S149
R
150

SHAW'S
PIONEERS SINCE 1831

account book S149

Available in 150 and 300 pages

0 78910 13932 5

CNWRA CONTROLLED COPY 057

No Units

Record Ruled, 27 Lines

Reorder number stamped on backbone of this book

WilsonJones®
Chicago, Illinois 60648

Made in Korea

PM Paula Muller Student Scientist

TG T ~~illegible~~ Student Scientist.

SS Stephen A. Sassman - Student Scientist

DRJ David R. Turner - Sr. Research Scientist

205704.072

Objectives of modeling experiments given on page 149, this volume. 8/11/94 DRJ

6/16 TG For the last couple weeks, May - June, the database of Mintegra, or Thermo.dbs, was being checked and rechecked for errors.

Many of the heats of formation and the stoichiometric coefficients were taken from EQ3/EQ6 and placed into a QuattroPro file which calculated the heats of reaction and the m.w.'s of the product. The m.w.'s are simply a check to ensure the the st. coefficients were put in properly and therefore provide Dave Turner and myself with reliable values for the heats of reaction.

See pg. 153 this volume for sample hand calculation DRJ 8/11/94

On the side, I was using a digitizing tablet to get data from other scientists work. These graphs coordinates can then supply Dave with more information on radionuclides such as Thorium, T.G. Americium, Plutonium, Neptunium, and Uranium and any other radioelements that need to be looked at.

6/16/92 T.G. Now that the thermo.dbs has been thoroughly established with the essential program data, I have been asked to practice setting up and running various solutions in the program.

In order to find a specific solution: for later refence, I decided on a certain nomenclature and or a page in this book where it can be found.

All runs with the major cation being Uranium will be logged on pg's 100-105.

Plutonium :	pg's 106-107
Neptunium :	pg's 108-109
Thorium :	pg's 110-111
Europium :	112-113
Americium :	114-115
Cobalt :	116-117
Cesium :	118-119
Technetium :	120-121
Zirconium :	122-123
Radium :	124-125

Ruthenium pg's 126-127

** File names will be written using T.G. DRJ 8/16/94 ~~with~~ T.G. DRJ 8/16/94 an 8 digit/letter ~~number~~ DRJ 8/16/94 system.

1st space : contains the 1st Letter of most positive species. e.g. U for uranium.

2nd space : contains the # of the charge on the Mintegrat2 component.
e.g. UO_2^{+2} would be 2.
U^{+4} would be 4.

3rd space : contains first letter of the major positive species in the salt.
For Na^+, the 3rd space is N.

Contains Ø for Uranium, or the 1st number in the power of the con

T.G. 6/24/92 4th space : contains the number of the charge on the major positive salt species.
For Na^{+1}, it would be 1

5th space : Contains just the number in the power of the radionuclide species.
e.g. $1\times10^{-5}M$ $UO_2(NO_3)_2$ T.G. would be 5.

6th space : If a Carbonate ion exists,
7th space : then the sign and power for the concentration are used. If not, use two underline bars.
e.g. $C_T = 0.001M$ Na_4CO_3, would be
-3. ~~Up till now, it would be~~ T.G. DRJ 8/16/94

DRJ 8/16/94

~~U2N15-3~~

8th space: contains the letter C if and only if the system is in the presence of CO_2. If not, it is left blank, which is represented with the symbol b.

For the system

1×10^{-5} M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$; ~~No CO_2~~ $C_T = 0.001$ M $NaHCO_3$; No CO_2 (DRJ 8/16/94)

the file name is U2N15-3b

6/17/91 T.6. Experimented with the Minteq A2 program to become acquainted withs its features and capabilities. Performed runs and looked at their outputs.

All runs are automatically put into another file with the same name, but using the subfilename .OUT

Output from above would appear as U2N15-3.OUT

6/18/91 T.6.

Decide to change the filename system. DRJ 8/16/94 T.6. ~~Upon~~ when modeling Plutonium, Neptunium, etc, the concentrations that are worked with are usually above 10^{-9}, which means that I will need two spaces for this number. Therefore, space 4 will now be a zero or one depending on the concentration. For the previous example using uranium, ~~the concentrations in~~ T.6. DRJ 8/16/94 the filename is:

U2N05-3b DRJ 8/16/94

6/24/9 A change was added to the filename system since the old and new data bases are being compared.

For Minteq A2 analysis on

2×10^{-6} M $U(VI)_T$

pH = 6.9

Variable CO_2

0.1 M $NaNO_3$

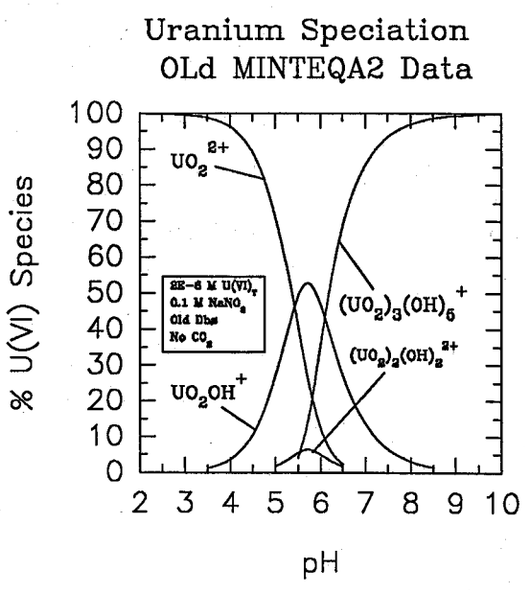
Revised (New) Data Base

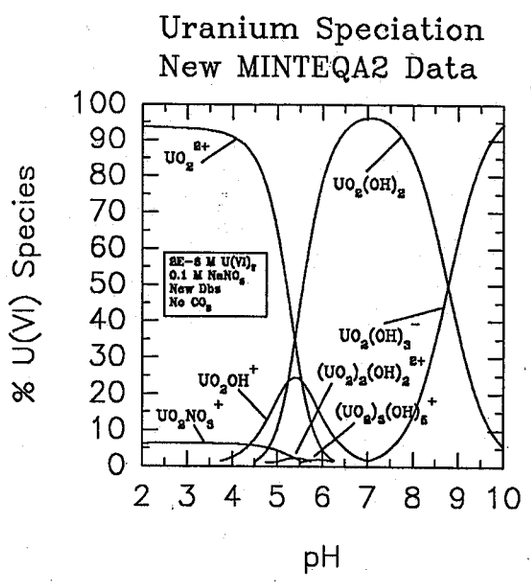
Filename is: UNN06_C

As output : UNNØ6_C.OUT
In WP : UNNØ6_C.WP
In SigmaPlot : UNNØ6_C.SP5

T.G. 6/29/92 Continued making and preparing graphs for % U(VI) speciation. Below is a comparison of the old and new MINTEQA2 database for the U(VI) system:

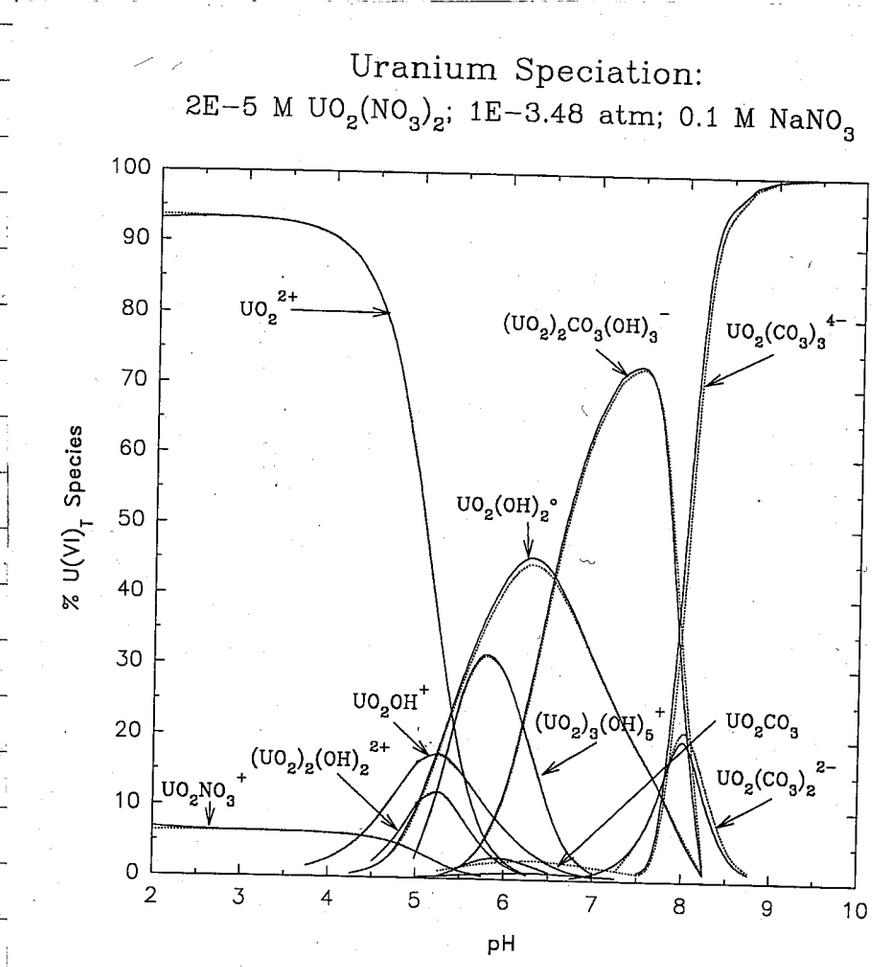
System
- 0.1 M $NaNO_3$
- No CO_2
- 2E-6 U(VI) or 2E-6 $UO_2(NO_3)_2$





These graphs can be found in filename : UONNØ6__.sp5

T.G. 7/14/92 Once all the MINTEQA2 runs were finished and graphed, Comparisons between MINTEQA2 and EQ3 were made. Dr. Bobby Tabalan provided the output from the EQ3 database. ~~and~~ T.G. 8/16/94 A graph was made and then overlayed onto the MINTEQA2 graph ~~with~~ of the same system.



The solid lines represent EQ3 output, while the dashed or dotted lines represent MINTEQA2 data.
These graphs can be found in the file, UMEØ5_C.SP5.

T.6. 7-22 DRJ

6. 7-22-92 ~~Made a run~~ Began using MINTEQA2 to model or make T.6. DRJ 8/16/94 sweeps of both pH and $p(CO_2)$ to determine what U(VI) species and concentrations existed under these concentrations.

7. 7-24-92 The MINTEQA2 output was imported into Word Perfect where Part 4 of the Output file was extracted. The output from the above run was originally DRJ 8/16/94 over 1200 pgs. and after recovering the T.6. DRJ 8/16/94 Part 4 files, it became ~ 130, however, the font size was changed to reduce the number of pages to just under 100.

This first 3-dimensional representation was created using T.6. DRJ 8/16/94 ~~an other~~ one of the earlier runs of just CO_2 and modified by adding a sweep of pH.

The concentration of the $U(VI)_T$

solution was 2E-6 M $UO_2(NO_3)_2$. The solute of the solution is 0.1 M $NaNO_3$ and we assumed that the temperature was 25°C.

The ~~section~~ Part DRJ 8/16/94 4 of the output was placed into a file named:
UPHCØ6_C.WP

T.6. 7-25-92 Went on vacation until Aug 1, 1992.

T.6. 8-3-92

UPHCØ6_C.wp was printed and labeled. The first page of this output can be seen on pg 10. Note that there DRJ 8/16/94 is a sweep of pH from DRJ 8/16/94 2-11.50 in increments of ~~0.5~~ 0.5 before there is ever a change in the $p(CO_2)$.

Only the Percent Bound In Species of UO_2^{2+} were taken out and placed into SigmaPlot 5.0 for analysis.
That file is:
UPHCØ6_C.SP5.

~~log~~ (ΔR) 8/16/94

$\log p(CO_2) = -4$ $pCO_2 = (\log) \cancel{=} 10^{-4} = -4$

PART 4 of OUTPUT FILE

PC MINTEQA2 v3.00 DATE OF CALCULATIONS: 22-JUL-92 TIME: 8:33: 6

pH = 2.00

PERCENTAGE DISTRIBUTION OF COMPONENTS AMONG TYPE I and TYPE II (dissolved and adsorbed) species

Na+1	100.0	PERCENT BOUND IN SPECIES # 500 Na+1
NO3-1	100.0	PERCENT BOUND IN SPECIES # 492 NO3-1
UO2+2	100.0	PERCENT BOUND IN SPECIES # 893 UO2+2
CO3-2	100.0	PERCENT BOUND IN SPECIES #3301401 H2CO3 AQ
H+1	100.0	PERCENT BOUND IN SPECIES # 330 H+1
H2O	99.8	PERCENT BOUND IN SPECIES #8933300 UO2OH +1

2.5 PC MINTEQA2 v3.00 DATE OF CALCULATIONS: 22-JUL-92 TIME: 8:33:10

pH

PERCENTAGE DISTRIBUTION OF COMPONENTS AMONG TYPE I and TYPE II (dissolved and adsorbed) species

Na+1	100.0	PERCENT BOUND IN SPECIES # 500 Na+1
NO3-1	100.0	PERCENT BOUND IN SPECIES # 492 NO3-1
UO2+2	99.6	PERCENT BOUND IN SPECIES # 893 UO2+2
CO3-2	99.9	PERCENT BOUND IN SPECIES #3301401 H2CO3 AQ
H+1	99.5	PERCENT BOUND IN SPECIES # 330 H+1
H2O	99.7	PERCENT BOUND IN SPECIES #8933300 UO2OH +1

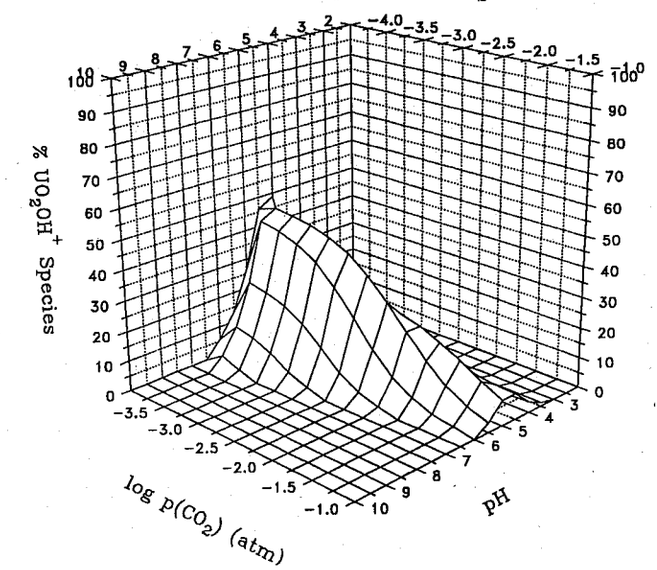
3.0 PC MINTEQA2 v3.00 DATE OF CALCULATIONS: 22-JUL-92 TIME: 8:33:12

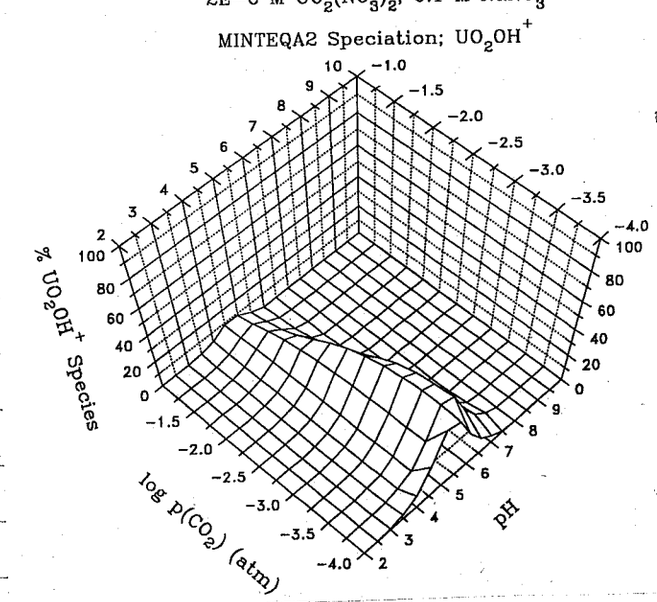
PERCENTAGE DISTRIBUTION OF COMPONENTS AMONG TYPE I and TYPE II (dissolved and adsorbed) species

Na+1	100.0	PERCENT BOUND IN SPECIES # 500 Na+1
NO3-1	100.0	PERCENT BOUND IN SPECIES # 492 NO3-1
UO2+2	98.8	PERCENT BOUND IN SPECIES # 893 UO2+2
	1.2	PERCENT BOUND IN SPECIES #8933300 UO2OH +1
CO3-2	99.8	PERCENT BOUND IN SPECIES #3301401 H2CO3 AQ
H+1	98.4	PERCENT BOUND IN SPECIES # 330 H+1
	1.6	PERCENT BOUND IN SPECIES #3301401 H2CO3 AQ
H2O	99.6	PERCENT BOUND IN SPECIES #8933300 UO2OH +1

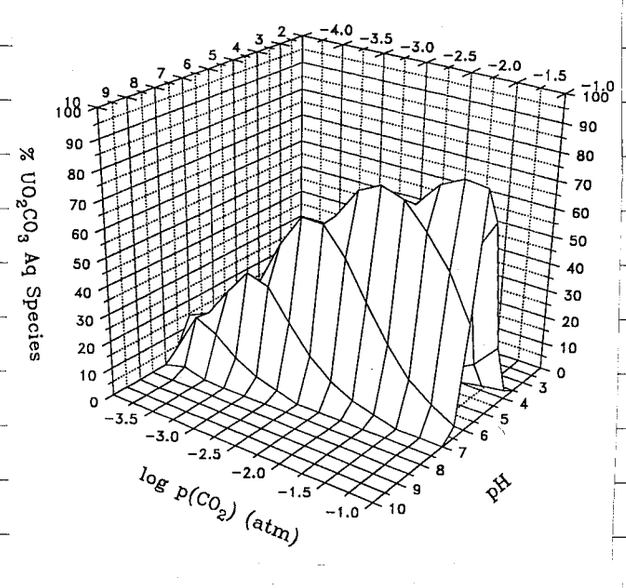
cont. Since SigmaPlot can only make one 3-D graph at a time, each ~~species~~ T.G. (ΔR) 8/16/94 of the studied species of U(VI)$_T$ were plotted

individually and after several days, the following graphs were generated.

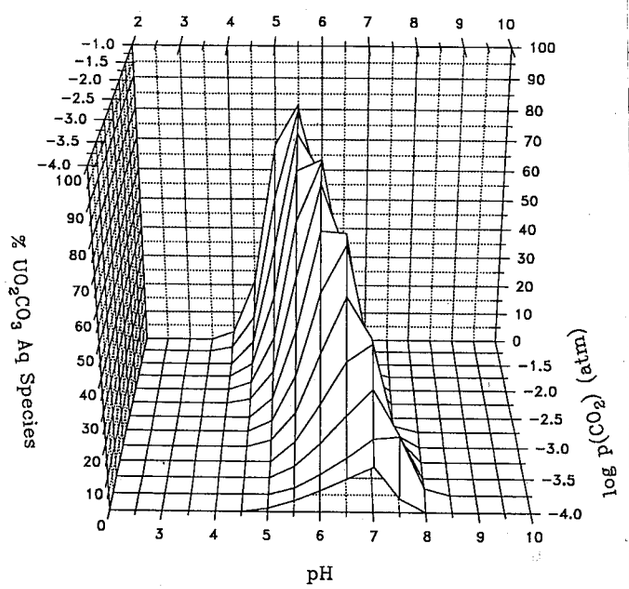




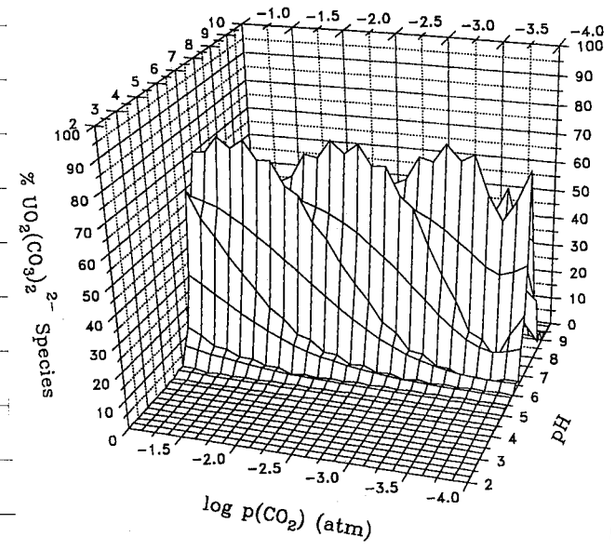
2E−6 M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$
MINTEQA2 Speciation; UO_2CO_3 Aq



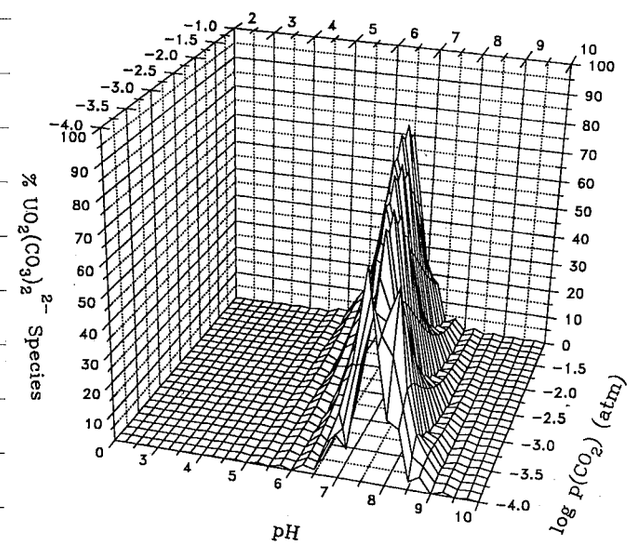
2E−6 M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$
MINTEQA2 Specieation; UO_2CO_3 Aq



2E−6 M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$
MINTEQA2 Speciation; $UO_2(CO_3)_2^{2-}$

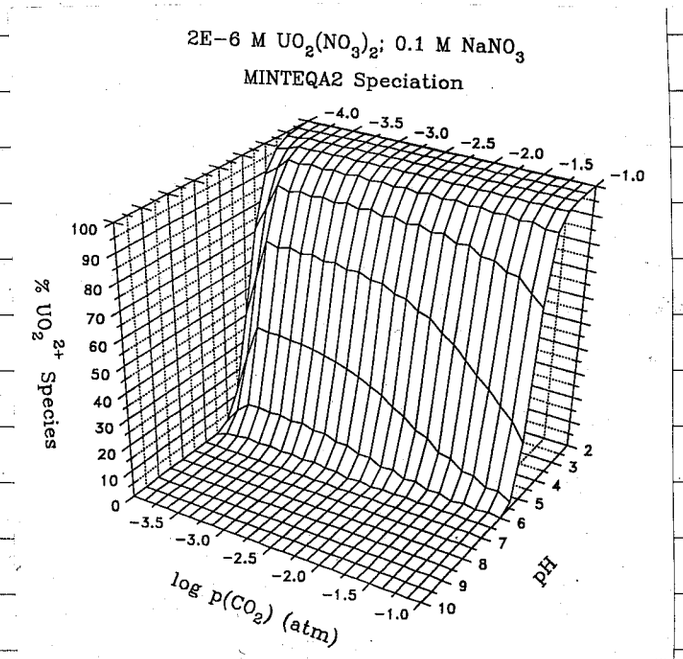


2E−6 M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$
MINTEQA2 Speciation; $UO_2(CO_3)_2^{2-}$



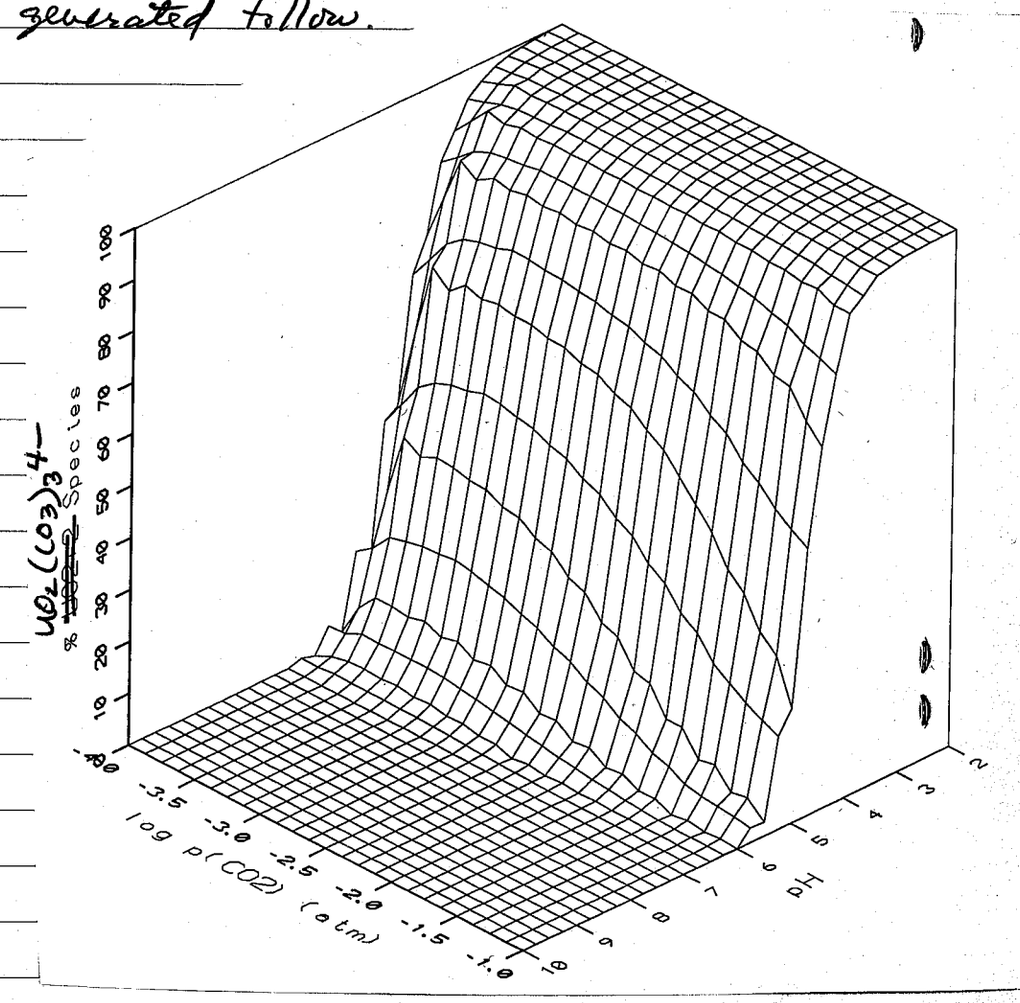
As one may note, each of the previous pages show a particular $U(VI)_T$ species ~~the~~ T.G. DRJ 8/16/94 from two views.

Although as seen below, some species don't require more than one view.

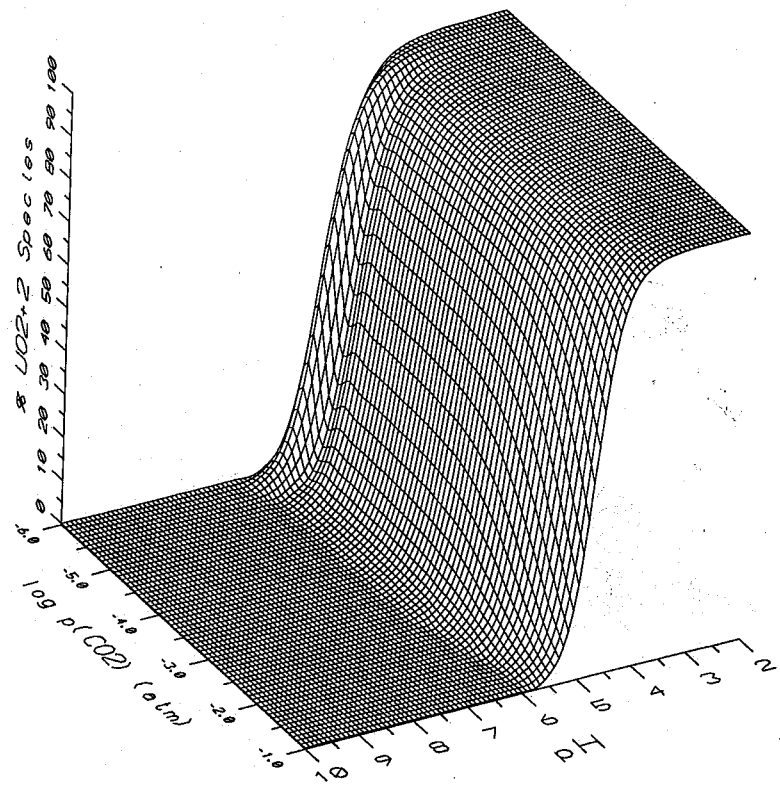


8-5-92 Because this is SigmaPlot's first version ~~at 3-D graphs~~ T.G. DRJ 8/16/94 capable of 3-D graphics, I wanted to compare them to a more popular 3-D graphing program known as Surfer, Version 4.

