)             1.074e-005        0.4681       1.0000       -4.9689
  HNO3(aq)            2.418e-006        0.1509       1.0000       -5.6165
  (UO2)3(OH)4++       7.883e-007        0.6855       0.4040       -6.4969
  UO2(OH)2(aq)        2.685e-007       0.08083       1.0000       -6.5711
  (UO2)3(OH)5+        6.991e-008       0.06196       0.7955       -7.2549
  N2(aq)              4.226e-008      0.001172       1.0000       -7.3741
   (only species > 1e-8 molal listed)

 Mineral saturation states
                      log Q/K                          log Q/K
-------------------------------------------------------------
  Ice                -0.1387        UO3:.9H2O(alpha)  -1.2732
  Schoepite          -1.0898        Schoepite-dehy(.  -1.3535
  UO3:2H2O           -1.0898        Schoepite-dehy(1  -1.3596
  UO2(OH)2(beta)     -1.2022        Schoepite-dehy(.  -2.4628
```

```
 Schoepite-dehy(.  -1.2732        Schoepite-dehy(.  -2.9808
  (only minerals with log Q/K > -3 listed)

Gases                 fugacity      log fug.
-----------------------------------------------
 O2(g)                    0.2000       -0.699
 H2O(g)                  0.02598       -1.585
 CO2(g)                0.0003162       -3.500
 N2(g)                 6.491e-005      -4.188
 NO2(g)                1.652e-012     -11.782
 NO(g)                 2.476e-018     -17.606
 H2(g)                 6.267e-042     -41.203
 CO(g)                 6.189e-049     -48.208
 NH3(g)                9.635e-062     -61.016
 UO3(g)                6.245e-068     -67.204
 UO2(g)                8.699e-121    -120.061
 CH4(g)                3.890e-146    -145.410
 C(g)                  3.246e-190    -189.489
 UO(g)                 4.812e-205    -204.318
 C2H4(g)               7.603e-239    -238.119
 U(g)                  6.242e-290    -289.205

                                      In fluid                  Sorbed
Original basis  total moles     moles       mg/kg        moles       mg/kg
-------------------------------------------------------------------------------
 H+                -0.04999     -0.04999      -49.89
 H2O                  55.46        55.46  9.893e+005
 HCO3-            1.075e-005   1.075e-005      0.6497
 NH3(aq)             0.05000      0.05000       843.2
 O2(aq)               0.1003       0.1003       3177.
 UO2++               0.02500      0.02500       6685.

Elemental composition                 In fluid                  Sorbed
                 total moles     moles       mg/kg        moles       mg/kg
-------------------------------------------------------------------------------
 Carbon           1.075e-005   1.075e-005      0.1279
 Hydrogen             111.0        111.0  1.108e+005
 Nitrogen            0.05000      0.05000       693.5
 Oxygen               55.71        55.71  8.826e+005
 Uranium             0.02500      0.02500       5892.
```

Note: Calculated pH = 2.90, consistent with previous result.

2. Calculate pH of 1.E-3 molal uranyl nitrate solution

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = .002
CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = .001
H+ charge balance

No reactants specified.

Output file:

```
Step #     0              Xi = 0.0000
Temperature =  25.0 C     Pressure =  1.013 bars
pH =  3.820               log fO2 =   -0.699
Eh =   0.9927 volts       pe =  16.7819
Ionic strength      =     0.002914
```

```
Activity of water   =    1.000000
Solvent mass        =    1.000000 kg
Solution mass       =    1.000411 kg
Solution density    =    1.013    g/cm3
Chlorinity          =    0.000000 molal
Dissolved solids    =        411 mg/kg sol'n
Rock mass           =    0.000000 kg
```

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
NO3-	0.001997	123.8	0.9417	-2.7256
UO2++	0.0008323	224.7	0.7904	-3.1818
O2(aq)	0.0002528	8.085	1.0000	-3.5973
H+	0.0001599	0.1611	0.9469	-3.8198
(UO2)2(OH)2++	5.538e-005	31.78	0.7904	-4.3588
UO2OH+	2.859e-005	8.204	0.9426	-4.5694
CO2(aq)	1.074e-005	0.4726	1.0000	-4.9689
(UO2)2OH+++	9.483e-006	5.281	0.5914	-5.2511
UO2NO3+	2.504e-006	0.8312	0.9426	-5.6270
UO2(OH)2(aq)	1.390e-006	0.4225	1.0000	-5.8569
(UO2)3(OH)5+	9.833e-007	0.8798	0.9426	-6.0330
(UO2)3(OH)4++	8.065e-007	0.7079	0.7904	-6.1955
HCO3-	3.403e-008	0.002075	0.9426	-7.4938
UO2CO3(aq)	3.025e-008	0.009978	1.0000	-7.5193
HNO3(aq)	1.419e-008	0.0008941	1.0000	-7.8479
(UO2)4(OH)7+	1.218e-008	0.01460	0.9426	-7.9399

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Ice	-0.1387	Schoepite-dehy(.	-0.6393
UO3:2H2O	-0.3756	Schoepite-dehy(1	-0.6454
Schoepite	-0.3756	Schoepite-dehy(.	-1.7486
UO2(OH)2(beta)	-0.4880	Schoepite-dehy(.	-2.2666
Schoepite-dehy(.	-0.5590	UO2CO3	-2.7292
UO3:.9H2O(alpha)	-0.5590	Rutherfordine	-2.7495

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	2.236e-009	-8.650
NO2(g)	9.697e-015	-14.013
NO(g)	1.453e-020	-19.838
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	5.655e-064	-63.248
UO3(g)	3.233e-067	-66.490
UO2(g)	4.504e-120	-119.346
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	2.491e-204	-203.604
C2H4(g)	7.603e-239	-238.119
U(g)	3.232e-289	-288.491

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-0.001989	-0.001989	-2.004		
H2O	55.51	55.51	9.996e+005		
HCO3-	1.081e-005	1.081e-005	0.6591		
NH3(aq)	0.002000	0.002000	34.05		
O2(aq)	0.004253	0.004253	136.0		
UO2++	0.001000	0.001000	269.9		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	1.081e-005	1.081e-005	0.1297		
Hydrogen	111.0	111.0	1.118e+005		
Nitrogen	0.002000	0.002000	28.00		
Oxygen	55.52	55.52	8.879e+005		
Uranium	0.001000	0.001000	237.9		

3. Calculate amount of HNO3 needed to bring the pH down to about 3.2

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = .002 (charge balance)
CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = .001
H+ pH = 3.2

No reactants specified.

Output file:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  3.200                log fO2 =   -0.699
Eh =   1.0294 volts        pe =  17.4017
Ionic strength       =     0.003633
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000449 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =          449 mg/kg sol'n
Rock mass            =     0.000000 kg

No minerals in system.

Aqueous species        molality    mg/kg sol'n    act. coef.     log act.
-----------------------------------------------------------------------------
NO3-                    0.002646         164.0       0.9355       -2.6063
UO2++                  0.0009735         262.7       0.7708       -3.1247
H+                     0.0006699        0.6749       0.9419       -3.2000
O2(aq)                 0.0002528         8.085       1.0000       -3.5973
CO2(aq)               1.074e-005        0.4726       1.0000       -4.9689
UO2OH+                7.877e-006         2.260       0.9367       -5.1320
(UO2)2(OH)2++         4.256e-006         2.442       0.7708       -5.4841
UO2NO3+               3.783e-006         1.256       0.9367       -5.4505
(UO2)2OH+++           3.130e-006         1.743       0.5594       -5.7567
UO2(OH)2(aq)          9.134e-008       0.02776       1.0000       -7.0393
HNO3(aq)              7.784e-008      0.004903       1.0000       -7.1088
  (only species > 1e-8 molal listed)

Mineral saturation states
                    log Q/K                          log Q/K
-------------------------------------------------------------
Ice                -0.1387        Schoepite-dehy(.  -1.7414
UO3:2H2O           -1.5580        Schoepite-dehy(.  -1.8217
Schoepite          -1.5580        Schoepite-dehy(1  -1.8278
UO2(OH)2(beta)     -1.6704        Schoepite-dehy(.  -2.9310
UO3:.9H2O(alpha)   -1.7414
  (only minerals with log Q/K > -3 listed)

Gases                 fugacity      log fug.
-----------------------------------------------
O2(g)                   0.2000       -0.699
H2O(g)                 0.02598       -1.585
CO2(g)               0.0003162       -3.500
N2(g)                6.725e-008       -7.172
NO2(g)               5.318e-014      -13.274
NO(g)                7.970e-020      -19.099
H2(g)                6.267e-042      -41.203
CO(g)                6.189e-049      -48.208
NH3(g)               3.101e-063      -62.508
```

```
UO3(g)              2.125e-068     -67.673
UO2(g)              2.959e-121    -120.529
CH4(g)              3.890e-146    -145.410
C(g)                3.246e-190    -189.489
UO(g)               1.637e-205    -204.786
C2H4(g)             7.603e-239    -238.119
U(g)                2.124e-290    -289.673

                                   In fluid                  Sorbed
Original basis  total moles     moles       mg/kg        moles       mg/kg
-------------------------------------------------------------------------------
H+               -0.001989    -0.001989      -2.004
H2O                  55.51        55.51  9.995e+005
HCO3-            1.075e-005   1.075e-005      0.6558
NH3(aq)            0.002650     0.002650       45.11
O2(aq)             0.005553     0.005553       177.6
UO2++              0.001000     0.001000       269.9

Elemental composition              In fluid                  Sorbed
                total moles     moles       mg/kg        moles       mg/kg
-------------------------------------------------------------------------------
Carbon           1.075e-005   1.075e-005      0.1291
Hydrogen             111.0        111.0  1.118e+005
Nitrogen           0.002650     0.002650       37.10
Oxygen               55.52        55.52  8.879e+005
Uranium            0.001000     0.001000       237.9
```

Note: Total moles of Nitrogen = 0.002650 (equal to total moles of NO3-). Initial moles of NO3- is 2.E-3. Therefore, moles on NO3- needed to adjust the pH to 3.2 is 0.00265 minus 0.002 = 0.00065 moles per liter. For a 450 mL of solution, need 0.000292 moles. Using a 1.0 molal HNO3 solution, need to add a little less than 300 microliters. (300 microliters of 1 molal HNO3 would have 0.0003 moles NO3-).

4. Calculate pH of 2.E-4 molal uranyl nitrate solution.

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = 4e-4
CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = 2e-4

No reactants specified.

Output file:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  4.256                log fO2 =   -0.699
Eh =   0.9669 volts        pe =  16.3456
Ionic strength       =     0.000564
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000094 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =           94 mg/kg sol'n
Rock mass            =     0.000000 kg

No minerals in system.
```

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
NO3-	0.0003999	24.79	0.9731	-3.4099
O2(aq)	0.0002528	8.088	1.0000	-3.5973
UO2++	0.0001471	39.73	0.8976	-3.8792
H+	5.692e-005	0.05736	0.9743	-4.2561
UO2OH+	1.518e-005	4.358	0.9733	-4.8304
(UO2)2(OH)2++	1.466e-005	8.414	0.8976	-4.8808
CO2(aq)	1.074e-005	0.4727	1.0000	-4.9689
UO2(OH)2(aq)	2.081e-006	0.6328	1.0000	-5.6816
(UO2)3(OH)5+	1.171e-006	1.048	0.9733	-5.9434
(UO2)2OH+++	7.865e-007	0.4381	0.7848	-6.2095
(UO2)3(OH)4++	3.196e-007	0.2807	0.8976	-6.5423
UO2NO3+	1.007e-007	0.03344	0.9733	-7.0086
HCO3-	9.000e-008	0.005491	0.9733	-7.0575
UO2CO3(aq)	4.529e-008	0.01494	1.0000	-7.3440
(UO2)4(OH)7+	2.171e-008	0.02604	0.9733	-7.6750

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Ice	-0.1387	Schoepite-dehy(.	-0.4640
UO3:2H2O	-0.2003	Schoepite-dehy(1	-0.4701
Schoepite	-0.2003	Schoepite-dehy(.	-1.5733
UO2(OH)2(beta)	-0.3127	Schoepite-dehy(.	-2.0913
Schoepite-dehy(.	-0.3837	UO2CO3	-2.5539
UO3:.9H2O(alpha)	-0.3837	Rutherfordine	-2.5742

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	1.283e-011	-10.892
NO2(g)	7.346e-016	-15.134
NO(g)	1.101e-021	-20.958
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	4.284e-065	-64.368
UO3(g)	4.841e-067	-66.315
UO2(g)	6.744e-120	-119.171
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	3.730e-204	-203.428
C2H4(g)	7.603e-239	-238.119
U(g)	4.839e-289	-288.315

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-0.0003891	-0.0003891	-0.3922		
H2O	55.51	55.51	9.999e+005		
HCO3-	1.088e-005	1.088e-005	0.6637		
NH3(aq)	0.0004000	0.0004000	6.812		
O2(aq)	0.001053	0.001053	33.68		
UO2++	0.0002000	0.0002000	54.00		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	1.088e-005	1.088e-005	0.1307		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	0.0004000	0.0004000	5.602		
Oxygen	55.51	55.51	8.881e+005		
Uranium	0.0002000	0.0002000	47.60		

5. Calculate amount of HNO3 needed to adjust the pH of a 2.E-4 molal uranyl nitrate solution from 4.26 to 3.2.

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp

Options: Debye-Huckel

Basis is:

H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = 4e-4 (charge balance)
CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = 2e-4
H+ pH = 3.2

No reactants specified.

Output file:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  3.200                log fO2 =   -0.699
Eh =   1.0294 volts        pe =  17.4017
Ionic strength       =     0.001250
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000134 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =          134 mg/kg sol'n
Rock mass            =     0.000000 kg

No minerals in system.

Aqueous species        molality    mg/kg sol'n    act. coef.     log act.
-------------------------------------------------------------------------
NO3-                   0.001052         65.25       0.9607       -2.9952
H+                    0.0006551        0.6602       0.9631       -3.2000
O2(aq)                0.0002528         8.087       1.0000       -3.5973
UO2++                 0.0001973         53.26       0.8537       -3.7736
CO2(aq)              1.074e-005        0.4727       1.0000       -4.9689
UO2OH+               1.723e-006        0.4945       0.9611       -5.7809
UO2NO3+              3.380e-007        0.1122       0.9611       -6.4883
(UO2)2(OH)2++        1.936e-007        0.1111       0.8537       -6.7818
(UO2)2OH+++          1.257e-007       0.07002       0.7018       -7.0544
HNO3(aq)             3.179e-008      0.002003       1.0000       -7.4977
UO2(OH)2(aq)         2.050e-008      0.006233       1.0000       -7.6882
 (only species > 1e-8 molal listed)

Mineral saturation states
                    log Q/K                          log Q/K
-------------------------------------------------------------
Ice                -0.1387        UO3:.9H2O(alpha)  -2.3903
Schoepite          -2.2069        Schoepite-dehy(.  -2.3903
UO3:2H2O           -2.2069        Schoepite-dehy(.  -2.4706
UO2(OH)2(beta)     -2.3193        Schoepite-dehy(1  -2.4767
 (only minerals with log Q/K > -3 listed)

Gases                 fugacity      log fug.
-----------------------------------------------
O2(g)                    0.2000       -0.699
H2O(g)                 0.02598       -1.585
CO2(g)               0.0003162       -3.500
N2(g)               1.122e-008       -7.950
NO2(g)              2.172e-014      -13.663
NO(g)               3.255e-020      -19.487
H2(g)               6.267e-042      -41.203
CO(g)               6.189e-049      -48.208
NH3(g)              1.267e-063      -62.897
UO3(g)              4.769e-069      -68.322
UO2(g)              6.642e-122     -121.178
CH4(g)              3.890e-146     -145.410
C(g)                3.246e-190     -189.489
UO(g)               3.674e-206     -205.435
C2H4(g)             7.603e-239     -238.119
U(g)                4.767e-291     -290.322

                                   In fluid                  Sorbed
```

Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-0.0003892	-0.0003892	-0.3923		
H2O	55.51	55.51	9.999e+005		
HCO3-	1.075e-005	1.075e-005	0.6559		
NH3(aq)	0.001053	0.001053	17.93		
O2(aq)	0.002358	0.002358	75.46		
UO2++	0.0002000	0.0002000	54.00		

Elemental composition		In fluid		Sorbed	
	total moles	moles	mg/kg	moles	mg/kg
Carbon	1.075e-005	1.075e-005	0.1291		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	0.001053	0.001053	14.75		
Oxygen	55.51	55.51	8.881e+005		
Uranium	0.0002000	0.0002000	47.60		

Note: Total moles of NO3- = 0.001052 (equal to total moles of NO3-). Initial moles of NO3- is 4.E-4. Therefore, moles on NO3- needed to adjust the pH to 3.2 is 0.001052 minus 0.0004 = 0.000652 moles per liter. For a 450 mL of solution, need 0.0002934 moles. Using a 1.0 molal HNO3 solution, need to add a little less than 300 microliters. (300 microliters of 1 molal HNO3 would have 0.0003 moles NO3-).

6. Calculate the pH of a 2.5E-5 molal uranyl nitrate solution.

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = 5e-5
CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = 2.5e-5
H+ charge balance

No reactants specified.

Output file:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  4.801                log fO2 =   -0.699
Eh =   0.9347 volts        pe =  15.8005
Ionic strength       =     0.000065
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000024 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =           24 mg/kg sol'n
Rock mass            =     0.000000 kg
```

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
O2(aq)	0.0002528	8.088	1.0000	-3.5973
NO3-	5.000e-005	3.100	0.9906	-4.3051
H+	1.595e-005	0.01608	0.9908	-4.8012
UO2++	1.290e-005	3.484	0.9632	-4.9057
CO2(aq)	1.074e-005	0.4728	1.0000	-4.9689
UO2OH+	4.924e-006	1.413	0.9907	-5.3118

UO2(OH)2(aq)	2.410e-006	0.7328	1.0000	-5.6179
(UO2)2(OH)2++	1.488e-006	0.8544	0.9632	-5.8436
(UO2)3(OH)5+	5.090e-007	0.4556	0.9907	-6.2973
HCO3-	3.102e-007	0.01893	0.9907	-6.5124
UO2CO3(aq)	5.244e-008	0.01731	1.0000	-7.2803
(UO2)3(OH)4++	3.758e-008	0.03300	0.9632	-7.4413
(UO2)2OH+++	2.086e-008	0.01162	0.9193	-7.7173
(UO2)4(OH)7+	1.094e-008	0.01311	0.9907	-7.9652

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Schoepite	-0.1366	Schoepite-dehy(.	-0.4003
UO3:2H2O	-0.1366	Schoepite-dehy(1	-0.4064
Ice	-0.1387	Schoepite-dehy(.	-1.5096
UO2(OH)2(beta)	-0.2490	Schoepite-dehy(.	-2.0276
Schoepite-dehy(.	-0.3200	UO2CO3	-2.4902
UO3:.9H2O(alpha)	-0.3200	Rutherfordine	-2.5105

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	1.689e-014	-13.772
NO2(g)	2.665e-017	-16.574
NO(g)	3.994e-023	-22.399
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	1.554e-066	-65.808
UO3(g)	5.606e-067	-66.251
UO2(g)	7.809e-120	-119.107
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	4.320e-204	-203.365
C2H4(g)	7.603e-239	-238.119
U(g)	5.604e-289	-288.252

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-3.889e-005	-3.889e-005	-0.03920		
H2O	55.51	55.51	1.000e+006		
HCO3-	1.111e-005	1.111e-005	0.6779		
NH3(aq)	5.000e-005	5.000e-005	0.8515		
O2(aq)	0.0003528	0.0003528	11.29		
UO2++	2.500e-005	2.500e-005	6.751		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	1.111e-005	1.111e-005	0.1334		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	5.000e-005	5.000e-005	0.7003		
Oxygen	55.51	55.51	8.881e+005		
Uranium	2.500e-005	2.500e-005	5.951		

7. Calculate amount of HNO3 needed to adjust the pH of a 2.5E-5 molal uranyl nitrate solution from 4.80 to 3.2.

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = 5e-5 (charge balance)

CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = 2.5e-5
H+ pH = 3.2

No reactants specified.

Output:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  3.200                log fO2 =   -0.699
Eh =   1.0294 volts        pe =  17.4017
Ionic strength       =     0.000724
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000065 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =          65 mg/kg sol'n
Rock mass            =     0.000000 kg
```

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
NO3-	0.0006994	43.37	0.9697	-3.1686
H+	0.0006497	0.6548	0.9711	-3.2000
O2(aq)	0.0002528	8.088	1.0000	-3.5973
UO2++	2.474e-005	6.679	0.8853	-4.6596
CO2(aq)	1.074e-005	0.4727	1.0000	-4.9689
UO2OH+	2.220e-007	0.06372	0.9699	-6.6669
UO2NO3+	2.921e-008	0.009698	0.9699	-7.5477
HNO3(aq)	2.132e-008	0.001344	1.0000	-7.6711

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Ice	-0.1387		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	5.047e-009	-8.297
NO2(g)	1.457e-014	-13.837
NO(g)	2.183e-020	-19.661
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	8.496e-064	-63.071
UO3(g)	6.200e-070	-69.208
UO2(g)	8.637e-123	-122.064
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	4.777e-207	-206.321
C2H4(g)	7.603e-239	-238.119
U(g)	6.198e-292	-291.208

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-3.925e-005	-3.925e-005	-0.03956		
H2O	55.51	55.51	9.999e+005		
HCO3-	1.075e-005	1.075e-005	0.6559		
NH3(aq)	0.0006995	0.0006995	11.91		
O2(aq)	0.001652	0.001652	52.85		
UO2++	2.500e-005	2.500e-005	6.750		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	1.075e-005	1.075e-005	0.1291		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	0.0006995	0.0006995	9.797		
Oxygen	55.51	55.51	8.881e+005		

```
Uranium          2.500e-005   2.500e-005       5.950
```

Note: Total moles of nitrogen = 0.0006995 (equal to total moles of NO3-). Initial moles of NO3- is 2.5E-5. Therefore, moles on NO3- needed to adjust the pH to 3.2 is 0.0006995 minus 0.000025 = 0.0006745 moles per liter. For a 450 mL of solution, need 0.000304 moles. Using a 1.0 molal HNO3 solution, need to add a little more than 300 microliters. (300 microliters of 1 molal HNO3 would have 0.000300 moles NO3-).

8. Calculate how much NaHCO3 or NaOH to add to a 2.5E-5 molal uranyl nitrate solution to adjust the pH from 4.80 to 6.3. Do by trial and error: balance on H+, change amount of Na+ until pH of 6.3 results from the calculation.