8-6-92 The purpose of using Surfer was to determine whether the peaks as seen in the $UO_2(CO_3)_2^{2-}$ ~~and~~ T.G. DRJ 8/16/94 and $UO_2(CO_3)_2^{2-}$ SigmaPlot 5.0 graphs were real or whether they could be smoothed. I also wanted to see ~~if that~~ T.G. DRJ 8/16/94 a topographical map could portray a more true representation of the speciation. The graphs that were generated follow.

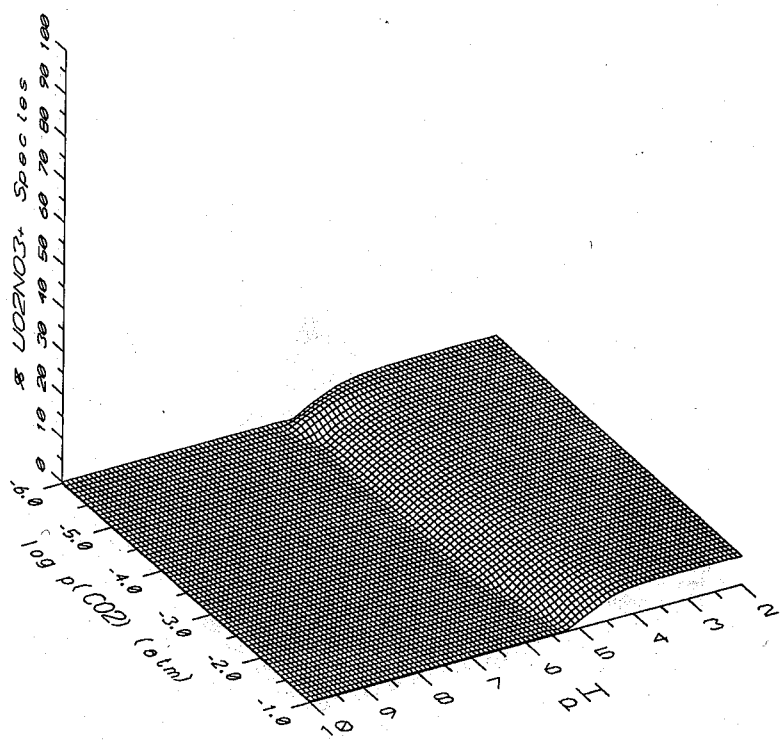


Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ J.O. DEJ 8/16/94



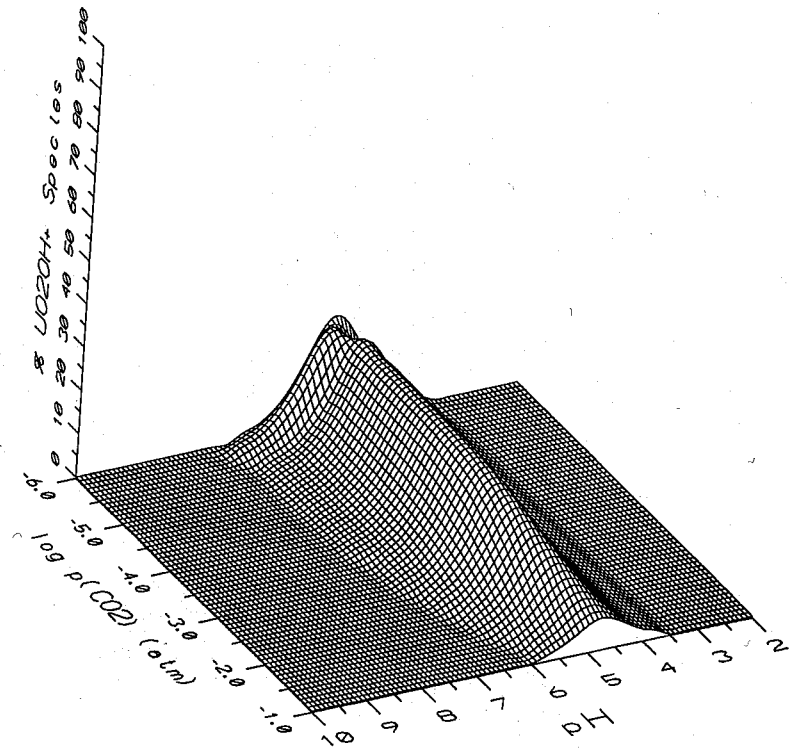
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ T.O. DEJ 8/16/94



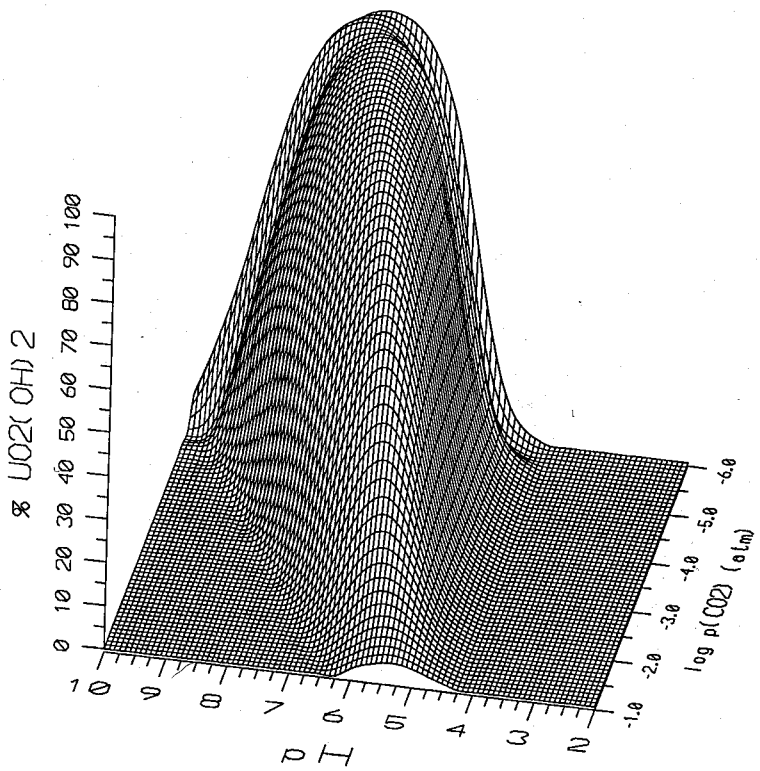
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ T.O. DEJ 8/16/19



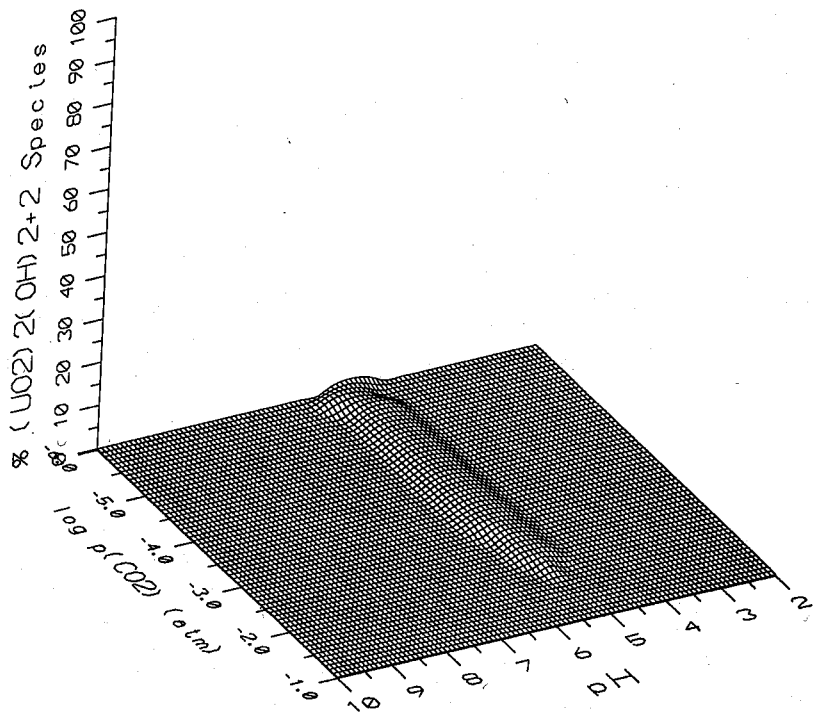
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ T.O. DCJ 8/16/99



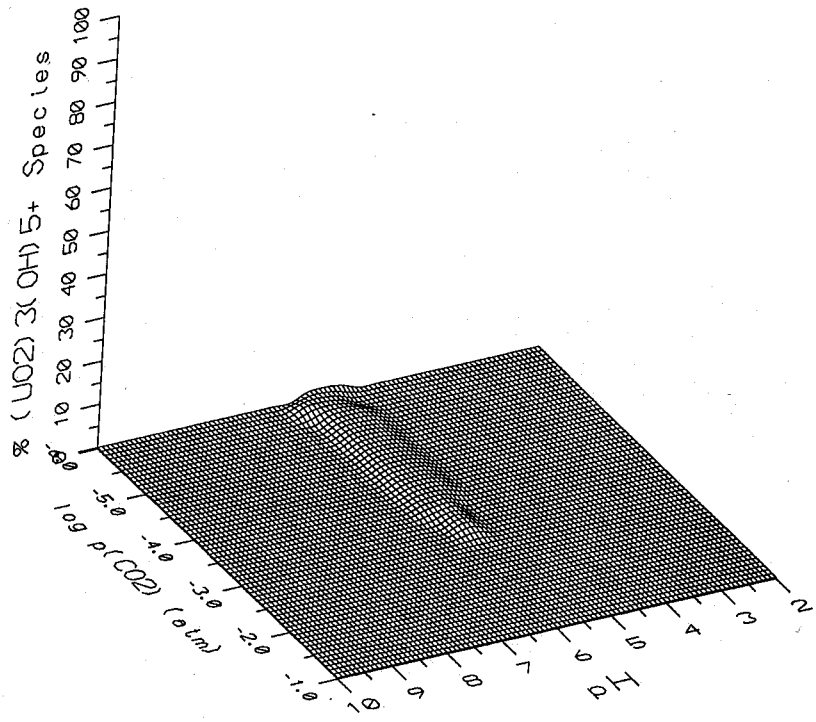
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ J.O. (DC) 08/16/94



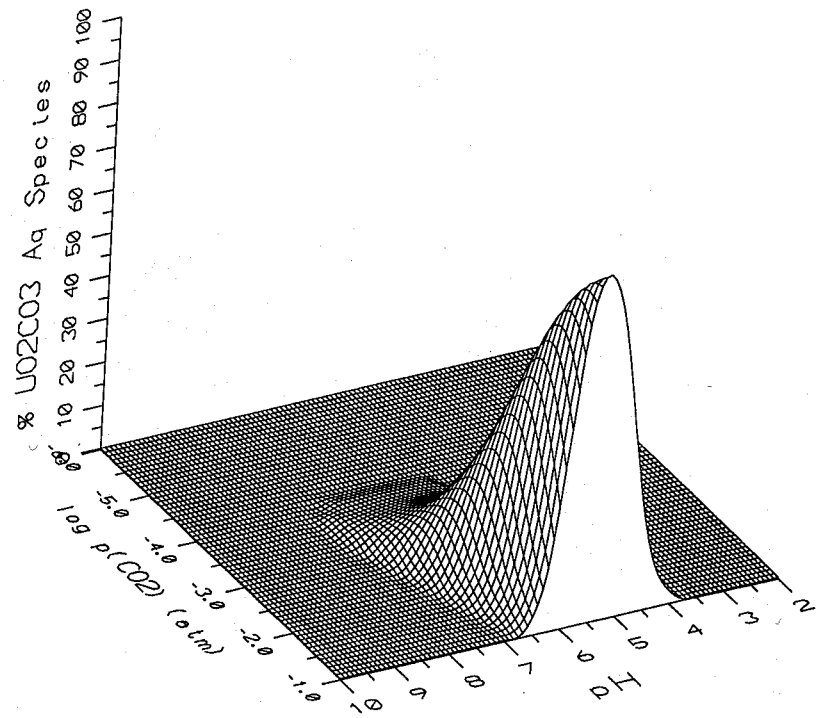
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ J.O. DEJ 8/16/94



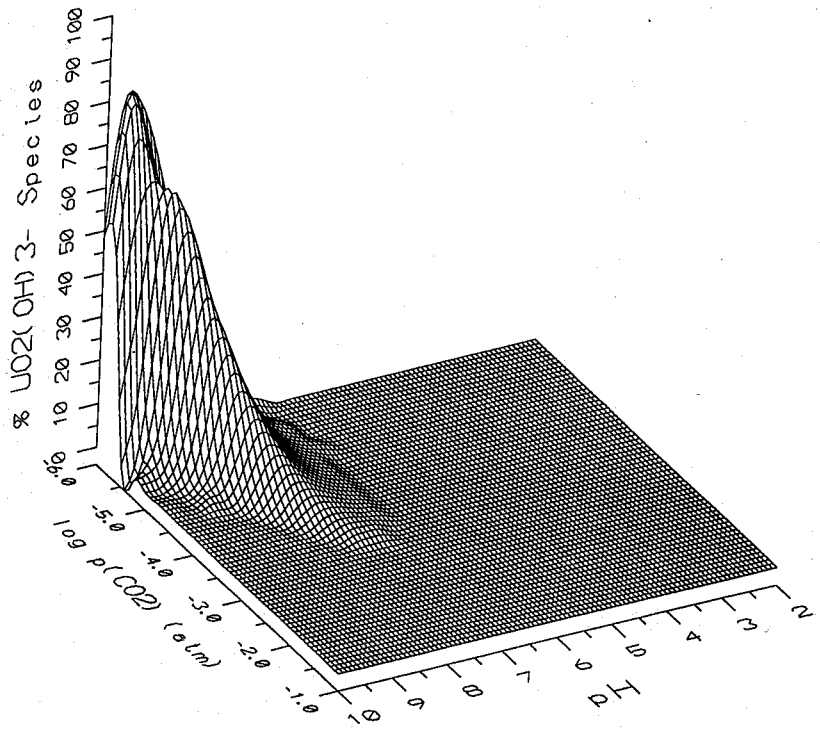
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ T.O. DEJ 8/16/94



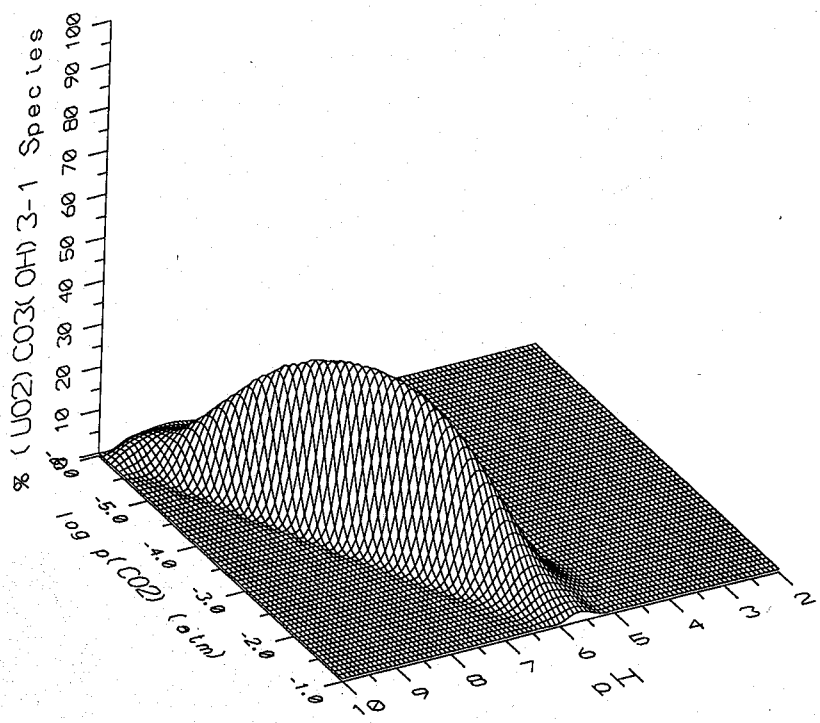
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ J.O. (DC) 8/16/94



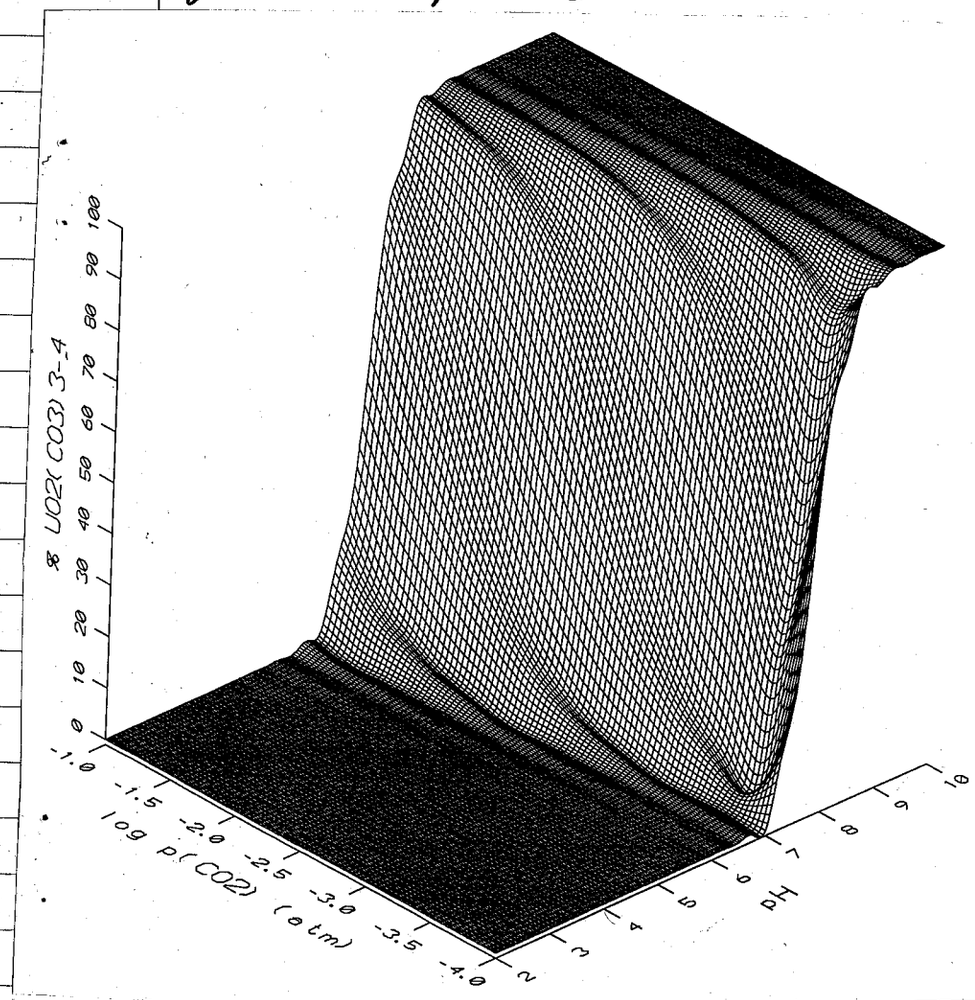
Although this graph is more pleasing to the eye, the ripples

Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ J.O. (DC) 8/16/94



Although this graph is more pleasing to the eye, the ripples

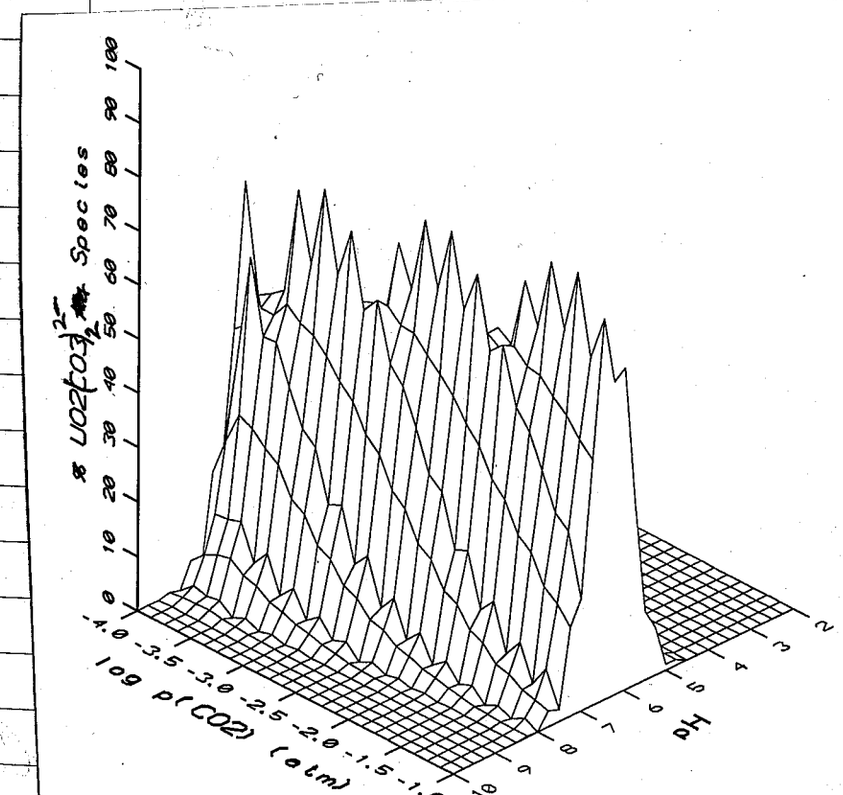
Analysis of the graph showed that something was still happening from about log $p(CO_2) = -3.75$ to -4.00. I then modified the graph by multiplying the grid by a factor of 6 and splining it. ~~Because~~ T.O. DEJ 8/16/94



Although this graph is more pleasing to the eye, the ripples

are not a true representation of $UO_2(CO_3)_3^{4-}$. T.O. ~~The~~ At 10^{-4} or log $p(CO_2) = -4$ atm and DEJ 8/16/94 pH = 7 we begin to see a wave. P/vs the ripples before and after the drastic changes in the slope arise from the fact that ~~the~~ T.O. DEJ 8/16/94 Surfer has generated points.

* One probable solution of getting rid of the ripples would be to go back into MINTEQA2 and ~~increase~~ DEJ 8/16/94 decrease the pH increment size to 0.25 instead of 0.5 and extend log $p(CO_2)$ to -6



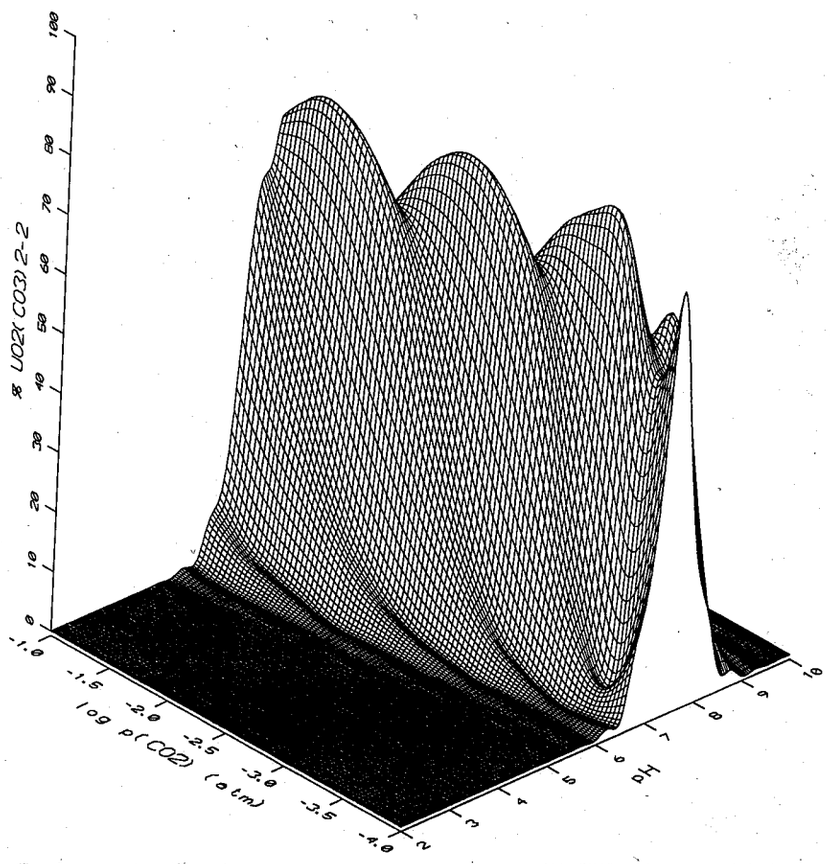
MINTEQA2 Speciation: UO2CO3 Aq

This graph above shows the true points of $UO_2(CO_3)_2^{2-}$. It was generated in Surfer and can be compared with the $UO_2(CO_3)_2^{2-}$ graphs on pg 13.

SigmaPlot

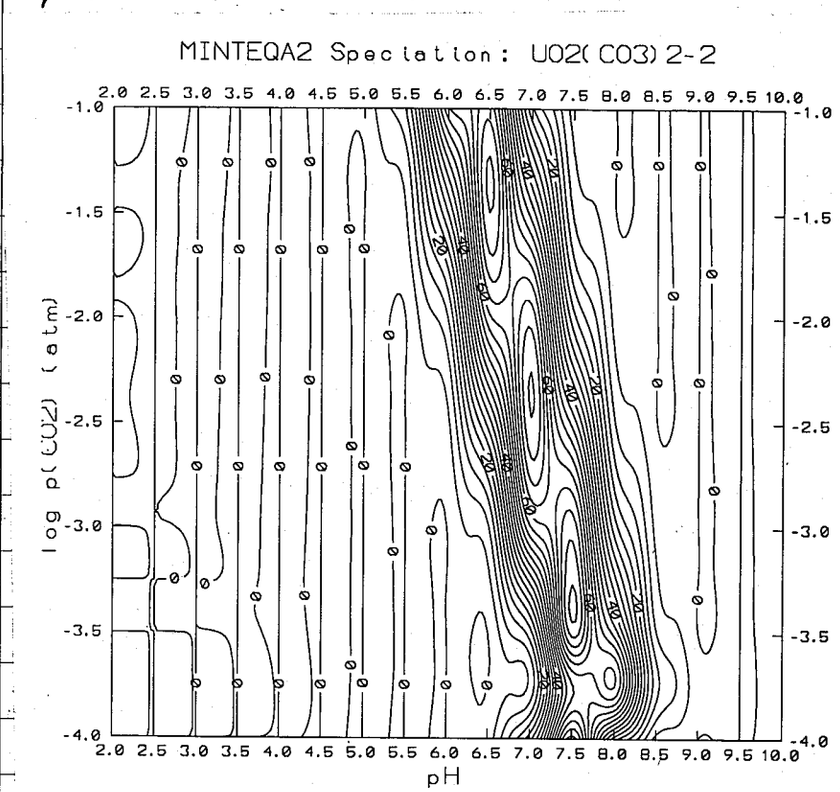
TB 9/16

Below is a spline of the above graph, which had the grid multiplied by six and smoothed.



Using the above 6X grid, a topographical map was generated so that the true shift across the range of

pH could be viewed.



8-10-92 T.G.

After viewing the surfer graphs in both 3-D and topographical, it was concluded that another run be made in MINTEQA2. This run would again use 2E-6 M $UO_2(NO_3)_2$ and 0.1 M $NaNO_3$ as the main components. However, the range of pressure for this run will span from 10^{-1} atm to 10^{-6} atm and the pH will again be from 2 to 10, but there will be a 0.25 increment

between pH units. Hopefully, Surfer will be able to better interpret the MINTEQA2 results when generating new points.

8-11-92 After creating the input file with PRODEFA2 and labelling the file:

UPHCO256,

U – 2E-6 M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$

T.6. pH – pH sweep from 2-11, 0.25 increment (36 divisions)

~~log~~ ~~CO~~ – log $p(CO_2)$ [atm], sweep from 10^{-1} to 10^{-4}, .25 increments.

(DR) 8/16/94 25 – represents increment size of pH

6 – represents the lowest concentration (or pressure) of CO_2.

it was then run in MINTEQA2.

Using a 386 SX IBM, the run lasted approximately 2 hrs. and the output (DR) 8/16/94 ~~that was~~ T.6. generated was 7.8 Megabytes on the hard drive.

* NOTE: The next revisions to MINTEQA2 will be from adding write statements to the Fortran source code ~~and~~ T.6. (DR) 8/16/94 that will automatically write Part 4 of the OUTPUT FILE to a file. It will reduce the time required by me to

reduce the data.

8-13-92 The first page of uPHCO256, as ~~the~~ T.6. (DR) 8/16/94 a MINTEQA2 input file, can be seen below.

```
2E-6 M U(VI)T; 0.1 M NaNO3; New Dbs; 3-D, Variable pH and CO2
Last Thermo.dbs revision, 6-22-93;
25.00 MOLAL  0.000
0 0 1 0 3 0 0 0 1 1 0 1 0
ACTIVITY   330 20
  0.25
0   0   0
    330  1.000E-02   -2.00                    /H+1
    140  0.000E-01  -16.00                    /CO3-2
    893  2.000E-06   -5.70                    /UO2+2
    492  1.000E-01   -1.00                    /NO3-1
    500  1.000E-01   -1.00                    /Na+1

  3   2
3301403    24.1600   -0.5300                  /CO2 (g)
    330     2.0000    0.0000                  /H+1
```

← 92

← Increment pH by .25 for 20 times

← pH = 2

$-\log K$ p_{CO_2} = 18.16 @ 1atm

$-18.16 + 6 = 24.16$

$p(CO_2) = 10^{-6}$ atm
last pH for sweep @ 10^{-6} is 6.75. Started @ 2.00.

```
2E-6 M U(VI)T; 0.1 M NaNO3; New Dbs; 3-D, Variable pH and CO2
Last Thermo.dbs revision, 6-22-93;
25.00 MOLAL  0.000
0 0 1 0 3 0 0 0 1 1 0 1 0
ACTIVITY   330 16
  0.25
0   0   0
    330  1.000E-07   -7.00                    /H+1
    140  0.000E-01  -16.00                    /CO3-2
    893  2.000E-06   -5.70                    /UO2+2
    492  1.000E-01   -1.00                    /NO3-1
    500  1.000E-01   -1.00                    /Na+1

  3   2
3301403    24.1600   -0.5300                  /CO2 (g)
    330     7.0000    0.0000                  /H+1
```

← pH = 7

$p(CO_2) = 10^{-6}$ atm
last pH for sweep @ 10^{-6} is 10.75. Started @ 7.00.

```
2E-6 M U(VI)T; 0.1 M NaNO3; New Dbs; 3-D, Variable pH and CO2
Last Thermo.dbs revision, 6-22-93;
25.00 MOLAL  0.000
0 0 1 0 3 0 0 0 1 1 0 1 0
ACTIVITY   330 20
  0.25
0   0   0
    330  1.000E-02   -2.00                    /H+1
    140  0.000E-01  -16.00                    /CO3-2
    893  2.000E-06   -5.70                    /UO2+2
    492  1.000E-01   -1.00                    /NO3-1
    500  1.000E-01   -1.00                    /Na+1

  3   2
3301403    23.9100   -0.5300                  /CO2 (g)
    330     2.0000    0.0000                  /H+1
```

← pH = 2

Once we have a pH sweep from 2-10, we then increase the $p(CO_2)$ conc. (DR) 8/16/94 by $10^{+0.25}$.

Now 18.16 + 5.75 = 23.91

```
2E-6 M U(VI)T; 0.1 M NaNO3; New Dbs; 3-D, Variable pH and CO2
Last Thermo.dbs revision, 6-22-93;
25.00 MOLAL  0.000
0 0 1 0 3 0 0 0 1 1 0 1 0
ACTIVITY   330 16
  0.25
0   0   0
    330  1.000E-07   -7.00                    /H+1
    140  0.000E-01  -16.00                    /CO3-2
    893  2.000E-06   -5.70                    /UO2+2
    492  1.000E-01   -1.00                    /NO3-1
    500  1.000E-01   -1.00                    /Na+1

  3   2
3301403    23.9100   -0.5300                  /CO2 (g)
    330     7.0000    0.0000                  /H+1
```

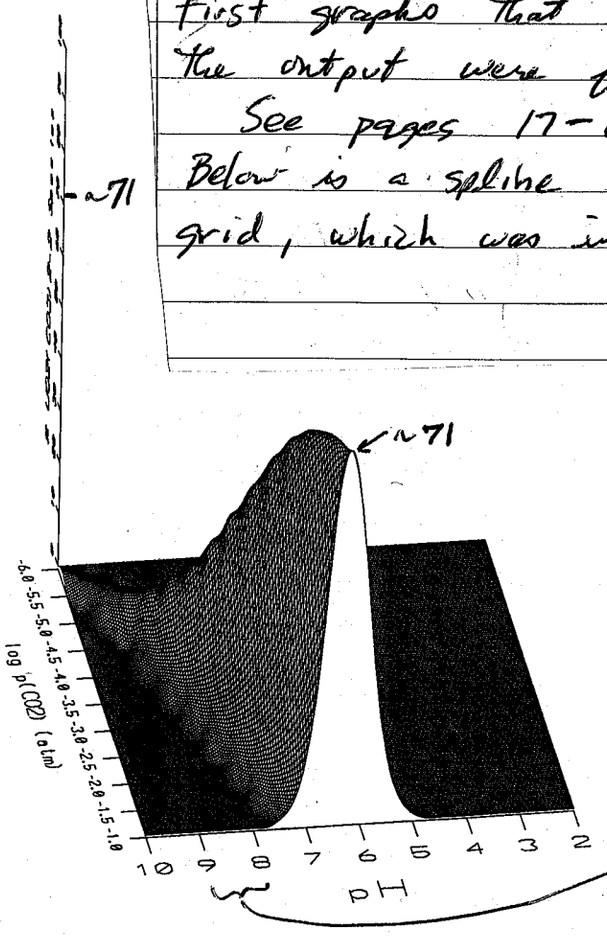
Note: the pH sweep begins all over again for log K = 23.91. First @ pH = 2, then @ pH = 7.

```
2E-6 M U(VI)T; 0.1 M NaNO3; New Dbs; 3-D, Variable pH and CO2
Last Thermo.dbs revision, 6-22-93;
25.00 MOLAL  0.000
0 0 1 0 3 0 0 0 1 1 0 1 0
ACTIVITY   330 20
  0.25
0   0   0
    330  1.000E-02   -2.00                    /H+1
    140  0.000E-01  -16.00                    /CO3-2
```

T.G. DRJ 8/16/94
from UPHCO256.out

8-13-92 T.G. The output was pulled up into
8-14-92 T.G. WordPerfect. Paula Muller assisted me obtaining the component concentrations. As I paged down through the 5000+ (plus) pages of output, she typed them into a Quattro Pro spreadsheet. The first graphs that were generated from the output were for $UO_2(CO_3)_2^{2-}$.

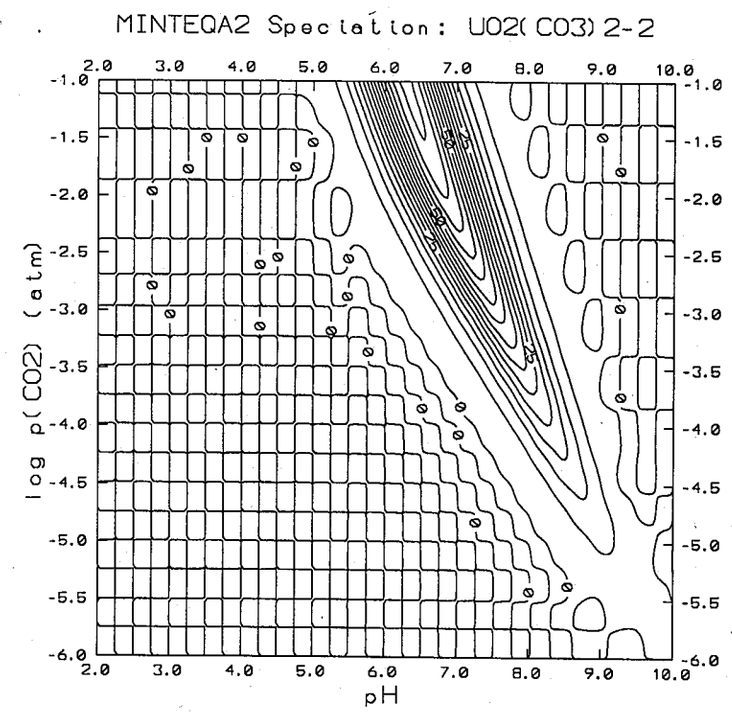
See pages 17-19 for comparison. Below is a spline of the $UO_2(CO_3)_2^{2-}$ grid, which was increased 6 fold.



for exact numbers, see UPHCO256.WQ1.

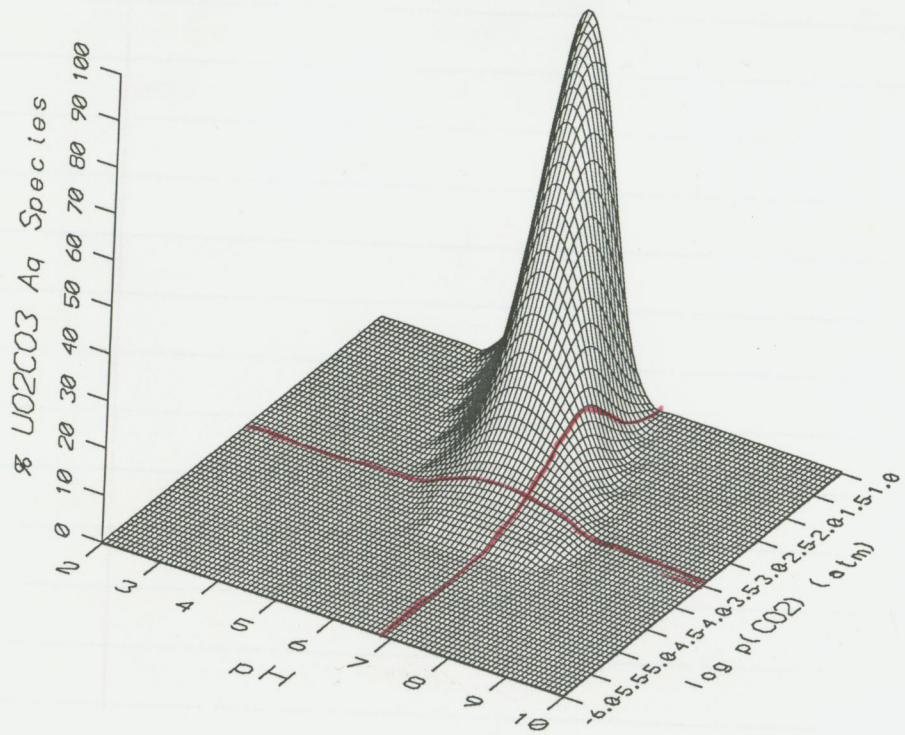
NOTE that there appear to be 4 ripples between each tick which occur at each increment of 0.25.

The next figure is another topographical view of $UO_2(CO_3)_2^{2-}$. Why the peak height has decreased from 75.6 ~~in figure~~ T.G. DRJ 8/16/94 (pg 19) to ~71, I am still not sure. However, there is a similar ~~shi~~ DRJ 8/16/94 shift across the pH range from 5-9.

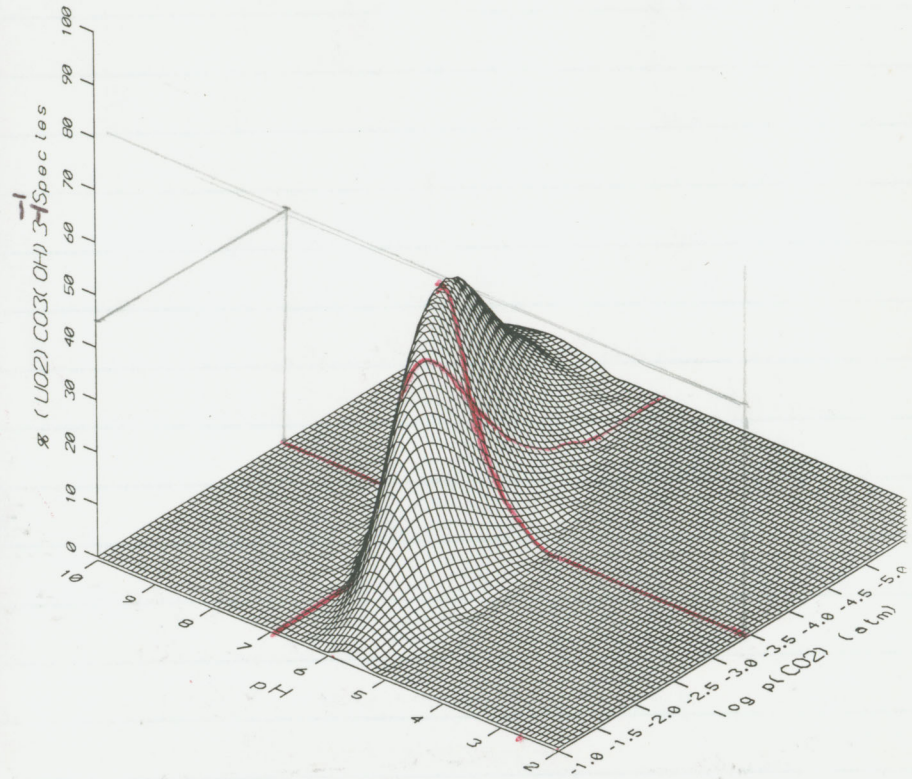


8-17-92 T.G. Continued making graphs from the UPHCO256.out file using Surfer 4.0. After looking at several variations of modifying the grid using the splining technique, T.G. DRJ 8/16/94 ~~It was~~ decided

that a factor of 2 (for grid increase) would be sufficient.



For comparative purposes one can go back to the old 2-D graphs for 2E-6 M $UO_2(NO_3)_2$; 0.1 M $NaNO_3$ and follow the red grid lines above to ensure that no errors in the runs existed.



8-20-92 T.G.

This morning, I began reading the PACKARD Liquid Scintillation Counter Manual.

Finished making 3-D graphs for every U(VI)$_T$ Species in the UPHCO256.out MINTEQA2 run.

Once again, at the lowest concentration, we see that there is a sudden shift or ripple in

JRW 8/16/94

cont. the surfaces that extended (or existed) into (or at) the 10^{-6} region.

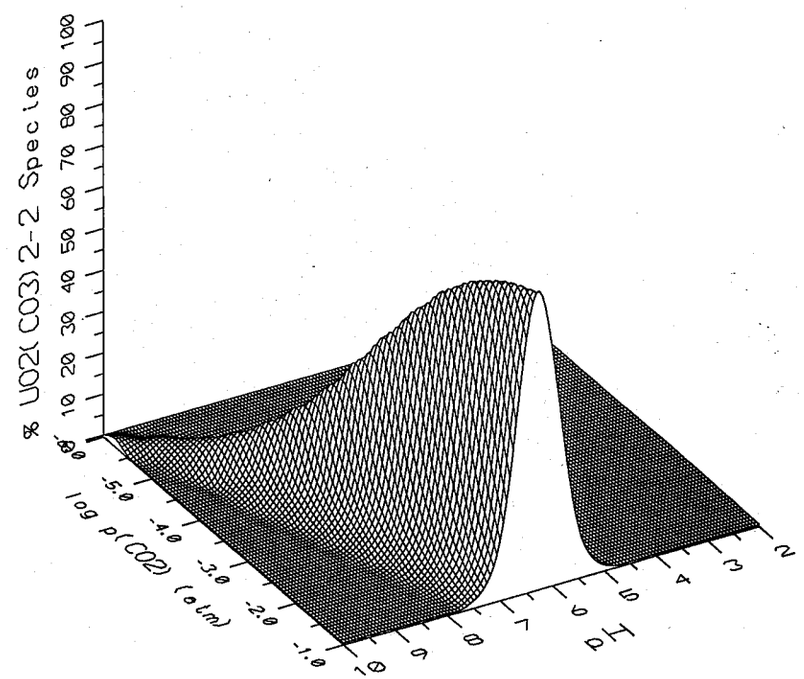
For now, these anomalies can only be associated with the splining of the grids in the SURFER program. My reasoning lies in the fact that similar deformaties were seen in UPHC06-C output, which was just a smaller file of ~~for~~ T.E. JRW 8/16/94 UPHC0256.

Each species can be seen in the graphs that follow on the next page. The grid files that were made of the data were labeled as: *.grd and *.plt

col #	e.g. Species	Filename
1	UO_2^{2+} :	UPHC06C. (grd, plt)
2	$UO_2NO_3^{+}$:	UPHC06D. (grd, plt)
3	UO_2OH^{+} :	UPHC06E. (grd, plt)
4	$UO_2(OH)_2^{0}$:	UPHC06F. (grd, plt)
5	$(UO_2)_2(OH)_2^{+2}$:	UPHC06G. (grd, plt)
6	$(UO_2)_3(OH)_5^{+1}$:	UPHC06H. (grd, plt)
7	UO_2CO_3Ag :	UPHC06I. (grd, plt)
8	$UO_2(OH)_3^{-}$:	UPHC06J. (grd, plt)
9	$(UO_2)_2CO_3(OH)_3^{-}$:	UPHC06K. (grd, plt)
10	$UO_2(CO_3)_2^{2-}$:	UPHC06L. (grd, plt)
11	$UO_2(CO_3)_4^{2-}$:	UPHC06M. (grd, plt)

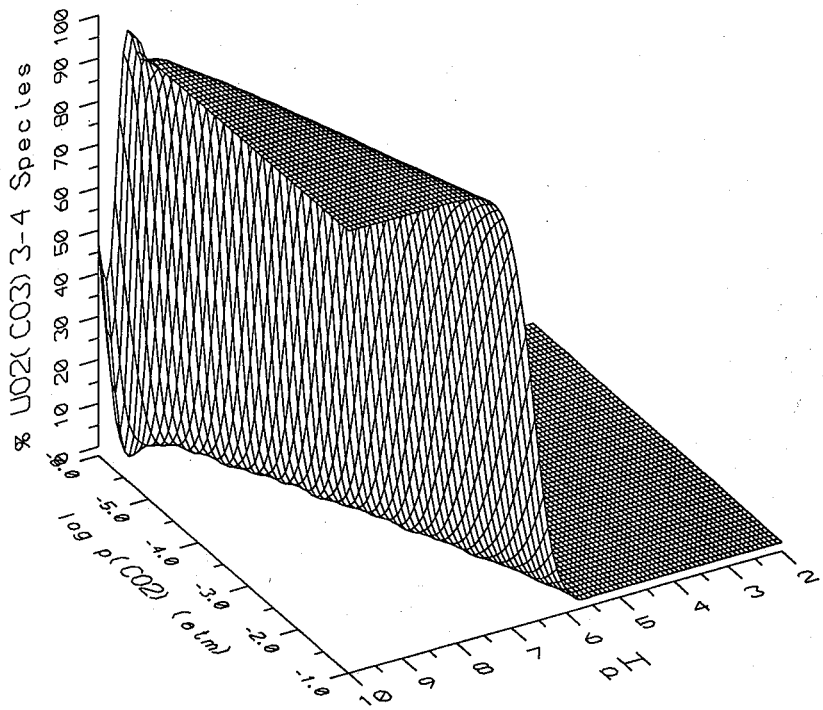
Filename: UPHC0256.WKI or (.WQ1)
2E-6 M $UO_2(NO_3)_2$; 0.1M $NaNO_3$

pH	log p(CO2)	UO2+2	UO2NO3+	UO2OH+	UO2(OH)2	UO2(OH)2+	UO2(OH)5+	UO2CO3	UO2(OH)3-	UO2(CO3)(OH)	UO2(CO3)2-2	UO2(CO3)3-4
2	-6	93.6	6.3	0	0	0	0	0	0	0	0	0
2.25	-6	93.6	6.3	0	0	0	0	0	0	0	0	0
2.5	-6	93.5	6.4	0	0	0	0	0	0	0	0	0
2.75	-6	93.4	6.4	0	0	0	0	0	0	0	0	0
3	-6	93.3	6.4	0	0	0	0	0	0	0	0	0
3.25	-6	93.1	6.4	0	0	0	0	0	0	0	0	0
3.5	-6	92.7	6.4	0	0	0	0	0	0	0	0	0
3.75	-6	92.1	6.3	1.5	0	0	0	0	0	0	0	0
4	-6	90.9	6.3	2.6	0	0	0	0	0	0	0	0
4.25	-6	88.8	6.1	4.5	0	0	0	0	0	0	0	0
4.5	-6	84.5	5.8	7.7	1.4	0	0	0	0	0	0	0
4.75	-6	77.4	5.3	12.5	4.1	0	0	0	0	0	0	0
5	-6	64.7	4.5	18.6	10.8	1.4	0	0	0	0	0	0
5.25	-6	46.2	3.2	23.6	24.4	2.2	0	0	0	0	0	0
5.5	-6	26.4	1.8	24.1	44.2	2.3	1.1	0	0	0	0	0
5.75	-6	12.1	0	19.6	63.9	1.5	1.8	0	0	0	0	0
6	-6	4.7	0	13.5	78.6	0	1.9	0	0	0	0	0
6.25	-6	1.7	0	8.5	87.6	0	1.5	0	0	0	0	0
6.5	-6	0	0	5	92.7	0	0	0	0	0	0	0
6.75	-6	0	0	2.9	95.2	0	0	0	0	0	0	0
7	-6	0	0	1.7	96.1	0	0	0	1.6	0	0	0
7.25	-6	0	0	0	95.7	0	0	0	2.8	0	0	0
7.5	-6	0	0	0	93.8	0	0	0	4.9	0	0	0
7.75	-6	0	0	0	90.2	0	0	0	8.4	0	0	0
8	-6	0	0	0	84.3	0	0	0	14	1.5	0	0
8.25	-6	0	0	0	75.5	0	0	0	22.2	2.1	0	0
8.5	-6	0	0	0	63.8	0	0	0	33.4	2.7	0	0
8.75	-6	0	0	0	50.2	0	0	0	46.7	3	0	0
9	-6	0	0	0	36.5	0	0	0	60.5	2.8	0	0
9.25	-6	0	0	0	24.6	0	0	0	72.5	2.3	0	0
9.5	-6	0	0	0	15.3	0	0	0	80.2	1.6	0	2.4
9.75	-6	0	0	0	8.3	0	0	0	76.9	0	0	13.2
10	-6	0	0	0	2.9	0	0	0	48.7	0	0	47.3
2	-5.75	93 6	6.3	0	0	0	0	0	0	0	0	0



Filename: UPHC0256.WKI or (.WQ1)
.2E-6 M $UO_2(NO_3)_2$; 0.1M $NaNO_3$

pH	log p(CO2)	UO2+2	UO2NO3+	UO2OH+	UO2(OH)2	UO2)2(OH)2+	UO2)3(OH)5+	UO2CO3	UO2(OH)3-	UO2)CO3(OH)	UO2(CO3)2-2	UO2(CO3)3-4
2	-6	93.6	6.3	0	0	0	0	0	0	0	0	0
2.25	-6	93.6	6.3	0	0	0	0	0	0	0	0	0
2.5	-6	93.5	6.4	0	0	0	0	0	0	0	0	0
2.75	-6	93.4	6.4	0	0	0	0	0	0	0	0	0
3	-6	93.3	6.4	0	0	0	0	0	0	0	0	0
3.25	-6	93.1	6.4	0	0	0	0	0	0	0	0	0
3.5	-6	92.7	6.4	0	0	0	0	0	0	0	0	0
3.75	-6	92.1	6.3	1.5	0	0	0	0	0	0	0	0
4	-6	90.9	6.3	2.6	0	0	0	0	0	0	0	0
4.25	-6	88.8	6.1	4.5	0	0	0	0	0	0	0	0
4.5	-6	84.5	5.8	7.7	1.4	0	0	0	0	0	0	0
4.75	-6	77.4	5.3	12.5	4.1	0	0	0	0	0	0	0
5	-6	64.7	4.5	18.6	10.8	1.4	0	0	0	0	0	0
5.25	-6	46.2	3.2	23.6	24.4	2.2	0	0	0	0	0	0
5.5	-6	26.4	1.8	24.1	44.2	2.3	1.1	0	0	0	0	0
5.75	-6	12.1	0	19.6	63.9	1.5	1.8	0	0	0	0	0
6	-6	4.7	0	13.5	78.6	0	1.9	0	0	0	0	0
6.25	-6	1.7	0	8.5	87.6	0	1.5	0	0	0	0	0
6.5	-6	0	0	5	92.7	0	0	0	0	0	0	0
6.75	-6	0	0	2.9	95.2	0	0	0	0	0	0	0
7	-6	0	0	1.7	96.1	0	0	0	1.6	0	0	0
7.25	-6	0	0	0	95.7	0	0	0	2.8	0	0	0
7.5	-6	0	0	0	93.8	0	0	0	4.9	0	0	0
7.75	-6	0	0	0	90.2	0	0	0	8.4	0	0	0
8	-6	0	0	0	84.3	0	0	0	14	1.5	0	0
8.25	-6	0	0	0	75.5	0	0	0	22.2	2.1	0	0
8.5	-6	0	0	0	63.8	0	0	0	33.4	2.7	0	0
8.75	-6	0	0	0	50.2	0	0	0	46.7	3	0	0
9	-6	0	0	0	36.5	0	0	0	60.5	2.8	0	0
9.25	-6	0	0	0	24.6	0	0	0	72.5	2.3	0	0
9.5	-6	0	0	0	15.3	0	0	0	80.2	1.6	0	2.4
9.75	-6	0	0	0	8.3	0	0	0	76.9	0	0	13.2
10	-6	0	0	0	2.9	0	0	0	48.7	0	0	47.3
2	-5.75	93.6	6.3	0	0	0	0	0	0	0	0	0



8-24-92 T.G.

If one looks close enough at the graphs on the previous page, they can see that a shift has occured in the log 10^{-5} to log 10^{-6} range. I believe that these shifts are due to the Surfer splining.

DRJ 8/16/94 ~~Also,~~ Today, Dave Turner returned from the MINTEQA2 seminar with the newest version, Ver. 3.11, and this version DRJ 8/16/94 ~~can~~ will, if specified, write up to 6 species' concentrations to T.G. DRJ 8/16/94 ~~an~~ spreadsheet file.

cont. 8-24-92 T.G.

After loading T.G. DRJ 8/16/94 ~~the~~ Ver. 3.11, I ran it using the thermo and type6 files that we revised (or modified). and used the same file UPHCO256 to create an new file,

UPHCO016, which has a $p(CO_2)$ increment of 0.1. I had to run it twice, since there are a total of 11 uranium species which exist over the pH range

These spreadsheet files need to be modified. That is, the input that they receive from MINTEQA2 is in the form of concentrations. Therefore, in order to determine the % bound for each species, one must divide by the Radioelement concentration, DRJ 8/16/94 ~~for this run it~~ T.G. it was 2E-6 for this run, and multiply by 100.

8-26-92 After the spreadsheet files had been generated and all the uranium species all brought into one file, new graphs were made.