The following is the input file that worked:

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
O2(g) (swapped for O2(aq)) fugacity = .2
NO3- (swapped for NH3(aq)) total molality = 5e-5
CO2(g) (swapped for HCO3-) fugacity = .000316227766
UO2++ total molality = 2.5e-5
H+ charge balance
Na+ total molality = 6e-5

Reactants:
Fix fugacity of CO2(g)

Output file:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  6.306                log fO2 =   -0.699
Eh =   0.8457 volts        pe =  14.2954
Ionic strength       =     0.000063
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000027 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =           27 mg/kg sol'n
Rock mass            =     0.000000 kg

                        moles        moles         grams          cm3
Reactants             remaining     reacted       reacted       reacted
-----------------------------------------------------------------------------
 CO2(g)              -- fixed fugacity buffer --

No minerals in system.

Aqueous species       molality    mg/kg sol'n    act. coef.     log act.
-----------------------------------------------------------------------------
 O2(aq)               0.0002528         8.088       1.0000       -3.5973
 Na+                 6.000e-005         1.379       0.9908       -4.2259
 NO3-                5.000e-005         3.100       0.9907       -4.3051
 UO2(OH)2(aq)        1.144e-005         3.477       1.0000       -4.9417
 CO2(aq)             1.074e-005        0.4728       1.0000       -4.9689
 HCO3-               9.923e-006        0.6055       0.9908       -5.0074
 (UO2)2CO3(OH)3-     3.308e-006         2.154       0.9908       -5.4845
```

(UO2)3(OH)5+	1.699e-006	1.521	0.9908	-5.7737
UO2OH+	7.301e-007	0.2096	0.9908	-6.1406
H+	4.986e-007	0.0005025	0.9909	-6.3062
UO2CO3(aq)	2.488e-007	0.08212	1.0000	-6.6041
(UO2)4(OH)7+	1.732e-007	0.2077	0.9908	-6.7654
UO2++	5.978e-008	0.01614	0.9636	-7.2395
(UO2)2(OH)2++	3.272e-008	0.01878	0.9636	-7.5012
UO2(OH)3-	2.893e-008	0.009288	0.9908	-7.5427
OH-	2.066e-008	0.0003514	0.9908	-7.6889

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
UO3:2H2O	0.5396s/sat	Ice	-0.1387
Schoepite	0.5396s/sat	Schoepite-dehy(.	-0.8334
UO2(OH)2(beta)	0.4272s/sat	Schoepite-dehy(.	-1.3514
Schoepite-dehy(.	0.3562s/sat	UO2CO3	-1.8140
UO3:.9H2O(alpha)	0.3562s/sat	Rutherfordine	-1.8343
Schoepite-dehy(.	0.2759s/sat	UO3(gamma)	-2.3344
Schoepite-dehy(1	0.2698s/sat	UO3(beta)	-2.9366

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	1.651e-017	-16.782
NO2(g)	8.332e-019	-18.079
NO(g)	1.249e-024	-23.904
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
UO3(g)	2.660e-066	-65.575
NH3(g)	4.859e-068	-67.313
Na(g)	1.305e-078	-77.884
UO2(g)	3.705e-119	-118.431
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	2.050e-203	-202.688
C2H4(g)	7.603e-239	-238.119
U(g)	2.659e-288	-287.575

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-8.577e-005	-8.577e-005	-0.08644		
H2O	55.51	55.51	1.000e+006		
HCO3-	2.423e-005	2.423e-005	1.479		
NH3(aq)	5.000e-005	5.000e-005	0.8515		
Na+	6.000e-005	6.000e-005	1.379		
O2(aq)	0.0003528	0.0003528	11.29		
UO2++	2.500e-005	2.500e-005	6.751		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	2.423e-005	2.423e-005	0.2910		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	5.000e-005	5.000e-005	0.7003		
Oxygen	55.51	55.51	8.881e+005		
Sodium	6.000e-005	6.000e-005	1.379		
Uranium	2.500e-005	2.500e-005	5.951		

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  6.328                log fO2 =   -0.699
Eh =   0.8444 volts        pe =  14.2739
Ionic strength       =     0.000061
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
Solution mass        =     1.000020 kg
Solution density     =     1.013    g/cm3
Chlorinity           =     0.000000 molal
Dissolved solids     =           21 mg/kg sol'n
Rock mass            =     0.000007 kg
```

Reactants	moles remaining	moles reacted	grams reacted	cm3 reacted
CO2(g)	-- fixed fugacity buffer --			

Minerals in system	moles	log moles	grams	volume (cm3)
Schoepite	2.069e-005	-4.684	0.006665	0.001368
(total)			0.006665	0.001368

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
O2(aq)	0.0002528	8.088	1.0000	-3.5973
Na+	6.000e-005	1.379	0.9910	-4.2258
NO3-	5.000e-005	3.100	0.9909	-4.3050
CO2(aq)	1.074e-005	0.4728	1.0000	-4.9689
HCO3-	1.043e-005	0.6361	0.9910	-4.9858
UO2(OH)2(aq)	3.301e-006	1.004	1.0000	-5.4813
H+	4.744e-007	0.0004781	0.9911	-6.3278
(UO2)2CO3(OH)3-	2.896e-007	0.1885	0.9910	-6.5421
UO2OH+	2.005e-007	0.05756	0.9910	-6.7018
UO2CO3(aq)	7.183e-008	0.02371	1.0000	-7.1437
(UO2)3(OH)5+	3.889e-008	0.03481	0.9910	-7.4141
OH-	2.171e-008	0.0003692	0.9909	-7.6673
UO2++	1.562e-008	0.004217	0.9644	-7.8222

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
UO3:2H2O	0.0000 sat	Schoepite-dehy(1	-0.2698
Schoepite	0.0000 sat	Schoepite-dehy(.	-1.3730
UO2(OH)2(beta)	-0.1124	Schoepite-dehy(.	-1.8910
Ice	-0.1387	UO2CO3	-2.3536
Schoepite-dehy(.	-0.1834	Rutherfordine	-2.3739
UO3:.9H2O(alpha)	-0.1834	UO3(gamma)	-2.8740
Schoepite-dehy(.	-0.2637		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	1.496e-017	-16.825
NO2(g)	7.930e-019	-18.101
NO(g)	1.189e-024	-23.925
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
UO3(g)	7.679e-067	-66.115
NH3(g)	4.625e-068	-67.335
Na(g)	1.372e-078	-77.863
UO2(g)	1.070e-119	-118.971
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
UO(g)	5.917e-204	-203.228
C2H4(g)	7.603e-239	-238.119
U(g)	7.676e-289	-288.115

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-8.847e-005	-4.708e-005	-0.04745		
H2O	55.51	55.51	1.000e+006		
HCO3-	2.153e-005	2.153e-005	1.314		
NH3(aq)	5.000e-005	5.000e-005	0.8515		

```
  Na+              6.000e-005   6.000e-005       1.379
  O2(aq)            0.0003528    0.0003528       11.29
  UO2++            2.500e-005   4.305e-006       1.163

 Elemental composition                In fluid                  Sorbed
                 total moles     moles       mg/kg        moles       mg/kg
-----------------------------------------------------------------------------
  Carbon          2.153e-005   2.153e-005      0.2586
  Hydrogen             111.0        111.0  1.119e+005
  Nitrogen        5.000e-005   5.000e-005      0.7003
  Oxygen               55.51        55.51  8.881e+005
  Sodium          6.000e-005   6.000e-005       1.379
  Uranium         2.500e-005   4.305e-006       1.025
```

Note: The amount of Na+ (as NaOH or NaHCO3) needed was 6.0E-5 moles per liter.

a) **For a 450 mL solution, need 0.0000270 moles. Using a 0.1 m NaHCO3 solution, need 270 microliters.**
b) **For a 250 mL solution, need 0.0000150 moles. Using a 0.1 m NaHCO3 solution, need 150 microliters.**

9. Calculate pH of 0.001 m HNO3 solution

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
NO3- (swapped for NH3(aq)) total molality = .001
O2(g) (swapped for O2(aq)) fugacity = .2
CO2(g) (swapped for HCO3-) fugacity = .000316227766
H+ charge balance

No reactants specified.

Output file:

```
        Step #     0              Xi = 0.0000
        Temperature =  25.0 C     Pressure =  1.013 bars
        pH =  3.015               log fO2 =   -0.699
        Eh =   1.0404 volts       pe =  17.5869
        Ionic strength      =     0.001000
        Activity of water   =     1.000000
        Solvent mass        =     1.000000 kg
        Solution mass       =     1.000077 kg
        Solution density    =     1.013    g/cm3
        Chlorinity          =     0.000000 molal
        Dissolved solids    =          77 mg/kg sol'n
        Rock mass           =     0.000000 kg

 No minerals in system.

 Aqueous species       molality    mg/kg sol'n    act. coef.     log act.
-----------------------------------------------------------------------------
  H+                    0.001000         1.008      0.9666       -3.0148
  NO3-                  0.001000         62.00      0.9646       -3.0157
  O2(aq)               0.0002528         8.088      1.0000       -3.5973
  CO2(aq)             1.074e-005        0.4727      1.0000       -4.9689
  HNO3(aq)            4.646e-008      0.002927      1.0000       -7.3329
   (only species > 1e-8 molal listed)

 Mineral saturation states
                   log Q/K                          log Q/K
```

```
-----------------------------------------------------------------
  Ice                -0.1387
   (only minerals with log Q/K > -3 listed)

 Gases                 fugacity      log fug.
-----------------------------------------------
  O2(g)                    0.2000      -0.699
  H2O(g)                  0.02598      -1.585
  CO2(g)                0.0003162      -3.500
  N2(g)                 2.396e-008      -7.621
  NO2(g)                3.174e-014     -13.498
  NO(g)                 4.757e-020     -19.323
  H2(g)                 6.267e-042     -41.203
  CO(g)                 6.189e-049     -48.208
  NH3(g)                1.851e-063     -62.733
  CH4(g)                3.890e-146    -145.410
  C(g)                  3.246e-190    -189.489
  C2H4(g)               7.603e-239    -238.119

                                      In fluid                  Sorbed
 Original basis  total moles     moles       mg/kg        moles       mg/kg
-------------------------------------------------------------------------------
  H+              1.075e-005   1.075e-005     0.01083
  H2O                  55.51        55.51  9.999e+005
  HCO3-           1.075e-005   1.075e-005      0.6557
  NH3(aq)           0.001000     0.001000       17.03
  O2(aq)            0.002253     0.002253       72.08

 Elemental composition                In fluid                  Sorbed
                 total moles     moles       mg/kg        moles       mg/kg
-------------------------------------------------------------------------------
  Carbon          1.075e-005   1.075e-005      0.1291
  Hydrogen             111.0        111.0  1.119e+005
  Nitrogen          0.001000     0.001000       14.01
  Oxygen               55.51        55.51  8.881e+005
```

10. Calculate how much NaOH or NaHCO3 to add to the 0.001 m HNO3 solution to bring the pH up to 8.2. Do by trial and error. Fix pH at 8.2; change inpout Na+ concentration until calculated pH is close to 8.2.

The following input file works:

Temperature is 25 C
Thermo dataset: C:\Program Files\Gwb\Gtdata\thermo.com.v8.r6+.dat
Working directory: d:\temp
Options: Debye-Huckel

Basis is:
H2O 1 kg solvent
NO3- (swapped for NH3(aq)) total molality = .001
O2(g) (swapped for O2(aq)) fugacity = .2
CO2(g) (swapped for HCO3-) fugacity = .000316227766
H+ charge balance
Na+ total molality = .002

No reactants specified.

Output file:

```
Step #      0              Xi = 0.0000
Temperature =  25.0 C      Pressure =  1.013 bars
pH =  8.281                log fO2 =   -0.699
Eh =   0.7288 volts        pe =  12.3206
Ionic strength       =     0.002008
Activity of water    =     1.000000
Solvent mass         =     1.000000 kg
```

```
Solution mass       =    1.000182 kg
Solution density    =    1.013    g/cm3
Chlorinity          =    0.000000 molal
Dissolved solids    =         182 mg/kg sol'n
Rock mass           =    0.000000 kg
```

No minerals in system.

Aqueous species	molality	mg/kg sol'n	act. coef.	log act.
Na+	0.001997	45.91	0.9516	-2.7211
NO3-	0.001000	61.99	0.9509	-3.0219
HCO3-	0.0009751	59.49	0.9516	-3.0325
O2(aq)	0.0002528	8.087	1.0000	-3.5973
CO2(aq)	1.074e-005	0.4727	1.0000	-4.9689
CO3--	1.013e-005	0.6078	0.8206	-5.0802
NaHCO3(aq)	2.515e-006	0.2112	1.0000	-5.5995
OH-	2.031e-006	0.03453	0.9512	-5.7140
NaCO3-	5.428e-008	0.004504	0.9516	-7.2869

(only species > 1e-8 molal listed)

Mineral saturation states

	log Q/K		log Q/K
Ice	-0.1387		

(only minerals with log Q/K > -3 listed)

Gases	fugacity	log fug.
O2(g)	0.2000	-0.699
H2O(g)	0.02598	-1.585
CO2(g)	0.0003162	-3.500
N2(g)	6.829e-019	-18.166
NO2(g)	1.695e-019	-18.771
NO(g)	2.540e-025	-24.595
H2(g)	6.267e-042	-41.203
CO(g)	6.189e-049	-48.208
NH3(g)	9.883e-069	-68.005
Na(g)	3.938e-075	-74.405
CH4(g)	3.890e-146	-145.410
C(g)	3.246e-190	-189.489
C2H4(g)	7.603e-239	-238.119

		In fluid		Sorbed	
Original basis	total moles	moles	mg/kg	moles	mg/kg
H+	-0.001001	-0.001001	-1.009		
H2O	55.51	55.51	9.998e+005		
HCO3-	0.0009985	0.0009985	60.92		
NH3(aq)	0.001000	0.001000	17.03		
Na+	0.002000	0.002000	45.97		
O2(aq)	0.002253	0.002253	72.07		

		In fluid		Sorbed	
Elemental composition	total moles	moles	mg/kg	moles	mg/kg
Carbon	0.0009985	0.0009985	11.99		
Hydrogen	111.0	111.0	1.119e+005		
Nitrogen	0.001000	0.001000	14.00		
Oxygen	55.52	55.52	8.880e+005		
Sodium	0.002000	0.002000	45.97		

Note: The amount of Na+ (as NaOH or NaHCO3) needed was 0.002 moles per liter.

c) For a 550 mL solution, need 0.00110 moles. Using a 1.0 m NaOH solution, need 1.1 mL.

July 10, 2002

1430 hrs: Using the 5.0E-3 molal uranyl nitrate solution that was prepared on July 16, 2001, by RTP (see ESN#278, page 323), uranium solutions of various concentrations were prepared.

1. Tared 100.18 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle. Added deionized H2O to a total mass of 500.02 g. Calculated [U] = 1.0E-3 molal.
2. Tared 20.11 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle. Added deionized H2O to a total mass of 500.02 g. Calculated [U] = 2.01E-4 molal.
3. Tared 2.52 g of 5.E-3 molal UO2(NO3) solution into a 500-mL polypropylene bottle. Added deionized H2O to a total mass of 500.02 g. Calculated [U] = 2.52E-5 molal.
4. Tared 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle. Added deionized H2O to a total mass of 500.00 g. Calculated [U] = 2.50E-5 molal.

July 11, 2002

1030 hrs – Prepared 1.0 m HNO3 solution from concentrated (15.8 N) HNO3 reagent by diluting ~31.65 mL of the reagent (measured with a 25 mL graduated cylinder) to 1000 mL with deionized H2O in a 1000-mL volumetric flask. Transferred solution to a 1000-mL polypropylene bottle.

pH measurements of uranium nitrate solutions:

--- electrode calibrated using pH 4 and 7 reference solutions; slope = 99.2
pH 7 standard check = 7.02
pH 4 standard check = 4.00

1) 5.0E-5 molal UO2(NO3)2 solution: measured pH = 3.34;
Value calculated using GWB is 3.38.

pH 7 standard recheck = 6.98
pH 4 standard recheck = 3.98

2) 1.E-3 molal UO2(NO3)2 solution: measured pH = 3.84
Value calculated using GWB is 3.82.

Added 300 microliters of 1.0 M HNO3 solution
Measured pH = 3.17

.pH 4 standard recheck = 3.95

3) 2.E-4 molal UO2(NO3)2 solution: measured pH = 4.36
Value calculated using GWB is 4.26.

Added 300 microliters of 1.0 m HNO3 solution
Measured pH = 3.17.

1250 hrs. – Recalibrate pH electrode with pH 4 and 7 reference standards: slope = 101%

Measured pH of 2.E-4 molal UO2(NO3)2 solution with added HNO3 solution = 3.22 (previous measurement is 3.17).

4) 2.5E-5 molal UO2(NO3)2 solution: measured pH = 4.90;
Value calculated using GWB is 4.80.

5) 2.5E-5 molal UO2(NO3)2 solution (second batch): measured pH = 4.83.
Value calculated using GWB is 4.80.

Preparation of EXAFS reactant mixtures:

1) Mixture EXAFS*3A

Transferred 49.94 g of the 1.E-3 molal uranyl nitrate solution into a 60-mL polypropylene bottle labeled EXAFS*3A*IU. The solution will be acidified later and submitted for analysis of initial U concentration.

To the remaining 450 g of U solution, added 1.001 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf prepared by Ajain on 5/2/01; SN 420/54-67). The Na-clinoptilolite was kept stored inside a dessicator over a saturated NaCl solution.

2) Mixture EXAFS*3B

Transferred 49.97 g of the 2.E-4 molal uranyl nitrate solution into a 60-mL polypropylene bottle labeled EXAFS*3B*IU. The solution will be acidified later and submitted for analysis of initial U concentration.

To the remaining 450 g of U solution, 1.001 g of Na-clinoptilolite was added.

Check pH standards: pH 7 std measured = 7.02; pH 4 std measured = 4.07

3) Mixture EXAFS*3C

Transferred 50.02 g of the 2.5E-5 molal uranyl nitrate solution into a 60-mL polypropylene bottle labeled EXAFS*3C*IU. The solution will be acidified later and submitted for analysis of initial U concentration.

To the remaining 450 g of U solution, 200 microliters of 0.1 m NaOH solution was added. The NaOH solution was prepared by B. Werling on 4/17/02 (SN 463/127). The pH was measured to be 6.10 ± 0.10 (relatively unstable). Added 1.003 g of Na-clinoptilolite.

4) Mixture EXAFS*3D

Transferred 250 g of the second batch of 2.5E-5 molal uranyl nitrate solution into a 250-mL polypropylene bottle labeled EXAFS*3D. Added 100 microliter of 0.1 m NaOH solution. Measured pH is 6.04. Added 0.999 g of Na-clinoptilolite.

Recheck pH standards: pH-7 std measured = 7.02; pH-4 std measured = 4.07.

1500 hrs – The four mixtures were shaken, then the caps were loosened to allow CO2(g) to enter, and the bottles placed on a gyratory shaker.

July 12, 2002

0915 hrs. – recheck pH 7 standard = 6.98; recheck pH 4 standard = 3.99

Measure pH of EXAFS uranium/clinoptilolite mixtures:

1) EXAFS*3A: pH = 3.81
   Added 25 microliters of 0.1 m HNO3 (prepared by B. Werling on 9/4/01; SN#463/94)
   PH = 3.78
   Added 50 microliters of 0.1 m HNO3; pH = 3.73
   Added 50 microliters of 1.0 m HNO3 (see page 351); pH = 3.47
   Added 50 microliters of 1.0 m HNO3; pH = 3.32
   Added 50 microliters of 1.0 m HNO3; pH = 3.25
   Mixture placed back on gyratory shaker; loosely capped.

2) EXAFS*3B: pH = 3.94
   Added 50 microliters of 1.0 m HNO3; pH = 3.55
   Added 50 microliters of 1.0 m HNO3; pH = 3.35
   Added 50 microliters of 1.0 m HNO3; pH = 3.26
   Mixture placed back on gyratory shaker; loosely capped.

3) EXAFS*3C pH = 6.6±0.1
   Added 50 microliters of 0.1 m HNO3; pH = 5.9±0.1
   Mixture placed back on gyratory shaker; loosely capped.

4) EXAFS*3D pH = 7.2±0.1
   Added 50 microliters of 0.1 m HNO3; pH = 5.9±0.1
   Mixture placed back on gyratory shaker; loosely capped.

Preparation of ^{237}Np-loaded clinoptilolite:

Yesterday, Paul Bertetti opened an ampoule containing neptunium nitrate in 4 M HNO3. The neptunium nitrate was purchased from Isotope Products and had 5.187 microcuries of Np-237 in 5.63897 g of solution (see scientific notebook #031, page 296). To reduce the HNO3 concentration, Paul evaporated the Np nitrate solution to near dryness, and redissolved it in several milliliters of 0.01 m HNO3. This was repeated two-three times. Several evaporation and dilution with pure deionized water followed. After the last dilution, the neptunium nitrate solution (about 100 mL) had a pH of ~4.0. This solution was transferred into a 1-L polycarbonate bottle, labeled EXAFS*Np*0.001 m HNO3, and diluted to 600 g with deionized water. Paul Bertetti took two 1-mL samples of the Np solution for analysis of Np concentration by liquid scintillation counting.

1030 hrs. – 0.600 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf prepared by A.Jain on 5/2/01; SN 420/54-67) was added to the neptunium nitrate solution. The Na-clinoptilolite was kept stored inside a dessicator over a saturated NaCl solution. The bottle was placed on a gyratory shaker, with the cap loosened to allow CO2(g) to enter.

1400 hrs. – pH 7 standard recheck = 6.99; pH 4 standard recheck = 4.02

Measured pH of 0.001 N HNO3 (prepared by B. Werling and used to dilute the neptunyl nitrate solution) = 3.02

EXAFS*3C measured pH = 6.4±0.05
EXAFS*3D measured pH = 6.3±0.05

July 15, 2002

0930 hrs. – measured pH of clinoptilolite/U-solution mixtures and clinoptilolite/Np-solution mixture

pH electrode recalibrated using pH 7 (value given 7.02) and 4: slope = 98.7

Mixture EXAFS*3A: pH = 3.41
Added 50 microliters of 1.0 m HNO3 solution; pH = 3.20

Mixture EXAFS*3B: pH = 3.46
Added 50 microliters of 1.0 m HNO3 solution; pH = 3.24

Mixture EXAFS*3C: pH = 6.7±0.05
Added 50 microliters of 1.0 m HNO3 solution; pH = 5.77±0.05

Mixture EXAFS*3D: pH = 6.7±0.05
Added 50 microliters of 1.0 m HNO3 solution; pH = 5.87

Mixture EXAFS*Np*0.001 m HNO3: pH = 3.65
Added 50 microliters of 1.0 m HNO3 solution; pH = 3.36
Added 50 microliters of 1.0 m HNO3 solution; pH = 3.20

1000 hrs. – A 1.0 N NaOH solution was prepared by diluting 1 unit of a NaOH DILUT-IT concentrate (Fisher Lot #H26123) with 18.2 megaohm deionized water in a 1-L volumetric flask. The 1 N NaOH solution was transferred into a 1-L polypropylene bottle and capped tightly.

1445 hrs – pH standard recheck: pH 7.02 = 7.01; pH 4.00 = 4.00

Measured pH of EXAFS mixtures:

EXAFS*3A = 3.23
EXAFS*3B = 3.26
EXAFS*3C = 6.12±0.02
EXAFS*3D = 6.30±0.02

EXAFS*Np*0.001 m HNO3 = 3.30
Added 50 microliters 0.1 m HNO3; pH = 3.30
Added 50 microliters of 1.0 m HNO3; pH = 3.15

Mixtures placed back on gyratory shaker with loose caps.

The weight of scintillation vials were measured empty and with 0.5 mL of 0.1 m HNO3 solution. Samples of the Np solutions will be placed in these vials for later liquid scintillation analysis.

Sample name	Wt. (g) scintillation vial empty	Wt. (g) w/ 500 µL 0.1 m HNO3	Wt (g) of HNO3 solution
Np*3F*1	7.3181	7.8135	0.4954
Np*3F*2	7.3322	7.8266	0.4944
Np*8F*1	7.3634	7.8510	0.4876
Np*8F*2	7.3896	7.8761	0.4865

July 16, 2002

0915 hrs. – measured the pH of EXAFS mixtures

pH 7 standard check: 7.04
pH 4 standard check: 4.01

EXAFS*3A: pH = 3.29
EXAFS*3B: pH = 3.33
EXAFS*3C: pH = 6.3±0.05
EXAFS*3D: pH = 6.54
Added 50 microliter of 1.0 m HNO3: pH = 4.36±0.05

EXAFS*Np*0.001 m HNO3: pH = 3.25

Preweighed 1-L polycarbonate bottle (labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2) that will be used to contain the filtrant solution from filtering the clinoptilolite/Np solution mixture. Weight = 172.3 grams.

1035 hrs. – Filtered the clinoptilolite/Np solution mixture contained in the polycarbonate bottle labeled EXAFS*Np*0.001 m HNO3 through a Fisher brand P5 qualitative glass fiber filter paper (medium porosity and medium flow rate, Cat. No. 09-801F, 19 cm diameter). The filtrant was collected into another polycarbonate bottle (labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2). Twenty milliliters of the filtrant were taken using a pipet and used to wash down the clinoptilolite powder on the sides of the filter paper towards the apex of the funnel so that collecting the clinoptilolite powder (paste) would be easier.

The clinoptilolite wet paste was scraped off the filter paper and transferred into a plastic vial labeled Np*IE. The vial was capped and covered with aluminum foil for later shipping to Argonne National Laboratory.

The weight of the bottle EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 was measured to be 765.3. Therefore, the weight of the filtrant is 593.0 grams.

Two 1-mL aliquots of the Np solution filtrant were taken. Each was transferred into the scintillation vials that have 0.5 mL of 0.1 m HNO3 solution each. The weight of vial labeled Np*3F*1 before addition of the Np sample is 7.8146 grams. After addition of the Np sample, the weight is 8.8108. Thus the Np sample weight is 0.9962 grams. The weight of the vial labeled Np*3F*2 before addition of the Np sample is 7.8262 grams. After addition of the Np sample, the weight is 8.8222. Thus the Np sample weight is 0.9960 grams.

The weight of the bottle EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 was re-measured to be 763.3. There is a difference of 2 grams from the previous reading. The two 1-mL samples are accounted for.

1050 hrs. – Recalibrate pH elecrode using pH standards 4 and 7; slope = 99.3.

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant = 3.35
Added 500 microliters of 1.0 m NaOH solution (see page 354); pH = 10+,overrange; **solution turned deep yellow in color**

Recalibrated pH electrode using pH 7 and 9 standards; slope = 100.1

Added 200 microliters of 1.0 m HNO3; **solution turned clear**; pH = 3.28
Added 100 microliters of 1.0 m NaOH solution
Added another 100 microliters of 1.0 m NaOH; pH = 3.75
Added another 100 microliters of 1.0 m NaOH; pH = 9.04; **yellow solution**
Added 50 microliters of 0.1 m HNO3 solution; pH = 7.84; **light yellow solution**

pH standard check: pH 9 = 8.99.

The weight of the bottle EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 was re-measured to be 764.9. Therefore the weight of the Np solution is 592.6 grams.

Added 0.592 grams of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf prepared by A.Jain on 5/2/01; SN 420/54-67) to the Np solution. The Na-clinoptilolite was kept stored inside a dessicator over a saturated NaCl solution.

1100 hrs. – the bottle EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 was placed on the gyratory shaker with the cap loosened to allow CO2(g) to enter.

1320 hrs. – pH standard check: pH 9 = 8.95; pH 7 = 6.97

Measured pH of EXAFS*3D = 4.51
Added 50 microliters of 0.1 m NaOH; pH = 5.35
Added another 50 microliters; pH = 6.10±0.05

1330 hrs. – Recalibrated pH electrode using pH 4 and 7 standards; slope = 98.9

Measured pH of EXAFS*3A = 3.27
Measured pH of EXAFS*3B = 3.31

1350 hrs. – The clinoptilolite/uranium-solution mixture (EXAFS*3A) was filtered through a Fisher brand P5 qualitative glass fiber filter paper (medium porosity and medium flow rate, Cat. No. 09-801F, 19 cm diameter). The filtrant was collected into another 500-mL polypropylene bottle (labeled EXAFS*3A*filtrant). Fourty milliliters of the filtrant were taken using a pipet and used to wash down the clinoptilolite powder on the sides of the filter paper towards the apex of the funnel so that collecting the clinoptilolite powder (paste) would be easier.

The clinoptilolite wet paste was scraped off the filter paper and transferred into a plastic vial labeled U*3A. The vial was capped and covered with aluminum foil for later shipping to Argonne National Laboratory.

The clinoptilolite/uranium-solution mixture (EXAFS*3B) was filtered and the filtrant collected into a 500-mL polypropylene bottle (labeled EXAFS*3B*filtrant). Some filtrant was used to wash down the clinoptilolite powder on the sides of the filter paper towards the apex of the funnel so that collecting the clinoptilolite powder (paste) would be easier.

The clinoptilolite wet paste was scraped off the filter paper and transferred into a plastic vial labeled U*3B. The vial was capped and covered with aluminum foil for later shipping to Argonne National Laboratory.

1430 hrs. – pH electrode was recalibrated using pH 7 and 9 standards; slope = 100.4

The bottle labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 was taken off the gyratory shaker. The color of the Np solution is only slightly yellow.

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 solution = 7.77
Added 50 microliters of 0.1 m NaOH; pH = 7.97
Added 50 microliters of 0.1 m NaOH; pH = 8.26

The bottle labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 was placed back on the gyratory shaker with the cap loose.

1445 hrs. – Paul Bertetti completed an analysis of the Np activity of the initial 600 mL neptunyl nitrate solution (EXAFS*Np*0.001 m HNO3) using liquid scintillation analysis. The calculated concentration is 12.2 ppm Np.

July 17, 2002

1330 hrs. – pH 7 standard check = 7.03

Measured pH of EXAFS*3C = 6.40±0.05
Measured pH of EXAFS*3D = 5.77
Added 50 microliters of 0.1 m NaOH; pH = 6.27±0.05

pH standard check = 9.03

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 solution = 7.34
Added 50 microliters of 0.1 m NaOH; pH = 7.41
Added 50 microliters of 1.0 m NaOH; pH = 8.46

EXAFS*3C, EXAFS*3D, and EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 mixtures placed back on gyratory shaker with caps loose.

July 18, 2002

0940 hrs. – pH 7 standard check = 7.03; pH 9 standard check = 9.05

Measured pH of EXAFS*3C = 6.50
Added ~50 microliters of 0.1 HNO3; pH = 6.10±0.05
Measured pH of EXAFS*3D = 5.93
Added ~20 microliters of 1.0 m NaOH; pH = 9.93
Added ~25 microliters of 1.0 m HNO3; pH = 5.53
Added ~75 microliters of 0.1 m NaOH; pH = 6.36

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.43
Added ~25 microliters of 1.0 m NaOH = 8.82

EXAFS*3C, EXAFS*3D, and EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 mixtures placed back on gyratory shaker with caps loose.

1500 hrs. – pH 7 standard check = 7.03; pH 9 standard check = 9.01

Measured pH of EXAFS*3C = 6.35±0.02
Measured pH of EXAFS*3D = 6.25±0.02

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.90±0.02
Added ~15 microliters of 1.0 m NaOH = 8.50

July 19, 2002

0945 hrs. – pH 7 standard check = 6.97; pH 9 standard check = 8.98

Measured pH of EXAFS*3C = 6.30±0.05
Measured pH of EXAFS*3D = 6.05
Added 10 microliters of 0.1 m NaOH; pH = 6.35

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.65
Added 10 microliters of 0.1 m NaOH = 7.67
Added 10 microliters of 1.0 m NaOH = 8.26

1000 hrs. – – The clinoptilolite/uranium-solution mixture (EXAFS*3C) was filtered through a Fisher brand P5 qualitative glass fiber filter paper (medium porosity and medium flow rate, Cat. No. 09-801F, 19 cm diameter). The filtrant was collected into another 500-mL polypropylene bottle (labeled EXAFS*3C*filtrant). Fourty milliliters of the filtrant were taken using a pipet and used to wash down the clinoptilolite powder on the sides of the filter paper towards the apex of the funnel so that collecting the clinoptilolite powder (paste) would be easier.

The clinoptilolite wet paste was scraped off the filter paper and transferred into a plastic vial labeled U*3C. The vial was capped and covered with aluminum foil for later shipping to Argonne National Laboratory.

Two empty 60-mL polypropylene bottles (labeled EXAFS*3C*FU*a and *b) were weighed. The weights are 11.4512 and 12.2016, respectively, for bottles EXAFS*3C*FU*a and EXAFS*3C*FU*b. To each bottle was added 100 microliters of 1.0 m HNO3 solution (see page 351). The weights after acid addition are 11.5596 and 12.3082 for *a and *b, respectively. Subsequently, to each bottle was added 25 mL of the EXAFS*3C*filtrant. The final weight of EXAFS*3C*FU*a and EXAFS*3C*FU*b are 36.6057 and 37.3477, respectively. The calculated weights of the added acid in *a and *b are 0.1084 and 0.1066 grams, respectively. The calculated weights of the EXAFS*3C*filtrant transferred into *a and *b, respectively, are 25.0461 and 25.0395 grams, respectively. The EXAFS*3C*FU*a and EXAFS*3C*FU*b samples will be sent for ICP analysis.

1410 hrs. – Recalibrate pH electrode using pH 7 and 9 standards; slope = 101.3

Measured pH of EXAFS*3D = 6.35±0.05

The clinoptilolite/uranium-solution mixture (EXAFS*3D) was filtered through a Fisher brand P5 qualitative glass fiber filter paper (medium porosity and medium flow rate, Cat. No. 09-801F, 19 cm diameter). Because the EXAFS*3D mixture is only about 250 mL in volume, a smaller glass funnel was used and the filter paper was cut with a pair of scissors to fit the size of the funnel.

The filtrant was collected into another 250-mL polypropylene bottle (labeled EXAFS*3D*filtrant). Ten milliliters of the filtrant were taken using a pipet and used to wash down the clinoptilolite powder on the sides of the filter paper towards the apex of the funnel so that collecting the clinoptilolite powder (paste) would be easier.

The clinoptilolite wet paste was scraped off the filter paper and transferred into a plastic vial labeled U*3D. The vial was capped and covered with aluminum foil for later shipping to Argonne National Laboratory.

Two empty 60-mL polypropylene bottles (labeled EXAFS*3D*FU*a and *b) were weighed. The weights are 11.5062 and 11.4323, respectively, for bottles EXAFS*3D*FU*a and EXAFS*3D*FU*b. To each bottle was added 100 microliters of 1.0 m HNO3 solution (see page 351). The weights after acid addition are 11.6155 and 11.5444 for *a and *b, respectively. Subsequently, to each bottle was added 25 mL of the EXAFS*3D*filtrant. The final weight of EXAFS*3D*FU*a and EXAFS*3D*FU*b bottles are 36.6352 and 36.5550, respectively. The calculated weights of the added acid in *a and *b are 0.1093 and 0.1121 grams, respectively. The calculated weights of the EXAFS*3D*filtrant transferred into *a and *b, respectively, are 25.0197and 25.0106 grams, respectively. The EXAFS*3D*FU*a and EXAFS*3D*FU*b samples will be sent for ICP analysis.

1515 hrs. – pH 7 standard check = 7.02; pH 9 standard check = 8.99

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.81
Added 10 microliters of 1.0 m NaOH = 8.45

July 21, 2002

1400 hrs. – pH 7 standard check = 7.04; pH 9 standard check = 9.01

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.67
Added 25 microliters of 1.0 m NaOH = 8.88

July 22, 2002

0950 hrs. – pH 7 standard check = 7.01; pH 9 standard check = 9.03

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.82
Added 25 microliters of 1.0 m NaOH = 8.81

1030 hrs. – FEDEX package containing eight XAFS sample cells were received. See the following email from Richard Reeder (SUNY) regarding the cells:

From: rjreeder@notes.cc.sunysb.edu
Sent: Monday, July 22, 2002 7:58 AM
To: rpabalan@cnwra.swri.edu
Subject: RE: sample holders

Bobby,

You should be receiving the sample holders this morning (Airborne Express). I sent 8 complete holders, with some extra o-rings and windows (so you should have extras of everything if anything looks problematic). The body and window are made of Kel-F and the o-ring material is neoprene; these should be resistant to acid and base. The seal is critical and requires that o-ring contact surfaces are wiped clean and visually inspected before assembly. It's also critical that no paste, particles, lint, etc., are inadvertently introduced to the o-ring area or to the top surface where the window and frame must compress. The bolts must be tightened in some even pattern so that the o-ring seals uniformly on the window material. It's also important to make sure that the assembly is fully compressed, but you also must avoid stripping the threads in the bottom aluminum plate. We have noticed that when fully compresse! d, the window material

tends to bulge out a tiny bit through the slot in the top frame. You might want to put a few drops of water in one holder and assemble it before loading hot material. If you have an questions, it might be a good idea for me to talk with you. I have loaded several cells here and they seal well as long as the contact surfaces are clean and the assembly is fully compressed.

The consistency of the paste is also a factor. When we last ran your samples extracted from centrifuge tubes, we found that they were too wet right out of the tube. We found that by scooping the paste onto a single layer of filter paper for just a few seconds, the excess liquid was wicked away leaving a moist (but not dripping wet) paste. The paste could then be scooped into the cavity. It's not essential to have the entire cavity filled to the top. And, it's almost impossible to avoid an air bubble; that's OK. We usually tap the holder to make sure that a homogeneous area exists so we can place the beam on that area (the beam is about 1 x 1 mm).

Finally, each assembled cell should be placed in a separate ziplock bag (the ones containing the cells are fine). That way, if anything leaks it will not affect other cells. You may want to double bag the Np samples.

I'll be around today if you have any questions.

Cheers,

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240

1300 hrs. – pH 7 standard check = 6.97; pH 9 standard check = 8.98

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.95
Added 25 microliters of 1.0 m NaOH = 8.85

1400 hrs. – Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 8.68

Wet paste was scooped out of the plastic vial marked U*3A using a flat stainless steel spatula. The spatula was contacted with a filter paper to soak up some of the uranium solution. Then the paste was transferred into the cavity of the XAFS sample cell. The cell was tapped on the side to allow the wet powder to settle into the cavity. In this process, some liquid is displaced towards to top of the cavity. This excess liquid was soaked up with a Kimwipe. Tapping and removal of excess liquid was done about three to four times. Subsequently, the cell was covered with its top, and the screws were tightened in a regular pattern. The tightening was repeated a few times until all screws are finger tight. The remaining paste was left in the capped vial. The sample cell was placed inside a ziploc bag and labeled U*3A.

The wet paste from vials U*3B, U*3C, U*3D, and Np*IE were also transferred into XAFS sample holders. The uranium cells were placed individually into ziploc bags. The neptunium cells were double-bagged.

The XAFS sample cell cavity holds approximately 100 microliters, measured by transferring deionized water with a 50 microliter pipet. For a mineral with a density of 2.1 g/mL, the cavity would contain approximately 0.21 grams of solid (compacted).

1658 hrs. – Recalibrate pH electrode using pH 7 and 9 standards; slope = 100.8

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 8.13
Added 10 microliters of 1.0 m NaOH = 8.74

July 23, 2002

1015 hrs. – Recalibrate pH electrode using pH 7 and 9 standards; slope = 100.9

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 7.92
Added 10 microliters of 1.0 m NaOH = 8.27

1325 hrs. – pH 7 standard check = 7.01; pH 9 standard check = 9.00

Measured pH of EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 = 8.16

1337 hrs. – Filtered the clinoptilolite/Np solution mixture contained in the polycarbonate bottle labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2 through a Fisher brand P5 qualitative glass fiber filter paper (medium porosity and medium flow rate, Cat. No. 09-801F, 19 cm diameter). The filtrant was collected into another polycarbonate bottle (labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2*filtrant). Twenty milliliters of the filtrant were taken using a pipet and used to wash down the clinoptilolite powder on the sides of the filter paper towards the apex of the funnel so that collecting the clinoptilolite powder (paste) would be easier.

The clinoptilolite wet paste was scraped off the filter paper and transferred into two XAFS sample cells, using a procedure described on page 363. The sample cells were placed individually into double bags of ziploc, labeled Np*SC*1 and Np*SC*2. The remaining wet paste was put into a plastic vial, which was capped and covered with aluminum foil.

Two 1-mL samples of labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2*filtrant were taken and each was transferred into a vial with 0.5 mL of 0.1 m HNO3 solution. The weight of vial Np8F1 before addition of the Np solution is 7.8532 grams; after addition of the Np solution, the weight is 8.8536. The sample weight is 1.0004 grams. The weight of vial Np8F2 before addition of the sample is 7.8758 grams; after the weight is 8.8775. The sample weight is 1.0017 grams.

1430 hrs. – The wet paste from vial Np*IE was transferred into an XAFS sample cell, using the procedure described on page 363. The clinoptilolite wet paste is drier than yesterday, and it was not necessary to wick some liquid from the paste. The loaded cell was placed inside two bags of ziploc labeled Np*IE*2. The first Np sample prepared yesterday was relabeled Np*Ie*1.

1530 hrs. – The 8 XAFS cells were given to John Hageman, SwRI radiological safety officer, for proper labeling and shipment to Argonne National Laboratory (c/o Andy Bracken/Lynne Soderholm). John's assistant, Ty Schraeder, will prepare the necessary paperwork.

July 24, 2002

1700 hrs. – The EXAFS sample cells were sent this afternoon to Argonne National Laboratory (Andy Bracken/Lynne Soderholm; Building 46 – Receiving) via FEDEX.

July 29, 2002

The following is an email from Rich Reeder (SUNY) regarding the analysis of the Np- and U-loaded clinoptilolite:

From: rjreeder@notes.cc.sunysb.edu
Sent: Monday, July 29, 2002 1:37 PM
To: rpabalan@cnwra.swri.edu
Subject: Update

Bobby,

I think I lost a more detailed description of what follows, so this is brief.

Some observations about what we're seeing in the samples. Only Np*IE showed enough Np to collect spectra. There's virtually no fluorescence to be seen in the SC sample. Also, we see what is almost certainly a Zr edge coming in at about 18.015 keV in both IE and SC. Must be Zr contamination in the Np source (Zr is important in both the waste cycle and in the separations process I think). But this introduces a feature in the spectrum for IE that only allows us to go out to 10 k (not far enough). The short k range broadens and smoothes the FT. We can see the expected axial and equatorial peaks, but they are very smoothed. We also note that the intensity of Zr peak is the same in both IE and SC, whereas the Np is very different. BTW, the Np fluorescence is very low in the IE sample. We can overcome the Zr problem by using a cleaner Np source, or by increasing the Np loading (but not the Zr loading).

Also some unexpected findings with the U samples. U3A has a very high loading, as your calcuations suggested. The FT shows no obvious splitting of the equatorial shell. U3C and U3D have loadings similar to one another, and about 1/2 or 2/5 of the amount on U3A. The FTs for both U3C and U3D are similar, but neither sho any obvious indication of equatorial splitting. In previous runs, we saw clear indication of eq. splitting for the pH 6 samples. So, I find this puzzling. Any ideas?

Currently scanning U3B; this has the lowest loading; only about 1/2 to 1/3 of the amount on U3C or U3D. Also interesting to note that the U-loaded samples all show the Zr peak. It does not have the same detrimetal effect, however, because it occurs at the end of the scan range for U.

Rich

The following is my reply to Rich Reeder's email:

From: R Pabalan [rpabalan@cnwra.swri.edu]
Sent: Monday, July 29, 2002 3:26 PM
To: 'rjreeder@notes.cc.sunysb.edu'
Subject: RE: Update

Rich,

Thanks for the update. The Zr concentration of the clinoptilolite is 97 ppm, based on ICP-MS analysis. Perhaps that could account for the Zr peak in the

spectra of all samples. Other trace elements in the clinoptilolite are Ba (123 ppm), Rb (54 ppm), Sr (20 ppm), Ce (44 ppm), La (31 ppm), Th (15 ppm), Nb (13 ppm), Ga (12 ppm), Zn (9 ppm), Y (8 ppm) and Ni (7 ppm).

The U loading of U3C and U3D is lower than that of previous pH6 samples you analyzed. Here is my take on the observed non-splitting of the equatorial shell: The total U on the clinoptilolite comes from an ion-exchanged component and a surface-complexed component. It is possible that at the U-loading of these samples, most of the U is due to the ion-exchanged component. Perhaps equatorial splitting is observed only when the contribution from the surface-complexed species exceeds a certain value. If the samples prepared at pH~3 also show no equatorial splitting at low U-loading or only weak splitting at moderate loading, then my interpretation would make sense. Note that equatorial splitting was observed in the ion-exchanged sample you analyzed in the previous run because of the very high U-loading for that sample. Let me know what you think.

For the Np samples, it is clear that I need to increase the Np-loading. I can increase the initial Np concentration of the solution, or can cut the amount of clinoptilolite reacted with the Np solution by half---I noticed that the sample cell cavity has a volume of about 100 microliters. For the Np sample prepared at pH~8, I suspect that not enough time was given to react the clinoptilolite with the Np solution. I was still making pH adjustments a few hours before I had to filter the mixture and load the sample cell.

bobby

July 30, 2002

The following table gives the calculate weights of the 0.5 m HNO3 and Np solution sample to be used in calculating the Np concentrations:

Sample name	Wt. (g) scintillation vial empty	Wt. (g) w/ 500 μL 0.1 m HNO3	Wt (g) of HNO3 solution	Wt. (g) of vial with acid	Wt. (g) of vial w/ Np sample	Wt. (g) Np sample
Np*3F*1	7.3181	7.8135	0.4954	7.8146	8.8108	0.9962
Np*3F*2	7.3322	7.8266	0.4944	7.8262	8.8222	0.9960
Np*8F*1	7.3634	7.8510	0.4876	7.8532	8.8536	1.0004
Np*8F*2	7.3896	7.8761	0.4865	7.8758	8.8775	1.0017

THE REST OF THIS PAGE IS LEFT BLANK.