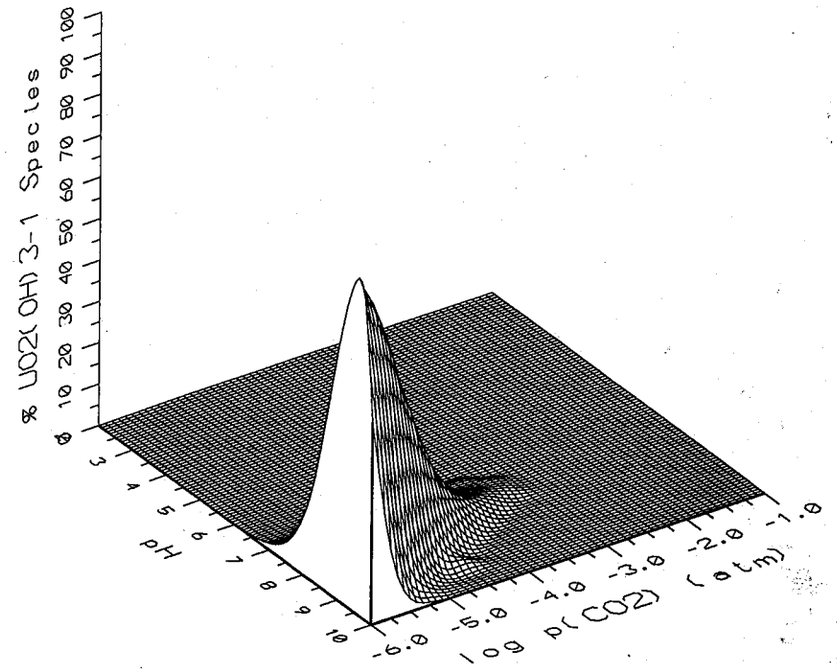
Below is a brief explanation of the new MINTEQA2 file, UPHCO016:

```
2E-6 M UO2(NO3)2; 0.1 M NaNO3; New Dbs; 3-D, Variable pH and CO2
Last Thermo.dbs revision, 6-22-92
25.00 MOLAL 0.000  0.00000E-01
0 0 1 0 3 0 0 0 1 1 1 5 3
ACTIVITY  330 20   → ends at pH = 6.75
 0.25              output spreadsheet file
uphco016.wk1 8933302 8931401 8931402 8931405   893
0  0  0                                  6 uranium output species
  330  0.000E-01  -2.00 y   starting pH.   /H+1
  893  2.000E-06  -5.70 y   U(VI) conc.    /UO2+2
  500  1.000E-01  -1.00 y                  /Na+1
  492  1.000E-01  -1.00 y                  /NO3-1
  140  0.000E-01  -16.00 y                 /CO3-2

 3  2                                  log K for p(CO2) = 10^-6
3301403  24.1600  -0.5300              /CO2 (g)
  330    2.0000   0.0000               /H+1
```

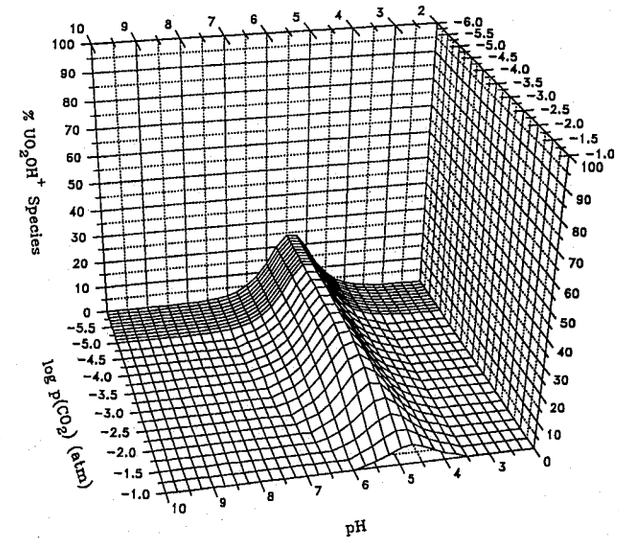
The first graph T.G. DRJ 8/16/94 ~~of~~ made from this run was $UO_2(OH)_3^-$. It follows.

and was done using SURFER 4.0.



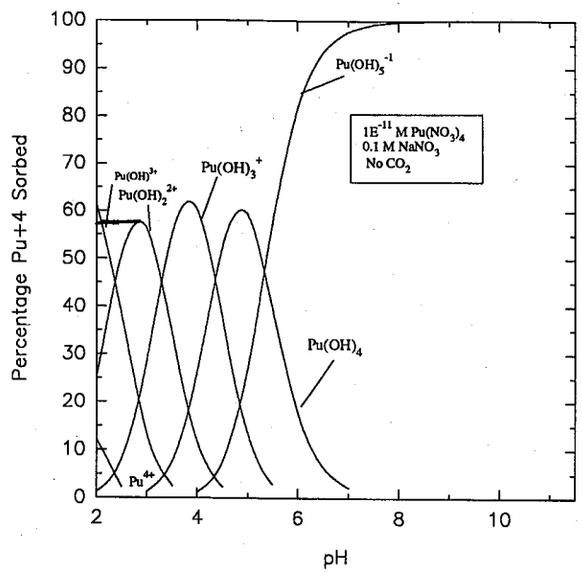
8-27-92 T.6.

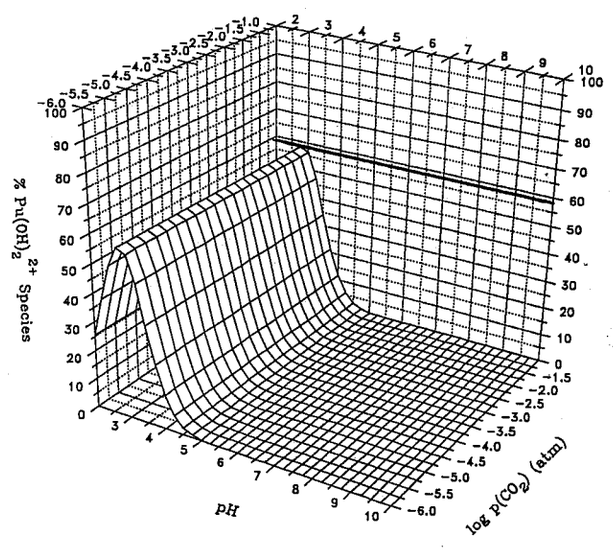
After looking at the above graph, I decided to retry SigmaPlot 5.0. Using just the MINTEQA2 generated points and no spline, the following graphs were made. Looking back at pg 27, one can compare both. However, using SigmaPlot, one can determine very easily the location of a surface point.



8-27-92 cont. T.6.

Because the graphs above were turning out very well, I decided that I would play with another radioelement. Looking back at earlier Plutonium runs, specifically P4N11 ... I then decided to run the same file above over a sweep of $p(CO_2)$ to determine whether any plutonyl carbonates would appear. Below are the results. The first graph is the original 2-D (for $1E^{-11}$ M $Pu(NO_3)_4$; 0.1 M $NaNO_3$; No CO_2). ~~and~~ ~~I decided~~ T.6. Just $Pu(OH)_2^{2+}$ was graphed.





One thing I didn't realize, but that Dave Turner pointed out, was that Pu^{4+} or Component #641 doesn't have any ~~solid~~ carbonate species associated with it. Consequently, there is no change in the plutonyl hydroxyl species ~~over the due to~~ arising from T.G. a change in $P(CO_2)$.

8-28-92 T.G.

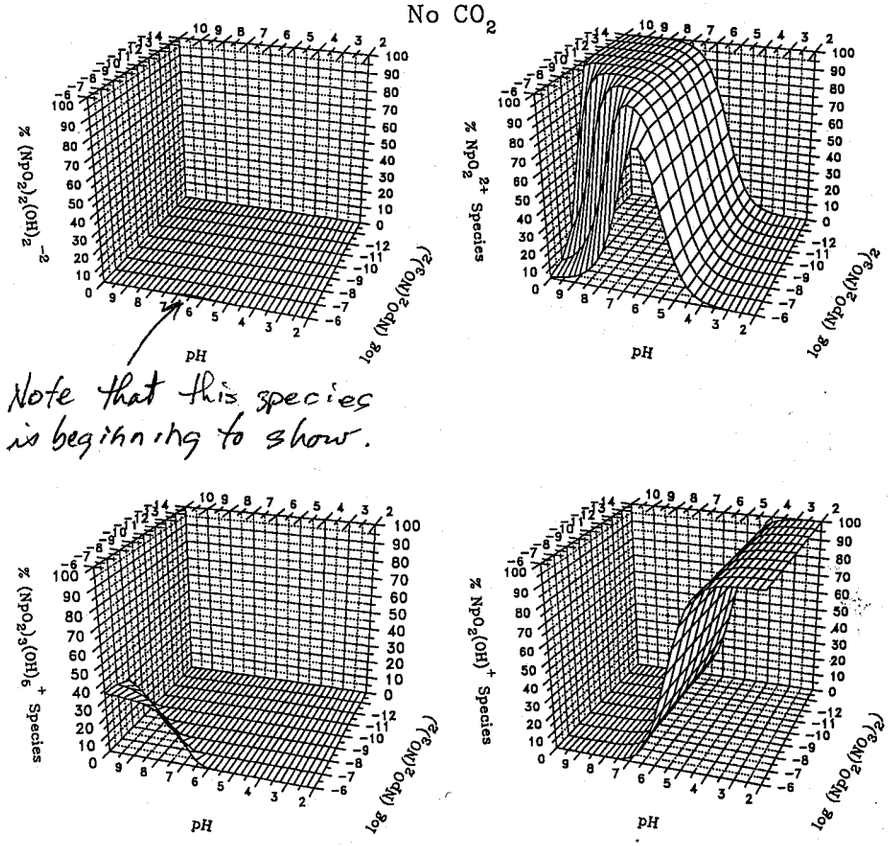
At the request of Dr. David Turner, a MINTEQA2 run was performed on Np(VI). However, unlike all previous radioelement runs, the system was isolated from atmospheric CO_2, and the concentration of $NpO_2(NO_3)_2$ varies from 10^{-6} M to 10^{-14} M. All four graphs, since only four species ~~are~~ are in the MINTEQA2 DBS, were ~~put on~~ T.G. DRJ 8/16/94 plotted on the same page. That page follows.

Analysis of the graphs show that something could be wrong. e.g. @ (10, -6), the total % of species that exist is just ~35%. It should be 100% total species at all times. See the graph for comparison.

Variable $NpO_2(NO_3)_2$

0.1 M $NaNO_3$

No CO_2



Note that this species is beginning to show.

8-31-92 T.G.

Went to see Dr. Turner in Bldg. 168 and discussed the graphs of the Neptunium and Plutonium runs. We reached no true conclusions for Np(VI). DRW 8/16/94

However, we did decide to ~~audit~~ T.G. revise the thermo.dbs. That is, all DRW 8/16/94 of the references for the log K's

were placed in column 82-90 and the program was tested or compiled to T.G. determine whether the program would crash. DRW 8/16/94

Dave also had me go through "Chemical Thermodynamics of Uranium," Vol 1., which is a 1992 copyright of the OECD Nuclear Energy Agency (NEA), and compare the log K's from EQ3/6 with the NEA's.

9-1-92 T.G.

After comparing log K's, I began adding references to the dbs. I also changed most of the log K's for the aqueous uranium species by replacing them with those found in NEA's Vol 1. They are referenced as NEA 1992. T.G. DRW 8/16/94

Please note that just because our T.G. DRW 8/16/94 log K's were changed don't mean that they were incorrect. Actually, all of them fell within the range of values offered by NEA.

Stephen Sassman 5-21-93

Analysis of Log K_+ and Log K_- values for Goethite at varying ionic strength were examined to determine "average" values. DLM & TLM Data were determined by a weighting factor method shown below. CCM data could not be "averaged" in this manner, because the ~~CDM~~ SSM CCM does not account for ionic strength. Therefore, Log K values are valid only at one ionic strength. Log K's for CCM were determined by an extrapolation method suggested by Dave: By plotting Log I versus log K and fitting a regression line to the data Log K's can be determined at any ionic strength. 81 hsi and 90 hayes data is located in the Digitized Data-Hsi (1981) folder

DLM & TLM should be corrected for ionic strength before use

CCM Good for only 0.1=I

Goethite (~~0.1=I~~) SS. 5/25/93

good for 39 all ionic strengths

DLM: $\text{Log}\,K_+^{0.1} = 7.73$ (SS. 5-21 93) $\text{Log}\,K_-^{0.1} = -9.27$ (SS 5-21 93)

CCM: $\text{Log}\,K_+^{0.1} = 7.37$ $\text{Log}\,K_-^{0.1} = -9.32$

TLM: $\text{Log}\,K_{cat}^{0.1} = -7.56$ (SS 5-21 93) $\text{Log}\,K_{an}^{0.1} = 9.15$ (SS 5-21 93)

Calculation of ~~K_+ &~~ $\text{Log}\,K_+$ and $\text{Log}\,K_-$ for TLM and DLM:

(All data from 81hsi & 90hayes)

$\text{Log}\,K_+^i$ (DLJ 8/16/94)	Inverse of $\text{sig}\,K_+^i$	w.f. = $\frac{\text{inv sig}\,K_{+,i}^i}{\sum_i \text{inv sig}\,K_+^i}$	$\text{Log}\,K_+^i \times$ w.f.
7.52	50.251	0.0827	0.6219
7.38	24.938	0.0411	0.3033
6.95	65.789	0.1083	0.7527
6.73	70.423	0.1160	0.7807
8.87	112.360	0.1850	1.6410
8.56	23.866	0.0393	0.3364
8.05	54.645	0.0900	0.7245
7.74	90.090	0.1483	1.1478
7.53	114.943	0.1893	1.4254

$\sum_i \text{sig}\,K_+^i = 607.305$

$\sum_i \text{Log}\,K_+^i \times \text{w.f.} = 7.734 = \text{Log}\,K_+^{0.1}$ (SS. 5-21 93)

5/23/93 Stephen Sassman

The log K values for Goethite that are listed on the previous page (after being corrected for ionic strength = 0.1M) were used with data from the Hsi dissertation to determine log K's for sorption of Uranium on Goethite using Fiteql. The following naming scheme was (DW) 8/16/94 used for all input and out put files

Input Files:

XOH2UO24.DGO

- XOH2 → # of H's on surface
- UO24 → # of hydroxyl groups
- D → D, C, or T for model used
- GO → Goethite

Output Files:

UOH4DDZ2.GO

- UOH4 → # of hydroxyl groups
- DDZ2 → # of H's on surface
- GO → Goethite

or

UO2ddz1.GO for no hydroxyls

These files are in the directory

K:/Turner/Fiteql/Input/CCM or DLM or TLM/Uranium/Hsi

Sorption of UO_2^{2+} on Goethite (I=0.1)

Species	DLM Log K	DLM 0.1 SOS/DF	DLM σ Log K	CCM Log K	CCM 0.1 SOS/DF	CCM σ Log K	TLM Log K	TLM 0.1 SOS/DF	TLM σ Log K
	S.S 5/28/93						S.S 5/28/93		
$XOUO_2^+$	3.120	1.158	4.233E-2	3.119	1.084	4.233E-2	-2.638	35.47	5.877E-2
	~~3.221~~	~~1.176~~	~~4.247E-2~~				~~-2.831~~	~~35.79~~	~~5.758E-2~~
$XOH \cdot UO_2^{+2}$	9.869	8.827	3.329E-2	10.58	3.108	4.065E-2	7.464	32.36	9.752E-2
	~~9.987~~	~~9.346~~	~~3.319E-2~~				~~7.410~~	~~32.65~~	~~9.308E-2~~
	S.S 5/28/93						S.S 5/28/93		
$XO \cdot UO_2OH^0$	-3.535	2.793	5.149E-2	-4.226	1.407	4.482E-2	-7.67	12.75	3.540E-2
	~~-3.450~~	~~2.867~~	~~5.188E-2~~				~~-7.835~~	~~13.19~~	~~3.515E-2~~
$XOH \cdot UO_2OH^+$	3.120	1.158	4.233E-2	3.119	1.084	4.233E-2	1.052	16.56	5.577E-2
	~~3.221~~	~~1.176~~	~~4.247E-2~~				~~1.134~~	~~16.48~~	~~5.429E-2~~
	S.S 5/28/93						S.S 5/28/93		
$XO \cdot UO_2(OH)_2^-$	-10.15	4.148	6.138E-2	-11.50	1.575	4.883E-2	-11.12	0.543	6.490E-2
	~~-10.08~~	~~4.249~~	~~6.204E-2~~				~~-11.19~~	~~0.4492~~	~~6.760E-2~~
$XOH \cdot UO_2(OH)_2^0$	-3.535	2.793	5.149E-2	-4.226	1.407	4.482E-2	-4.146	1.318	4.423E-2
	~~-3.450~~	~~2.867~~	~~5.188E-2~~				~~-4.078~~	~~1.311~~	~~4.420E-2~~
$XOH_2 \cdot UO_2(OH)_2^+$	3.120	1.158	4.233E-2	3.119	1.084	4.233E-2	4.500	0.8289	6.361E-2
	~~3.221~~	~~1.176~~	~~4.247E-2~~				~~4.854~~	~~0.7451~~	~~6.290E-2~~
	S.S 5/28/93						S.S 5/28/93		
$XOH \cdot UO_2(OH)_3^-$	-10.15	4.148	6.138E-2	-11.50	1.575	4.883E-2	-7.496	0.8666	7.137E-2
	~~-10.08~~	~~4.249~~	~~6.204E-2~~				~~-7.292~~	~~0.7148~~	~~7.105E-2~~
$XOH_2 \cdot UO_2(OH)_3^0$	-3.535	2.793	5.149E-2	-4.226	1.407	4.482E-2	-0.4950	3.207	5.297E-2
	~~-3.450~~	~~2.867~~	~~5.188E-2~~				~~-0.1061~~	~~3.242~~	~~5.305E-2~~
	S.S 5/28/93						S.S 5/28/93		
$XOH \cdot UO_2(OH)_4^{-2}$	-16.72	4.935	7.234E-2	-18.70	1.470	5.465E-2	-9.843	0.5707	1.092E-1
	~~-16.67~~	~~5.030~~	~~7.350E-2~~				~~-9.350~~	~~0.4070~~	~~1.200E-1~~
$XOH_2 \cdot UO_2(OH)_4^{-1}$	-10.15	4.148	6.138E-2	-11.50	1.575	4.883E-2	-3.825	1.990	8.037E-2
	~~-10.08~~	~~4.242~~	~~6.204E-2~~				~~-3.340~~	~~1.744~~	~~8.286E-2~~

Surface Area

Goethite	$50 m^2/g$
zeol	$50 m^2/g$
am Fe	$600 m^2/g$
Hematite (syn & nat)	$2 m^2/g$

Log K_+ and K_- values for Ferrihydr (Hsi) were determined by the method described on pages 37 and 39.

SS 5/25/93

Ferrihydr ~~(I = 0.1)~~ -8.93 ~~-8.82~~

SS 6-3-93 ~~7.18~~ 7.29

DLM: Log K_+^0 = ~~7.348~~ Log K_-^0 = ~~-8.776~~

DLM & TLM should be corrected for ionic strength before use.

CCM: Log $K_+^{0.1}$ = 7.073 Log $K_-^{0.1}$ = -9.078

TLM: Log K^0_{cat} = -7.652 Log K^0_{an} = 8.306

SS 5/24/93

These Log K values after being corrected for ionic strength = 0.1 were used* with data from the Hsi disertation to determine Log K's for the sorption of Uranium on Ferrihydr using Fiteql. Input & Output files are in the directory

K:/Turner/Fiteql/Input/TLM or CCM or DLM/Uranium/Hsi

*Note: For The TLM Log K_{an} & Log K_{cat} were taken from Dzombak & Morel Not the table above

Log K_{an} = 7.18

Log K_{cat} = -8.82

Input Files:

XOH2U023.T FH

2H's on surface — 3OH groups — TLM — ferrihydrite

Output Files:

U02DDZ1.FH For No OH groups attatched

ferrihydrite

UOH2DDZ3.FH ← ferrihydrite

OH groups — # H attached to surf +1

DRJ 8/16/94

Sorption of UO_2^{2+} on Ferrihydrite (I=0.1)

Species	DLM Log K$^{0.1}$	DLM SOF/DF	DLM σLogK	CCM Log K$^{0.1}$	CCM SOF/DF	CCM σLogK	TLM Log K$^{0.1}$	TLM SOF/DF	TLM σLogK
	S.S. 5/27/93						S.S 5/28/93		
	3.032	8.241	4.151E-2				-5.501	59.92	3.833E-2
$XO \cdot UO_2^+$	~~3.027~~	~~8.195~~	~~4.124E-2~~	3.179	8.461	4.287E-2	~~-5.842~~	~~59.90~~	~~3.926E-2~~
	10.88	7.980	4.013E-2				2.968	59.01	5.826E-2
$XOH \cdot UO_2^{2+}$	~~10.90~~	~~7.901~~	~~3.945E-2~~	11.25	8.368	4.290E-2	~~2.998~~	~~58.91~~	~~6.010E-2~~
	S.S. 5/27/93						S.S 5/28/93		
	-4.813	8.519	4.301E-2				-9.055	20.35	2.681E-2
$XO \cdot UO_2OH^0$	~~-4.839~~	~~8.523~~	~~4.303E-2~~	-4.881	8.500	4.290E-2	~~-9.212~~	~~20.36~~	~~2.679E-2~~
	3.032	8.241	4.151E-2				-0.8446	24.96	5.258E-2
$XOH \cdot UO_2OH^+$	~~3.027~~	~~8.195~~	~~4.124E-2~~	3.179	8.461	4.287E-2	~~-0.9511~~	~~25.31~~	~~3.312E-2~~
	S.S. 5/27/93						S.S 5/28/93		
	-12.66	8.793	4.460E-2				-12.52	7.459	4.709E-2
$XO \cdot UO_2(OH)_2^-$	~~-12.70~~	~~8.852~~	~~4.493E-2~~	-12.93	8.483	4.299E-2	~~-12.44~~	~~7.441~~	~~4.780E-2~~
	-4.813	8.519	4.301E-2				-4.834		
$XOH \cdot UO_2(OH)_2^0$	~~-4.839~~	~~8.523~~	~~4.303E-2~~	-4.881	8.500	4.290E-2	~~-4.760~~	8.500	4.290E-2
	3.032	8.241	4.151E-2				3.367	7.456	4.713E-2
$XOH_2 \cdot UO_2(OH)_2^+$	~~3.027~~	~~8.195~~	~~4.124E-2~~	3.179	8.461	4.287E-2	~~3.475~~	~~7.447~~	~~4.774E-2~~
	S.S 5/27/93						S.S 5/28/93		
	-12.66	8.793	4.460E-2				-8.211	9.166	5.959E-2
$XOH \cdot UO_2(OH)_3^-$	~~-12.70~~	~~8.852~~	~~4.493E-2~~	-12.93	8.483	4.299E-2	~~-7.905~~	~~8.990~~	~~6.000E-2~~
	-4.813	8.519	4.301E-2				-0.5196	11.80	
$XOH_2 \cdot UO_2(OH)_3^0$	~~-4.839~~	~~8.523~~	~~4.303E-2~~	-4.881	8.500	4.290E-2	~~-0.2315~~	~~11.81~~	5.877E-2
	S.S 5/27/93								
$XOH \cdot UO_2(OH)_4^{-2}$	-20.50	9.045	4.625E-2				-11.26	8.582	8.084E-2
$XOH_2 \cdot UO_2(OH)_4^-$	~~-12.70~~	~~8.852~~	~~4.493E-2~~	-12.93	8.483	4.299E-2	~~-3.392~~	~~11.53~~	~~7.612E-2~~
	S.S. 5/27/93 -12.66	8.793	4.460E-2				-3.192	11.76	7.585E-2
							S.S. 5/28/93		

These Values are for 0.1 ionic strength

S.S. 6/3/93

DLM

Species	Log K$^{0.1}$	SOF/DF	σLogK
$XO \cdot UO_2^+$	3.027	8.195	4.124 E-2
$XOH \cdot UO_2^{2+}$	10.90	7.901	3.945 E-2
$XO \cdot UO_2OH^0$	-4.839	8.523	4.303 E-2
$XOH \cdot UO_2OH^+$	3.027	8.195	4.124 E-2
$XO \cdot UO_2(OH)_2^-$	-12.70	8.852	4.493 E-2
$XOH \cdot UO_2(OH)_2^0$	-4.839	8.523	4.303 E-2
$XOH_2 \cdot UO_2(OH)_2^+$	3.027	8.195	4.124 E-2
$XOH \cdot UO_2(OH)_3^-$	-12.70	8.852	4.493 E-2
$XOH_2 \cdot UO_2(OH)_3^0$	-4.839	8.523	4.303 E-2
$XOH \cdot UO_2(OH)_4^{-2}$	-20.56	9.157	4.690 E-2
$XOH_2 \cdot UO_2(OH)_4^-$	-12.70	8.852	4.493 E-2

Corrected Log K values for zero ionic strenth. Absorption of UO_2^{2+} on Goethite & Ferrihydrite

Species	Goethite Log K° DLM	Goethite CCM	Goethite TLM	Ferrihydrite DLM	Ferrihydrite CCM	Ferrihydrite TLM
$XO \cdot UO_2^+$	3.44	3.34	-2.61	3.25	3.40	-5.62
$XOH \cdot UO_2^{2+}$	9.99	10.58	7.41	10.90	11.25	2.92
$XO \cdot UO_2OH^0$	-3.23	-4.01	-7.61	-4.62	-4.66 ~~-8.99~~	-8.99
$XOH \cdot UO_2OH^+$	3.44	3.34	1.35	3.25	3.40 ~~-0.73~~	-0.73
$XO \cdot UO_2(OH)_2^-$	-10.08	-11.50	-11.19	-12.70	-12.93 ~~-12.44~~	-12.44
$XOH \cdot UO_2(OH)_2^0$	-3.23	-4.01	-3.81	-4.62	-4.66 ~~-4.55~~	-4.55
$XOH_2 \cdot UO_2(OH)_2^+$	3.44	3.34	5.07	3.25	3.40 ~~2.87~~ ~~3.705~~	3.70
$XOH \cdot UO_2(OH)_3^-$	-10.08	-11.50	-7.29	-12.70	-12.93 ~~-7.90~~	-7.90
$XOH_2 \cdot UO_2(OH)_3^0$	-3.23	-4.01	0.106	-4.62	-4.66	-0.01
$XOH \cdot UO_2(OH)_4^{-2}$	-17.11	-19.14	-9.79	~~-12.70~~ SS 5/28/93		
$XOH_2 \cdot UO_2(OH)_4^-$	-10.08	-11.50	-3.35	-12.70	-12.93	-3.39

$$* \; Log K^{o} = Log K^{corr} + Log \gamma_{prod} - Log \gamma_{react}$$

S.S
5/27/93

I Found A mistake in the way that I have been correcting for ionic strength. Need to go back and check Goethite and Ferrihydrite Models. TLM & CCM should be correct....Goethite and Ferrihydrite models have been corrected. Need to create new output files for DLM of Goethite and Ferrihydrite. Also changed TLM for Goethite and Ferrihydrite (mistake with Log Kan & Log Kcat) Need to make new Output Files for TLM-Go & FH Models have been created for Synthetic and Natural Hematite using these parameters.

$N_s = 2.31 \frac{sites}{nm^2}$ $SA = 2 m^2/g$ $C = 1 g/L \times 1 L$

S.S 5/31/93 ~~Acidity constants were determined from titration data and are on the next page~~

$$[XOH] = \frac{(2.31)(1)(2)(10^{18})}{6.02 \times 10^{23}} = 7.67 \times 10^{-4}$$

$\Sigma U = 1 \times 10^{-5}$ $I = 0.1$

Log K°

UO_2^{2+} on Goethite and Ferrihydrite

I = 0

Species	Goethite DLM	Goethite CCM	Goethite TLM	Ferrihydrite DLM	Ferrihydrite CCM	Ferrihydrite TLM
$XO \cdot UO_2^+$	3.45 S.S 5/28/93; ~~3.55~~	3.45	-2.31	3.36 S.S 6-3 93; ~~3.36~~	3.51	-5.17
$XOH \cdot UO_2^{2+}$	10.31	10.14	7.90	11.34; ~~11.32~~	11.69	3.41
$XO \cdot UO_2OH^0$	-3.31	-3.09	-7.45	-4.59 S.S 6-3 93; ~~-4.59~~	-4.66	-8.83
$XOH \cdot UO_2OH^+$	3.45	3.45	1.38	3.36; ~~3.36~~	3.51	-0.5146
$XO \cdot UO_2(OH)_2^-$	-10.04	-11.39	-11.01	-12.59 S.S 6-3 93; ~~-12.55~~	-12.82	-12.41
$XOH \cdot UO_2(OH)_2$	-3.31	-4.01	-3.93	-4.59; ~~-4.59~~	-4.66 SS ~~-4.10~~ 5/28/93	-4.61
$XOH_2 \cdot UO_2(OH)_2^+$	3.45	3.45	4.83	3.36; ~~3.36~~	3.51	3.70
$XOH \cdot UO_2(OH)_3^-$	-10.04	-11.39	-7.39	-12.59 S.S 6-3 93; ~~-12.55~~	-12.82	-8.10
$XOH_2 UO_2(OH)_3^0$	-3.31	-4.01	-0.28	-4.59; ~~-4.59~~	-4.66	-0.300
$XOH \cdot UO_2(OH)_4^{-2}$	-16.72	-18.70	-9.84	-20.56 S.S 6-3 93; ~~-20.50~~		-11.26
$XOH_2 \cdot UO_2(OH)_4^-$	-10.04	-11.39	-3.71	-12.59; ~~-12.55~~	-12.82	-3.08

$$\text{Log } K^\circ = \text{Log } K^{corr} + \text{Log } \gamma_{prod} - \text{Log } \gamma_{react}$$

Log γ = correction factor (Dzombak & Morel Table 2.13)

Natural & Synthetic Hematite input and output files are in the directory:

K:/TURNER/FITEQL/INPUT/DLM/CCM/TLM/

URANIUM/HSI/*.*

Input Files:

XOH2U023.THS → HS or HN for natural or synthetic

↑ #OH groups ↑ model type

Output Files:

U02TDZ1.HMN For NoOH's attached

↑ # H's+1 on surface ↑ HMS or HMN for natural or synthetic

U0H3TDZ3.HMS For OH's attached

↑ #OH groups ↑ #H+1 on surf ↑ HMS or HMN

Note: Pages 49-50 had been removed from this notebook before it was closed out. RBrient 12/31/03

5/31/93 SS

Input files have been created for adsorption of ~~Thorium~~ **Neptunium** SS 5/31/93 on Goethite using the digitized Girvin data. Parameters used were:

$SA = 50 m^2/g$

$N_s = 2.31$ sites/nm^2

$C_s = 6.25 \times 10^{-3}$ g/L

$[XOH] = 1.20 \times 10^{-6}$ m

$Th(tot) = 10^{-13}$ M

0.1 M $NaNO_3$

DLM

K:/TURNER/FITEQL/INPUT/DLM/Neptunium S.S 5/31/93

XONPO2.D11 → Np = 4.5×10^{-11} M

" .D12 → Np = 4.7×10^{-12} M

" .D13 → Np = 4.5×10^{-13} M

XOHNPO21.D11

" .D12 } XOH-NpO_2(OH)

" .D13

2XONPO2.D11

" .D12 } XO, XO > NpO_2

" .D13

XONPO21.D11

" .D12 } XO-NpO_2(OH)

" .D13

XOHNPO2.D11

" .D12 } XOH-NpO_2

" .D13

S.S 6-4-93 ~~XOH2NPO21~~.

XOH2NO21.D11

" .D12 } $XOH_2NpO_2(OH)^+$

" .D13

S.S 5/13/93 CCM

K:/~~FITEQ~~ TURNER/FITEQL/INPUT/CCM/

Neptunum

XO·NPO$_2$.C11 → 4.5×10^{-11} m = Np(+) ~~XOHNpO2(OH)~~ DRJ 9/16/94

" .C12 → Np = 4.7×10^{-12} XOHNPO2.C11

" .C13 → Np = 4.5×10^{-13} " .C12 } XOH·NpO$_2$(OH)

" .C13

2XO·NPO$_2$.C11 } XO$^-$, XO$^-$ } NpO$_2^+$ XONPO2.C11

" .C12 " .C12 } XO·NpO$_2$(OH)

" .C13 " .C13

XOHNPO2.C11 } XOH·NPO$_2$ " .C13

" .C12

" .C13

5/31/93 TLM DRJ for SS

K:/TURNER/FITEQL/INPUT/~~DLM~~/Neptunum

TLM

XONPO2.T11 → 4.5×10^{-11} = Np(+) XOHNPO2.T11

" .T12 → Np = 4.7×10^{-12} " .T12 } XOH·NpO$_2$(OH)

" .T13 → Np = 4.5×10^{-13} " .T13

2XONPO2.T11 } XO, XO } NpO$_2$ XONPO2.T11

" .T12 " .T12 } XO·NpO$_2$(OH)

" .T13 " .T13

XOHNPO2.T11 } XOH·NPO2. XOH$_2$NO2.T11

" .T12 " .T12 } XOH$_2$·NpO$_2$(OH)

" .T13 " .T13

S 6/4/93

OH$_2$NO2.C11

" .C12

" .C13

OH$_2$·NpO$_2$(OH)

Input Files Have Been created for absorption of Thorium on Goethite using the digitized data of LaFlamme & Murray (87) Parameters used were:

SA = 50 m^2/g

Ns = 2.31 sites/nm^2

S.S 5/31/93

Cs = ~~1.2×10^{-6} M~~/L 6.25×10^{-3} g/L

[XOH] = 1.2×10^{-6} M/L

Th(tot) = 10^{-13} M

0.1M Na NO$_3$

Directory:

K:/TURNER/FITEQL/INPUT/TLM CCM or DLM/Thorium

XOTH.T60 — T, C, or D for model

XOTH1.T60

XOTH2.T60 → XOTh(OH)$_2^+$

XOTH3.T60 → XOTh(OH)$_3^0$

XOTH4.T60 → XOTh(OH)$_4^-$ → More

Deleted:

~~XOTH22.T60 → XOTh$_2$(OH)$_2^{+5}$~~

~~XOTH48.T60 → XOTh$_4$(OH)$_8^{+7}$~~

~~XOTH615.T60 → XOTh$_6$(OH)$_{15}^{+8}$~~

S.S 6-4-93

6-4-93 Stephen Sassman

DRJ 8/16/94

$XOHTh(OH)_2^{+2} \rightarrow$ XOHTh2.TGO

$XOHTh(OH)_3^{+} \rightarrow$ XOHTh3.TGO

$XOHTh(OH)_4^{0} \rightarrow$ XOHTh4.TGO

$XOH_2Th(OH)_4^{+} \rightarrow$ XOH2Th4.TGO

Fiteql was run on the neptunium and thorium input files. Most of the species did not converge. All of the obvious mistakes have been corrected. Changes will be neccessary.

Titration Data

6/9/93

DRJ p. 55

Data has been digitized from various sources. Sources and filenames are listed on the next page.

Sample hand calculations are shown on pp. 151-152. DRJ 8/11/94

The data was then incorporated into a spreadsheet. DRJ 8/16/94 Charge was converted to TOTH when necessary, and the data was printed. Filenames are listed on the next page with the appropriate set of data files. Printouts may be found in the appropriate file.

Directory:

K:/TURNER/FITEQL/INPUT/TITRATE/

Balmurra	Stumm
PulsPow	Mesuere
DZM	James
Murray	
Sprycha	

Directory	Filename	Source
Balmurra /	FeOOH01.PRN	Balisterieri & Murray
	FeOOH7.PRN	(Am. Jour. Sci 81)
	FeOOH1.PRN	Pg 795 Fig 2
PulsPow /	01Fe2O3.PRN	Puls & Powell
	05Fe2O3.PRN	(Eviron Sci Tech 92) 26 Pg 617 Fig 2
DZM /	Fig1T1.PRN	Dzombak & Morel
	Fig1T2.PRN	
	Fig1T3.PRN	Surf. Comp.
	Fig1T4.PRN	Modelling
	Fig1T5.PRN	
	Fig1T6.PRN	
	Fig1T7.PRN	
Murray /	001 MnO2.PRN	Murray (73)
	010 MnO21.PRN	"Surf Chem of MnO_2"
	010 MnO22.PRN	Jour. Coll. & Int. Sci.
	010 SiO2.PRN ←	Vol 46 (74)
Sprycha /	TiO2001.PRN	Sprycha
	TiO2010.PRN	Jour. Coll. Int Sci (84)
	TiO2100.PRN	Vol. 102

Stumm /	0025Lian.PRN	Aquatic Sciences 52/1 1990
	1000Lian.PRN	Liang & Morgan "Chemia" S.S. 6-9-93
	0120Lian.PRN	by way of Stumm
Mesuere /	Goth005.PRN	Dissertation
	Goth010.PRN	Mesuere, Karen L. G.
	Goth100.PRN	
	Goth0050.PRN	
	Goth0100.PRN	
	Goth1000.PRN	
Sprycha /	AL203001.PRN	Jour. Coll. Int. Sci
	AL20301.PRN	1989 Vol 127 No. 1
	AL2031.PRN	
	AL2031M.PRN	
	AL203CL1.PRN	
	AL203BR1.PRN	
	AL203I1.PRN	
	AL203Na1.PRN	
	AL203K1.PRN	
	AL203Cs1.PRN	
James /	AL203001.PRN	Robert. O. James &
	AL203010.PRN	George O. Parks
	AL203100.PRN	Surface & Colloid
		Science Vol. 12
		Ch. 2

James Cont.

GOTH001.PRN
GOTH010.PRN
GOTH100.PRN

SIO2010.PRN
SIO2100.PRN
SIO21.PRN

TIO2001.PRN
TIO2010.PRN
TIO2100.PRN
TIO2001L.PRN
TIO2010L.PRN
TIO2100L.PRN
TIO21L.PRN

TIO2001C.PRN
TIO2010C.PRN
TIO2100C.PRN
TIO2001N.PRN
TIO2010N.PRN
TIO2100N.PRN
TIO21N.PRN

TIO2001S.PRN
TIO2010S.PRN
TIO2100S.PRN

6/11/93

DB in S.S.

The titration data was loaded into a spreadsheet for conversion of μcoul/cm² to TOTH. Filenames are listed below.

Turner/Fiteql/Input/Titrate/

Abendrot/SIO21.WKQ
SIO22.WKQ

SS. 6/11/93
~~DZM~~/James/
AL203.WKQ
Goethite.WKQ
SIO2.WKQ
TIO2L.WKQ
TIO2S.WKQ
TIO2C.WKQ
TIO2N.WKQ
TIO2.WKQ

Murray/MnO2.WKQ
SiO2.WKQ

Sprycha/TIO2.WKQ
AL2031.WKQ
AL2032.WKQ

Balmurra/Goth.WKQ

Mesuere/ Goethite1.WKQ

Pulspow/ Fe2O3.WKQ

Stumm/ Fe2O3.WKQ

Input Files for Fiteql have been created. The following site density and surface areas were used and will be used in all models. S.S 6/11/93

N_s = 2.31 sites /nm^2 for all minerals

Mineral	Surface Area (m^2/g)
Gothite	50
γAl_2O_3	120
αAl_2O_3	~~15~~ 12 S.S 6/21/93
δMnO_2	270
amSiO_2	175
Anatase	125
Rutile	30
Hematite	40
Nat. Hematite	2
~~α-AlZ~~ S.S 6/11/93	
Magnetite	**5**

Directories and Filenames for Input Files are as follows:

K:/TURNER/FITEQL/INPUT/TITRATE/

Abendrot/
- SIO2O1.DDZ
- SIO21.DDZ
- SIO21M.DDZ
- SIO21CS.DDZ
- SIO21K.DDZ
- SIO21LI.DDZ

} Also CDZ. & TDZ

DZM/
- FIG1T1.DDZ
- FIG1T2.DDZ
- FIG1T3.DDZ
- FIG1T4.DDZ
- FIG1T5.DDZ
- FIG1T6.DDZ
- FIG1T7.DDZ

} Also CDZ & TDZ

JAMES/
- AL2O3001.DDZ
- AL2O301.DDZ
- AL2O31.DDZ

} Also CDZ, TDZ

S.S. 6/14/93
~~FEOOH.DDZ~~
- FEOOH001.DDZ
- FEOOH01.DDZ
- FEOOH1.DDZ

} Also CDZ, TDZ

JAMES / SIOZ01.DDZ
SIOZIM.DDZ
} Also CDZ, TDZ

TIOZ001.DDZ
TIOZ001A.DDZ
TIOZ001B.DDZ
TIOZ001C.DDZ
TIOZ001D.DDZ
TIOZ01.DDZ
TIOZ01A.DDZ
TIOZ01B.DDZ
TIOZ01C.DDZ
TIOZ01D.DDZ
TIOZ1.DDZ
TIOZ1A.DPZ
TIOZ1B.DDZ
TIOZ1C.DDZ
TIOZ1D.DDZ
TIOZIMA.DDZ
} Also CDZ & TDZ

ARRAY / MNOZ001.DDZ
MNOZ01.DDZ
} Also CDZ & TDZ

RYCA / TIOZ001.DDZ
TIOZ01.DDZ
TIOZ1.DDZ
} Also CDZ & TDZ

AL203001.DDZ
AL20301.DDZ
AL2031.DDZ
AL203BR.DDZ
AL203CL.DDZ
AL203CS.DDZ
AL203I.DDZ
AL203K.DDZ
AL203Na.DDZ
} Also CDZ & TDZ

BALMURRA / FEOOH01.DDZ
FEOOH1.DDZ
FEOOH7.DDZ
} Also CDZ and TDZ

MESUERE / FEOOH005.DDZ
FEOOH01.DDZ
FEOOH1.DDZ
} Also CDZ & TDZ

PULSPOW / FE20301.DDZ
FE20305.DDZ
} Also CDZ & TDZ

Stumm / Fe203002.DDZ
FE203012.DDZ
FE2031.DDZ
} Also CDZ & TDZ

Batch Files were created for the input files listed above. They were then run with the Fiteql executable. Some of them converged, these were entered into a spreadsheet. Others will need more work.

6-16-93 Stephen Sassman

More of the input files converged after several mistakes (typos, units) were corrected.

I will now try new initial guesses for MnO_2, TiO_2 & SiO_2 because of the low pzc. The new guesses are basically two to either side of the PZC. i.e.:

$$PZC = 6 \Rightarrow K^+ \approx 4 \quad K^- \approx -8$$

For those that were close to converging the error estimate for TOTH will be

relaxed to try to induce convergence.

6/17/93 Stephen Sassman

Data has been digitized from Regazoni et al, Jour of Coll. Int Sci Vol 91 No. 2 Feb 83. The Data files are

K:\TURNER\FITEQL\INPUT\TITRATE\REGAZONI\

Fe304001.PRN
FE304010.PRN
FE304100.PRN

The data was then pulled into a spreadsheet as before. The spreadsheet is

Fe304.WKQ

In put Files have been created for the Regazoni Data.

FE304001.TDZ
FE30401.TDZ
FE3041.TDZ
} Also CDZ & DDZ

6-21-93 Stephen Sassman

Data has been digitized, pulled into a spread sheet, and input files Created for:

S.S 6/21/93

Balistrieri & Murray

Geochimica et Cosmochimica Acta Vol46 pp1041-1052

The files that have been created are on the next page.

Data Files:

MN0201.PRN
~~MN020~~
MN021.PRN
MN021M.PRN

Spreadsheet
MN02.WKQ

INPUT FILES:

MN0201.TDZ
MN021.TDZ
MN021M.TDZ
} Also CDZ & DDZ

Tables have been created that list all sources of data (Table 1) and all parameters for the models. (Table 2). The tables are in the directory:

K:\TURNER\FITEQL\INPUT\TIRATE\

TABLE.WP

& SOURCES

6-23-93 Stephen Sassman

PulsPow Hematite will not be used because it will not converge.

The original Murray Data for Manganeese has been replaced by more data from the same source. It converged.

The Balmurra Manganeese data was not used because it would not converge.

For models that could not be made to converge ~~any other way beside~~ by making new initial guesses or relaxing error estimates, The method of Dzombak and Morel was used. One of the Log K Values was fixed while the other was adjusted with Fiteql. S.S 6-23-93

Manganees & Silica DLM & CCM were modelled using only K- because almost all of the data was above the zero point of charge S.S 6/23/93 ~~Modelel~~

Manganeese TLM were done similarly using only Kcat & K-
Silica TLM's converged with both Kcat & Kan & K+ & K-.

6-28-93 Stephen Sassman

After all of the input files converged, they were entered into two tables. One table, TITRATE.WP was used in the report that Dave wrote. The other table, TITRATE2.WKQ was used to compute the average ~~tok~~ Log K values for each mineral. The method described on pg. 39 was used to compute averages for the DLM and TLM. For CCM that had a Log K at 0.MM Ionic strength, that number was used.

For CCM's that had no 0.1M Ionic Strength value, the extrapolation technique described on pg. 37 was used.

Confidence Limits for the average Log K values were computed by the method of Dzombak & Morrel (Surf. Comp. Modeling Pg 84)
The computations for ave Log K values and confidence limits can be found in the spreadsheet TITRATE 2.WKQ

All Input Files, Data Files, and spreadsheets can be found on disk. The disks are labelled Fiteql Input Files - Titration Data.

7-8-93 Stephen Sassman

One more Titration data set has been added for MnO2. The Source is:

Catts & Langmuir. Applied Geochemistry Vol. 1 Fig. 2 (1986)

The results were entered into the spreadsheet that calculates ave. Log K's and confidence Limits. TABLE.WP & TITRATE.WP were also updated.

7-16-93 Stephen Sassman

The next task was to begin modelling sorption data. The first step was to digitize data from various sources. Sources are listed below. File-names are also listed. The Scientific Measurement System Sigma Scan was used with the digitizing tablet. The second step was to create the input files for Fiteql. These files, when executed, will produce Log K values for sorption of radionuclides onto various minerals. These log K values, along with other previously determined ~~para~~ parameters, will be used later to produce a graph of pH vs. % sorbed. This will be accomplished using the computer code Minteq.

S.S. ~~[illegible]~~ heumonics 2200

~~As of today, two data sets have been completed:~~

S.S 8/4/93

S.S 8/4/93

Digitized Data:

Directory: D:\FITEQL\SORPTION\...

Directory	Filename	Source
SILVA	USIOZ.PRN	Silva, Robert "Mechanisms for The Retardation of U(VI) Migration" Mat. Res. Soc. Symn. Proc Vol 257 (1992)

Fiteql Input Files:

8-4-93 Stephen Sassman

The following is a list of Files including digitized data, and Fiteql Input Files. For the sake of simplicity, a description of the method used to name the input files is given rather than a complete listing.

S.S. 8/4/93

~~Digitized Data~~: Input Files:

D:\FITEQL\SORPTION\Subdirectory\Model Type

Subdirectories:

Payne	Mea
Nakayama	Righetto
Kohler	Ohe
Moulin	Silva

Model Types:

TLM
CCM
DLM

Figure #

XOUOZ.T11

surface complex — model type

Surface Complexes Considered:

Uranium

$XOUOZ^{+}$	→	XOUOZ
$XOHUOZ^{+2}$		XOHUOZ
$XOUOZOH^{0}$		XOUOZ1
$XOHUOZOH^{+}$		XOHUOZ1
$XOUOZ(OH)_2^{-}$		XOUOZ2
$XOHUOZ(OH)_2^{0}$		XOHUOZ2
$XOH2UOZ(OH)_2^{+}$		XOH2UOZ2
$XOHUOZ(OH)_3^{-}$		XOHUOZ3
$XOH2UOZ(OH)_3^{0}$		XOH2UOZ3
$XOHUOZ(OH)_4^{-2}$		XOHUOZ4
$XOH2UOZ(OH)_4^{-}$		XOH2UOZ4
$XOHUOZCO_3^{0}$		XOHUOZC1
$XOH_2UO_2(CO_3)_2^{-}$		XOH2UC2
$XOH_2UO_2(CO_3)_3^{-3}$		XOH2UC3
~~$XOH_2(UO_2)(CO_3)_4$~~	→	XOH2UCO
$XOH_2(UO_2)_2(CO_3)(OH)_3^{0}$		

Neptunium:

$XONpO_2^0$	→ XONPO2
$XOHNpO_2^+$	XOHNPO2
$2XONpO_2^-$	2XONPO2
$XONpO_2OH^-$	XONPO21
$XOHNpO_2OH^0$	XOHNPO21
$XOH_2NpO_2OH^+$	XOH2NPO21
$XOH_2NpO_2(CO_3)^0$	XOH2NC1
$XOH_2NpO_2(CO_3)_2^{-2}$	XOH2NC2

Thorium:

$XOTh^{+3}$	XOTH
$XOThOH^{+2}$	XOTH1
$XOTh(OH)_2^+$	XOTH2
$XOHTh(OH)_2^{+2}$	XOHTH2
$XOTh(OH)_3^0$	XOTH3
$XOHTh(OH)_3^+$	XOHTH3
$XOTh(OH)_4^-$	XOTH4
$XOHTh(OH)_4^0$	XOHTH4
$XOH_2Th(OH)_4^+$	XOH2TH4

Americium:

$XOAm^{+2}$	XOAM
$XOAmOH^+$	XOAM1
$XOHAm(OH)_2^+$	XOHAM2
$XOAm(OH)_2^0$	XOAM2
$XOAm(OH)_3^-$	XOAM3
$XOHAm(OH)_3^0$	XOHAM3
$XOH_2Am(OH)_3^+$	XOH2AM3
$XOAmCO_3^0$	XOAMC1
$XOH_2Am(CO_3)_2^0$	XOH2AMC2
$XOH_2Am(CO_3)_3^{-2}$	XOH2AMC3

Data Files:

D:\FITEQL\SORPTION\SUBDIRECTORY

Subdirectories:

Kohler	Meq
Moulin	Nakayama
OHE	Payne
RIGHETTO	Silva

Kohler:

NpHem005.PRN	NPHEM.WKQ
NPHEM05. "	NPHEM01.PRN
NPKA01. "	NPHEM1. "
UGOCO2Ø. "	NPKA01.WP
UGOCO24. "	UGOCO22.PRN

Moulin:

~~AMØ~~	AM.WP
AMALØ1.PRN	AMAL1.PRN
AMSIOZØ1.PRN	AMSIOZ1.PRN

OHE:

	NABENTON.PRN
NPKUZ62.PRN	NPKUZ620. "
NPKU4OZ "	NPKU4020. "
NPKU7OZ "	NPKU7020. "

RIGHETTO:

	NPALZ03.PRN
AMALZ01.PRN	NPSIOZ "
AMALCOZ. "	NPALCOZ "
AMALZØØ1. "	THALCOZ "
AMALZ03. "	THALZ03. "
AMSIOZ. "	THSIOZ. "

MEA:

	UMUSC100.PRN
UMUSC250.PRN	USIOZ001.PRN
USIOZ100.PRN	USIOZ250.PRN

NAKAYAMA

	NPALZ03.PRN
NPBOEHMI.PRN	NPNBIOTI. "
NPNGOETH. "	NPNHEMAT. "
NPNMAGNE. "	NPSHEMAT. "
NPSLEPID. "	NPSMAGNE. "

PAYNE*:

UKAO.WP	UKAO4.PRN
UKAO1.PRN	UKAO16.PRN

SILVA:

SIOZ.WP	USIOZØ1.PRN
USIOZ.PRN	USIOZIM.PRN
USIOZ1.PRN	

Note - Some of the Payne digitized data files were accidentally erased, the data still exists in the Input Files only.

8-11-93 Stephen Sassman

For the past week, I have been working on some Minteq runs for Dave. The runs will be done to produce graphs of pH vs/ TOT H and pH vs/ %sorbed for titration and sorption graphs respectively. The graphs will be incorporated into the Semi-Annual report due at the end of this month. The graphs, input files, and worksheets

are currently located on my machine at:

D:\FITEQL\SORPTION\PLOT

&

D:\FITEQL\TITRATE\PLOT

these files will probably be transfered to disk at a later time

8-20-93 Stephen Sassman (SKS)

Sorption Data has been digitized for three more ~~data~~ sources.

The digitized data is located at:

D:\FITEQL\SORPTION\VOCHTEN,
D:\FITEQL\SORPTION\VENKAT,
and D:\FITEQL\SORPTION\LIESER.

8-24-93 Stephen Sassman

An analysis of the Tripathi (1984), ~~and~~ Hsi (1981) and Hsi & Langmuir (1985) data has been completed. Results will appear in the semi-annual report.

Analysis of the data included:

Fitting of Log K's for Goethite ΔFH CO_2 data.

Modelling of all data sets with Minteq

Graphing of Minteq results using Sigma Plot.

Computation of Average Log K's and confidence limits for Tripathi CO_2 data.

Creation of tables listing Log K's & standard deviations for Hsi data, and Log K's & confidence limits for Tripathi data. These tables will appear in the semi annual report

The input files that were used to fit Log K's for Hsi CO_2 data can be found at:

Hsi (1981) w/ CO_2 Input Files

D:\FITEQL\SORPTION\HSI\TLM \
CCM
& DLM

Model Type ↓

Surface Complex . T 28 ← Figure #

Note:

Fig28#1= 28
Fig28#2=29
Fig64#1=64
Fig64#2=65

Minteq Input Files and graphs of results can be found at:

Minteq Runs (Tripathi & Hsi data)

D:\FITEQL\SORPTION\PLOT

Graphs are in the above directory

Input Files are in the subdirectory Input.

Sample Calculation shown on pg. 150 This volume DRJ 8/11/94

The Spreadsheet containing the computations for average Log K's and confidence limits are Located at:

Tripathi & Hsi Ave LogK's & conf. lim.

K:\TURNER\QUATTRO\TRIPAVEK.WKQ

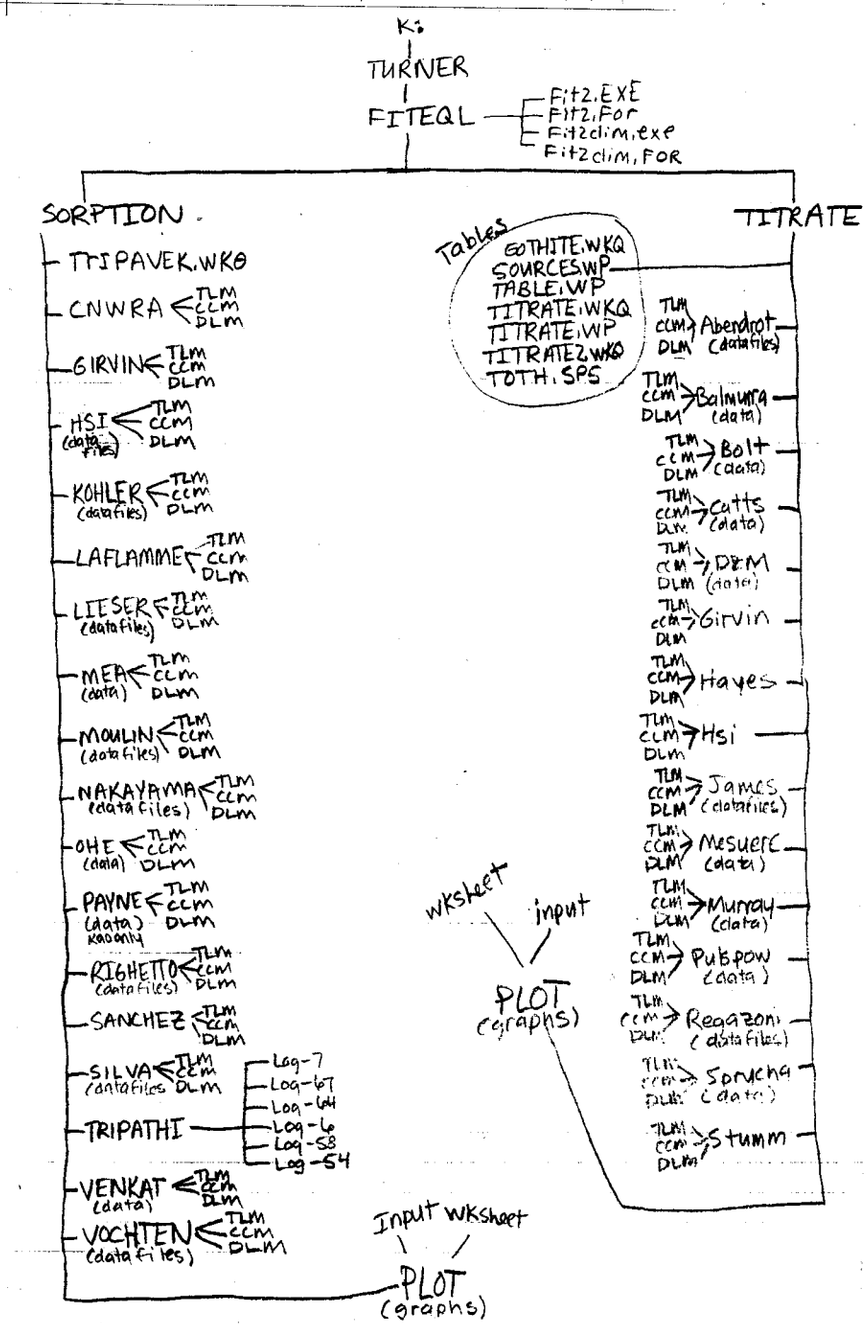
and at D:\FITEQL\SORPTION\TRIPAVEK.WKQ

8-27-93 Stephen Sassman

The directory structure for the K: drive has been changed. I found this necessary in order to copy data from my machine to the K drive. The directory structure is on the next page. None of the filenames have been changed.