August 5, 2002

The following is an email received from Rich Reeder (SUNY) regarding the XAFS work on the clinoptilolite samples:

From: rjreeder@notes.cc.sunysb.edu
Sent: Monday, August 05, 2002 12:20 PM
To: rpabalan@swri.edu
Subject: samples

Bobby,

I am not certain as to when Lynne will ship the samples back to you; perhaps they are en route already.

There are two other observations that we made in our run at the APS, and they do not bode well. First, we observed a systematic increase in fluorescence yield as a function of time scanned for each of the sorption samples. This includes the Np as well as U-loaded samples. At first, we thought it was a program problem, e.g., a channel not being reset with each scan. But the software person finally determined that this was not the case. The only other possible explanations were an increase in the beam intensity (which did not occur), and change in beam position on the sample over time, or a change in the sample itself. The puzzling factor was that, after normalization, each of the successive spectra looked similar, so initially we discounted the latter. When we finished our session, we opened two of the U-loaded samples to check them for wetness. In each case, the area where the beam hit, showed! dry material, some of which was caked onto the window. With that insight, I now think that the progressive increase in fluorescence yield is explained by gradual drying of the sample hit by the beam. As the water content decreased in the effected area, the absorption of the fluoresced X-rays decreased, resulting in an increased signal at the detector. The fact that the increase was uniform over time meant that the drying was ongoing. It's possible that the slurries were too dry on loading. There is some sample heating because of the brightness of the beam, although it should not be much. But if the moisture content is low to begin with, the moisture could easily be redistributed away from the irradiated region. I will compare first and last scans to see if there are observable differences. But right now, I would not want to attach much significance to the U data. That's a shame, too, because the signal/noise was e! xcellent for the U samples. Let me look at these data s! ome more to get a better idea of what's happened.

I am pretty certain that I will have another chance to redo these in the fall. I've also got some ideas about a single crystal experiment that would allow us to evaluate if and how actinyl species enter the channels. Let's plan to get together at Goldschmidt. I'm staying at Hotel Sunstar Park.

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240

The following is my reply:

From: R Pabalan [rpabalan@cnwra.swri.edu]
Sent: Monday, August 05, 2002 1:42 PM
To: 'rjreeder@notes.cc.sunysb.edu'
Subject: RE: samples

Hi, Rich,

I got the samples back on Friday. Do you want me to ship the samples cells back to you, or keep them until next time?

When I loaded the samples, I made sure the paste is not too wet by wicking excess solution onto a filter paper, as you suggested. After loading onto the cell, I noticed that when I tap the cell to make the paste settle into the cavity, excess solution came up to cover the surface of the sample. I wicked most of the excess solution off with a Kimwipe, but only sufficiently to remove the liquid layer but keeping the paste still moist. I agree that under the beam the top layer could have gradually become dry. The problem may have been solved by tapping the cell a few times to further expel excess solution, if any is left, and force it to the top. I'll check this possibility when I open up the cell for cleaning.

With respect to the Zr inference for the Np samples, I can either double the initial Np concentration that I used previously or cut the clinoptilolite mass by 50%. I'll need to buy additional Np spikes if I do the former (funding is no problem right now). Also, I can allow the sample at pH ~ 8 to react longer than I previously did.

The single crystal experiments would be great if I can load enough Np or U onto the mineral. I also plan to do batch experiments on powder samples in the next few months that focus on the ion exchange mechanism of Np and U uptake.

bobby

August 7, 2002

1100 hrs. – The XAFS sample cells were opened for cleaning and disposal of the radioactive samples. The following are my observations regarding the sample cells as I opened them. For close-up observation, a binocular microscope located in the petrography room of Bldg. 57 was used.

Sample 3A: Sample cell cavity not fully filled with clinoptilolite sample. The sample was dry. Minimal caking on the window material.

Sample 3B: Droplets of solution evident on window material before the cell was opened. After opening, the cell cavity is observed to be completely filled with clinoptilolite sample. Portions of the sample is moist and other portions are dry, as evident from the difference in visual appearance of different portions. Lateral microcracks(?) appear to be present on the window material. Tapping the opened cell caused some shifting of the clinoptilolite mass, indicating the sample is not fully dry.

Sample 3C: Before opening – apparent droplets of solution on window. After opening – some amount of caked material along center of window; possible lateral microcrack along the center of window; cavity fully filled but vuggy in appearance; tapping the cell does not cause shifting of clinoptilolite mass; sample appears mostly dry but retains some amount of moisture as evidenced by droplets on window.

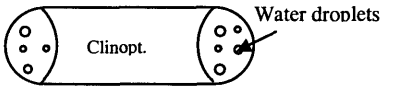
Sample 3D: Before opening – looks homogeneous, no visible cracking of window nor vuggy appearance; looks moist throughout. After opening – Very flat surface, fully filled cavity; moist appearance, but extensive dry caking on window.

Sample SC-1: Before opening – appears homogeneous and moist throughout, apparent droplets of solution near the center of the window; possible microcracks are evident along the center of the window. After opening – very flat homogeneous surface with moist appearance; no apparent caking on window

Sample SC-2: Before opening – appears homogeneous; no apparent cracking of window. After opening – window is fairly clean; the cell cavity is full and the sample surface flat; the sample appears moist but not wet.

Sample IE-1: Before opening – appears homogeneous, no apparent caking of clinoptilolite paste, some water droplets observed, possible lateral microcracks on window. After opening – microcracks on window more apparent; no dried cake on window but some powder sticking to window; flat surface of sample in the cavity; appears moist but not wet. Tapping the cell does not shift the sample mass.

Sample IE-2: Before opening – appears moist with water droplets on window, but sample mass not homogeneous. After opening – sample looks moist but cell cavity not full: empty on one end and about one-third full on the other end (see figure); window has moist cake with hour-glass shape (as in figure).



August 8, 2002

The following is an email from Paul Bertetti regarding the Np liquid scintillation analysis he conducted on the XAFS samples:

```
From: Paul Bertetti [pbertetti@cnwra.swri.edu]
Sent: Thursday, August 08, 2002 9:49 AM
To: rpabalan@cnwra.swri.edu
Subject: RE: Np sample weights

Bobby,

The calculated Np concentrations are provided in the attached excel
worksheet. I would use the averaged values from all three counting efforts
(columns N-O). Note that values are in ppb. The 'initial' is the sample I
took after the initial dilution of the spike. I had calculated 12.25 ppm
based on mass, which agrees well with the measured 12.27 ppm based on
activity. You can reference notebook #369 pages 47-53 for the original data
locations.

If you have any questions, let me know.

Paul
```

The following is a copy of the Excel worksheet:

8/8/02

sample name	sample weight	cpm alpha	efficiency correct	mass correct	atom convert	mole convert	Np-237 ppb		exp soln	average ppb Np237	average molality
Np*8F*1	1.0004	17118.420	17118.420	17111.575	2.78E+16	4.61E-08	10937.96		Np*8F	10795.20	4.55E-05
Np*8F*2	1.0017	16693.220	16693.220	16664.890	2.71E+16	4.49E-08	10652.43		Np*3F	11441.56	4.83E-05
Np*3F*1	0.9962	17761.180	17761.180	17828.930	2.90E+16	4.81E-08	11396.50		initial	12397.27	5.23E-05
Np*3F*2	0.9960	17898.020	17898.020	17969.900	2.92E+16	4.85E-08	11486.61				
initial1	0.6244	12227.820	12227.820	19583.312	3.18E+16	5.28E-08	12517.93				
initial2	0.6248	11999.780	11999.780	19205.794	3.12E+16	5.18E-08	12276.61				

7/29/02

sample name	sample weight	cpm alpha	efficiency correct	mass correct	atom convert	mole convert	Np-237 ppb		exp soln	average ppb Np237	average molality
Np*8F*1	1.0004	16993.130	16993.130	16986.335	2.76E+16	4.58E-08	10857.90		Np*8F	10977.86	4.63E-05
Np*8F*2	1.0017	17391.180	17391.180	17361.665	2.82E+16	4.68E-08	11097.82		Np*3F	11442.69	4.83E-05
Np*3F*1	0.9962	17862.960	17862.960	17931.098	2.91E+16	4.84E-08	11461.81		initial	12265.80	5.17E-05
Np*3F*2	0.9960	17799.800	17799.800	17871.285	2.90E+16	4.82E-08	11423.58				
initial1	0.6244	11843.330	11843.330	18967.537	3.08E+16	5.11E-08	12124.32				
initial2	0.6248	12127.510	12127.510	19410.227	3.15E+16	5.23E-08	12407.29				

7/30/02

sample name	sample weight	cpm alpha	efficiency correct	mass correct	atom convert	mole convert	Np-237 ppb		exp soln	average ppb Np237	average molality
Np*8F*1	1.0004	17074.420	17074.420	17067.593	2.77E+16	4.60E-08	10909.85		Np*8F	10956.84	4.62E-05
Np*8F*2	1.0017	17243.910	17243.910	17214.645	2.80E+16	4.64E-08	11003.84		Np*3F	11463.69	4.84E-05
Np*3F*1	0.9962	17705.110	17705.110	17772.646	2.89E+16	4.79E-08	11360.52		initial	12160.89	5.13E-05
Np*3F*2	0.9960	18023.060	18023.060	18095.442	2.94E+16	4.88E-08	11566.86				
initial1	0.6244	11773.100	11773.100	18855.061	3.06E+16	5.08E-08	12052.42				
initial2	0.6248	11992.700	11992.700	19194.462	3.12E+16	5.18E-08	12269.37				

Columns N, O, and P of the worksheet are shown below. These give the average values of the triplicate measurements.

exp soln	average ppb Np237	average molality
Np*8F	10909.97	4.60E-05
Np*3F	11449.31	4.83E-05
initial	12274.66	5.18E-05

The following is the calculated amount of Np lost from solution:

Calc. %Np lost from soln:	
pH~3	6.7
pH~8	11.1

The calculated %Np lost from solution at pH~8 is surprising because the amount of radioactivity on the pH~8 sample is much lower than that of the pH~3 sample, as determined using a ratemeter by T. Shraeder (SwRI radiological safety office), who measured the samples prior to shipping to Argonne National Laboratory. The lower Np concentration of the pH~8 versus the pH~3 sample was also observed by Rich Reeder while doing the XAFS analysis. Most likely, Np was lost from solution due to sorption onto the filter paper during filtration of the mixture reacted at pH~8. The sample aliquots were taken after filtration.

Entries into Scientific Notebook No. 278 for the period March 26, 2002, through September 5, 2002, have been made by

R T Pabalan 9/5/02

Roberto T. Pabalan / Date

No original entry into this Scientific Notebook has been removed.

R T Pabalan 9/5/02

Roberto T. Pabalan / Date

The entries in this electronic scientific notebook #278 document activities conducted during the Period September 6, 2002, through March 6, 2003, under the Radionuclide Transport Key Technical Issue (Project Number 20.06002.01.141).

October 15, 2002

Below is an email from Richard Reeder (SUNY) regarding the initial interpretations of the XAS spectra on the uranium-loaded and neptunium-loaded samples he analyzed last July at the APS facility (Argonne National Laboratory):

From: rjreeder@notes.cc.sunysb.edu
Sent: Tuesday, October 15, 2002 2:02 PM
To: rpabalan@cnwra.swri.edu
Subject: July data issues

Bobby,

Thanks for the info re U-exchange solutions. At first glance it looks like the Al/Si ratio in your crystals is a bit higher than in some other reported samples, which should be good for exchange. The Ba and Sr are also high; if you did Na-exchange these, is it possible that some of the Ba or Sr was removed?

I have attached a powerpoint file showing some of the individual scans we took in July at the APS. Look at slides 3 and 4 first; these are two of the U samples. The spectra are all normalized by Izero (incident beam current). The individual scans have two numbers: the scan set and the scan number. For example 12_1 indicates this is scan 1 in set number 12. Everything goes in sequence, so 12_1 preceded 12_2, which preceded 13_2, etc. I have selected scans covering the entire duration of data collection on a particular sample, typically spread out over several sets. The legend indicates this sequence. For both U samples, it's evident that the fluorescence yield increased systematically over time (note the consistent baselines at the beginning of the scans). This trend was also found for the other U samples. This is also evident in sample position scans. After collecting multipl! e datasets, we scanned the sample x-y position, consistently finding a peak where the beam had been during data collection. Our interpretation is that the area irradiated by the beam gradually lost water, presumably because of localized heating. The concern is that the coordination could have changed during drying.

The Np samples are a different story. Slide 2 is for the Zr K-edge in Np SC, which you will recall we found as an impurity in these samples. (There was essentially no fluorescence from Np.) There's no change in the fluorescence signal over time. That's what we should see since the spectra are normalized by Izero. There's nothing else significant here other than the constant signal. Slide 1 is for the Np L3-edge in Np IE. Again the time sequence is indicated by the two numbers. Here, we see a decrease in the fluorescence signal over time, rather than an increase. We are at a loss to explain that, unless the sample in the holder moved as a result of local heating (pure speculation). The feature at just over 18 keV is the Zr edge. Oddly, it's apparent in some channels and not others. That might reflect differences in sensitivity among the individual channels. Greater! Np loading should avoid

this problem, becaue the fluorescence yield will increase with absorber concentration, hopefully drowning out the Zr edge.

I am still uncertain what role the new sample holders may have played. I spoke with Lynne Soderholm about her experience using this same window material. She says it's worked well, with no indication of leakage or damage from the beam. But she reminded me that she has only run liquids, not pastes, in which case drying is not an issue. I also recall that one the window material is compressed against the o-ring, a slight budge in the window is evident. What's good about that is that is is evidence of a good seal. But I wonder if that budge might also create a slight space in fron of the sample, which if not fully saturated, could allow it to dry prematurely. That's all speculation, of course. My other option is to go with the old style mounts, in which I use Kaption tape to make the seal. Generally these stay tight, but I have had a few leak. So, they may not be a good chice for the ! Np samples. Health physics people at ANL wipe these before and after placing them in the sample stage. Any thought? Are you still up for some more experiments? If so, let me think about what ones are best repeated for this run.

Cheers,

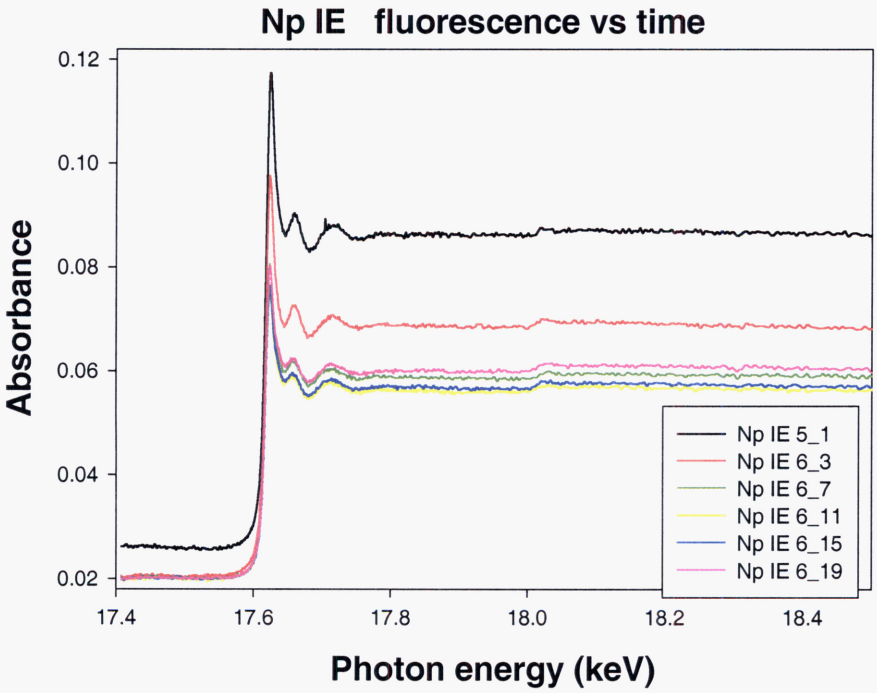
Rich

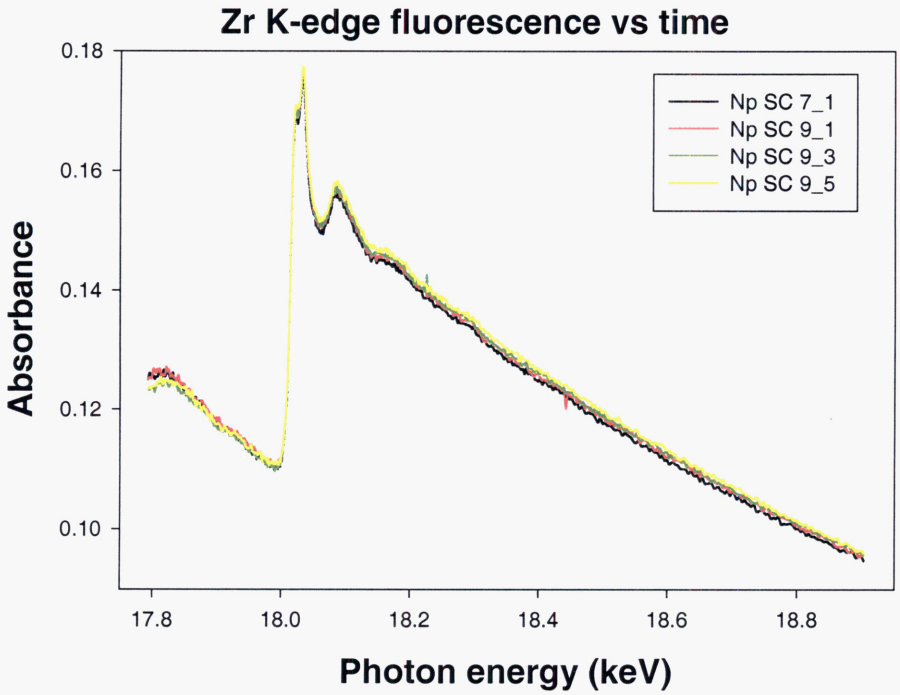
PS. I recall that you use a given size fraction for the clinoptilolite. Is it possible to use a finer fraction, thereby providing a greater surface area? That could help for the Np sorption.

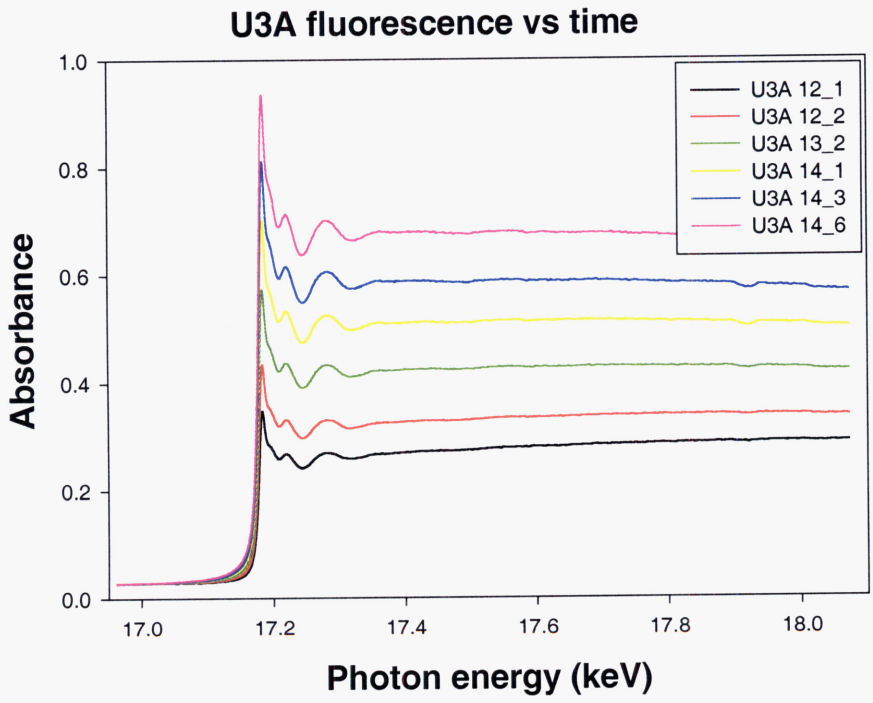
Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

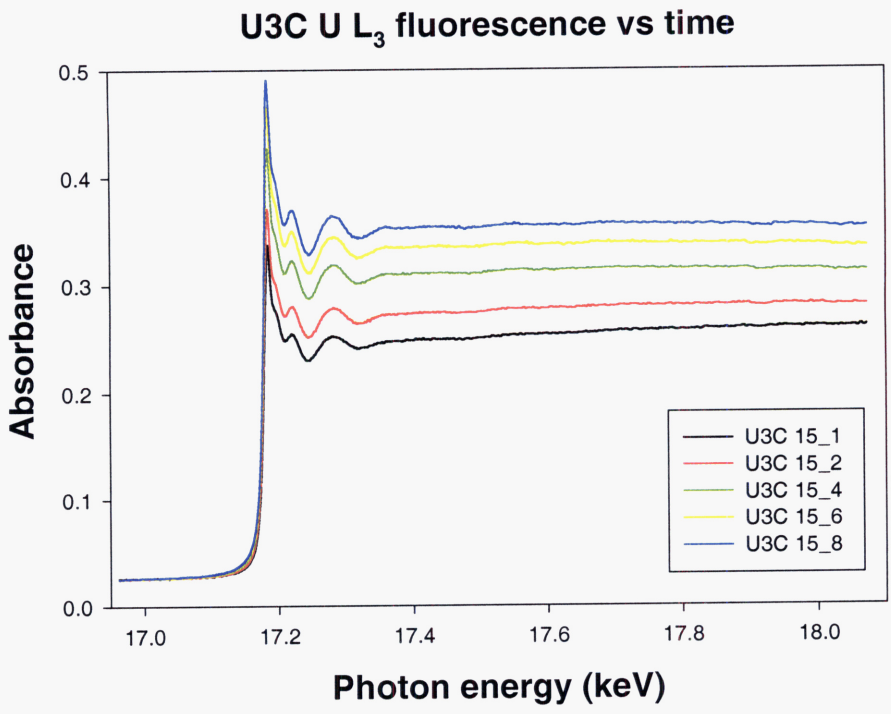
Tel. (631) 632-8208
Fax (631) 632-8240

The following are the figures in the Powerpoint file that R. Reeder attached to his email:






U3A fluorescence vs time
Absorbance
Photon energy (keV)
U3A 12_1
U3A 12_2
U3A 13_2
U3A 14_1
U3A 14_3
U3A 14_6
0.0
0.2
0.4
0.6
0.8
1.0
17.0
17.2
17.4
17.6
17.8
18.0


U3C U L3 fluorescence vs time
Absorbance
Photon energy (keV)
U3C 15_1
U3C 15_2
U3C 15_4
U3C 15_6
U3C 15_8
0.0
0.1
0.2
0.3
0.4
0.5
17.0
17.2
17.4
17.6
17.8
18.0

October 17, 2002

The following is an email from Richard Reeder (SUNY):

From: rjreeder@notes.cc.sunysb.edu
Sent: Thursday, October 17, 2002 4:42 PM
To: rpabalan@swri.edu
Subject: old and new experiments

Bobby,

Last evening I looked at raw spectra that we've collected over the last few years, particularly to see if the effect of increasing fluorescence with scan time is evident. Interestingly, I only see this in the clinoptilolite samples run at the APS. The spectra I collected in August of 2001 (which gave results that differed from our first samples) also show this effect (and I didn't spot it at the time).