All digitized data, Fiteql input files, tables, Minteq runs, and graphs were copied to diskettes. I will give these to Dave before I leave.

Directory Structure for Location of Fiteql and Minteq related files:



1-14-93 Stephen Sassman

Over Christmas vacation I spent time modelling some of the data sets. The following procedure was used on all of the data that I have modelled.

Procedure:

1. Create Fiteql Input Files by plugging parameters and serial data into template.
2. Run Input Files from OS2 window
3. Record LogK's using "List Files" on WP5.1
4. Create Minteq Input File Using Fiteql-determined LogK's & mineral specific parameters.
5. Run Minteq
6. Import Minteq data output into Sigma Plot & Plot against experimental data.

I have also gone through all of the folders and arrange all of the contents ~~so that~~ S.S 1/14 in a logical manner. Then I made a list of which data have already been modelled, And which data still need to be looked at. Dave has a copy of this list & a table w/ the same information.

The following is a list of all of the sorption data that I have modelled over the last ~~four~~ S.S 1/14 weeks. A capital letter for the source indicates that the data was successfully modelled. A lowercase letter indicates that problems were encountered. A brief description of the problem is given in each case.

~~Uranium~~

~~Source:~~

~~Goethite~~

~~U(VI) on Goethite~~ S.S 1/14 ~~A~~

~~Ferrihydrite~~

See Next Page for table

Uranium

Goethite	Ferrihydrite	Magnetite
A	B	C

Nat. Hematite	Synth. Hematite
b probably because of low surf area & site conc. [redacted]	B

Plutonium IV

Goethite
d probably because of uncertainty of Hydrolysis Log K's Also had probs w/ Th^{+4}

Plutonium V

Goethite	γAl_2O_3
D	E

Neptunium

Kaolinite	Synt. Hematite	γAl_2O_3
A	A	E

Americium

γAl_2O_3
E

Thorium

γAl_2O_3
e Bad Fit to the data. May be due to lack of thermodynamic data.

Sources

A Kohler et al (92)

B Hsi (81)

C Venkataramani & Gupta (91)

D Sanchez et al (85)

E Righetto et al (91) or (88)

Pamela Muller
PM

1/20/94

I went to building 189 to speak with Dr. David Turner about working on the CNWRA geochemical modelling project. This will involve using the FITEQL program for determining chemical equilibrium constants, and the PRODEFA2/MINTEQA2 programs for predicting amounts of species sorbed to a solid surface in an aqueous environment. I will begin reading the user's manuals for these as well as Dzomback and Morel's Surface Complexation Modelling.

1/25/94

I will start working with the LaFlamme and Murray data for sorption of thorium on goethite, and the Righetto et. al. data for thorium and americium on silica or alumina. First of all, however, I need to better understand the input files for FITEQL and what information goes where. Below is a printout from an input file using Tripathi's data. What the different numbers mean is written in in the margins. This file is used as an example because the results from this data set are modelled successfully using MINTEQA2, as I've been told by Stephen Sassman. Steve's work on modelling these systems is referred to previously in this notebook.

(See pages 37-88 for Steve's work in this notebook.)

PM 1/25/94

```
 0 print data in def. of chemical prob                          U-Goethi
 1  "     "    "  experimental data                              4.2e-6
# group I components
 0  "  speciation at each pH
 0  "  details of numerical optimization                         2.3si/nm
 1  "  summary of numerical optimization                         00000040
90  # of iterations allowed                                      00000050
 3  #group II  1  #group III  3                                   00000060
                                                                 00000070
                                                                 00000080
00001  -4.1   7.86E-5   XOH                                       00000090
00160  -1.0   0.00E00   PSI0                                      00000098
00032  -5.4   4.20E-6   UO2+2                                     00000100
00033   0.0   0.00E00   UO2ads                                    00000102
00050   0.0   0.00E00   H+                                        00000104
00003  -1.0   0.00E00   NA+                                       00000106
00005  -1.0   0.00E00   NO3-                                      00000110
                                                                 00000120
00050   0.00  050  1                                             00000124
00032   0.00  032  1                                             00000126
03201  -5.43  032  1 050 -1                                      00000127
03202 -10.54  032  1 050 -2                                      00000128
03203 -19.22  032  1 050 -3                                      00000129
03204 -32.59  032  1 050 -4                                      00000130
03205  -2.49  032  2 050 -1                                      00000131
03206  -5.42  032  2 050 -2                                      00000132
03207 -12.37  032  3 050 -4                                      00000133
03208 -16.25  032  3 050 -5                                      00000134
03209 -31.49  032  3 050 -7                                      00000135
03210 -22.83  032  4 050 -7                                      00000137
03211  -0.05  032  1 005  1                                      00000139
00100 -13.78  050 -1                                             00000140
01050   7.24  001  1 160  1 050  1                               00000145
01100  -9.06  001  1 160 -1 050 -1                               00000150
00001   0.00  001  1                                             xouo2
03301  -7.60  001  1 160  1 050 -1 032  1 033  1
```

ID# for components

log of free [ ]

total [mol/l]

ID# for species

log K of stability constant

protonation + deprotonation

surface rxn

1 adsorption on a charged surface is to be considered

```
                DLM   Asp   solid(s)/Fe2(L)
electrolyte     00002  50.0  0.410                    00000174
[ ]             0.1    1.0  ← charge of elec. ions    00000175
                       1                              00000180
                       3301                           00000185
                       15    1    x → 1    0 ← no dil. factor for ser. data   00000190
                       33                             00000195
                 2.271E-6                             00000230
                 2.380E-6                             00000240
                 2.618E-6                             00000260
                 2.676E-6                             00000270
                 2.690E-6                             00000280
                 2.777E-6                             00000290
                 2.887E-6                             00000295
                 3.160E-6                             00000300
                 3.359E-6                             00000310
                 3.517E-6                             00000320
                 3.719E-6                             00000330
                 3.790E-6                             00000340
                 3.793E-6                             00000350
                 3.840E-6                             00000360
                 4.014E-6                             00000370
                       50                             00000380
                 -6.45                                00000390
                 -6.46                                00000580
                 -6.49                                00000590
                 -6.54                                00000600
                 -6.54                                00000610
                                                      00000620
                                                      00000630
```

(there is T serial data for 1 comp)

digitized data UO2ads

ID# → 50

$-p^H$

(This file can be found on the CNWRA K drive,

under K:\TURNER\FITEQL\SORPTION\TRIPATHI\LOG-54\XOUO2.DDZ .)

```
-6.54                                                 00000640
-6.52                                                 00000650
-6.58                                                 00000660
-6.64                                                 00000670
-6.63                                                 00000680
-6.72                                                 00000690
-6.71                                                 00000700
-6.75                                                 00000710
-6.79                                                 00000720
-6.91                                                 00000730
                                                      00000760
                1        1                            00000770
               33     0.08    3.4E-07                 00000790
               50     0.05    0.0E-00
```

will be SD estimates ln [T] + ln [x]

ID#s

end of file.

1/26/94 PM Things to remember if using WP to edit FITEQL files:

Wordperfect Note: If FITEQL or other input files get the right margin shifted when the file is retrieved, select the File\Print\Select printer\Edit\courier 12cpi option. The FITEQL (and PRODEFA2) input files cannot be used unless each part of the numerical information for input is located with the proper spacing.

Also - FITEQL files must be saved as DOS text files, NOT using just the 'Save' command but Text In/Out (Control - F5), or FITEQL cannot read them.

1/31/94 PM

The K drive used by CNWRA is currently unavailable (SYS0015). In the future, I need to copy files I might be using onto my PC or a floppy disk if possible. That way, if the drive is not working, at least part of the information will still be available.

I have copies of both the May '82 (version 1.2) and the October '82 (version 2.0) user's guides for FITEQL. The older version has listings of error messages in section 6.2, which are not in the newer version.

2/3/94 PM Below is a copy of Dave Turner's explanation of the calculation for ionic strength for equilibrium constants, followed by examples:

Generally, the correction for ionic strength is

$$\text{Log } K_I = \text{Log } K_0 - \sum^{prod} \nu_i \log \gamma_i + \sum^{react} \nu_j \log \gamma_j$$

$\text{Log } K_I$ = Equil. constant at ionic strength = I PM 10/6/94

$\text{Log } K_0$ = Equil constant at $I = 0$

ν_i = stoichiometry of products

ν_j = stoichiometry of reactant

$\log \gamma_i$ = Activity coeff for products from table 2.13 (Drever, 1982)

$\log \gamma_j$ = " " " reactants

As an example:

$$UO_2^{2+} + H_2O = UO_2OH^+ + H^+ \qquad \log K_0 = -5.208$$

for $I = 0.1$ m

$$\log K_I = -5.208 - \left[\overbrace{(1)(-0.11)}^{UO_2OH^+} + \overbrace{(1)(-0.11)}^{H^+}\right] + \overbrace{(1)(-0.44)}^{UO_2^{2+}}$$

$$= -5.43$$

example 2:

PM for sorption reactions, $\gamma=1$ is assumed for surface species (i.e. $\log\gamma=0$)

$XOH^{o} + H^{+} = XOH_2^{+}$ $\quad \frac{\log K_o}{7.29}$ ← Ferrihydrite w/ DLM

for $I=0.1$ m

$$\log K_I = 7.29 + \overbrace{(1)(-0.11)}^{H^+} - \overbrace{(1)(0)}^{XOH_2^+}$$
$$= 7.18$$

02/07/94 PM

Only one of the LaFlamme Th on goethite CCM files will converge - the AXOTH.cgo file, for the XO-Th^{+3} species. The rest either don't converge, or give an error #1, which is a warning that the number of iterations allowed has been exceeded.

Example of how to calculate g/L from ppm concentration:

given 10ppm Al_2O_3, find g/L

$ppm = M(gam)1000$

$10 = M(101.9613)1000$

$M = .00010 = mol/L$

$$mol/L = \frac{g/gam}{L}$$

$$\frac{g/gam}{L} \cdot gam = \frac{g}{L}$$

$.00010(101.9613) = 0.010$

Example of how to calculate [XOH], given g/L

given g/L = 0.010 and A_{sp} for Al_2O_3 = 120 m^2/g

PM $$[XOH] = \frac{2.31(120)(.010 g/L)(10^{18})}{6.023\times10^{23}} = 4.6\times10^{-6}$$

2/14/94 PM Example: calculation of the k_i value for $ThOH^{+3}$, as seen in C:\model\thsif5\xoh2th4.c11

$$Th^{+4}+H_2O \rightleftharpoons ThOH^{+3}+H^+$$
$$\log k_1=-3.8874-[1(ThOH^{+3})+1(H^+)]+1(Th^{+4})]$$
$$\log k_1=-3.8874-[-.99+(-.11)]+(-1.76)$$
$$\log k_1=-4.5474$$

The FITEQL chemical equations on lines 5101-5107 were checked in this manner.

activity coefficient calculations:

PM $$\log f_i=(-A)\left[\left(\frac{\sqrt{I}}{1+\sqrt{I}}\right)-CI\right](z_i^2)$$

where:

f_i is the activity coefficient of species i
A is the constant .508
C is the constant .24
I is the ionic strength 0.1

Refer to the FITEQL version 2.0 user's handbook for this equation. The Davies equation was used to calculate the activity coefficients of species with a charge of 6 or greater; other coefficients were taken from table 2.13 in Dzombak and Morel's Surface Complexation Modelling.

'Species with a charge of 6 or greater' includes the Th ionic species $Th_2(OH_2)^{+8}$, $Th_4(OH)_8^{+8}$, + $Th_6(OH)_{15}^{+9}$.

Example of activity coefficient calculation, using the Davies equation:

PM where the charge of the ionic species equals 6,

$$\log\gamma_i=-(0.508)(6)^2\left[\frac{\sqrt{.1}}{1+\sqrt{.1}}-0.24(0.1)\right]$$
$$\log\gamma_i=-3.9548$$

where charge = 8, log γ_i = -7.0308, and
where charge = 9, log γ_i = -8.8984 .

2/17/94 PM The Righetto et. al. experiments from the '91 journal article are currently being

worked on. Copies of the batch file (for the Am(III) data from graph 5 in the article) + the FITEQL input file follow.

FITEQL file names + corresponding species:

DM

Am(III)	
XOAM	$XO\text{-}Am^{2+}$
XOAM1	$XO\text{-}AmOH^{+}$
XOAM2	$XO\text{-}Am(OH)_2^{0}$
XOH2AM3	$XOH\text{-}Am(OH)_2^{+}$
XOAM3	$XO\text{-}Am(OH)_3^{-}$
XOHAM3	$XOH\text{-}Am(OH)_3^{0}$
XOHAM2	$XOH_2\text{-}Am(OH)_3^{+}$

switch DM 10/26/94

batch file C:\MODEL\AMSIF5\AMS102C.BAT :

```
fit2    <xoam.c51 >xoam.cut
fit2    <xoam1.c51 >xoam1.cut
fit2    <xoam2.c51 >xoam2.cut
fit2    <xoam3.c51 >xoam3.cut
fit2    <xoham2.c51 >xoham2.cut
fit2    <xoham3.c51 >xoham3.cut
fit2    <xoh2am3.c51 >xoh2am3.cut
```

input file C:\MODEL\AMSIF5\XOAM.C51
(.C51 => C for constant capacitance model, 51 for graph #5, Am data)

JM

K val = -6.94 for deprotonation rxn is not correct: -7.05 is right value for 0.1 M electrolyte + -6.94 is over corrected.
DM 6/9/94

```
                0
                1
                0

                0
                1
               90
                3          1          3          1
00001       -3.09   8.06E-4   XOH
00160       -2.0    0.00E00   PSI0
00031       -9.3    5.0E-10   Am+3
00032        0.0    0.00E00   Am(ads)
00050        0.0    0.00E00   H+
00003       -1.0    0.00E00   NA+
00005       -1.0    0.00E00   ClO4-

00050        0.00   050  1
00031        0.00   031  1
03100       -7.03   031  1 050 -1
03101      -16.65   031  1 050 -2
03102      -24.45   031  1 050 -3
00100      -13.78   050 -1
01100       -6.94   001  1 160 -1 050 -1
00001        0.00   001  1
03201        1.00   001  1 160  2 050 -1 031  1 032  1
```

```
Americum
  on
 silica

2.3s\nm2
00000050
00000060
00000070
00000080
00000090
00000098
00000100
00000102
00000104
00000106
00000110
00000120
00000124
00000126
00000126
00000126
00000139
00000145
00000150
 xoam
00000174
00000175
```


JM

```
00001   175.0 1.202     1.0                                  00000180
0.1     1.0                                                  00000185
          1                                                  00000190
       3201                                                  00000195
         15          1         1         0                  00000230
         32                                                  00000240
1.4362E-11                                                   00000260
5.8873E-11                                                   00000280
1.1438E-10                                                   00000300
1.8634E-10                                                   00000320
2.3679E-10                                                   00000330
2.8423E-10                                                   00000340
2.7923E-10                                                   00000350
3.3540E-10                                                   00000260
3.4299E-10                                                   00000280
3.7051E-10                                                   00000300
4.2646E-10                                                   00000320
4.5608E-10                                                   00000330
4.7653E-10                                                   00000340
4.8117E-10                                                   00000350
4.6909E-10                                                   00000350
        50                                                   00000560
-3.91                                                        00000580
-4.51                                                        00000600
-5.17                                                        00000620
-5.60                                                        00000640
-6.17                                                        00000650
-6.44                                                        00000660
-6.70                                                        00000670
-7.18                                                        00000580
-7.43                                                        00000600
-7.69                                                        00000620
-8.25                                                        00000640
-8.78                                                        00000650
-9.23                                                        00000660
-9.65                                                        00000670
-9.98                                                        00000670

                                                             00000880
          1         1                                        10%error
         32      0.10   5.0E-11                             +/- 0.02
         50      0.05   0.0E-00
```

JM

end of file

2/22/94 DM Source, Righetto et al, 1991
Am(III) on silica
0.1 M $NaClO_4$
~~1.0 g/L~~ DM 2/22/94 1.202 g/L
$[Am(III)]_T = 5.0 \times 10^{-10}$ M

Model-specific values (0.1 M ionic strength)

	CCM	DLM	TLM
$LogK_1$	—	—	.79
K_2	see pg 94 note DM 6/9/94 -6.94	-7.10	-4.79
K_{cat}	—	~~-6.22~~ DM 2/22/94	4.44
K_{an}	—	—	-6.22
C_1 (Farads/m^2)	1.00	—	.80
C_2	—	—	.20
N_s (sites/nm^2)	2.31	2.31	2.31

This value is generated by FITEQL & is found in the output file under "Standard Deviation" PM 6/1/94

2/28/94 PM Righetto '91 Am(III) on Silica - continued

Following is a table with FITEQL output results. The ** values are back-corrected to ionic strength = 0 for MINTEQ use later. The $\sigma_{\log K}$ spaces are blank because I don't know how to calculate them yet.

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: silica A_{sp}: 175 m^2/g Data Source: Righetto 91 Concentration: $[Am^{3+}] = 5.0 \times 10^{-10}$	Rel Error (pH): .05 Abs Error (pH): .0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 5.0e-11	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_S = 2.31 sites/nm^2 1.202 g/L, 0.020 mol/L, 1200 ppm SiO_2, [XOH] = 8.06e-4

no CO_2	DLM (I=0.1 M)			CCM (I=0.1 M)			TLM (I=0.1 M)		
	Log K_1 = - Log K_2 = -7.10 {-7.21}			Log K_1 = - Log K_2 = -6.94 {-7.05} C = 1.0 F/m^2			Log K_1 = 0.79 {0.90} Log K_2 = -4.79 {-4.90} Log $K_{1,w}$ = -6.22 {-6.22} Log $K_{2,w}$ = 4.44 {4.66}		
	Log K	V_T	$\sigma_{\log K}$	Log K	V_T	$\sigma_{\log K}$	Log K	V_T	$\sigma_{\log K}$
$XO\text{-}Am^{2+}$ {-3.20}	-4.08	1.58		-4.08	1.63		-5.58 {-4.70}	5.03	
$XO\text{-}AmOH^{+}$ {-9.21}	-9.98	1.91		-9.98	1.92		-10.83 {-10.06}	1.82	
$XO\text{-}Am(OH)_2^{o}$ {-15.18}	NC -15.84	2.12		-15.82 {15.16}	2.11		-15.25 {-14.59}	1.94	
$XOH\text{-}Am(OH)_2^{+}$ {-9.21}	-9.98	1.91		-9.98	1.92		NC -12.25 {-11.48}	2.08	
$XO\text{-}Am(OH)_3^{-}$ {-18.83}	-19.38	3.02		-19.37 {-18.82}	3.02		-19.39 {-18.84}	2.15	
$XOH\text{-}Am(OH)_3^{o}$ {-15.18}	-15.84	2.12		NC -15.83 {-15.17}	2.11		-15.81 {-15.15}	2.12	
$XOH_2\text{-}Am(OH)_4^{+}$ {-9.21}	-9.98	1.91		-9.98	1.92		-11.68 {-10.91}	1.89	
$XO\text{-}AmCO_3^{o}$									
$XOH\text{-}AmCO_3^{+}$									
$XOH_2\text{-}Am(CO_3)_2^{o}$									
$XOH_2\text{-}Am(CO_3)_3^{+}$									
OTHER SPECIES:									
NC: no convergence									
**: values for I = 0 are	in { }.								

3/1/94 PM Righetto '91 Np(V) on Silica
FITEQL filenames + corresponding species:

PM

Np (V)	
XONPO2	$XO\text{-}NpO_2^{0}$
XOHNPO2	$XOH\text{-}NpO_2^{+}$
XONPO21	$XO\text{-}NpO_2OH^{-}$
XOHNPO21	$XOH\text{-}NpO_2OH^{0}$
XOH2NO21	$XOH_2\text{-}NpO_2OH^{+}$

input file C:\MODEL\NPSIE5\XOH2NO21.DS1

space should NOT be there PM 3/1/94

```
                                                                    Neptunum
                                                                       on
                                                                     silica
                0                                                   2.3s\nm2
                1                                                   00000050
                0                                                   00000060
                0                                                   00000070
                1                                                   00000080
               90                                                   00000090
                3          1          3          1                 00000098
00001  -3.09  8.07E-4   XOH                                        00000100
00160  -2.0   0.00E00   PSI0                                       00000102
00031  -14.0  1.0E-14   NpO2+                                      00000104
00032   0.0   0.00E00   NpO2(ads)                                  00000106
00050   0.0   0.00E00   H+                                         00000110
00003  -1.0   0.10E00   Na+                                        00000120
00005  -1.0   0.10E00   ClO4-                                      00000124
                                                                   00000126
00050    0.00  050  1                                              00000139
00031    0.00  031  1                                              00000145
03100   -8.90  031  1 050 -1                                       00000150
00100  -13.78  050 -1                                              XOH2NO21
01100   -6.99  001  1 160 -1 050 -1                                00000174
00001    0.00  001  1                                              00000175
03206    5.00  001  1 160  1 050  0 031  1 032  1                  00000180
                                                                   00000185
00002   175.0 1.202                                                00000190
0.1    1.0                                                         00000195
         1                                                         00000230
        3206                                                       00000240
         15         1         1         0                         00000260
         32                                                        00000280
4.5489E-16                                                         00000300
5.5698E-16                                                         00000320
6.1346E-16                                                         00000660
8.0280E-16                                                         00000680
1.6107E-15                                                         00000700
2.2342E-15                                                         00000710
2.8760E-15                                                         00000660
3.5406E-15                                                         00000680
4.2281E-15                                                         00000700
5.0530E-15                                                         00000710
5.9557E-15                                                         00000660
6.3178E-15                                                         00000680
7.2208E-15                                                         00000700
7.0425E-15                                                         00000710
6.5255E-15                                                         00000660
         50                                                        00000680
-6.21                                                              00000700
-6.64                                                              00000560
-7.11                                                              00000580
-7.62                                                              00000600
-8.24                                                              00000620
-8.49                                                              00000640
-8.74                                                              00000660
-8.96                                                              00000680
-9.18                                                              00000700
-9.38                                                              00000710
-9.61                                                              00000660
-9.74                                                              00000680
-10.04                                                             00000700
-10.13                                                             00000710
-10.24                                                             00000680
                                                                   00000700
                                                                   00000710
                                                                   00000660
                                                                   00000680
                                                                   00000700
                                                                   00000710
                                                                   00000880
  1        1                                                       10%error
 32      0.10   1.0E-15
```

PM

```
 50      0.05   0.0E-00                                             +/- 0.02
```

end of file

Source: Righetto et al, 1991
Np(V) on silica
0.1 M $NaClO_4$
1.202 g/L
$[Np(V)]_T = 1.0 \times 10^{-14}$ M

Model-specific values; see table, page 95.

3/8/94 PM Righetto '91 Np(V) on Silica - continued

Following is an abbreviated output file showing a typical result in this set of data: the program runs through 90 iterations + still doesn't converge. I am going on to the Righetto Th data + will come back to this problem later.

PM

FILE: X0H2N021.DUT

INPUT DATA FOR VERIFICATION

SERIAL DATA AND ERROR ESTIMATES

TOTAL CONCENTRATION FOR COMPONENT:

	32
1	4.549E-16
2	5.570E-16
3	6.135E-16
4	8.028E-16
5	1.611E-15
6	2.234E-15
7	2.876E-15
8	3.541E-15
9	4.228E-15
10	5.053E-15
11	5.956E-15
12	6.318E-15
13	7.221E-15
14	7.043E-15
15	6.525E-15

LOG FREE CONCENTRATION FOR COMPONENT:

	50
1	-6.21
2	-6.64
3	-7.11
4	-7.62
5	-8.24
6	-8.49
7	-8.74
8	-8.96
9	-9.18
10	-9.38
11	-9.61
12	-9.74
13	-10.04
14	-10.13
15	-10.24

STANDARD DEVIATION OF FREE CONCENTRATION:

COMPONENT 50 RELATIVE 0.0500 ABSOLUTE 0.000E+00

STANDARD DEVIATION OF TOTAL CONCENTRATION:

COMPONENT 32 RELATIVE 0.1000 ABSOLUTE 1.000E-15

VALUES OF ADJUSTABLE PARAMETERS AT EACH ITERATION: LOG K, T, LOG X

PM

I	V(Y): SOS/DF 3206	
0	5.000E+00	5.673E-02
1	4.000E+00	2.783E-02
2	3.000E+00	3.946E+00
3	2.000E+00	2.768E-03
4	1.462E+00	9.156E-01
5	2.337E+00	3.799E-02
6	1.337E+00	2.866E-04
7	1.338E+00	2.047E-03
8	1.620E+00	4.066E-02
9	2.376E+00	2.602E-02
10	2.664E+00	2.177E-04
11	1.664E+00	1.428E-03
12	1.901E+00	1.120E-03
13	1.895E+00	3.121E-03
14	4.424E-01	4.382E-03
15	1.266E+00	6.356E-03
16	2.729E+00	7.484E-01
17	2.594E+00	1.780E-04
18	1.594E+00	6.925E-04
19	1.240E+00	2.279E-03
20	1.818E+00	7.218E-02
21	2.684E+00	4.230E-02
22	1.684E+00	1.023E-01
23	9.031E-01	2.594E-03
24	1.832E+00	6.291E-02
25	2.457E+00	9.070E-04
26	1.457E+00	6.996E-01
27	1.668E+00	9.518E-04
28	1.682E+00	3.833E-03
29	1.184E+00	3.375E-03
30	1.490E+00	4.657E-02

— file continued —

PM

31	1.558E+00	1.475E-01
32	1.333E+00	1.558E-01
33	1.619E+00	1.447E-01
34	1.154E+00	2.927E-03
35	1.579E+00	1.805E-01
36	2.604E+00	4.327E-02
37	1.604E+00	5.552E-04
38	1.626E+00	5.106E-03
39	2.028E+00	3.597E-01
40	2.750E+00	8.529E-02
41	1.750E+00	2.506E-03
42	2.120E+00	7.594E-03
43	2.326E+00	3.694E-04
44	1.326E+00	4.803E-01
45	2.291E+00	4.246E-01
46	2.733E+00	3.938E-04
47	1.733E+00	2.604E-04
48	1.419E+00	8.279E-03
49	1.586E+00	4.665E-03
50	1.860E+00	5.518E-03
51	1.626E+00	2.123E-03
52	7.029E-01	2.368E-01
53	2.357E+00	5.208E-04
54	1.376E+00	6.518E-03
55	1.341E+00	1.247E-03
56	1.448E+00	1.317E-02
57	1.795E+00	2.443E-03
58	7.949E-01	1.985E-03

PM

59	1.308E+00	9.623E-03
60	1.876E+00	8.924E-03
61	1.341E+00	6.237E-03
62	2.147E+00	1.269E-02
63	1.147E+00	1.577E-03
64	1.732E+00	1.632E-02
65	1.421E+00	6.330E-03
66	2.015E+00	3.856E-03
67	2.436E+00	8.336E-04
68	1.436E+00	9.723E-03
69	1.774E+00	7.912E-03
70	2.100E+00	6.765E-03
71	2.281E+00	4.536E-03
72	2.656E+00	8.819E-03
73	1.656E+00	2.312E-01
74	2.072E+00	5.001E-02
75	2.634E+00	1.075E-03
76	1.634E+00	1.010E-01
77	1.757E+00	1.735E-02
78	2.529E+00	3.877E-03
79	1.529E+00	2.282E-03
80	1.841E+00	2.865E-04
81	1.489E+00	1.971E-03
82	1.841E+00	1.672E-02
83	1.283E+00	9.623E-01
84	2.543E+00	3.323E-03
85	1.543E+00	5.147E-03
86	1.030E+00	2.722E-02
87	1.949E+00	1.279E-03
88	1.728E+00	1.763E-03
89	1.575E+00	1.337E-03
90	1.479E+00	2.212E-02

**** NO COVERGENCE OF OPTIMIZATION PROCEDURE

90	1.479E+00	2.213E-02

PM

OPTIMIZATION PROCEDURE: FINAL STEP

NORMAL MATRIX: SUM OF U*V*(DY/DU)*(DY/DV)/(SY*SY)

	3206
3206	6.684E-03

RIGHT HAND SIDE OF NORMAL EQUATIONS: SUM OF U*(DY/DU)*Y/(SY*SY)

3206
-3.427E-02

INVERSE OF NORMAL MATRIX = COVARIANCE MATRIX: S(U,V)*S(U,V)/(U*V)

	3206
3206	1.496E+02

SOLUTION TO NORMAL EQUATIONS: DELTA K/K, DELTA T/T, DELTA X/X

3206
-5.128E+00

STATISTICS:

STANDARD DEVIATION: S(LOG K), S(T), S(LOG X)

3206
5.312E+00

LINEAR CORRELATION COEFFICIENTS: S(U,V)*S(U,V)/(S(U,U)*S(V,V))

	3206
3206	1.000E+00

end of file

Continued on page 101.

Minteq A2 | See page 2 DM 6/1/94

Uranium

Filename	Date	Description of System.
U2N15-3t	6-17-92	1×10^{-5} $UO_2(NO_3)_2$; 0.1M $NaNO_3$; ~~No CO_2~~ C_T = 0.001 $NaHCO_3$; No CO_2
U2N15__t	6-17-92	1×10^{-5}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; No CO_2
U2N16__t	6-17	1×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; No CO_2
U2N15-2t	6-18	1×10^{-5}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; C_T = 0.01M $NaHCO_3$; No CO_2
U2N16-3t	6-18	1×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; C_T = 0.001M $NaHCO_3$; No CO_2
UNNØ6__t	6-28	2×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Data; No CO_2; Range 2-10 pH
UONØ6__t	6-28	2×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; Old Data; No CO_2; 2-10 pH
UONØ5__t	6-26	2×10^{-5}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; Old Dbs; No CO_2; pH = 2-10
UNNØ5__t	6-26	2×10^{-5}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Dbs; No CO_2; pH = 2-10
UNNØ6__C	6-26	2×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Dbs; w/E-3.48 CO_2; pH = 2-10
UONØ6__C	6-26	2×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; Old Dbs; w/E-3.48 CO_2; pH = 2-10
UNNØ6_C	7-1	2×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Dbs; pH = 6.9; CO_2, 10^{-4} to 10^{-1}
UONØ6_C	7-6	2×10^{-6}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; Old Dbs; pH = 6.9; CO_2, 10^{-4} to 10^{-1}
UONØ5_C	7-6	2×10^{-5}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; old Dbs; pH = 6.9; CO_2: .0001-.1 atm
UNNØ5_C	7-6	2×10^{-5}M $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Dbs; pH = 6.9; CO_2: .0001-.1 atm
UNNØØ__t	7-6	Variable T.C. ~~6×10^{-6}~~ $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Dbs; pH = 6.9; No CO_2
UNNØØ__C	7-6	Variable $UO_2(NO_3)_2$; 0.1M $NaNO_3$; New Dbs; pH = 6.9; w/ $10^{-3.48}$ CO_2
UONØØ__t	7-6	Variable $UO_2(NO_3)_2$; 0.1M $NaNO_3$; Old Dbs; pH = 6.9; No CO_2
UONØØ__C	7-6	Variable $UO_2(NO_3)_2$; 0.1M $NaNO_3$; Old Dbs; pH = 6.9; w/ $10^{-3.48}$ CO_2

3/14/94 DM Righetto '91 Th(IV) sorption on silica

FITEQL file names and corresponding species:

Th(IV)	
XOTH	$XO\text{-}Th^{3+}$
XOTH1	$XO\text{-}ThOH^{2+}$
XOTH2	$XO\text{-}Th(OH)_2^{+}$
XOHTH2	$XOH\text{-}Th(OH)_2^{2+}$
XOTH3	$XO\text{-}Th(OH)_3^{0}$
XOHTH3	$XOH\text{-}Th(OH)_3^{+}$
XOTH4	$XO\text{-}Th(OH)_4^{-}$
XOHTH4	$XOH\text{-}Th(OH)_4^{0}$
XOH2TH4	$XOH_2\text{-}Th(OH)_4^{+}$

Input file c:\MODEL\THSIF5\XOHTH4.T51

```
                                                                                  Thorium
                                                                                    on
                                                                                  silica

                                                                                 2.3s\nm2
    0                                                                            00000050
    1                                                                            00000060
    0                                                                            00000070
                                                                                 00000080
    0                                                                            00000090
    1                                                                            00000091
   90                                                                            00000092
    5          1          3          1                                          00000098
00001  -6.17  6.72E-7  XOH                                                       00000100
00160  -1.0   0.00E00  PSI0                                                      00000102
00161  -0.5   0.00E00  PSIB                                                      00000104
00162  -0.1   0.00E00  PSID                                                      00000106
00051  -11.0  1.0E-11  Th+4                                                      00000110
00052   0.0   0.00E00  Thads                                                     00000120
00050   0.0   0.00E00  H+                                                        00000124
00003  -1.0   0.00E00  NA+                                                       00000126
00005  -1.0   0.00E00  ClO4-                                                     00000127
                                                                                 00000128
00050   0.00  050  1                                                             00000129
00051   0.00  051  1                                                             00000130
05101  -4.55  051  1 050 -1                                                       00000131
05102  -8.21  051  1 050 -2                                                       00000132
05103 -13.02  051  1 050 -3                                                       00000139
05104 -17.22  051  1 050 -4                                                       00000140
05105  -5.81  051  2 050 -2                                                       00000145
05106 -20.89  051  4 050 -8                                                       00000147
05107 -37.72  051  6 050-15                                                       00000149
00100 -13.78  050 -1                                                             00000150
01050   0.79  001  1 160  1 050  1                                               xohth4
01100  -4.79  001  1 160 -1 050 -1
01103  -6.22  001  1 160 -1 161  1 050 -1 003  1                                 00000174
01055   4.44  001  1 160  1 161 -1 050  1 005  1                                 00000175
00001   0.00  001  1                                                             00000180
05208  -3.10  001  1 160  0 161  0 050 -4 051  1 052  1                          00000185
                                                                                 00000190
                                                                                 00000195
00004  175.0  0.060  0.8    0.2                                                  00000230
0.1    1.0                                                                       00000240
         1                                                                       00000380
      5208                                                                       00000400
        11          1          1          0                                     00000420
        52                                                                       00000421
1.3936E-14                                                                       00000422
1.2875E-12                                                                       00000423
5.2172E-12                                                                       00000424
6.2482E-12                                                                       00000425
6.9084E-12                                                                       00000426
7.4908E-12                                                                       00000427
7.6344E-12                                                                       00000428
7.6308E-12                                                                       00000560
7.4897E-12                                                                       00000580
7.6279E-12                                                                       00000600
7.4694E-12                                                                       00000620
        50                                                                       00000640
-0.063                                                                           00000660
-0.285                                                                           00000670
-0.610                                                                           00000680
-0.871                                                                           00000700
-1.144                                                                           00000720
-1.430
-1.935
-2.245
-2.512

-2.760
-3.308                                                                           00000740
         1          1                                                            00000741
        52       0.10    1.0E-12                                                 00000880
        50       0.05    0.0E-00                                                 10%error
                                                                                 +/- 0.02
```

end of file

3/21/94 PM Righetto '91 Th(IV) on Silica - Continued
Table of FITEQL output:

DM

Thorium (IV) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: silica A_{sp}: 175 m^2/g Data Source: Righetto 91 Concentration:[Th^{++}] = 1e-11	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1e-12	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 0.060 g/L, 0.001 mol/L, 60 ppm SiO_2, [XOH]=6.72e-7

no CO_2	DLM (I=0.1 M)			CCM (I=0.1 M)			TLM (I=0.1 M)		
	Log K_1 = - Log K_2 = -7.10			Log K_1 = - Log K = -6.94 C = 1.0 F/m^2			Log K_1 = 0.79 Log K = -4.79 Log K_{1a} = -6.22 Log K_{2a} = 4.44		
	Log K	V_T	$\sigma_{Log K}$	Log K	V_1	$\sigma_{Log K}$	Log K	V_T	$\sigma_{Log K}$
XO-Th^{++}	14.0 [15.65]**	18.9		5.28 [6.93]	1.15		7.92 [9.57]	1.33	
XO-$Th(OH)^{+}$	4.73 [6.27]	1.34		4.73	1.34		7.36 [8.90]	1.62	
XO-$Th(OH)_2^{+}$	4.18 [5.61]	1.62		4.18	1.62		6.81 [8.24]	1.84	
XOH-$Th(OH)_2^{++}$	4.73 [6.27]	1.34		4.73	1.34		7.49 [9.03]	1.62	
XO-$Th(OH)_3^{-}$	3.63 [4.95]	1.84		3.63	1.84		6.26 [7.58]	1.98	
XOH-$Th(OH)_3^{+}$	4.18 [5.61]	1.62		4.18	1.62		6.94 [8.37]	1.84	
XO-$Th(OH)_4$	-9.78 [-8.37]	13.7		-9.78	1.37		-9.54 [-8.33]	1.37	
XOH-$Th(OH)_4^{-}$	3.63 [4.95]	1.84		3.63	1.84		6.39 [7.71]	1.99	
XOH_2-$Th(OH)_4^{+}$	4.18 [5.61]	1.62		4.18	1.62		7.07 [8.50]	1.84	
OTHER SPECIES:	-								

**: values for I=0 are in [ ]

Results from the log K values generated by FITEQL (back-corrected to ionic strength = 0) will be used in the PRODEFA2 / MINTEQA2 program to determine % of the different ionic species sorbed in a given system, e.g. Americium species on silica.

3/29/94 PM MINTEQ runs of Righetto-based Data
The output files from MINTEQA2 are huge. Below are reduced copies of pH vs % sorbed Am species used in the SIGMAPLOT software program to build graphs.
Data in the C1.SP5 + C2.SP5 sets are from the constant capacitance model.

R91AMC1.SP5: Tue, 29-Mar-94

DM

1	SiOAm+2 2	3	4	5
Am(III)onSilica Am=5E-10	Righetto 91 Si=.06g/L		--	--
pH	% sorbed		Experimental pH	% sorbed
2	0		3.9	2.9
2.25	0		4.5	11.8
2.5	0		5.2	22.9
2.75	0		5.6	37.3
3	0		6.2	47.4
3.25	0		6.4	56.8
3.5	0		6.7	55.8
3.75	0		7.2	67.1
4	0		7.4	68.6
4.25	0.1		7.7	74.1
4.5	0.1		8.2	85.3
4.75	0.3		8.8	91.2
5	0.5		9.2	95.3
5.25	1		9.7	96.2
5.5	2.1		10	93.8
5.75	4.9			
6	11.5			
6.25	26.1			
6.5	50.1			
6.75	74.5			
7	89.4			
7.25	95.9			
7.5	98.4			
7.75	99.4			
8	99.7			
8.25	99.9			
8.5	99.9			
8.75	100			
9	100			
9.25	100			
9.5	100			
9.75	100			
10	100			
10.25	99.9			

DM

R91AMC1.SP5: Tue, 29-Mar-94

7	SiOAm(OH)+ 8	9	10
--	--		--
--	--		--
pH	% sorbed		pH
2	0		2
2.25	0		2.25
2.5	0		2.5
2.75	0		2.75
3	0		3
3.25	0		3.25
3.5	0		3.5
3.75	0		3.75
4	0		4
4.25	0		4.25
4.5	0		4.5
4.75	0		4.75
5	0		5
5.25	0.1		5.25
5.5	0.5		5.5
5.75	1.8		5.75
6	6.2		6
6.25	20.1		6.25
6.5	49.1		6.5
6.75	78.5		6.75
7	93		7
7.25	97.9		7.25
7.5	99.3		7.5
7.75	99.8		7.75
8	99.9		8
8.25	100		8.25
8.5	100		8.5
8.75	100		8.75
9	100		9
9.25	100		9.25
9.5	100		9.5
9.75	100		9.75
10	100		10
10.25	100		10.25

DM

R91AMC1.SP5: Tue, 29-Mar-94

SiOHAm(OH)2+ 11	12	SiOAm(OH)2 13	14
--		--	--
--		--	--
pH	% sorbed	pH	% sorbed
2	0	2	0
2.25	0	2.25	0
2.5	0	2.5	0
2.75	0	2.75	0
3	0	3	0
3.25	0	3.25	0
3.5	0	3.5	0
3.75	0	3.75	0
4	0	4	0
4.25	0	4.25	0
4.5	0	4.5	0
4.75	0	4.75	0
5	0	5	0
5.25	0.1	5.25	0.2
5.5	0.5	5.5	0.6
5.75	1.8	5.75	2.4
6	6.2	6	9.5
6.25	20.1	6.25	31.4
6.5	49	6.5	67
6.75	78.5	6.75	89.9
7	93	7	97.4
7.25	97.9	7.25	99.3
7.5	99.3	7.5	99.8
7.75	99.8	7.75	99.9
8	99.9	8	100
8.25	100	8.25	100
8.5	100	8.5	100
8.75	100	8.75	100
9	100	9	100
9.25	100	9.25	100
9.5	100	9.5	100
9.75	100	9.75	100
10	100	10	100
10.25	100	10.25	100

PM

R91AMC2.SP5: Tue, 29-Mar-94

SiO-Am(OH)3-

1	2	3	4	5
m(III)onSilica	Righetto 91		--	--
Am=5E-10	Si=.06g/L		Experimental	--
pH	% sorbed		pH	% sorbed
2	0		3.9	2.9
2.25	0		4.5	11.8
2.5	0		5.2	22.9
2.75	0		5.6	37.3
3	0		6.2	47.4
3.25	0		6.4	56.8
3.5	0		6.7	55.8
3.75	0		7.2	67.1
4	0		7.4	68.6
4.25	0		7.7	74.1
4.5	0		8.2	85.3
4.75	0		8.8	91.2
5	0.1		9.2	95.3
5.25	0.8		9.7	96.2
5.5	6.3		10	93.8
5.75	36.1			
6	81.7			
6.25	97.1			
6.5	99.6			
6.75	99.9			
7	100			
7.25	100			
7.5	100			
7.75	100			
8	100			
8.25	100			
8.5	100			
8.75	100			
9	100			
9.25	100			
9.5	100			
9.75	100			
10	100			
10.25	100			

PM

R91AMC2.SP5: Tue, 29-Mar-94

SiOHAm(OH)3

6	7	8	9
	--	--	
	--	--	
	pH	% sorbed	
	2	0	
	2.25	0	
	2.5	0	
	2.75	0	
	3	0	
	3.25	0	
	3.5	0	
	3.75	0	
	4	0	
	4.25	0	
	4.5	0	
	4.75	0	
	5	0	
	5.25	0	
	5.5	0.1	
	5.75	0.7	
	6	3.7	
	6.25	16.8	
	6.5	50.9	
	6.75	83.6	
	7	95.9	
	7.25	99	
	7.5	99.7	
	7.75	99.9	
	8	100	
	8.25	100	
	8.5	100	
	8.75	100	
	9	100	
	9.25	100	
	9.5	100	
	9.75	100	
	10	100	
	10.25	100	

R91AMC2.SP5: Tue, 29-Mar-94

SiOH2Am(OH)3+

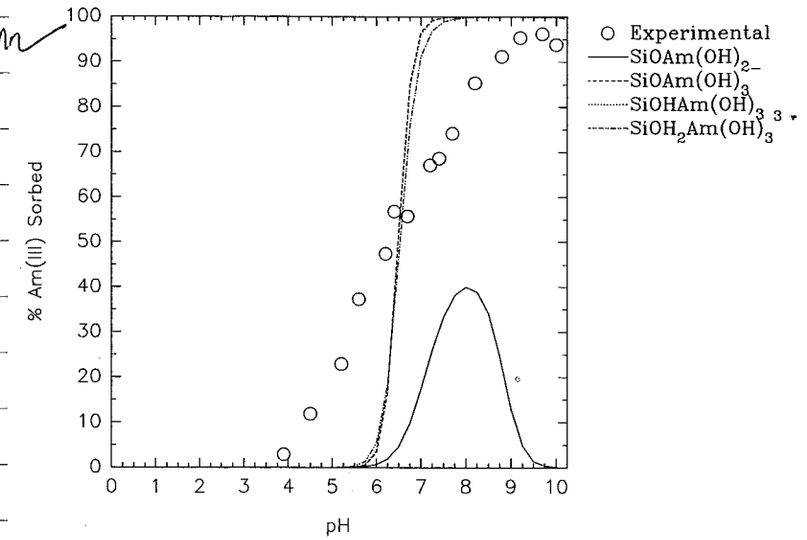
10	11
--	--
--	--
pH	% sorbed
2	0
2.25	0
2.5	0
2.75	0
3	0
3.25	0
3.5	0
3.75	0
4	0
4.25	0
4.5	0
4.75	0
5	0
5.25	0.1
5.5	0.5
5.75	1.7
6	6.2
6.25	20
6.5	48.9
6.75	78.4
7	93
7.25	97.9
7.5	99.3
7.75	99.8
8	99.9
8.25	100
8.5	100
8.75	100
9	100
9.25	100
9.5	100
9.75	100
10	100
10.25	100

PM

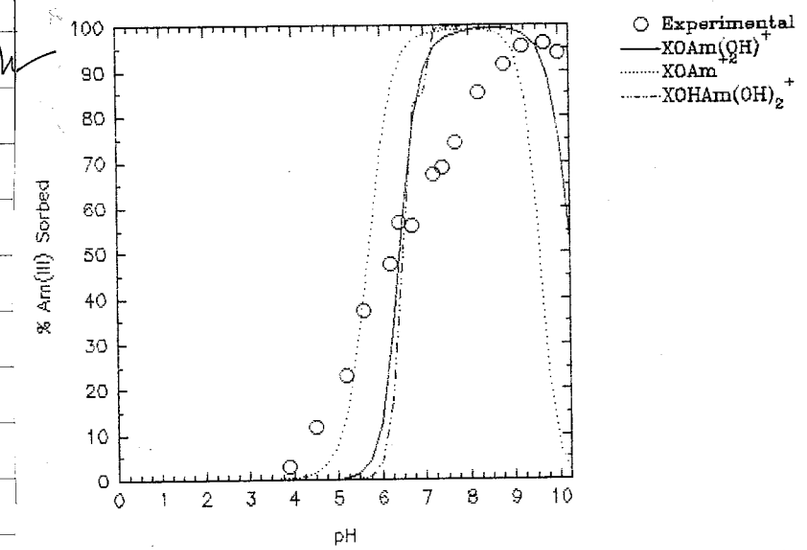
PM

The graps show the sorption of the Am(III) species (O marks experimental data) versus pH.

PM



PM



Continued on page 107

See page 2
PM 6/1/94

Plutonium

Filename	Date	Description of System.
P4N11__t	6/18	1×10^{-11} M $Pu(NO_3)_4$; 0.1 M $NaNO_3$; No CO_2
P1N11__t	6/18	1×10^{-11} M $PuO_2(NO_3)$; 0.1 M $NaNO_3$; No CO_2
P1N10__t	6/18	1×10^{-10} M $PuO_2(NO_3)$; 0.1 M $NaNO_3$; No CO_2
P4N11A1~~__~~t	6/18	1×10^{-11} M $Pu(NO_3)_4$; 0.1 M $NaNO_3$;
P1N11A1~~__~~t	6/18	Alk = 10, 30, 100

4/6/94 PM MINTEQA2 screens:

Following are reduced screen printouts showing the different editing levels used in MINTEQA2. The file shown is the Am(III)-Silica sorption, CCM, $Si{:}OAm^{+2}$ species from the Righetto '91 data.

PM

```
PROGEFAZERE

                                   EDIT LEVEL 1                    PROB # 1

 1 Title 1: Am(III)-Silica Sorption, CCM, SiOAm-2
 2 Title 2:
 3 Temperature (Celsius):  25.00
 4 Units of concentration: MOLAL
 5 Ionic strength:  TO BE COMPUTED
 6 Inorganic carbon is not specified.
 7 Terminate if charge imbalance exceeds 30% ? NO
 8 Oversaturated solids ARE NOT ALLOWED to precipitate. EXCEPTIONS: Solids
   listed in this file as TYPE -III (Infinite), -IV (Finite) or -V (Possible).
 9 The maximum number of iterations is: 200
10 The method used to compute activity coefficients is: Davies equation
11 Level of output: ABBREVIATED
12 The pH is: FIXED at  2.00
13 The pe and Eh are: UNDEFINED
99 Choose a different file to modify OR return to output filename prompt.

To change any of the above entries or to explore other possible values,
enter the number to the left of the entry.  Press ENTER to accept all settings

 ENTER CHOICE > _
```

PM

```
PROGEFAZERE

                                   EDIT LEVEL I                    PROB # 1
                              Adsorbing Surface Number 1

1 Adsorption site densities:

        Site Type Number   Component Id number   Site Density (mol/l)
                1                  S11                0.300E-03

2 Concentration of the adsorbing surface (g/l):    1.202
3 Specific surface area pertaining to this surface (sq. meters/gr):  175.00
4 Inner layer capacitance (farads/sq. meters): 1.000

To change any of the above entries or to explore other possible values,
enter the number to the left of the entry.  Press ENTER to accept all settings

 ENTER CHOICE >
```

PM

```
PROGEFAZERE

                                   EDIT LEVEL III                  PROB # 1
                         Verify or change listing of COMPONENTS

Entry     I.D.     Name          Total Conc.    Log Act.(GUESS)   Improve ?
  1        330     H+1          0.00000E-01        -2.00           YES
  2        500     Na+1         1.00000E-01        -1.00           YES
  3        131     ClO4-        1.00000E-01        -1.00           YES
  4         40     Am+3         5.00000E-10        -9.30           YES
  5        813     ADS1PSIo     0.00000E-01         0.00           YES
  6        811     ADS1TYP1     8.06000E-04        -3.09           YES

Enter entry # to change or delete (press ENTER to accept all entries) > _
```

← Edit level II is summarized + checked in Edit level III + so is not shown here. PM 4/6/94

```
PROGEFAZERE

                                   EDIT LEVEL III                  PROB # 1
                         Verify or change listing of ADDED SPECIES

Entry     I.D.      Name        Log K   Enthalpy   Charge   gfw    Mik. Factor
  1    8113300  SiO-           -7.050     0.00      0.0    0.000     0.00
       Stoichiometry: ( 1.0001811  (-1.0001330  (-1.0001613
  2    8110400  SiOAm+2        -1.200     0.00      0.0    0.000     0.00
       Stoichiometry: ( 1.0001811  ( 1.0001 40  (-1.0001330  ( 2.0001613

Enter entry # to change or delete (press ENTER to accept all entries) >
```

Continued on page 109.

See page 2
PM 6/1/94

~~Neptunium~~

4/8/94 PM Sorption Constants - Compilation
Steve Sassman's FITEQL generated sorption constants for the data sets will be entered into tables and then the input/output files will be checked to make certain the values are correct.

JM 4/11/94 JM
I entered the sorption constants into WP tables today for the U(VI) and Np(V) data. They are saved under C:\model\binding*.* and on a floppy disk with a yellow label under BINDING.**. Presently, the tables are named BINDING.U** or BINDING.N** with a number in the ** position. The files will be renamed later so that it will be possible to know which author's data is in which table from the file name alone.

4/18/94 PM More log K values are being entered into the tables.

JM 5/18/94 JM
Today I took a plot from Waite et. al. (1993) and digitized it. The data is saved on a floppy disk with a red label marked "Digitized Data" under the file name QUARTZ.PRN. The copy of the journal article an a blow-up of the plot is in a manila folder marked "Waite 93".

5/20/94 PM Analog Data Plotting
I will be plotting data received from English Pearcy from the Analog project. The original data files are ANALOG5.inp & ANALOG5.123, which contains pH, C, + S values from MINTEQA2 output. C = concentration + S = amount sorbed of the species. Plots will be 3-D, with (x, y, z) coordinates = (C_t, pH, Kd). Continued on pg 111.

See page 2
PM 6/1/94

Thorium

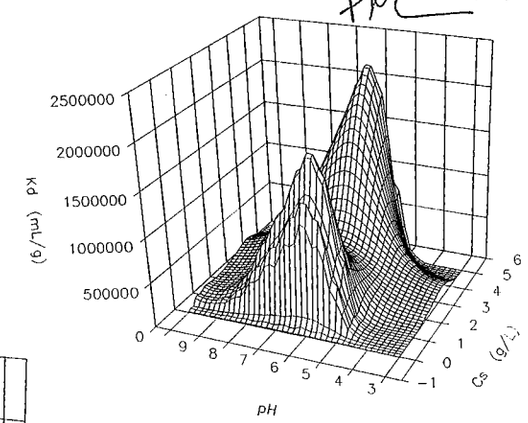
Filename	Date	Description of System.
T4N13__C	6/18	1×10^{-13} M $Th(NO_3)_4$; 0.1 M $NaNO_3$; w/ CO_2
T4N13__t	6/18	1×10^{-13} M $Th(NO_3)_4$; 0.1 M $NaNO_3$; NO CO_2

PM $Kd = \frac{S}{C} \times \frac{1000}{C_s}$

Following are graphs of the data, showing the smoothing effect of using a larger-valued mesh. The 'saddle' seen is probably an artifact.

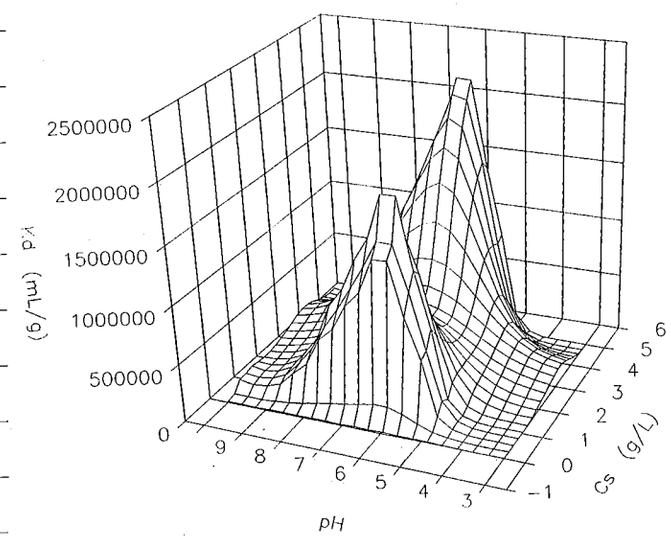
PM

PM 5/20/94



A:\ANALOG1.SPS
40 x 40 mesh
wt of 4

PM



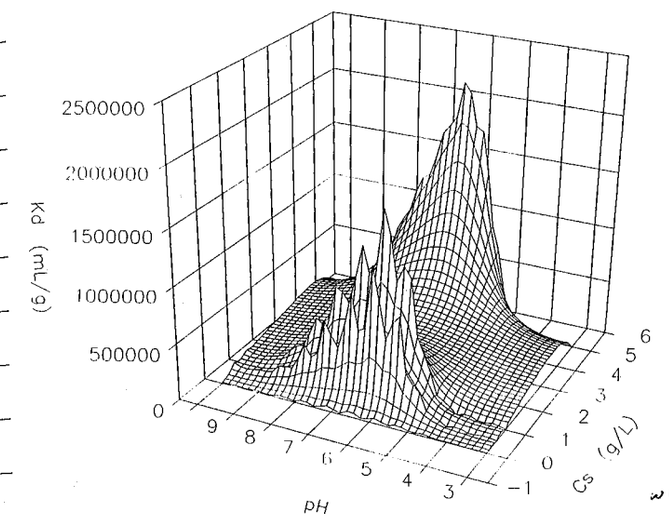
20 x 20 mesh - 400 (x,y,z) points

Continued on pg 113.