Over the same general time frame and using similar sample holders, our wet pastes samples for uranyl adsorption on calcite don't show this effect. I then went back to look at the original spectra we collected at NSLS (when Melissa was with you). These spectra are fine; no change in fluorescence over time. These sample holders were the old design, but the same as what I used at the APS in Aug 2001. The differences b/t NSLS and the APS are beam size (significantly smaller at the APS) and intensity (much greater at the APS). So, it's perhaps not surprising that the problems we just encountered at the APS do not show up at X11A at NSLS.

At this point, my idea is to try to maintain a higher water content in the sample holder, so it literally cannot dry out. But that also increases the potential for the dissolved uranium to contrinute to the spectrum. I'm wondering if there would be a benefit to using a smaller size fraction for the clinoptilolite, possibly to retain moisture better. Have you experimented with different size fractions? I see inone of your papers that the particle size is 75-150 uM. If the particle size were smaller, and the surface area greater, then additional water (and the dissolved uranium in it) should have less of an effect on the total signal.

Mostly this is just thinking out loud. Any thoughts? Right now, I'm leaning toward just uranyl sorption samples for the upcoming run. That is Nov. 7th. That way, I can mount and remount if necessary in Lynne Soderholm's lab at ANL.

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240

October 24, 2002

The following is an email from Richard Reeder (SUNY):

From: rjreeder@notes.cc.sunysb.edu
Sent: Thursday, October 24, 2002 2:17 PM
To: rpabalan@cnwra.swri.edu
Subject: RE: old and new experiments

Bobby,

That sounds fine.

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240

R Pabalan <rpabalan@cnwra.swri.edu>
10/24/02 02:36 PM
Please respond to rpabalan

To: rjreeder@notes.cc.sunysb.edu
cc:
Subject: RE: old and new experiments

Rich,

I plan to prepare six samples with the following parameters:

1. U*4A ---- pH~3, U-loading~2000 ppm, 200/325 mesh size clinoptilolite
2. U*4B ---- pH~3, U-loading~1000 ppm, 200/325 mesh size clinoptilolite
3. U*4C ---- pH~3, U-loading~1000 ppm, <400 mesh size clinoptilolite
4. U*4D ---- pH~6, U-loading~2000 ppm, 200/325 mesh size clinoptilolite
5. U*4E ---- pH~6, U-loading~1000 ppm, 200/325 mesh size clinoptilolite
6. U*4F ---- pH~6, U-loading~1000 ppm, <400 mesh size clinoptilolite

I have the 200/325 mesh size available, but not the <400. I'll grind up some of the 200/325 powder, but may not have time to sieve to less than 400 mesh.

With regards to pH changes in the tube (vial), the U-solution/clinoptilolite mixtured will be equilibrated under atmospheric pCO2 conditions. I don't expect pH changes of the slurry to be significant during storage. The vial will be tightly capped so no significant evaporation and drying will occur. I'll run a parallel experiment and measure the pH of the slurry with pH paper after a few days to check for any pH change.

Let me know if the above parameters are okay.

bobby

-----Original Message-----
From: rjreeder@notes.cc.sunysb.edu [mailto:rjreeder@notes.cc.sunysb.edu]
Sent: Thursday, October 24, 2002 9:21 AM
To: rpabalan@cnwra.swri.edu
Subject: RE: old and new experiments

Bobby,

I think for both pH 3 and pH 6 it would be good to try for loadings of about 2000 and 1000 ppm U. These would match at least some of the previous samples. The loading for U3A was just too high.

Another thought.... is there any chance that the pH of the slurry retained after centrifuging could change while it's sitting in the tube. If some CO2 were evolved, particularly for the pH 3 sample, would that be likely to have an effect on sorption in the remaining slurry? Once it's mounted in a sample holder, I'm less concerned.

Let me know what you think?

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240

October 24, 2002

Preparation of Uranium-loaded Clinoptilolite for EXAFS Analysis
(scheduled for November 7, 2002)

1) Using the 5.0E-3 molal uranyl nitrate solution that was prepared on July 16, 2001, by RTP (see ESN#278, page 323), uranium solutions of various concentrations were prepared.

   a) Procedure: Tare 20 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4A). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.0E-4 molal.

      Wt. Bottle w/ cap: 54.70 g
      Plus 20 g U soln: 74.67 g
      Plus H2O: 574.66 g

   b) Procedure: Tare 10 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4B). Add deionized H2O to a total mass of 500 g. Calculated [U] = 1.0E-4 molal.

      Wt. Bottle w/ cap: 54.75 g
      Plus 10 g U soln: 64.71 g
      Plus H2O: 564.72 g

   c) Procedure: Tare 10 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4C). Add deionized H2O to a total mass of 500 g. Calculated [U] = 1.0E-4 molal.

      Wt. Bottle w/ cap: 54.54 g
      Plus 10 g U soln: 64.52 g
      Plus H2O: 564.58 g

   d) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3) solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4D). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

      Wt. Bottle w/ cap: 54.56 g
      Plus 2.5 g U soln: 57.05 g
      Plus H2O: 557.03 g

   e) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4E). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

      Wt. Bottle w/ cap: 54.68 g
      Plus 2.5 g U soln: 57.18 g
      Plus H2O: 557.19 g

f) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4F). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

Wt. Bottle w/ cap: 54.69 g
Plus 2.5 g U soln: 57.15 g
Plus H2O: 557.14 g

g) Procedure: Tare 10 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4G). Add deionized H2O to a total mass of 500 g. Calculated [U] = 1.0E-4 molal.

Wt. Bottle w/ cap: 54.49 g
Plus 10 g U soln: 64.44 g
Plus H2O: 564.47 g

h) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*4H). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

Wt. Bottle w/ cap: 54.47 g
Plus 2.5 g U soln: 56.97 g
Plus H2O: 556.99 g

2) Prepare 8 U-solution/clinoptilolite mixtures

a) Mixture EXAFS*U*4A

Procedure: Transfer 50 mL of the 2E-4 molal U nitrate solution (EXAFS*U*4A) into a 60-mL PP bottle labeled EXAFS*U*4A*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/cap: 14.079
Plus 50 mL U soln: 63.898

To the remaining 450 g of U solution, added 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf prepared by A. Jain on 5/2/01; SN 420/54-67). The Na-clinoptilolite was kept stored inside a dessicator over a saturated NaCl solution

b) Mixture EXAFS*U*4B

Procedure: Transfer 50 mL of the 1E-4 molal U nitrate solution (EXAFS*U*4B) into a 60-mL PP bottle labeled EXAFS*U*4B*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/cap: 14.970 g
Plus 50 mL U soln: 64.495 g

To the remaining 450 g of U solution, added 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf).

c) Mixture EXAFS*U*4C

Procedure: Transfer 50 mL of the 1E-4 molal U nitrate solution (EXAFS*U*4C) into a 60-mL PP bottle labeled EXAFS*U*4C*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/cap: 14.098
Plus 50 mL U soln: 63.546

d) Mixture EXAFS*U*4D

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*4D) into a 60-mL PP bottle labeled EXAFS*U*4D*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/ cap: 14.202
Plus 50 mL U soln: 63.534

To the remaining 450 g of U solution, added 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf).

e) Mixture EXAFS*U*4E

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*4E) into a 60-mL PP bottle labeled EXAFS*U*4E*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/cap: 14.957
Plus 50 mL U soln: 64.163

Transferred (tared) 250 g of the U solution into a 250-mL PP bottle labeled EXAFS*U*4E*a, added 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf).

f) Mixture EXAFS*U*4F

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*4F) into a 60-mL PP bottle labeled EXAFS*U*4F*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/ cap: 14.767 g
Plus 50 mL U soln: 63.886 g

g) Mixture EXAFS*U*4G

Procedure: Transfer 50 mL of the 1E-4 molal U nitrate solution (EXAFS*U*4G) into a 60-mL PP bottle labeled EXAFS*U*4G*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/ cap: 14.269 g
Plus 50 mL U soln: 63.537 g

To the remaining 450 g of U solution, add 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe; label dated 4/17/01 by A. Jain; lab notebook page number 420/63)

h) Mixture EXAFS*U*4H

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*4H) into a 60-mL PP bottle labeled EXAFS*U*4H*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Bottle w/ cap: 14.915 g
Plus ~ 50 g U soln: 64.122 g

Transferred (tared) 250 g of the U solution into a 250-mL PP bottle labeled EXAFS*U*4H*a, added 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe; label dated 4/17/01 by A. Jain; lab notebook page number 420/63).

3) Procedure: Measure and adjust pH of U-solution/clinoptilolite mixtures. Adjust pH of *4A, *4B, *4C, and *4G to ~3.2 by adding ~300 microliters of 1.0 M HNO3. Adjust pH of *4D to ~6.2 by adding ~450 mL of 0.1 M NaOH, and that of *4E*a, *4F*a, and *4H*a to ~6.2 by adding ~150 mL of 0.1 M NaOH. Place in gyratory shaker, and unloosen the bottle caps.

1530 hrs. – pH electrode was calibrated using pH standards 4.00 and 7.01; slope = 98.3%

EXAFS*U*4A

Measured pH = 4.81
Added 300 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.50
Plus 50 microL 1 M HNO3; pH = 3.45
Plus 100 microL 1 M HNO3; pH = 3.40
Plus 100 microL 1 M HNO3; pH = 3.37
Placed back on gyratory shaker with cap loosened

EXAFS*U*4B

Measured pH = 5.17
Added 300 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.46
Plus 225 microL 1 M HNO3; pH = 3.36
Placed back on gyratory shaker with cap loosened

EXAFS*U*4D

Measured pH = 5.51
Added 100 microL of 0.1 M NaOH prepared by B.Werling on 4/17/02; pH = 7.22
Placed back on gyratory shaker with cap loosened

EXAFS*U*4E*a

Measured pH = 5.82
Added 50 microL of 0.1 M NaOH prepared by B.Werling on 4/17/02; pH = 7.50
Placed back on gyratory shaker with cap loosened

EXAFS*U*4G

Measured pH = 5.14
Added 300 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.45
Plus 200 microL of 1 M HNO3; pH = 3.31
Placed back on gyratory shaker with cap loosened

EXAFS*U*4H*a

Measured pH = 6.30
Placed back on gyratory shaker with cap loosened

1625 hrs. – pH standard check: pH4 = 4.01; pH 7= 7.04

October 28, 2002

0900 hrs. – pH electrode calibrated; slope = 97.1 %

EXAFS*U*4A

Measured pH = 3.49
Added 100 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.30
Placed back on gyratory shaker with cap loosened

EXAFS*U*4B

Measured pH = 3.54
Added 100 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.34
Placed back on gyratory shaker with cap loosened

EXAFS*U*4D

Measured pH = 6.57±0.05
Added 10 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 6.13±0.03
Placed back on gyratory shaker with cap loosened

EXAFS*U*4E*a

Measured pH = 6.88
Added 10 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 6.04±0.05
Placed back on gyratory shaker with cap loosened

EXAFS*U*4G

Measured pH = 3.46
Added 50 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.35
Placed back on gyratory shaker with cap loosened

EXAFS*U*4H*a

Measured pH = 6.64±0.05
Added 10 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 5.97±0.05
Placed back on gyratory shaker with cap loosened

1250 hrs. – Calibrate a different electrode (also different pH meter); slope = 99.4%

EXAFS*U*4A

Measured pH = 3.44
Added 75 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.32
Placed back on gyratory shaker with cap loosened

EXAFS*U*4B

Measured pH = 3.46
Added 100 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.33
Placed back on gyratory shaker with cap loosened

EXAFS*U*4D

Measured pH = 6.19±0.03
Placed back on gyratory shaker with cap loosened

EXAFS*U*4E*a

Measured pH = 6.30±0.05
Placed back on gyratory shaker with cap loosened

EXAFS*U*4G

Measured pH = 3.47
Added 75 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.34
Placed back on gyratory shaker with cap loosened

EXAFS*U*4H*a

Measured pH = 6.23±0.05
Placed back on gyratory shaker with cap loosened

Check pH standards: pH4 = 4.03; pH7 = 7.03

1100 hrs – Procedure: Take 2.0 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf) from the stock kept in the dessicator (over saturated NaCl solution). Place in a mortar and grind with a pestle until the powder has a consistency of flour. Transfer the powder into two centrifuge tubes, add water and place in a beaker in an ultrasonic cleaner. Turn on ultrasonic cleaner for about 3 to 5 minutes. Transfer the tubes into a centrifuge. Centrifuge the mixtures for about 3 to 4 minutes, with speed setting of 10. Decant the solution phase. **This procedure was done about six times to eliminate much of the highly reactive surfaces and fine particles that were generated by the grinding process.**

It is hoped that this procedure would minimize any potential dissolution of the clinoptilolite during the sorption procedure.

The clinoptilolite powder (sample name CDV*200/325*UC*WA*RC*HL*Rfe*Naf*<325) was collected by filtration using a #5 qualitative paper filter. The wet filtrate and filter paper were placed in an oven at 50 C for about two hours. Then the dry filter paper with clinoptilolite was transferred into the dessicator overnight to equilibrate with water vapor over a saturated NaCl solution.

October 29, 2002

0914 hrs. -- Measure and adjust pH of U-solution/clinoptilolite mixtures:

EXAFS*U*4A

Measured pH = 3.40
Added 20 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.30
Placed back on gyratory shaker with cap loosened

EXAFS*U*4B

Measured pH = 3.41
Added 70 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.31
Placed back on gyratory shaker with cap loosened

EXAFS*U*4D

Measured pH = 6.43±0.05
Added 10 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 6.01±0.05
Placed back on gyratory shaker with cap loosened

EXAFS*U*4E*a

Measured pH = 6.7±0.1
Added 10 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 6.00±0.05
Placed back on gyratory shaker with cap loosened

EXAFS*U*4G

Measured pH = 3.40
Added 70 microL of 1 M HNO3 prepared by RTP on 7/11/02; pH = 3.30
Placed back on gyratory shaker with cap loosened

EXAFS*U*4H*a

Measured pH = 6.3±0.1
Placed back on gyratory shaker with cap loosened

Check pH standards: pH4 = 4.06; pH7 = 7.06

0945 hrs. – recalibrate the pH electrode; slope = 99.2
Measured the pH of EXAFS*U*4C (no zeolite added yet) = 4.46

Measured the pH of EXAFS*U*4F (no zeolite added yet) = 4.88

Check pH standard: pH4 = 3.98

1015 hrs. -- The weight of the CDV*200/325*UC*WA*RC*HL*Rfe*Naf*<325 collected yesterday and measured to be 1.90 g. Of that amount, 0.90 g was transferred into the EXAFS*U*4C bottle. The bottle was shaken to mix the solution and powder, and then was placed on the gyratory shaker with its cap loosened.

250 g of the EXAFS*U*4F solution was transferred into a 250-mL bottle labeled EXAFS*U*4F*a (wt of btl empty = 30.58; plus U solution = 280.59 g). Then 1.00 g of CDV*200/325*UC*WA*RC*HL*Rfe*Naf*<325 was transferred into the EXAFS*U*4F*a bottle. The bottle was shaken to mix the solution and powder, and then was placed on the gyratory shaker with its cap loosened.

1412 hrs. -- calibrated pH electrode with pH 4 and 7 standards; slope = 99.1 %

Measured pH of EXAFS*U*4C = 5.17; added 600 microL of 1 M HNO3; pH = 3.28

Measured pH of EXAFS*U*4F*a = 6.35±0.05

Measured pH of EXAFS*U*4D = 6.32±0.03

Measured pH of EXAFS*U*4E*a = 6.25±0.03

Measured pH of EXAFS*U*4H*a = 6.27±0.03

October 30, 2002

1030 hrs. -- calibrated pH electrode with pH 4 and 7 standards; slope = 99.2 %

Measured pH of EXAFS*U*4A = 3.34; added 20 microL of 1 M HNO3; pH = 3.30

Measured pH of EXAFS*U*4B = 3.34; added 20 microL of 1 M HNO3; pH = 3.31

Measured pH of EXAFS*U*4C = 3.46; added 100 microL of 1 M HNO3; pH = 3.30

Measured pH of EXAFS*U*4D = 6.32±0.05

Measured pH of EXAFS*U*4E*a = 6.30±0.05

Measured pH of EXAFS*U*4F*a = 6.8±0.1; added 10 microL of 1M HNO3; pH = 5.6±0.1

Measured pH of EXAFS*U*4G = 3.33; added 20 microL of 1 M HNO3; pH = 3.29

Measured pH of EXAFS*U*4H*a = 6.33±0.03

Check pH of standards: pH4 = 4.04; pH7 = 7.03

October 31, 2002

0950 hrs. – check pH standards: pH4 = 4.00, pH7 = 7.01

Measured pH of EXAFS*U*4A = 3.31

Measured pH of EXAFS*U*4B = 3.32

Measured pH of EXAFS*U*4C = 3.37; added 30 microL of 1 M HNO3; pH = 3.31

Measured pH of EXAFS*U*4D = 6.30±0.03

Measured pH of EXAFS*U*4E*a = 6.29±0.01

Measured pH of EXAFS*U*4F*a = 6.21±0.01

Measured pH of EXAFS*U*4G = 3.31

Measured pH of EXAFS*U*4H*a = 6.24±0.03

Check pH of standards: pH4 = 4.03; pH7 = 7.04

November 1, 2002

1230 hrs. – check pH standards: pH4 = 4.00, pH7 = 7.00

Measured pH of EXAFS*U*4A = 3.30

Measured pH of EXAFS*U*4B = 3.29

Measured pH of EXAFS*U*4C = 3.32

Measured pH of EXAFS*U*4D = 6.25±0.01

Measured pH of EXAFS*U*4E*a = 6.23±0.03

Measured pH of EXAFS*U*4F*a = 6.46±0.03; added 5.5 microL of 1 M HNO3; pH = 4.3±0.1

Measured pH of EXAFS*U*4G = 3.28

Measured pH of EXAFS*U*4H*a = 6.20

Check pH of standards: pH4 = 4.06; pH7 = 7.05

1600 hrs. – check pH standards: pH4 = 3.99; pH7 = 6.99

Measured pH of EXAFS*U*4F*a = 4.65±0.03; added 140 microL of 0.1 M NaOH prepared by B. Werling on 4/17/02 (lab notebook 463 p.127); pH = 6.28±0.01

November 4, 2002

1030 hrs. – calibrated pH electrode using pH4 and pH7 standards; slope = 99.0%

Measured pH of EXAFS*U*4A = 3.30

Measured pH of EXAFS*U*4B = 3.30

Measured pH of EXAFS*U*4C = 3.35

Measured pH of EXAFS*U*4D = 6.29±0.02

Measured pH of EXAFS*U*4E*a = 6.26±0.05

Measured pH of EXAFS*U*4F*a = 6.31±0.02

Measured pH of EXAFS*U*4G = 3.28

Measured pH of EXAFS*U*4H*a = 6.28±0.03

Check pH of standards: pH4 = 4.02; pH7 = 7.04

From each of the U-solution/clinoptilolite mixtures, ~20 mL of aqueous sample was taken (unfiltered) and transferred onto a 60-mL PP bottle for later analysis of U concentration. The following are the weights of the samples taken:

Sample	Wt. Bottle empty	Wt. Bottle+sample	Wt. Sample
EXAFS*U*4A*FU	14.24	34.39	20.15
EXAFS*U*4B*FU	14.28	34.35	20.07
EXAFS*U*4C*FU	14.89	34.92	20.03
EXAFS*U*4D*FU	14.89	34.93	20.04
EXAFS*U*4E*a*FU	14.11	34.07	19.96
EXAFS*U*4F*a*FU	14.89	34.87	19.98
EXAFS*U*4G*FU	14.10	34.10	20.00
EXAFS*U*4H*a*FU	14.93	34.90	19.97

For each U-solution/clinoptilolite mixture, the solid phase was separated from the U solution using filtration (200-325 mesh clinoptilolite) or centrifugation (<325 mesh clinoptilolite). For filtration, Fisherbrand qualitative P8 filter (catalog # 09-795G) was placed on a glass filter funnel. The filter paper has a fast flow rate and did not require vacuum assistance. Samples EXAFS*U*4C and EXAFS*U*4F*a have clinoptilolite that is too fine-grained to be effectively separated by filtration. For these samples, a centrifuge was used, set to between 9000 and 12600 rpm (repetitive steps were necessary to decrease the volume).

Sample	Wt. Bottle empty	Wt. Bottle+filtrant	Wt. Filtrant
EXAFS*U*4A*filtrant	55.06	493.26	438.2
EXAFS*U*4B*filtrant	54.74	488.77	434.03
EXAFS*U*4C*filtrant	54.95	487.48	432.53
EXAFS*U*4D*filtrant	54.81	480.64	425.83
EXAFS*U*4E*a*filtrant	30.50	254.07	223.57
EXAFS*U*4F*a*filtrant	30.53	Not measured	---
EXAFS*U*4G*filtrant	53.96	488.14	434.18
EXAFS*U*4H*a*filtrant	30.62	254.14	223.52

After filtration, the wet pastes were transferred into plastic vials that will be shipped to Argonne National Laboratory. The vials were labeled as 4A, 4B, 4C....

Approximately 20 mL of the filtrant solutions were taken and transferred into 60-mL PP bottles for later analysis of U concentration:

Sample	Wt. Bottle empty	Wt. Bottle+filtrant	Wt. Filtrant
EXAFS*U*4A*filtrant*FU	14.03	33.94	19.91
EXAFS*U*4B*filtrant*FU	14.92	34.77	19.85
EXAFS*U*4C*filtrant*FU	14.26		
EXAFS*U*4D*filtrant*FU	14.20	33.99	19.79
EXAFS*U*4E*a*filtrant*FU	14.12	34.00	19.88
EXAFS*U*4F*a*filtrant*FU	14.28	34.31	20.03
EXAFS*U*4G*filtrant*FU	12.40	32.43	20.03
EXAFS*U*4H*a*filtrant*FU	12.29	32.22	19.93

NOTE: Separation of the solids in mixtures EXAFS*U*4C and EXAFS*U*4F*a were not done today due to time constraint. Will be done tomorrow.

1620 hrs. -- After filtration, the pH of the filtrant solutions were measured:

check pH standards: pH4 = 4.02; pH7 = 7.01

pH of EXAFS*U*4A*filtrant = 3.35
pH of EXAFS*U*4B*filtrant = 3.29
pH of EXAFS*U*4D*filtrant = 6.27±0.03
pH of EXAFS*U*4E*a*filtrant = 6.30±0.03
pH of EXAFS*U*4G*filtrant = 3.28
pH of EXAFS*U*4H*filtrant = 6.31±0.03

November 5, 2002

0930 hrs. – check pH of standards: pH4 = 4.00; pH7 = 7.02

pH of EXAFS*U*4C = 3.36

pH of EXAFS*U*4F*a = 6.42

recheck standards: pH7 = 7.06; pH4 = 4.03

0945 hrs. – Separation of the solids from mixtures EXAFS*U*4C and EXAFS*U*4F*a was initiated. The mixtures were decanted into 40-mL centrifuge tubes and placed in a centrifuge. A speed between 9,000 and 12,600 rpm was used for periods between 5 and 10 minutes. The clear solutions were decanted into a PP bottle (labeled EXAFS*U*4C*filtrant or EXAFS*U*4F*a*filtrant). The remaining solid and liquid were shaken and mixed, and then transferred into another centrifuge tube, in order to eventually collect as much of the the solid phase into a single tube. The process was repeated until most of the zeolite was collected as a wet paste in a single tube. The excess solution was pipetted out, leaving a wet cake. A spatula was used to retrieve the wet cake for transfer into a plastic vial (labeled 4C and 4F) which will be shipped to Argonne National Laboratory.

(In the process of processing sample EXAFS*U*4F*a, approximately 20mL of deionized water were inadvertently mixed with the decanted solution. Thus, the total weight of the aqueous separate for this sample was not measured. Approximately 20-mL of the clear solution was taken from one of the centrifuge tubes, for later U analysis.)

1430 hrs. – Uranium-loaded clinoptilolite samples 4A, 4B, 4C, 4D, 4E, and 4F, in plastic vials covered with aluminum foil, were shipped to Dr. Lynne Soderholm at Argonne National Laboratory via FEDEX next day air service.

Samples 4G and 4H were kept in the lab. The pH of the wet paste will be monitored.

November 17, 2002

The following is an email from Richard Reeder (SUNY) regarding the uranium-loaded samples he analyzed at the APS facility (Argonne National Laboratory):

From: rjreeder@notes.cc.sunysb.edu
Sent: Sunday, November 17, 2002 11:26 AM
To: rpabalan@cnwra.swri.edu
Subject: RE: sample holders

Hi Bobby,

Sorry that I haven't updated you about the recent beamtime at the APS. I was running alone this time, and I spent all my "free time" talking with members of various CATs about future opportunities. The APS has just made a change-over to a new system of beamtime proposals. This heralds a potentially major change in how beamtime will be made available, and I felt it was important for me to be "in" on current thinking there.

Anyway, I have good and bad news about the samples. Four of the sample were dry when I opened them for loading; these were 4A, 4B, 4D, and 4E. They had dried into a rigid mass in the sample vial, but probably not completely free of moisture. After prodding with a spatula, the material came free in chunks. I loaded these anyway. When I scanned them, however, I found the same result as last time. The fluorescence yield increased with time, indicating that the remaining moisture was being lost.

Samples 4C and 4F were moist in the sample vials, with a small amount of liquid visible. I had to stir these up to homogenize them them before loading into the sample holders; but they were clearly wet in the sense that water was present b/t particles after I got them into the holders. I spent most of the beamtime on these and collected good scans. There was no change in fluorescence yield over 15 or scans, so I'm confident that our prior experience was simply sample dryness. I haven't had a chance to analyze the data on these sample yet, but will try to get to them soon.

I see the good samples were the ones that you separated by centrifugation, leaving a bit of supernatant. I recall that being the case for previous samples that we ran at NSLS. So, I think this is the best procedure to use in the future. But it's also possible that some of the moisture was lost while in the sample vials (can't be sure of that). I also wonder if it's possible that clinoptilolite absorbs or exchanges water over the time frame of the samples sitting in the vials(?).

I'll update you when I've got some fits.

Best,

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

February 3, 2003

The following is an email from Richard Reeder (SUNY) advising of beamtime availability on March 12, 2003:

From: rjreeder@notes.cc.sunysb.edu
Sent: Monday, February 03, 2003 2:33 PM
To: rpabalan@swri.edu
Subject: Update and plans

Bobby,

Just a few things. Aaron has taken a few of the U-exchanged single crystals to the ESRF to see if we can get exchange. We should have some idea of what he gets in a week or so. I also want to revisit the Np sorption at beamtime coming up March 12 at the APS. When we ran Np on clinoptilolite before you will recall that we had this problem of sample drying. I'm confident that we can get around that by just keeping the sample wetter. But I'm wondering if we might not be better off trying Np sorption on montmorillonite instead of (or in addition to) clinop. Are you aware if Np on mont has been looked at using a surface spectroscopic method? If it's been done, then I don't want to duplicate that. And I still think we can get good Np on clinop spectra.

What do you think, and would you be able to put samples together for a March 13 run?

Rich

Richard J. Reeder, Professor
Dept. of Geosciences
SUNY at Stony Brook
Stony Brook NY 11794-2100

Tel. (631) 632-8208
Fax (631) 632-8240

February 17, 2003

Preparation of Uranium- and Neptunium-loaded Clinoptilolite and Montmorillonite Samples for XAS Analysis

Objective: Clinoptilolite and montmorillonite sorbed with Np(V) at pH~3 and clinoptilolite with sorbed U(VI) at pH~3 and ~6.5 will be prepared for XAS analysis at Argonne National Laboratory.

1400 hrs. –

Procedure:

1. Prepared $5x10^{-3}$ m uranium nitrate solution

Weighed 1.2550 g $^{238}UO_2(NO_3)_2 \cdot 6H_2O$ reagent (Mallinkrodt Lot # 8640KCAP) in a weighing boat. Transferred the solids into a 500-mL polypropylene bottle. (Weighing boat had a tared weight of 0.0000 g after the transfer).

Tared 500 g of deionized water into the bottle. The bottle was capped, then shaken to dissolve the uranium solid.

After calibration of the pH electrode (pH 4 and 7 standards, slope=99.4%), the pH of the $5x10^{-3}$ m uranium solution was measured to be 3.40. The value calculated using GWB is 3.378 (SEE NOTEBOOK #278 PAGE 324). The measured pHs of standard solutions are 3.98 (pH 4 standard) and 7.01 (pH 7 standard).

February 18, 2003

1330 hrs. –

Preparation of Uranium-loaded Clinoptilolite for EXAFS Analysis (scheduled for March 12, 2003)

2. Using the 5.0E-3 molal uranyl nitrate solution that was prepared yesterday, uranium solutions of various concentrations were prepared.

   a) Procedure: Tare 20 g of 5.E-3 molal $UO_2(NO_3)_2$ solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5A). Add deionized H_2O to give a total solution mass of 500 g. Calculated [U] = 2.0E-4 molal.

Wt. Bottle w/ cap:	54.70 g
Plus 20 g U soln:	74.70 g
Plus H_2O:	554.72 g
Mass of U soln:	**500.02 g**

   b) Procedure: Tare 10 g of 5.E-3 molal $UO_2(NO_3)_2$ solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5B).). Add deionized H_2O to give a total solution mass of 500 g. Calculated [U] = 1.0E-4 molal.

Wt. Bottle w/ cap:	54.69 g
Plus 10 g U soln:	64.70 g

Plus H2O:	554.78 g
Mass of U soln:	**500.09 g**

c) Procedure: Tare 10 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5C).). Add deionized H2O to give a total solution mass of 500 g. Calculated [U] = 1.0E-4 molal.

Wt. Bottle w/ cap:	54.52 g
Plus 10 g U soln:	64.52 g
Plus H2O:	554.51 g
Mass of U soln:	**499.99 g**

d) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3) solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5D). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

Wt. Bottle w/ cap:	54.55 g
Plus 2.5 g U soln:	57.06 g
Plus H2O:	554.56 g
Mass of U soln:	**500.01 g**

e) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5E). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

Wt. Bottle w/ cap:	54.69 g
Plus 2.5 g U soln:	57.17 g
Plus H2O:	554.69 g
Mass of U soln:	**500.00 g**

f) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5F). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

Wt. Bottle w/ cap:	54.63 g
Plus 2.5 g U soln:	57.12 g
Plus H2O:	554.66 g
Mass of U soln.:	**500.03 g**

g) Procedure: Tare 10 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5G). Add deionized H2O to a total mass of 500 g. Calculated [U] = 1.0E-4 molal.

Wt. Bottle w/ cap:	54.48 g
Plus 10 g U soln:	64.48 g
Plus H2O:	554.49 g
Mass of U soln.:	**500.01 g**

h) Procedure: Tare 2.50 g of 5.E-3 molal UO2(NO3)2 solution into a 500-mL polypropylene bottle (labeled EXAFS*U*5H). Add deionized H2O to a total mass of 500 g. Calculated [U] = 2.5E-5 molal.

Wt. Bottle w/ cap:	54.49 g
Plus 2.5 g U soln:	56.98 g
Plus H2O:	554.57 g
Mass of U soln.:	**500.08 g**

3. Take 50 mL aliquots of EXAFS*U*5A through EXAFS*U*5H using a 10-mL Oxford pipet (variable volume pipet, set to 10-mL). Transfer the aliquots into a 60-mL PP bottles labeled EXAFS*U*5A*IU, EXAFS*U*5B*IU, EXAFS*U*5C*IU, ... Set the solution aside for later analysis of U concentration (acidify later). Reweigh the bottles EXAFS*U*5A through EXAFS*U*5H:

EXAFS*U*5A:	503.24 (solution left = 448.54 g)
EXAFS*U*5B:	503.47 (solution left = 448.78 g)
EXAFS*U*5C:	503.33 (solution left = 448.81 g)
EXAFS*U*5D:	503.31 (solution left = 448.76 g)
EXAFS*U*5E:	503.47 (solution left = 448.78 g)
EXAFS*U*5F:	503.49 (solution left = 448.86 g)
EXAFS*U*5G:	503.41 (solution left = 448.93 g)
EXAFS*U*5H:	503.44 (solution left = 448.65 g)

For mixture EXAFS*U*5E, transferred (tared) 250 g of the U solution into a 250-mL PP bottle labeled EXAFS*U*5E*a:

Wt. EXAFS*U*5E*a bottle empty:	30.41 g
Wt. EXAFS*U*5E*a bottle w/ U soln:	280.40 g
Wt. Of U solution:	249.99 g

Wt. Of EXAFS*U*5E bottle with leftover U soln:	253.41 g
Wt. Of U soln. Left in EXAFS*U*5E bottle:	198.72 g

For mixture EXAFS*U*5H, transferred (tared) 250 g of the U solution into a 250-mL PP bottle labeled EXAFS*U*5H*a:

Wt. EXAFS*U*5H*a bottle empty:	30.37 g
Wt. EXAFS*U*5H*a bottle w/ U soln:	280.40 g
Wt. Of U solution:	250.03 g

Wt. Of EXAFS*U*5H bottle with leftover U soln:	253.39 g
Wt. Of U soln. Left in EXAFS*U*5H bottle:	198.90 g

February 19, 2003

0915 hrs. – Prepared six U-solution/clinoptilolite mixtures (two will be prepared later)

a) Mixture EXAFS*U*5A

Procedure: Transfer 50 mL of the 2E-4 molal U nitrate solution (EXAFS*U*5A) into a 60-mL PP bottle labeled EXAFS*U*5A*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5A: 503.24 g

To the remaining 450 g of U solution, added 1.00 g of pretreated clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe prepared by A. Jain on 5/2/01; SN 420/63). The clinoptilolite was kept stored in a glass jar. The same material previously was used to prepare the homoionic forms of clinoptilolite (e.g., Na-clinoptilolite, SN 420/54-67).

The bottle was placed on a gyratory shaker with the cap loose to allow CO2(g) to enter.

b) Mixture EXAFS*U*5B

Procedure: Transfer 50 mL of the 1E-4 molal U nitrate solution (EXAFS*U*5B) into a 60-mL PP bottle labeled EXAFS*U*5B*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5B: 503.47 g

To the remaining 450 g of U solution, added 1.00 g of pretreated clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe).

The bottle was placed on a gyratory shaker with the cap loose to allow CO2(g) to enter.

c) Mixture EXAFS*U*5C

Procedure: Transfer 50 mL of the 1E-4 molal U nitrate solution (EXAFS*U*5C) into a 60-mL PP bottle labeled EXAFS*U*5C*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5C: 503.33 g

d) Mixture EXAFS*U*5D

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*5D) into a 60-mL PP bottle labeled EXAFS*U*5D*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5D: 503.31 g

To the remaining 450 g of U solution, added 1.00 g of pretreated clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe).

The bottle was placed on a gyratory shaker with the cap loose to allow CO2(g) to enter.

e) Mixture EXAFS*U*5E

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*5E) into a 60-mL PP bottle labeled EXAFS*U*5E*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5E: 503.47 g

Transferred (tared) 250 g of the U solution into a 250-mL PP bottle labeled EXAFS*U*5E*a, added 1.00 g of pretreated clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe).

The bottle was placed on a gyratory shaker with the cap loose to allow CO2(g) to enter.

Wt. Of bottle labeled EXAFS*U*5E*a empty:	30.41 g
Wt. with 250-mL of U solution:	280.40 g
Calculated weight of U solution added:	249.99 g

Wt. Of EXAFS*U*5E bottle with leftover solution: 253.41 g

f) Mixture EXAFS*U*5F

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*5F) into a 60-mL PP bottle labeled EXAFS*U*5F*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5F: 503.49 g

g) Mixture EXAFS*U*5G

Procedure: Transfer 50 mL of the 1E-4 molal U nitrate solution (EXAFS*U*5G) into a 60-mL PP bottle labeled EXAFS*U*5G*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5G: 503.41 g

To the remaining 450 g of U solution, add 1.00 g of pretreated clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe).

The bottle was placed on a gyratory shaker with the cap loose to allow CO2(g) to enter.

h) Mixture EXAFS*U*5H

Procedure: Transfer 50 mL of the 2.5E-5 molal U nitrate solution (EXAFS*U*5H) into a 60-mL PP bottle labeled EXAFS*U*5H*IU. Set the solution aside for later analysis of U concentration (acidify later).

Wt. Of Bottle labeled EXAFS*U*5H: 503.44

Transferred (tared) 250 g of the U solution into a 250-mL PP bottle labeled EXAFS*U*5H*a, added 1.00 g of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe).

The bottle was placed on a gyratory shaker with the cap loose to allow CO2(g) to enter.

Wt. Of bottle labeled EXAFS*U*5H*a empty:	30.37 g
Wt. with 250-mL of U solution:	280.40 g
Calculated weight of U solution added:	250.03 g

Wt. Of EXAFS*U*5H bottle with leftover solution: 253.39 g

Measured weights of bottles with U-solution/clinoptilolite mixtures:

EXAFS*U*5A:	504.23
EXAFS*U*5B:	504.45
EXAFS*U*5D:	504.29
EXAFS*U*5E*a:	281.43
EXAFS*U*5G:	504.39
EXAFS*U*5H*a:	281.39

1250 hrs. –

The pH of the U-solution/clinoptilolite mixtures was measured and adjusted:

pH4 standard check: 4.01
pH7 standard check: 7.01

pH of EXAFS*U*5A: 4.73
added 300 microliters of 1-N HNO3; pH = 3.28

pH of EXAFS*U*5B: 4.98
added 300 microliters of 1-N HNO3; pH = 3.40
added 100 microliters of 1-N HNO3; pH = 3.26

pH of EXAFS*U*5D: 5.52
added 100 microliters of 0.1-N NaOH (prepared by B. Werling on 4/17/02; SN 463/127); measured pH is 6.80 and decreasing.

pH of EXAFS*U*5E*a: 6.03±0.05
Added 25 microliters of 0.1-N NaOH; pH = 6.87 and decreasing

pH of EXAFS*U*5G: 5.07
Added 300 microliters of 1-N HNO3; pH = 3.69 and increasing
Added 100 microliters of 1-N HNO3; pH = 3.49
Added 100 microliters of 1-N HNO3; pH = 3.34

pH of EXAFS*U*5H: 6.02±0.03
Added 10 microliters of 0.1-N NaOH; pH = 6.59±0.05

pH7 standard check = 7.04
pH4 standard check = 4.03

1330 hrs. –

To each of the 50 mL U solution aliquots in bottles labeled EXAFS*U*5A*IU, EXAFS*U*5B*IU, EXAFS*U*5C*IU, EXAFS*U*5D*IU, EXAFS*U*5E*IU, EXAFS*U*5F*IU, EXAFS*U*5G*IU, and EXAFS*U*5H*IU, 1 mL of 1-N HNO3 solution (prepared by B.Werling on 2/18/03, notebook 556, p. 22) was added using a 1000 microliter Eppendorf pipet to acidify the solution prior to chemical analysis.

In addition, 1-N HNO3 solutions were also added to the samples of U solutions prepared for the previous EXAFS run (see page 384 and succeeding pages). 400 microliters of 1-N HNO3 solution were added to the bottles labeled EXAFS*U*4A*IU through EXAFS*U*4H*IU, EXAFS*U*4A*FU through EXAFS*U*4H*FU, and EXAFS*U*4A*filtrant*FU through EXAFS*U*4H*filtrant*FU.

1400 hrs. –

This morning, Paul Bertetti opened two ampoules containing neptunium nitrate in 4 M HNO3. The neptunium nitrate was purchased from Isotope Products and had ~5 microcuries in ~5 g of solution. To reduce the HNO3 concentration, Paul evaporated the Np nitrate solution to near dryness, and redissolved it in several milliliters of 0.001 m HNO3. This was repeated several times. After the last redissolution, the neptunium nitrate solution was transferred into a volumetric flask and diluted to 500 mL with 0.001 m HNO3. Paul took one 1-mL sample of the Np solution for analysis of Np concentration by liquid scintillation counting. The preparation of the Np solution is documented in Paul's scientific notebook (SN 369, p. 62).

0.500 grams of Na-clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf prepared by A. Jain on 5/2/01; SN 420/54-67) was tared onto a weighing paper, then transferred into a 500-mL PP bottle labeled EXAFS*Np*1A. The Na-clinoptilolite was kept stored inside a dessicator over a saturated NaCl solution.

The Np solution was transferred from the volumetric flask into the PP bottle labeled EXAFS*Np*1A. The bottle was placed on a gyratory shaker with its cap loose.

1500 hrs. –

2.50 grams of pretreated clinoptilolite powder (CDV*200/325*UC*WA*RC*HL*Rfe prepared by A. Jain on 5/2/01; SN 420/63) was ground in a mortar and pestle until the powder had the

consistency of flour. The powder was transferred into two centrifuge tubes (1.25 grams of powder in each tube). Water was added to each tube. Both tubes were placed in a beaker two-thirds full of water. The beaker was placed in an ultrasonic cleaner. The ultrasonic cleaner was turned on for about 3 to 5 minutes. The tubes were then placed into a centrifuge, which was run for 3 to 5 minutes, with speed setting of 10. The solution phase was decanted from each tube and the water replaced. **This procedure was done six times to eliminate much of the highly reactive surfaces and fine particles that were generated by the grinding process. It is hoped that this procedure would minimize any potential dissolution of the clinoptilolite during the sorption procedure.**

The ground clinoptilolite powder was collected by filtration using a #1 qualitative paper filter placed in a funnel. Because of the slow filtration process, much of the solution was removed by absorbing onto Kimwipes. The filter paper with wet paste of clinoptilolite powder was placed on a watch glass, and placed inside a dessicator over a saturated NaCl solution and left overnight. The same dessicator is where the homoionic forms of clinoptilolite are stored.

February 20, 2003

0830 hrs. –

Measure the pH of the U- or Np-solution/clinoptilolite mixtures:

pH4 standard check = 4.01
pH7 standard check = 7.01

EXAFS*U*5A: pH = 3.68
Added 200 microliters of 1-N HNO3; pH = 3.38
Added 100 microliters of 1-N HNO3; pH = 3.28

EXAFS*U*5B: pH = 3.59
Added 100 microliters of 1-N HNO3; pH = 3.41
Added 100 microliters of 1-N HNO3; pH = 3.29

EXAFS*U*5D: pH = 6.57
Added 40 microliters of 0.1-N HNO3; pH = 6.37±0.05

EXAFS*U*5E*a: pH = 6.64±0.05
Added 40 microliters of 0.1-N HNO3; pH = 6.35±0.03

EXAFS*U*5G: pH = 3.51
Added 100 microliters of 1-N HNO3; pH = 3.33

EXAFS*U*5H*a: pH = 6.55±0.03
Added 100 microliters of 1-N HNO3; pH = 4.35
Added 300 microliters of 0.1-N NaOH; pH = 5.00
Added 50 microliters of 1-N NaOH; pH = 6.54
Added 10 microliters of 0.1-N HNO3; pH = 6.07±0.03

EXAFS*Np*1A: pH = 3.44
Added 200 microliters of 1-N HNO3; pH = 3.29

0920 hrs. –

pH4 standard check = 4.02
pH standard check = 7.02

The bottles of U-solution/clinoptilolite mixtures were placed back on the gyratory shaker.

0930 hrs. –

The wet paste of ground clinoptilolite left in the dessicator overnight was taken out of the dessicator. It was observed to be dry. The powder was scraped off the filter paper and transferred onto a weighing

paper. The measured mass is 1.80 grams. Of that amount, 0.90 grams were transferred into the bottle labeled EXAFS*U*5C.

250 g of the EXAFS*U*5F solution was transferred into a 250-mL bottle labeled EXAFS*U*5F*a (wt of btl empty = 30.48; plus U solution = 280.50 g). Then the remaining 0.90 g of the ground clinoptilolite powder was transferred into the EXAFS*U*5F*a bottle.

Both bottles were shaken to mix the solution and powder, and then placed on the gyratory shaker with their caps loose to allow CO2(g) in.

1515 hrs. –

Measure the pH of the U- or Np-solution/clinoptilolite mixtures:

pH4 standard check = 3.99
pH7 standard check = 7.00

EXAFS*U*5A: pH = 3.33

EXAFS*U*5B: pH = 3.32

EXAFS*U*5C: pH = 5.21
Added 2 mL of 1-N HNO3, pH = 2.49
Added 500 microliters of 1-N NaOH; pH = 2.69
Added 500 microliters of 1-N NaOH; pH = 2.93

EXAFS*U*5D: pH = 6.57±0.05
Added 30 microliters of 0.1-N HNO3; pH = 6.33±0.03

EXAFS*U*5E*a: pH = 6.66±0.03
Added 30 microliters of 0.1-N HNO3; pH = 6.32±0.03

EXAFS*U*5F*a: pH = 6.79±0.03
Added 50 microliters of 0.1-N HNO3; pH = 6.37±0.03

EXAFS*U*5G: pH = 3.37
Added 10 microliters of 0.1-N HNO3 and 40 microliters of 1-N HNO3; pH = 3.32

EXAFS*U*5H*a: pH = 6.07
Added 40 microliters of 0.1-N NaOH; pH = 6.26±0.03

EXAFS*Np*1A: pH = 3.27

1400 hrs. –

pH4 standard check = 4.01
pH7 standard check = 7.01

The bottles were placed back on the gyratory shaker.

February 21, 2003

0820 hrs. –

Measure the pH of the U- or Np-solution/clinoptilolite mixtures:

pH4 standard check = 4.01
pH7 standard check = 7.01

EXAFS*U*5A: pH = 3.39
Added 50 microliters of 1-N HNO3; pH = 3.31

EXAFS*U*5B: pH = 3.38
Added 50 microliters of 1-N HNO3; pH = 3.31

EXAFS*U*5C: pH = 2.94
Added 250 microliters of 1-N NaOH; pH = 3.27

EXAFS*U*5D: pH = 6.57
Added 25 microliters of 0.1-N HNO3; pH = 6.35

EXAFS*U*5E*a: pH = 6.53±0.03
Added 25 microliters of 0.1-N HNO3; pH = 6.24

EXAFS*U*5F*a: pH = 6.65±0.05
Added 45 microliters of 0.1-N HNO3; pH = 6.32

EXAFS*U*5G: pH = 3.41
Added 50 microliters of 1-N HNO3; pH = 3.31

EXAFS*U*5H*a: pH = 6.12
Added 25 microliters of 0.1-N NaOH; pH = 6.27

EXAFS*Np*1A: pH = 3.39
Added 60 microliters of 1-N HNO3; pH = 3.31

0905 hrs. –

pH4 standard check = 4.04
pH7 standard check = 7.04

1510 hrs. –

Measure the pH of the U- or Np-solution/clinoptilolite mixtures:

pH4 standard check = 4.00
pH7 standard check = 7.00

EXAFS*U*5A: pH = 3.32

EXAFS*U*5B: pH = 3.32

EXAFS*U*5C: pH = 3.20

EXAFS*U*5D: pH = 6.36±0.02

EXAFS*U*5E*a: pH = 6.43
Added 40 microliters of 0.001-N HNO3 and 20 microliters of 0.1-N HNO3; pH = 6.21

EXAFS*U*5F*a: pH = 6.45±0.03
Added 20 microliters of 0.1-N HNO3; pH = 6.32

EXAFS*U*5G: pH = 3.27

EXAFS*U*5H*a: pH = 6.17

EXAFS*Np*1A: pH = 3.25

1535 hrs. –

pH4 standard check = 4.02
pH7 standard check = 7.02

February 23, 2003 (Sunday)

1700 hrs. –

Calibrated the pH electrode with pH 4 and pH 7 standards; slope = 99.9%

Measured the pH of the U- or Np-solution/clinoptilolite mixtures:

EXAFS*U*5A: pH = 3.38
Added 100 microliters of 1-N HNO3; pH = 3.22

EXAFS*U*5B: pH = 3.37
Added 100 microliters of 1-N HNO3; pH = 3.23±0.03

EXAFS*U*5C: pH = 3.21

EXAFS*U*5D: pH = 6.47±0.03
Added 50 microliters of 0.1-N HNO3; pH = 6.20±0.03

EXAFS*U*5E*a: pH = 6.40±0.02
Added 25 microliters of 0.1-N HNO3; pH = 6.17±0.03

EXAFS*U*5F*a: pH = 6.55
Added 35 microliters of 0.1-N HNO3; pH = 6.25±0.02

EXAFS*U*5G: pH = 3.34
Added 50 microliters of 1-N HNO3; pH = 3.26

EXAFS*U*5H*a: pH = 6.15

EXAFS*Np*1A: pH = 3.34
Added 100 microliters of 1-N HNO3; pH = 3.21

February 27, 2003

1012 hrs. –

Check pH standards: pH 4 = 4.01; pH 7 = 7.00

Measured the pH of the U- or Np-solution/clinoptilolite mixtures:

EXAFS*U*5A: pH = 3.25

EXAFS*U*5B: pH = 3.29

EXAFS*U*5C: pH = 3.21

EXAFS*U*5D: pH = 6.36

EXAFS*U*5E*a: pH = 6.31

EXAFS*U*5F*a: pH = 6.52
Added 25 microliters of 0.1-N HNO3; pH = 6.29±0.01

EXAFS*U*5G: pH = 3.28

EXAFS*U*5H*a: pH = 6.16

EXAFS*Np*1A: pH = 3.28

Check pH standards: pH 4 = 4.04; pH 7 = 7.04

1400 hrs. –

Prepared 0.1-N HNO3 solution by diluting 100 mL of the 1.0-N HNO3 solution (prepared by B.Werling on 2/18/03, notebook 556, p. 22) to 1000 mL with ultrapure water in a volumetric flask.

Two 1-mL samples of the solution in the bottle labeled EXAFS*Np*1A were taken. Each sample was transferred into sample vials containing 500 microliters of the newly prepared 0.1-N HNO3 solution. The vials were shaken to mix the solutions and set aside for later liquid scintillation analysis.

Sample Name	Wt. Scint. Vial empty	Wt. With 500 microL 0.1 N HNO3	Wt. With 1-mL Np solution	Wt. Of Np sample
Np*1A*F1*a	7.2077	7.7073	8.7067	0.9994
Np*1A*F1*b	7.2532	7.7496	8.7508	1.0012

Weight of bottle EXAFS*Np*1A = 548.18 g.

The clinoptilolite in the EXAFS*Np*1A bottle was separated by centrifugation. The process involved transferring the clear Np solution from the polypropylene bottle into six 35-mL centrifuge bottles and centrifuging at 12,700 rpm for 10 mins. The clear solutions in the tubes were transferred into another 500-mL polypropylene bottle labeled EXAFS*Np*1B. When the solution volume in the EXAFS*Np*1A bottle was low, the mixture was shaken to get the clinoptilolite to rise and the slurry was transferred into the centrifuge tubes. Centrifugation and decantation of clear solutions was repeated until the clinoptilolite powder was contained in one centrifuge tube only. A final centrifugation was done, and the clear solution was transferred into the EXAFS*Np*1B bottle, leaving about 5 mL of Np solution in the tube. The tube was set aside covered with an aluminum foil, for later loading of the clinoptilolite powder into the EXAFS sample cell.

Weight of EXAFS*Np*1B bottle empty = 54.70 g
Weight of EXAFS*Np*1B bottle with decanted Np solution = 537.74 g
Weight of Np solution = 483.04 g

0.5 g of montmorillonite (Saz*Na*<2*FD*scr, prepared by P. Bertetti and used in previous U and Np sorption experiments) was added to the EXAFS*Np*1B bottle. The mixture was shaken and the bottle placed on the gyratory shaker, with the cap loose to allow CO2(g) to enter.

Weight of weighing boat empty = 0.5655 g
Weight with montmorillonite = 1.0653 g (observed to decrease to 1.0628 while waiting to transfer, approx. 10 minutes)
Weight of weighing boat after transfer = 0.5696 g
Calc. weight of montmorillonite transferred = 0.4957 g

1440 hrs. –

Measured the pH of Np-solution in the bottle labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2*filtrant (see notebook #278 volume 8, page 31):

Check pH standards: pH7 = 6.99; pH10 = 10.02
pH of Np solution = 7.53
Added 200 microliters of 0.1-N NaOH; pH = 8.02

1500 hrs. –

Measured weight of bottle labeled EXAFS*Np*0.001 m HNO3*filtrant*pH 8.2*filtrant = 747.3 g (initial weight of bottle was not measured; see notebook #278 volume 8).

Re-labeled the bottle EXAFS*Np*1C.

Added 0.500 g of Na-clinoptlilolite (CDV*200/325*UC*WA*RC*HL*Rfe*Naf prepared by A. Jain on 5/2/01; SN 420/54-67) to the bottle. The bottle was shaken then placed on a gyratory shaker with the cap loose.

February 28, 2003

0845 hrs. –

Measured the pH of some U- and Np-bearing mixtures:

Check pH standards: pH 4 = 4.01; pH 7 = 7.01; pH 10 = 10.06; Temp = 18.9 C

EXAFS*Np*1B: pH = 3.62
Added 50 microliters of 0.1-N HNO3; pH = 3.61
Added 200 microliters of 1.0-N HNO3; pH = 3.24

EXAFS*U*5F*a: pH = 6.40

EXAFS*Np*1C: pH = 7.94
Added 200 microliters of 0.1-N NaOH; pH = 8.21

Check pH standards: pH 4 = 4.06; pH 7 = 7.05; pH 10 = 10.10; Temp = 19.1 C.

1350 hrs. –

Calibrated pH electrode using pH 4, 7, and 10 standards, slope = 99.4 %; Temp = 20.9 C.

Measure pH of Np- or U-bearing mixtures:

EXAFS*Np*1B: pH = 3.23
EXAFS*U*5F*a: pH = 6.53±0.03
Added 25 microliters of 0.1-N HNO3; pH = 6.19
EXAFS*Np*1C: pH = 7.94
Added 100 microliters of 0.1-N NaOH; pH = 8.21

Check pH standards: pH 4 = 4.03; pH 7 = 7.04; pH 10 = 10.08; Temp = 21 C.

1440 hrs. –

Check pH standards: pH 4 = 4.01; pH 7 = 7.01

Measured pH of some U-bearing mixtures:

EXAFS*U*5A: pH = 3.25
EXAFS*U*5B: pH = 3.27
EXAFS*U*5A: pH = 3.20

Measured weight of bottle labeled EXAFS*U*5A = 504.17 g.

Filtered the solution through a Fisher brand P8 qualitative filter paper (fast flow rate, Cat. No. 09-795G, 19 cm diameter). A small volume of U solution remaining in the polypropylene bottle was transferred into a plastic LSA vial (labeled U*5A), and the wet clinoptilolite paste on the filter paper was

scooped with a teflon coated spatula and transferred into the vial. The vial was tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil.

Measured weight of bottle labeled EXAFS*U*5B = 504.06 g.

Filtered most of the solution through a Fisher brand P8 qualitative filter paper (fast flow rate, Cat. No. 09-795G, 19 cm diameter). When only a small volume of U solution remained in the polypropylene bottle, the bottle was swirled to suspend most of the clinoptilolite and the mixture was transferred into a plastic LSA vial (labeled U*5B). A small volume of filtrant was taken with a micropipet, transferred into the polypropylene bottle, the bottle swirled, and the suspended clinoptilolite transferred also into the vial. This procedure effectively transferred most of the clinoptilolite from the bottle into the vial without filtration. Some clinoptilolite that was transferred onto the filter paper was scooped and transferred also into the vial. The vial was tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil.

1530 hrs. –

Check pH standards: pH 4 = 4.01; pH 7 = 7.01

Measured the pH of some U-bearing mixtures:

EXAFS*U*5D: pH = 6.37±0.01
EXAFS*U*5E*a: pH = 6.33
EXAFS*U*5F*a: pH = 6.27

Check pH standards: pH 4 = 4.03; pH 7 = 7.03

Measured weight of bottle labeled EXAFS*U*5D = 503.13 g.

Filtered most of the solution through a Fisher brand P8 qualitative filter paper (fast flow rate, Cat. No. 09-795G, 19 cm diameter). When only a small volume of U solution remained in the polypropylene bottle, the bottle was swirled to suspend most of the clinoptilolite and the mixture was transferred into a plastic LSA vial (labeled U*5D). A small volume of filtrant was taken with a micropipet, transferred into the polypropylene bottle, the bottle swirled, and the suspended clinoptilolite transferred also into the vial. This procedure effectively transferred almost all of the clinoptilolite from the bottle into the vial without filtration. The vial was tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil.

Measured weight of bottle labeled EXAFS*U*5E*a = 280.48 g.

Filtered most of the solution through a Fisher brand P8 qualitative filter paper (fast flow rate, Cat. No. 09-795G, 19 cm diameter). When only a small volume of U solution remained in the polypropylene bottle, the bottle was swirled to suspend most of the clinoptilolite and the mixture was transferred into a plastic LSA vial (labeled U*5E). A small volume of filtrant was taken with a micropipet, transferred into the polypropylene bottle, the bottle swirled, and the suspended clinoptilolite transferred also into the vial. This procedure effectively transferred almost all of the clinoptilolite from the bottle into the vial without filtration. The vial was tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil.

1600 hrs. –

Measured the pH of EXAFS*Np*1C = 7.92. Added 100 microliters of 0.1-N NaOH. Measured pH = 8.26.

March 1, 2003

1320 hrs. –

Check pH standards: pH 4 = 4.01; pH 7 = 7.01; pH 10 = 10.06

Measured the pH of U- and Np-bearing mixtures:

EXAFS*U*5C: pH = 3.21

EXAFS*U*5F*a: pH = 6.27

EXAFS*U*5G: pH = 3.30

EXAFS*U*5H*a: pH = 6.22

EXAFS*Np*1B: pH = 3.28

EXAFS*Np*1C: pH = 7.92
Added 100 microliters 0.1-N NaOH; pH = 8.22

1340 hrs. – check pH standards: pH 4 = 4.04; pH 7 = 7.04; pH 10 = 10.08

Filtered most of the solution in bottles labeled EXAFS*U*5G and EXAFS*U*5H*a through a Fisher brand P8 qualitative filter paper (fast flow rate, Cat. No. 09-795G, 19 cm diameter). When only several mL of solution was left in each bottle, the bottle was swirled to suspend the clinoptilolite powder and the slurry was poured into a plastic LSA vial (labeled EXAFS*U*5G and EXAFS*U*5H). This procedure effectively transferred almost all of the clinoptilolite from the bottle into the vial without filtration. The vials were tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil.

The clinoptilolite in the bottles labeled EXAFS*U*5C and EXAFS*U*5F*a was separated by centrifugation using a procedure similar to the one on page 410 of this notebook. Centrifugation and decantation of clear solutions was repeated until the clinoptilolite powder was contained in one centrifuge tube only. A final centrifugation was done, and a 10-mL Oxford pipet was used to uptake most of the clear solution, which was discarded, leaving only about 1 to 2-mL of solution in the tube. The tube containing the EXAFS*U*5C mixture was swirled to suspend the clinoptilolite, and the mixture was poured into a plastic LSA vial labeled EXAFS*U*5C. The vial was tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil. The tube containing the EXAFS*U*5F*a mixture was centrifuged an additional 45 minutes because some fine particles were observed still suspended. However, because of lack of time to complete the process, the centrifuge tube was left unattended for the rest of the day.

1445 hrs. –

Checked the pH standards: pH 7 = 7.01; pH 10 = 10.05; temp = 21.2 C.

Measured the pH of EXAFS*Np*1C = 7.95; added 200 microliters of 0.1-N NaOH; pH = 8.59.

1550 hrs. –

Checked the pH standards: pH 7 = 6.99; pH 10 = 10.04; temp = 21.2 C

Measured the pH of EXAFS*Np*1C = 8.29.

March 3, 2003

0835 hrs. –

Checked the pH standards: pH 4 = 4.00; pH 7 = 7.01; pH 10 = 10.05; temp = 21.2 C

Measured the pH of Np-bearing mixtures:

EXAFS*Np*1B: pH = 3.30

EXAFS*Np*1C: pH = 7.92
Added 200 microliters of 0.1-N NaOH; pH = 8.58 and decreasing

Centrifuged the mixture EXAFS*U*5F*a for 15 min; pipetted the clear solution, discarding ~23 mL. The remaining mixture was shaken, and the slurry was poured into a plastic LSA vial labeled EXAFS*U*5F. The vial was tightly capped, sealed with a piece of parafilm, and covered with an aluminum foil.

Fifty milliliters of EXAFS*U*5A*filtrant, EXAFS*U*5B*filtrant, EXAFS*U*5D*filtrant, EXAFS*U*5E*filtrant, EXAFS*U*5G*filtrant, and EXAFS*U*5H*filtrant each were transferred into a 60-mL PP bottle (labeled EXAFS*U*5A*FU, etc.). One-mL of 1.0-N HNO3 solution was added to each PP bottle, and the solutions set aside for later analysis of U concentration.

Two ~30-mL aliquots of EXAFS*U*5C*filtrant were transferred into two centrifuge tubes. The same was done for EXAFS*U*5F*a*filtrant. The four tubes were centrifuged for 12 minutes at 12,700 rpm. Then, a total of 50 mL were taken from the two tubes containing the EXAFS*U*5C*filtrant and transferred into a 60-mL PP bottle (labeled EXAFS*5C*FU); 1-mL of 1.0-N HNO3 was added. The same was done for the EXAFS*U*5F*a*filtrant.

The measured weights of the PP bottles are as follows:

Bottle (EXAFS*U*)	Wt. Empty	Plus 50 mL soln.	Plus 1 mL HNO3	Wt. Solution only
5A*FU	12.38	62.40	63.43	50.02
5B*FU	12.38	62.36	63.38	49.98
5C*FU	12.40	62.41	63.42	50.01
5D*FU	12.36	62.41	63.41	50.05
5E*FU	12.41	62.43	63.42	50.02
5F*FU	12.38	62.40	63.42	50.02
5G*FU	12.40	62.37	63.39	49.97
5H*FU	12.33	62.37	63.36	50.04

1015 hrs. –

Checked pH standards: pH 7 = 7.00; pH 10 = 10.04

Measured pH of EXAFS*Np*1C = 8.17; added 100 microliters of 0.1-N NaOH.

1338 hrs. –

Checked pH standards: pH 7 = 7.00; pH 10 = 10.04

Measured pH of EXAFS*Np*1C = 7.98; added 200 microliters of 0.1-N NaOH.

1710 hrs. –

Checked pH standards: pH 7 = 6.99; pH 10 = 10.06

Measured pH of EXAFS*Np*1C = 8.06; added 200 microliters of 0.1-N NaOH.

March 4, 2003

0900 hrs. –

Checked pH standards: pH 7 = 7.00; pH 10 = 10.06.

Measured pH of EXAFS*Np*1B = 3.29.

Measured pH of EXAFS*Np*1C = 8.00; added 200 microliters of 0.1-N NaOH.

1330 hrs. –

Checked pH standards: pH 7 = 6.99; pH 10 = 10.05.

Measured pH of Np*1C = 8.09; added 200 microliters of 0.1-N NaOH.

1705 hrs. –

Checked pH standards: pH 7 = 7.00; pH 10 = 10.06.

Measured pH of EXAFS*Np*1C = 8.14; added 200 microliters of 0.1-N NaOH solution.

March 5, 2003

1030 hrs. – pH of EXAFS*Np*1C measured by B. Werling. PH electrode calibrated with pH 7 and pH 10 standards (slope = 101.5%). PH of Np solution is 8.01. B. Werling added 200 microliters of 0.1-N NaOH solution. (see B. Werling's notebook #463, page 127).

1300 hrs. –

Calibrated pH electrode with 3 pH standards (4, 7 and 10). Slope = 99.5%.

Measured pH of EXAFS*Np*1B = 3.33.

Measured pH of EXAFS*Np*1C = 8.14

Checked pH standards: pH 4 = 4.04; pH 7 = 7.03; pH 10 = 10.08.

Measured weight of polypropylene bottle (empty) labeled EXAFS*Np*1B*soln = 54.69.

The LSA vials with samples EXAFS*Np*1A*F1*a and EXAFS*Np*1A*F1*b, which had label stickers on them, were relabeled. The stickers were removed because those will interfere with the LSA counting. The sample names (Np*1 and Np*2) were written on the plastic caps. The weight of the stickers were measured, and are 0.0928 g (for the label EXAFS*Np*1A*F1*a) and 0.0917 g (for the label EXAFS*Np*1A*F1*b).

March 6, 2003

0830 hrs. –

Checked pH standards: pH 7 = 7.01; pH 10 = 10.08; Temp = 19.9 C

Measured pH of EXAFS*Np*1C = 8.10.

Transferred most of the EXAFS*Np*1C solution into a polypropylene bottle labeled EXAFS*Np*1C*soln. The remaining were poured into 4 centrifuge tubes, and centrifuged for 12 minutes at 12,700 rpm. Most of the clear solution was uptaken using an Oxford pipet and transferred into the PP bottle. A spatula was used to scoop some of the Np-loaded clinoptilolite into an XAS sample cell, labeled Np*1C. Excess solution was uptaken with an Eppendorf micropipet, but the clinoptilolite was maintained quite wet to prevent drying out during the XAS analysis as observed in previous XAS runs. The cell was closed and sealed with 10 screws. Another XAS sample, Np*1C*dup, was prepared.

Two XAS samples of EXAFS*Np*1B (Np-loaded montmorillonite) were prepared: Np*1B and Np*1B*dup.

A second XAS sample of Np-loaded clinoptilolite (pH~3.3) was also prepared. The XAS cell was labeled Np*1A*dup.

All the XAS cells were covered with aluminum foil. The estimated loading on the six samples are (assuming 0.2 g of wet solid in each cell):

1. Np*1A: 0.23 microCi
2. Np*1A*dup: 0.23 microCi
3. Np*1B: 0.77 microCi
4. Np*1B*dup: 0.77 microCi
5. Np*1C: 0.38 microCi
6. Np*1C*dup: 0.38 microCi

The remaining Np-loaded clinoptilolite and montmorillonite slurries were transferred into plastic LSA vials labeled EXAFS*Np*1A, EXAFS*Np*1B, and EXAFS*Np*1C.

The six XAS samples were given to John Hageman, SwRI radiological safety officer, who will ship the samples to Andrew Bracken/Lynda Soderholm [Building 46—Receiving, Argonne National Laboratory 9700 South Cass Ave., Argonne, IL. 60439; telephone (630) 252-7388].

Solutions were transferred from the bottle EXAFS*Np*1B*soln into two centrifuge tubes. The same thing was done for the solution in the bottle EXAFS*Np*1C*soln. The solutions were centrifuged for 20 min at 27,000 rpm. One-mL samples were taken from each centrifuge tube and transferred into LSA vials containing 0.5-mL of 0.1-N HNO3. The two LSA samples from EXAFS*Np*1B*soln were labeled Np*3 and Np*4. The two LSA samples from EXAFS*Np*1C*soln were labeled Np*5 and Np*6. The samples were set aside for later LSA analysis. The measured weights of the samples are given below:

Sample Name	Wt. Scint. Vial empty	Wt. With 500 microL 0.1 N HNO3	Wt. With 1-mL Np solution	Wt. Of Np sample
Np*1	See page 410			0.9994
Np*2	See page 410			1.0012
Np*3	7.1326	7.6310	8.6373	1.0063
Np*4	7.1680	7.6649	8.6726	1.0077
Np*5	7.1734	7.6690	8.6797	1.0107
Np*6	7.1540	7.6305	8.6282	0.9977

Entries into Scientific Notebook No. 278 for the period September 6, 2002, through March 6, 2003, have been made by

RT Pabalan 3/7/03

Roberto T. Pabalan / Date

No original entry into this Scientific Notebook has been removed.

RT Pabalan 3/7/03

Roberto T. Pabalan / Date

The entries in this electronic scientific notebook #278 document activities conducted during the Period March 7, 2003, through August 31, 2003, under the Radionuclide Transport Key Technical Issue (Project Number 20.06002.01.141).

March 10, 2003

0830 hrs. –

The remaining solutions in the bottle EXAFS*Np*1C*soln were transferred into centrifuge tubes. Those solutions, and those that were transferred on March 6 (see page 418), were centrifuged for 15 min at 27,000 rpm. The top ~25 mL of the centrifuged solutions were taken up with an Oxford pipet and transferred into a PP bottle labeled EXAFS*Np*1C*soln*cent. The process was repeated until all EXAFS*Np*1C*soln solutions have been transferred, except for ~5 mL in three centrifuge tubes, which were discarded. The EXAFS*Np*1C*soln*cent will be saved for other experiments to be conducted later.

The same process was done for the solutions in the bottle EXAFS*Np*1B*soln. After centrifugation, the clear solutions were transferred into a PP bottle labeled EXAFS*Np*1B*soln*cent. The EXAFS*Np*1B*soln*cent will be saved for other experiments to be conducted later.

April 4, 2003

The following table gives Paul Bertetti's results of liquid scintillation analyses of the Np samples (see his notebook #369).

4/1/03

sample name	sample weight	cpm alpha	efficiency correct	mass correct	atom convert	mole convert	Np-237 ppb	exp soln	Ave ppb Np237	average molality
Np*1	0.9994	42163.2	42163.2	42188.51	6.85E+16	1.14E-07	26967	Np*1-2	26911	1.14E-04
Np*2	1.0012	42061.8	42061.8	42011.39	6.82E+16	1.13E-07	26854	Np*3-4	5696	2.40E-05
Np*3	1.0063	8953.2	8953.2	8897.15	1.44E+16	2.40E-08	5687	Np*5-6	10820	4.56E-05
Np*4	1.0077	8993.6	8993.6	8924.88	1.45E+16	2.41E-08	5705	Initial	28707	1.21E-04
Np*5	1.0107	17178.0	17178.0	16996.14	2.76E+16	4.58E-08	10864			
Np*6	0.9977	16819.1	16819.1	16857.87	2.74E+16	4.55E-08	10776			
Initial*1 p. 403	0.4985	22364.2	22364.2	44862.99	7.29E+16	1.21E-07	28677			
Initial*2 p. 403	0.4994	22450.7	22450.7	44955.35	7.30E+16	1.21E-07	28736			

The initial sample taken on 2/19/03, page 403, was run twice on the LSA.

The following are the calculated %Np sorbed and Np-loading on the mineral samples:

Sample	% Np sorbed	Np loading (ppm)
EXAFS*Np*1A Clinop pH~3	100*(1.21E-4–1.14E-4)/1.21E-4 = 5.8%	(1.21E-4-1.14E-4)*237.048*0.5L/0.5g*1E6 = 1,659 ppm
EXAFS*Np*1B Mont pH~3	100*(1.14E-4–2.40E-5)/1.14E-4 = 79%	(1.14E-4-2.40E-5)*237.048*.483L/0.5g*1E6 = 20,609 ppm
EXAFS*Np*1C Clinop pH~8	100*(4.60E-5–4.56E-5)/4.60E-4 = 0.87%	(4.60E-5-4.56E-5)*237.048*.575L/0.5g*1E6 = 109 ppm

Wt. Of EXAFS*Np*1C solution is 747.3 g (bottle w/ solution) minus 172.3 g (bottle empty, see page 356) = 575.0 grams

The following are the calculated loading and activity of Np-237 in the clinoptilolite or montmorillonite. The XAS samples that were sent to Argonne National Laboratory in sealed XAS sample cells had approximately 0.2 grams of clinoptilolite or montmorillonite. Thus the XAS sample activity would be 40 percent of the value listed in the third column.

Sample	Loading (ppm)	Activity (microCuries)
EXAFS*Np*1A, dup Clinoptilolite pH~3	1,660	0.59
EXAFS*Np*1B, dup Montmorillonite pH~3	20,600	7.3
EXAFS*Np*1C, dup Clinoptilolite pH~8	109	0.039

July 10, 2003

Preparation of Uranium-loaded Clinoptilolite and Montmorillonite for EXAFS Analysis (scheduled for July 24, 2003)

Objective: Clinoptilolite sorbed with U(VI) at pH~3.1 and ~6.2 and montmorillonite sorbed with Np(V) at pH~3.1 and ~8.1 will be prepared for XAS analysis at Argonne National Laboratory.

0900 hrs. – Two 1-mL samples each were taken from the solutions EXAFS*Np*1B*soln*cent and EXAFS*Np*1C*soln*cent (see page 430) for LSA analysis of Np activity. The samples were added to 0.5-mL 0.1 N HNO3 solutions in glass scintillation vials. The 0.1 N HNO3 solution was prepared by J. Prikryl on 7/9/03 (see notebook 556, page 89). The measured weights are as follows:

Sample Name	Wt. Scint. Vial empty	Wt. With 500 microL 0.1 N HNO3	Wt. With 1-mL Np solution	Wt. Of Np sample
Np*1a	7.1591	7.6595	8.6661	
Np*1b	7.1706	7.6698	8.6742	
Np*2a	7.1962	7.6954	8.6999	
Np*2b	7.1339	7.6328	8.6384	

A 500-mL PP bottle labeled EXAFS*Np*2A was weighed: 54.43 grams.

~400-mL of the EXAFS*Np*1B*soln*cent solution was transferred into the EXAFS*Np*2A bottle. Total weight = 454.52 (calculated weight of transferred solution is 400.09 grams. The weight of the EXAFS*Np*1B*soln*cent bottle after transfer is 115.61 grams.

A 1-L PP bottle labeled EXAFS*Np*2B was weighed: 101.05 grams.

The EXAFS*Np*1C*soln*cent solution was all transferred into the EXAFS*Np*2B bottle. Total weight of EXAFS*Np*2B bottle after transfer is 626.04 grams (calculated weight of EXAFS*Np*1C*soln*cent transferred is 524.99 grams.

The remaining EXAFS*Np*1B*soln*cent solution was also transferred into the EXAFS*Np*2B bottle. Total weight of EXAFS*Np*2B after transfer is 686.95 (calculated weight of EXAFS*Np*1B*soln*cent solution transferred is 60.91 grams.

1330 hrs. – Measure pH of Np solutions.

Check pH standards (prepared by J. Prikryl 7/3/03)
pH4 = 4.01
pH7 = 6.99
pH10 = 9.99

EXAFS*Np*2A pH = 3.33±0.02
EXAFS*Np*2B pH = 7.41±0.02

Montmorillonite added: 0.50 g of montmorillonite (batch SAz*Na<2*FD*SCR) was added to EXAFS*Np*2A solution. 0.30 g of montmorillonite was added to EXAFS*Np*2B solution. Both

mixtures were placed on a gyratory shaker with the caps loose to allow CO2 to equilibrate with the solution.

1350 hrs – Procedure: Take 3.5 g of purified clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe) from the stock kept in a glass jar. Place in a mortar and grind with a pestle until the powder has a consistency of flour (estimated mesh size <400). Transfer the powder into four centrifuge tubes, add water and place in a beaker in an ultrasonic cleaner. Turn on ultrasonic cleaner for about 3 to 5 minutes. Transfer the tubes into a centrifuge. Centrifuge the mixtures for about 5 to 7 minutes, with speed setting of 10. Decant the solution phase. **This procedure was done six times to eliminate much of the highly reactive surfaces and fine particles that were generated by the grinding process. It is hoped that this procedure would minimize any potential dissolution of the clinoptilolite during the sorption procedure.**

After the last centrifugation step, most of the aqueous phase from each of the centrifuge tubes was extracted with the aid of an Oxford pipet. The remaining slurry was transferred into a glass petri dish, which was placed in a drying oven with the temperature set to 56 C.

The dried clinoptilolite (CDV*200/325*UC*WA*RC*HL*Rfe*<400) was recovered and disaggregated using a spatula. The amount recovered was weighed:

Wt. Of glass vial empty = 16.82 g
Wt. Of vial plus recovered clinoptilolite powder = 19.86 g
Amount clinoptilolite powder = 3.04 g

1550 hrs. - Check pH of Np/montmorillonite mixtures

pH4 standard = 4.02
pH10 standard = 9.95

EXAFS*Np*2A pH = 3.70
Added 300 microL of 1-N HNO3, pH = 3.14
(HNO3 solution prepared by B. Werling 2/18/03, notebook 556, p. 22)

EXAFS*Np*2B pH = 7.54
Added 250 microL of 0.1 N NaOH, pH = 8.18
(NaOH solution prepared by B. Werling 7/10/01, notebook 463, page 41)

July 11, 2003

0850 hrs. – Measure pH of Np/montmorillonite mixtures.

pH4 standard = 4.03
pH7 standard = 7.03
pH10 standard = 9.99

EXAFS*Np*2A pH = 3.20
Added 100 microL of 1-N HNO3, pH = 3.09
EXAFS*Np*2B pH = 8.05
Added 100 microL of 0.1-N NaOH, pH = 8.30

1310 hrs. – Measure pH of Np/montmorillonite mixtures

pH4 standard = 4.03
pH10 standard = 10.02

EXAFS*Np*2A pH = 3.10
EXAFS*Np*2B pH = 8.20

1330 hrs. – Prepare 1 L each of 2E-4M and 5E-4 M UO2(NO3)2 solution using the 0.005 M UO2(NO3)2 solution prepared by R. Pabalan on 2/17/03.

Wt. Of 1-L PP bottle-1 empty = 101.27 g
Wt. With ~40 g U solution = 141.40 g
Calculated mass of U solution = 40.13 g
Wt. With H2O to make up ~ 1L = 1,101.51
Calculated mass of solution = 1,000.24 g
Calculated U concentration = 2.0E-4 M

Wt. Of 1-L PP bottle-2 empty = 100.76
Wt. With ~100 g U solution = 201.06
Calculated mass of U solution = 100.30 g
Wt. With H2O to make up ~ 1L = 1,100.93
Calculated mass of solution = 1,000.17 g
Calculated U concentration = 5.0E-4 M

Transferred ~450 mL of 2.0E-4 M U solution into 500-mL PP bottle labeled EXAFS*U*6A and another ~450 mL into EXAFS*U*6B. Transferred ~450 mL of 5.0E-4 M U solution into 500-mL bottle labeled EXAFS*U*6C and another ~450 mL into EXAFS*U*6D. Measured weights are as follows:

Bottle label	Wt. Btl. Empty	Wt. With U solution	U conc. (M)	Calc. soln. Wt.
EXAFS*U*6A	53.92	503.99	2.0E-4	
EXAFS*U*6B	54.54	504.75	2.0E-4	
EXAFS*U*6C	54.80	504.82	5.0E-4	
EXAFS*U*6D	54.79	504.77	5.0E-4	

1345 hrs. – Added 0.45 g of CDV*200/325*UC*WA*RC*HL*Rfe*<400 to each of the U solutions.

1400 hrs. – Measure pH of U solutions.

pH 4 standard = 4.03
pH 7 standard = 7.03

EXAFS*U*6A pH = 4.61
Added 500 microL of 1-N HNO3; pH = 3.15
EXAFS*U*6B pH = 4.67
Added 100 microL of 0.1 NaOH; pH =4.76

Added 100 microL 1.0-N NaOH (prepared by RTP 7/15/02; notebook 278, p. 354)

pH = 5.75

Added 50 microL 1.0-N NaOH; pH = 7.1±0.1; **yellowish solution**

EXAFS*U*6C pH = 4.35

Added 500 microL 1-N HNO3; pH = 3.22

EXAFS*U*6D pH = 4.34

Added 100 microL 1.0-N NaOH; pH = 4.55

Added 200 microL 1.0–N NaOH; pH = 5.4±0.1 **(yellowish solution)**

July 14, 2003

0910 hrs. – Check pH of Np and U mixtures

pH 4 standard = 4.02

pH 7 standard = 6.99

pH 10 standard = 10.02

EXAFS*Np*2A pH = 3.16

EXAFS*Np*2B pH = 8.09

EXAFS*U*6A pH = 3.44

Added 100 microL 1.0-N HNO3; pH = 3.27

Added 100 microL 1.0-N HNO3; pH = 3.15

EXAFS*U*6B pH = 6.23

Added 50 microL 1.0-N NaOH; pH = 9.5±0.1

EXAFS*U*6C pH = 3.36

Added 100 microL 1.0-N HNO3; pH = 3.21

Added 100 microL 1.0-N HNO3; pH = 3.09

EXAFS*U*6D pH = 5.37

Added 50 microL 1.0-N NaOH; pH = 6.44

Added 150 microL 1.0-L NaOH; pH = 9.0±0.1

1400 hrs. – Measure pH of U-clinoptilolite and Np-montmorillonite mixtures

Temp = 22.4 C

pH 4 standard = 4.01

pH 7 standard = 7.01

pH 10 standard = 10.01

EXAFS*Np*2A pH = 3.14±0.02

EXAFS*U*6A pH = 3.15

EXAFS*U*6C pH = 3.18

EXAFS*U*6F pH =

EXAFS*Np*2B pH = 8.01

Added 50 microL 0.1-N NaOH, pH = 8.14

EXAFS*U*6B pH = 7.17

EXAFS*U*6D pH = 7.14

EXAFS*U*6E pH = 6.23

July 15, 2003

0927 hrs. – Measure pH of U-clinoptilolite mixtures

Temp = 21.5 C
pH 4 standard = 4.03
pH 7 standard = 7.02
pH 10 standard = 10.04

EXAFS*U*6A pH = 3.16
EXAFS*U*6C pH = 3.12±0.01
EXAFS*U*6F pH = 3.29±0.01
Added 150 microL 1.0-N HNO3, pH = 3.18±0.01
EXAFS*U*6B pH =7.05±0.05
EXAFS*U*6D pH = 7.14±0.05

July 16, 2003

0930 hrs. – Measure pH of U-clinoptilolite mixtures

pH 4 standard = 4.03
pH 7 standard = 7.04

EXAFS*U*6A pH = 3.17
EXAFS*U*6C pH = 3.18
EXAFS*U*6F pH = 3.22
Added 50 microL 1.0-N HNO3; pH = 3.18
EXAFS*U*6B pH = 7.15±0.03
EXAFS*U*6D pH = 7.11±0.02
EXAFS*U*6E pH = 6.30

July 17, 2003

1040 hrs. – Measure pH of Np-montmorillonite and U-clinoptilolite samples

pH 4 standard = 4.02
pH 7 standard = 7.03
pH 10 standard = 10.03

EXAFS*U*6A pH = 3.19
EXAFS*U*6C pH = 3.18
EXAFS*U*6F pH = 3.19

EXAFS*Np*2A = 3.18
EXAFS*Np*2B = 8.02
Added 50 microL 0.1-N NaOH; pH = 8.14

EXAFS*U*6B pH = 7.20
EXAFS*U*6D pH = 7.11
EXAFS*U*6E pH = ??

July 18, 2003

0900 hrs. – Measure pH of Np- and U-mineral mixtures.

pH 4 standard = 4.01
pH 7 standard = 7.03
pH 10 standard = 10.03

EXAFS*U*6A pH = 3.18
EXAFS*U*6C pH = 3.19
EXAFS*U*6E pH = 3.19

EXAFS*Np*2A pH = 3.16
EXAFS*Np*2B pH = 7.97
Added 100 microL 0.1-N NaOH; pH = 8.21

EXAFS*U*6E pH = 6.33±0.01
EXAFS*U*6B pH = 7.17
EXAFS*U*6D pH = 7.08

0915 hrs. – Take 2 1-mL samples of EXAFS*Np*2A for LSA analysis.

Sample Name	Wt. Scint. Vial empty	Wt. With 500 microL 0.1 N HNO3	Wt. With 1-mL Np solution	Wt. Of Np sample
Np*3a	7.1604	7.6583	8.6664	1.0081
Np*3b	7.1537	7.6538	8.6601	1.0063

The remaining montmorillonite/Np-solution mixtures in the bottle EXAFS*Np*2A were transferred into centrifuge tubes. Those solutions were centrifuged for 20 min at 27,000 rpm. The top ~25 to ~30 mL of the centrifuged solutions were taken up with an Oxford pipet and transferred into a PP bottle labeled EXAFS*Np*2A*cent. The process was repeated until all EXAFS*Np*2A solutions have been transferred, except for ~10 mL in one centrifuge tube with the separated montmorillonite. The EXAFS*Np*2A*cent will be saved for other experiments to be conducted later. The Np-loaded montmorillonite in the centrifuge tube will be used later to prepare a sample for EXAFS analysis.

Wt. Of EXAFS*Np*2A*cent bottle empty = 54.48 g; total weight with transferred Np solution = 429.78 g. Calculated weight of EXAFS*Np*2A*cent solution = 375.3 g.

Two 1-mL samples of the EXAFS*Np*2A*cent solution were taken:

Sample Name	Wt. Scint. Vial empty (g)	Wt. With 500 microL 0.1 N HNO3 (g)	Wt. With 1-mL Np solution (g)	Wt. Of Np sample (g)
Np*4a	7.1508	7.6485	8.6489	1.0004
Np*4b	7.1842	7.6822	8.7000	1.0178

1430 hrs. – Measure pH of EXAFS*Np*2B solution.

pH 7 standard = 7.03

pH 10 standard = 10.01
EXAFS*Np*2B pH = 8.10

1440 hrs. – Centrifuge EXAFS*U*6B and EXAFS*U*6D.

Weight of bottle labeled EXAFS*U*6B*cent (empty) = 54.68 g

Weight of bottle labeled EXAFS*U*6D*cent (empty) = 54.62 g.

July 20, 2003

1415 hrs. – Measure pH of Np-montmorillonite mixture
pH 4 standard = 4.03
pH 7 standard = 7.02
pH 10 standard = 10.03
EXAFS*Np*2B pH = 8.09

Centrifugation of EXAFS*U*6B was completed. .

Wt. Of bottle EXAFS*U*6B*cent empty = 54.68 g
Wt. Of bottle EXAFS*U*6B*cent with U solution = 428.01 g
Calc. wt. Of EXAFS*U*6B*cent solution = 373.33 g

The same procedure was done for EXAFS*U*6D. The glass vial was labeled U*6D.
Wt. Of bottle EXAFS*U*6D*cent empty = 54.62 g
Wt. Of bottle EXAFS*U*6D*cent with U solution = ???? g
Calc. wt. Of EXAFS*U*6D*cent solution = ????? g

Centrifugation of EXAFS*Np*2B was done.
Wt. Of bottle EXAFS*Np*2B*cent empty = 101.19 g
Wt. With separated Np solution = 655.03 g
Calc. wt. Of EXAFS*Np*2B*cent solution = 553.84 g

Two 1-mL samples of EXAFS*Np*2B*cent were taken for LSA analysis.

Sample Name	Wt. Scint. Vial empty (g)	Wt. With 500 microL 0.1 N HNO3 (g)	Wt. With 1-mL Np solution (g)	Wt. Of Np sample (g)
Np*5a	7.3703	7.8668	8.8687	1.0019
Np*5b	7.1524	7.6494	8.6575	1.0081

July 21, 2003

0900 hrs. – Four XAS sample cells were examined under a binocular microscope to make sure there is no damage to the O-ring that could cause a leak. The cells were labeled using a fine-point Sharpie permanent marker with the labels EXAFS*Np*2A, EXAS*Np*2A*dup, EXAFS*Np*2B, and EXAFS*Np*2B*dup.

Using a spatula, a scoop of the EXAFS*Np*2A wet paste (Np-loaded montmorillonite) was transferred into the XAS cell EXAFS*Np*2A to cover the cell cavity. The side of the cell was tapped with a pen to make sure no air bubbles are trapped. A thin aqueous film was allowed to cover the top of the cavity to minimize potential drying of the sample during XAS analysis. The cell was sealed by tightening the screws on the cell cover.

Another sample of EXAFS*Np*2A was placed into the cell EXAFS*Np*2A*dup.

The same procedure was used to prepare two samples of EXAFS*Np*2B.

The estimated loading and activity the four samples are (assuming 0.2 g of wet solid in each cell):

1. Np*2A:	3,600 ppm Np	0.5 microCi Np-237
2. Np*2A*dup:	3,600 ppm Np	0.5 microCi Np-237
3. Np*2B:	2,000 ppm Np	0.3 microCi Np-237
4. Np*2B*dup:	2,000 ppm Np	0.3 microCi Np-237

The estimated loading and activity of the two U samples (assuming 0.45 g of solid in the glass vial) are:

5. U*6B	38,000 ppm U-238	0.0057 microCi U-238
6. U*6D	38,000 ppm U-238	0.0057 microCi U-238

The four XAS cells (in double zip-locked plastic bags) and the two glass vials (in a zip-locked plastic bag) with U samples were given the SwRI radiological safety office for FEDEX shipment to Andrew Bracken/Lynda Soderholm [Building 46—Receiving, Argonne National Laboratory, 9700 South Cass Ave., Argonne, IL. 60439; telephone (630) 252-7388].

1315 hrs. – Centrifugation of EXAFS*U*6E and EXAFS*U*6F were initiated (~20 min at 12,500 rpm several times until the wet paste was concentrated in one centrifuge tube).

July 22, 2003

1030 hrs. – Centrifugation of EXAFS*U*6E was completed. The centrifugation was not completely effective, and a colloidal suspension is evident. Approximately 30 mL of the colloidal suspension in the centrifuge tube was discarded and the wet paste (with approx. 2-mL solution) was transferred into a glass LSA vial. The vial was labeled U*6E and set aside for later shipment to Argonne National Laboratory.

The separated U solution was collected in a 1000-mL PP bottle labeled EXAFS*U*6E*cent. The initial weight was not measured.

1400 hrs. – Centrifugation of EXAFS*U*6F was completed. The centrifugation was not completely effective, and a colloidal suspension is evident. Approximately 30 mL of the colloidal suspension in the centrifuge tube was discarded and the wet paste (with approx. 2-mL solution) was transferred into a glass LSA vial. The vial was labeled U*6F and set aside for later shipment to Argonne National Laboratory.

The separated U solution was collected in a 1000-mL PP bottle labeled EXAFS*U*6F*cent. The initial weight was not measured.

1530 hrs. – The solution in the vials U*6E and U*6F were reduced to about 1 mL to minimize the time required for the colloids/particles to settle after the samples are received at Argonne. A Fedex package

was prepared containing the glass vials U*6A, U*6C, U*6E, and U*6F, for shipment to Lynda Soderholm [Building 200—Argonne National Laboratory, 9700 South Cass Ave., Argonne, IL. 60439; telephone (630) 252-4364].

Entries into Scientific Notebook No. 278 for the period March 7, 2003, through August 31, 2003, have been made by

9/15/03

Roberto T. Pabalan / Date

No original entry into this Scientific Notebook has been removed.

9/15/03

Roberto T. Pabalan / Date