See page 2
PM 6/2/94

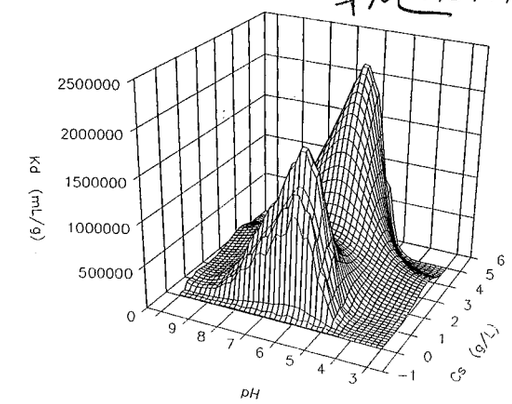
Europium

PM 5/20/94



wt of 2

PM 5/20/94



A:\ANALOG1.SP5
40x40 mesh
wt of 4

wt determines effect of distant pts on interpolation values
larger wt → less emphasis on distant pts → smoother curve which passes closer to the original data

The graph above shows the smoothing effect of a weight of 4 vs a weight of 2. For an explanation of the math involved, refer to page 13-38 in the Math Menu section of the Sigma Plot User's manual.
Interpolation has been done in Sigma Plot with 1600 pts generated.

5/24/94 PM Analog Plot - continued
The midsection of the plot will be filled in

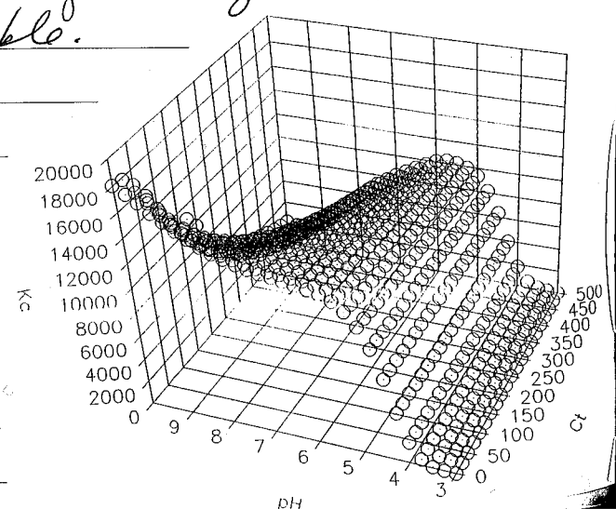
See page 2
PM 6/2/94

~~Americium~~

with more values from MINTEQA2.
The OS2 editor can be used to change the MINTEQA2 input file - type START e filename.ext from the OS2 window.
The modified data in SIGMAPLOT now has
pH = 3.00, 3.25, . . . , 10.00
C_t = 0, 25, 50, . . . , 500
C_s is constant at C_s = 0.01
There are problems with the MINRUN; the program is finished after 0 iterations.
The file type is not wrong, so that's not the problem.

5/25/94 PM Analog plot - continued
The problem is found: pulling the file into WP moved the file's right margin in, making it unreadable.

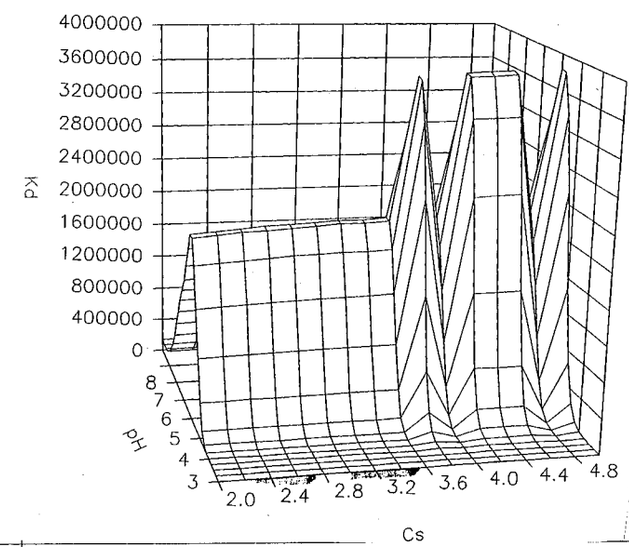
Scatterplot of → the data



5/30/94 PM The plot below shows odd spikes at $C_s \simeq$ 3.8, 4.2-4.6, + 5.0. They are due to an extra zero in the MINTEQA2 input file on the 9th line, C_s value
i.e, 3.8000E+00 vs 3.800E+00.

PM

PM 5/30/94
ANALOG2.SPS
16x29 mesh
wt = 4
C_s =
2.0, 2.2, . . . , 5.0
pH = 3 - 10

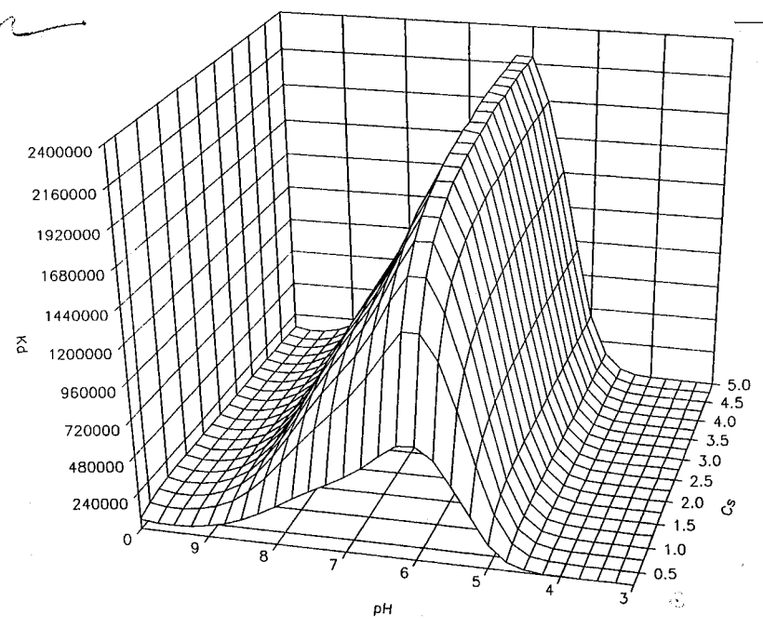


5/31/94 PM The plot on the following page shows the plot with the offending 0s removed and the C_s values taken down to 0.2. This is a much more satisfactory plot

See page 2
PM 6/2/94

~~Cobalt~~

PM



6/3/94 PM Sorption Constants - Compilation - Continued from 4/18/94 (see pg 109 this notebook).

PM

PM 6/3/94
The FITEQL-generated sorption binding constants for the data from Payne, Nakayama & Sakamoto, Righetto, Moulin, Kohler, Hsi, Tripathi, Sanchez, and Venkataramani & Gupta have been entered into the Wordperfect tables. They are saved both on the c: drive of my PC (in C:\model\) and on a floppy disk.

There are currently 72 tables:

BINDING.A1 - A9 for Americium
.N1 - N18 for neptunium
.T1, T2 for thorium
.U1 - U34 for uranium
BINDING4.P1 for plutonium(IV)
5.P1, P2 for plutonium(V)
BINDING U.AS1 - 6 for uranium on Kaolinite,

modelled using a 1:1 AlOH/SiOH ratio.

6/6/94 PM SC compilation continued
In the WP table used for finding model specific values, the CCM values are already adjusted to 0.1M ionic strength. When I ran FITEQL, I readjusted these values further than needed.
All the input & output files from FITEQL need to be checked to see if the K_+, K_-, K_{an}, + K_{cat} values are correct. Also the absolute error associated with each needs to be made

See page 2
PM 6/2/94

Cesium

uniform as this parameter was changed back + forth during earlier work with the files.

6/9/94 PM The updated K tables will be copied reduced, + put in the notebook as they are completed.

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: gamma (γ) alumina A_{sp}: 120 m²/g Data Source: Righetto et al Concentration: [Am(III)] = 5e-10 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 5.0e-13	Ionic Strength (electrolyte): 0.01 M $NaClO_4$ N_s = 2.31 sites/nm²

fig 2.1 C_s = .2 g/L	DLM			CCM			TLM		
	Log K_+ = 6.84 Log K_- = -9.18			Log K_+ = 6.98 Log K_- = -9.06			Log K_+ = 6.35 Log K_- = -10.35 Log K_{Cs} = -7.81 Log K_{As} = 8.23		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	3.474*	4.802	.04823	3.257	12.38	.03597	-1.586	32.35	.02118
XO-$AmOH^+$	-3.901	12.16	.03654	-3.882	11.56	.03759	-7.415	19.04	.02808
XO-$Am(OH)_2^o$	-11.09	10.82	.03927	-10.91*	3.428	.05400	-13.19	14.01	.03335
XOH-$Am(OH)_2^+$	-3.753*	3.997	.05100	-3.934	11.60	.03756	-3.260	11.25	.03821
XO-$Am(OH)_3^-$	-18.29	9.82	.04236	-18.12	10.27	.04072	-18.98	10.66	.03963
XOH-$Am(OH)_3^o$	-11.09	10.82	.03927	-11.03	10.87	.03922	-11.12	10.97	.03885
XOH_2-$Am(OH)_3^+$	-3.753*	3.997	.05100	-3.934	11.60	.03756	-5.334	14.43	.03284
XO-$AmCO_3^o$									
XOH-$AmCO_3^+$									
XOH_2-$Am(CO_3)_2^o$									
XOH_2-$Am(CO_3)_2^{2-}$									
OTHER SPECIES:									

*: Abs Error = 5.0e-12

A:\BINDING.A1

Note that where absolute errors are relaxed, the original error (above, = 5.0e-13) did not allow convergence. In most cases, the value was enlarged by 5.0e1 until convergence occurred.

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: gamma (γ) alumina A_{sp}: 120 m²/g Data Source: Righetto et al Concentration: [Am(III)] = 5e-10	Rel Error (pH): .05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 5.0e-13	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm²

fig 2.2	DLM			CCM			TLM		
	Log K_+ = 6.78 Log K_- = -9.12			Log K_+ = 6.92 Log K_- = -9.00			Log K_+ = 6.29 Log K_- = -10.29 Log K_{Cs} = -7.81 Log K_{As} = 8.11		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	2.314	10.08	.04276	2.820	11.62	.04852	-3.118	42.27	.02127
XO-$AmOH^+$	-3.901	12.16	.03654	-4.353	12.60	.05216	-8.411	7.706	.03115
XO-$Am(OH)_2^o$	-11.47	14.19	.05884	-11.76*	3.142	.08179	-13.92	9.976	.04232
XOH-$Am(OH)_2^+$	-4.580	12.19	.04938	-4.353	12.60	.05216	-3.617	13.11	.05423
XO-$Am(OH)_3^-$	-18.37	16.39	.06791	-18.70	14.89	.06017	-19.44	13.96	.05737
XOH-$Am(OH)_3^o$	-11.47	14.19	.05884	-11.76*	3.142	.08179	-11.55	14.01	.05468
XOH_2-$Am(OH)_3^+$	-4.580	12.19	.04938	-4.353	12.60	.05216	-6.009	9.654	.04104
XO-$AmCO_3^o$									
XOH-$AmCO_3^+$									
XOH_2-$Am(CO_3)_2^o$									
XOH_2-$Am(CO_3)_2^{2-}$									
OTHER SPECIES:									

* Abs Error = 5.0e-12

A2

PM

1988 paper
PM 11/15/94

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: gamma (γ) alumina A_{sp}: 120 m²/g Data Source: Righetto et al Concentration: [Am(III)] = 5e-10 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 5.0e-13	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm² 10 ppm Al_2O_3

fig 1.2	DLM			CCM			TLM		
	Log K_+ = 6.78 Log K_- = -9.12			Log K_+ = 6.92 Log K_- = -9.00			Log K_+ = 6.29 Log K_- = -10.29 Log K_{Cs} = -7.81 Log K_{As} = 8.11		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	2.801	7.510	.05394	3.123	11.03	.06106	-2.239	69.16	.03446
XO-$AmOH^+$	-4.335	12.20	.06360	-4.176	13.41	.06645	-7.531	1.044	.03447
XO-$Am(OH)_2^o$	-11.48	16.30	.07385	-11.48	15.67	.07208	-13.41	6.786	.05256
XOH-$Am(OH)_2^+$	-4.335	12.20	.06360	-4.176	13.41	.06645	-3.502	14.62	.06941
XO-$Am(OH)_3^-$	-18.65	19.91	.08496	-18.78	17.78	.07797	-19.43	16.26	.07372
XOH-$Am(OH)_3^o$	-11.48	16.30	.07385	-11.48	15.67	.07208	-11.47	15.44	.07171
XOH_2-$Am(OH)_3^+$	-4.335	12.20	.06360	-4.176	13.4	.06645	-5.540	5.751	.05066
XO-$AmCO_3^o$									
XOH-$AmCO_3^+$									
XOH_2-$Am(CO_3)_2^o$									
XOH_2-$Am(CO_3)_2^{2-}$									
OTHER SPECIES:									

A3

See page 2
PM 6/2/94

~~Technetium~~

6/13/94 PM K tables continued

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: amorphous silica A_{sp}: 175 m^2/g Data Source: Moulin et al 92 Concentration: [Am(III)] = 1e-8 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-11	Ionic Strength (electrolyte): 0.01 M $NaClO_4$ N_s = 2.31 sites/nm^2

fig 2.1 modelled using pyro-SiO_2 mass\volume ratio: 1g/100mL	DLM Log K_+ = Log K_- = -7.16			CCM Log K_+ = Log K_- = -7.05			TLM Log K_+ = .85 Log K_- = -4.85 Log K_{Cs} = -6.22 Log K_{As} = 4.56		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	-3.023	7.073	.05352	-2.959	6.345	.05199	-5.330	26.90	.02730
XO-$AmOH^+$	-7.514	10.61	.0102	-7.480	10.59	.07006	-10.80	17.56	.03726
XO-$Am(OH)_2^o$	-11.94	10.78	.08409	-11.94	10.78	.08270	-16.96	14.58	.06456
XOH-$Am(OH)_2^+$	-7.514	10.61	.0102	-7.480	10.59	.07006	-5.825	16.25	.08435
XO-$Am(OH)_3^-$	-21.93	20.54	.1497	-22.20	20.54	.1500	-23.09	16.38	.08693
XOH-$Am(OH)_3^o$	-11.94	10.78	.08409	-11.94	10.78	.08270	-14.46	16.32	.08568
XOH_2-$Am(OH)_3^+$	-7.514	10.61	.0102	-7.480	10.59	.07006	-8.322	14.39	.06329
XO-$AmCO_3^o$	-10.41	21.56	.06129	-12.37	21.40	.05995	-15.06	40.14	.03647
XOH-$AmCO_3^+$									
XOH_2-$Am(CO_3)_2^o$	-8.885	24.26	.05797	-8.885	24.26	.06030	-8.352	34.32	.06132
XOH_2-$Am(CO_3)_2^{2-}$	-21.78	44.74	.08544	-22.32	44.80	.08545	-8.434	1427	.09726
OTHER SPECIES:									

A4

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: amorphous silica A_{sp}: 175 m^2/g Data Source: Moulin et al 92 Concentration: [Am(III)] = 1e-8 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-11	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2

fig 2.2 modelled using pyro-SiO_2	DLM Log K_+ = Log K_- = -7.10			CCM Log K_+ = Log K_- = -7.05			TLM Log K_+ = .79 Log K_- = -4.79 Log K_{Cs} = -6.22 Log K_{As} = 4.44		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	-3.411	10.89	.06921	-3.418	10.30	.06937	-5.945	12.16	.06140
XO-$AmOH^+$	-7.434	16.42	.07112	-7.433	16.40	.07112	-8.896	16.17	.06879
XO-$Am(OH)_2^o$	-11.56	16.62	.08547	-11.56	16.61	.08546	-11.87	16.58	.07866
XOH-$Am(OH)_2^+$	-7.434	16.42	.07112	-7.433	16.40	.07112	-8.166	16.62	.08348
XO-$Am(OH)_3^-$	-21.04	15.93	.1205	-21.07	15.93	.1206	-14.85	16.62	.08926
XOH-$Am(OH)_3^o$	-11.56	16.62	.08547	-11.56	16.61	.08546	-11.51	16.62	.09789
XOH_2-$Am(OH)_3^+$	-7.434	16.42	.07112	-7.433	16.40	.07112	-8.529	16.49	.05643
XO-$AmCO_3^o$	-10.03	33.23	.06032	-10.03	33.22	.06032	-10.35	33.12	.05309
XOH-$AmCO_3^+$									
XOH_2-$Am(CO_3)_2^o$	-8.510	36.23	.05908	-8.510	36.24	.05908	-8.096	36.28	.06596
XOH_2-$Am(CO_3)_2^{2-}$	-21.49	38.66	.07976	-21.55	38.65	.08008	-18.58	38.80	.08122
OTHER SPECIES:									

A5

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: alpha-alumina (α Al_2O_3) A_{sp}: 12 m^2/g Data Source: Moulin et al 92 Concentration: [Am(III)] = 1e-8 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-11	Ionic Strength (electrolyte): 0.01 M $NaClO_4$ N_s = 2.31 sites/nm^2 Cs = 10 g/L

fig 1.1 mass to vol ratio 1g/100 mL log pCO_2 = -3.5 atm	DLM Log K_+ = 8.28 Log K_- = -9.68			CCM Log K_+ = 9.08 Log K_- = -8.32			TLM Log K_+ = 6.75 Log K_- = -10.75 Log K_{Cs} = -7.73 Log K_{As} = 10.02		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	2.586	13.51	.1196	2.964	13.52	.1223	-3.970	29.63	.03424
XO-$AmOH^+$	-6.032	13.52	.1217	-5.538	13.53	.1229	-8.820	8.857	.06041
XO-$Am(OH)_2^o$	-9.290	9.821	.09487	-9.125	9.833	.08594	-13.47	9.779	.07254
XOH-$Am(OH)_2^+$	-6.032	13.52	.1217	-5.538	13.53	.1229	-6.123	13.53	.1233
XO-$Am(OH)_3^-$	-16.41	9.840	.1083	-17.41	9.835	.08823	-18.12	9.834	.08749
XOH-$Am(OH)_3^o$	-9.290	9.821	.09487	-9.125	9.833	.08594	-9.739	9.832	.08570
XOH_2-$Am(OH)_3^+$	-6.032	13.52	.1217	-5.538	13.53	.1229	-5.088	9.760	.07109
XO-$AmCO_3^o$	-7.765	19.68	.06697	-7.601	19.67	.06067	-12.09	19.54	.05122
XOH-$AmCO_3^+$									
XOH_2-$Am(CO_3)_2^o$	-6.241	20.19	.06649	-6.077	20.17	.06016	-2.958	20.19	.07047
XOH_2-$Am(CO_3)_2^{2-}$	-19.95	20.37	.08574	-22.74	38.81	.07519	-10.73	20.37	.09550
OTHER SPECIES:									

A6

PM

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: alpha alumina (α Al_2O_3) A_{sp}: 12 m^2/g Data Source: Moulin et al 92 Concentration: [Am(III)] = 1e-8 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-11	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2

fig 1.2 mass to volume ratio 1g/100mL log pCO_2 = -3.5 atm	DLM Log K_+ = 8.22 Log K_- = -9.62			CCM Log K_+ = 9.08 Log K_- = -8.32			TLM Log K_+ = 6.69 Log K_- = -10.69 Log K_{Cs} = -7.73 Log K_{As} = 9.90		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	1.605	14.93	.06832	2.741	16.10	.08200	-5.330	26.90	.02730
XO-$AmOH^+$	-6.558	15.99	.08035	-5.742	16.22	.08419	-10.80	17.56	.03726
XO-$Am(OH)_2^o$	-14.67	16.56	.09061	-14.22	16.33	.08587	-16.96	14.58	.06456
XOH-$Am(OH)_2^+$	-6.558	15.99	.08035	-5.742	16.22	.08419	-5.825	16.25	.08435
XO-$Am(OH)_3^-$	-22.75	16.90	.09980	-22.70	16.43	.08770	-23.09	16.38	.08693
XOH-$Am(OH)_3^o$	-14.67	16.56	.09061	-14.22	16.33	.08587	-14.46	16.32	.08568
XOH_2-$Am(OH)_3^+$	-6.558	15.99	.08035	-5.742	16.22	.08419	-8.322	14.39	.06329
XO-$AmCO_3^o$	-12.91*	28.99	.05794	-12.45	31.40	.05328	-15.10	40.16	.03633
XOH-$AmCO_3^+$	-11.05	45.95	.04694	-10.57	45.52	.04305	-8.352	34.32	.06010
XOH_2-$Am(CO_3)_2^o$									
XOH_2-$Am(CO_3)_2^{2-}$	-25.40	49.97	.04923	-25.58	61.34	.03742	-8.434	142.7	.09726
OTHER SPECIES:									

* Abs Error CO_3 (ads)
= 2.0 e-11

A7

See page 2
PM 6/2/94

~~Zirconium~~

6/14/94 PM K tables continued

Americium (III) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: silica A_{sp}: 175 m^2/g Data Source: Righetto 91 Concentration: $[Am^{+3}] = 5.0 \times 10^{-10}$ M	Rel Error (pH): .05 Abs Error (pH): .0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 5.0e-13	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 1.202 g/L, 0.020 mol/L, 1200 ppm SiO_2, [XOH]=8.06e-4

no CO_2 Fig 5	DLM			CCM			TLM		
	Log K_+ = - Log K_- = -7.10			Log K_+ = - Log K_- = -7.05 C = 1.0 F/m^2			Log K_+ = 0.79 Log K_- = -4.79 Log K_{Cu} = -6.22 Log K_{An} = 4.44		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-Am^{2+}	-2.872	21.90	.04680	-2.835	21.14	.04574	-5.656**	31.90	.04857
XO-$AmOH^+$	-9.844	29.34	.07665	-9.787	29.30	.07673	-10.78	27.82	.06542
XO-$Am(OH)_2^0$	-15.76*	16.37	.09901	-15.77*NC	16.25	.09806	-15.15	29.77	.07800
XOH-$Am(OH)_2^+$	-9.844	29.34	.07665	-9.787	29.30	.07673	-12.17	30.86	.08382
XO-$Am(OH)_3^-$	-19.25	36.13	.1032	-19.28	36.16	.1039	-19.31	31.40	.08870
XOH-$Am(OH)_3^0$	-15.76*	16.37	.09856	-15.73**	27.52	.08622	-15.82++NC	2.095	.2239
XOH_2-$Am(OH)_3^+$	-9.844	29.34	.07665	-9.787	29.30	.07673	-11.68++NC	1.886	.2010
XO-$AmCO_3^-$									
XOH-$AmCO_3^0$									
XOH_2-$Am(CO_3)_2^0$									
XOH_2-$Am(CO_3)_3^{3-}$									
OTHER SPECIES:	*: Abs Err	=5e-12	NC : no convergence	**: Abs Err	=1e-12		++: Abs Err	=5e-11	

A8

Np(V)
PM 6/14/94

The tables with the K values for sorption of ~~Am(III)~~ on kaolinite (an aluminosilicate) will be next. Since this mineral has both AlOH + SiOH sites, it was modelled differently from other minerals. In .N1 + .N2, the results from one file using two site types are listed. In .N3, only one site was used (AlOH for the DLM + TLM models, + SiOH for the CCM). The (*) here shows different initial guess values for Log K and the resulting error messages - no convergence was possible.

PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: Kaolinite A_{sp}: 11 m^2/g Data Source: Kohler 92 Concentration: [Np(V)] = 1.25e-7 M	Rel Error (pH): 0.05 Abs Error (pH):0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.2e-10	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 5g/L

Fig 3 AlOH & SiOH (one input file, ext TKO)	DLM - alpha Al_2O_3 values			CCM			TLM		
	Log K_+ = 8.22 Log K_- = -9.62			Log K_+ = 9.08 Log K_- = -8.32			Log K_+ = 6.69 Log K_- = -10.69 Log K_{Cu} = -7.73 Log K_{An} = 9.90		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-NpO_2^0	-4.4983	19.54	1.264	-3.484	19.94	.6580	-2.418	10.35	.04805
XOH-NpO_2^+	4.298	6.923	.1325	5.243	13.84	.1030	3.018	24.72	.03327
XO-NpO_2OH^-	*			*			**		
XOH-NpO_2OH^0	-4.498	19.54	1.264	-3.484	19.94	.6580	-3.230	19.94	.4425
XOH_2-NpO_2OH^+	4.298	6.923	.1325	5.243	13.84	.1030	3.197	16.12	.1012
XOH_2-$NpO_2CO_3^0$									
XOH_2-$NpO_2(CO_3)_2^{2-}$									
XOH_2-$NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$(XO)_2$-NpO_2^-	-8.568	22.12	1.542	-7.450	21.58	1.545	-2.382	9.449	.07034

* -6.8-4 => fatal error - normal equations are singular
4.8-2 => Error #1
1.6 2 => Error #2

** no convergence

A:\BINDING.N1

PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: kaolinite A_{sp}: 11 m^2/g Data Source: Kohler 92 Concentration:[Np(V)] = 1.25e-7 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 5e-11	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 5 g/L

Fig 3 AlOH & SiOH (one input file, ext TKO)	DLM - pyroSiO_2 values			CCM			TLM		
	Log K_+ = Log K_- = -7.10			Log K_+ = Log K_- = -7.05			Log K_+ = Log K_- = Log K_{Cu} = -4.79 Log K_{An} = -6.22		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
XO-NpO_2^0	-2.412	19.54	.03949	-2.824	19.94	.04025	-3.811	10.35	.06268
XOH-NpO_2^+	4.425	6.923	.04500	4.511	13.84	.05998	6.333	24.72	.1481
XO-NpO_2OH^-	*			*			**		
XOH-NpO_2OH^0	-2.912	19.54	.03949	-2.824	19.94	.04025	-2.651	19.94	.03823
XOH_2-NpO_2OH^+	4.425	6.923	.04500	4.511	13.84	.05998	2.455	16.12	.06515
XOH_2-$NpO_2CO_3^0$									
XOH_2-$NpO_2(CO_3)_2^{2-}$									
XOH_2-$NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$(XO)_2$-NpO_2^-	-5.896	22.12	.03481	-5.772	21.58	.03363	-4.134	9.449	.03909

* see .N1

** no convergence

.N2

Seepage 2
PM 6/2/94

Radium

PM 6/15/94

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: kaolinite A_{sp}: 11 m^2/g Data Source: Kohler 92 Concentration: [Np(V)] = 1.25e-7M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.2e-11	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 5 g/L

Fig. 3	DLM - AlOH only			CCM - SiOH only			TLM - AlOH only		
	Log K_+ =6.78 Log K_- =-9.12 (delta-Al_2O_3)			Log K_+ =-6.94 Log K_- =-7.05 (py SiO_2)			Log K_+ = 6.69 Log K_- = -10.69 Log K_{Cu} = -7.73 Log K_{As} = 9.90 (alpha Al_2O_3)		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-3.697	19.91	.03663	-3.365	18.49	.03456.	-5.359	53.23	.02587
$XOH\text{-}NpO_2^+$	3.984	16.11	.03030	2.865	23.63	.02260	3.198	58.35	.02704
$XO\text{-}NpO_2OH^-$	-11.39	23.59	.04197	-9.818	24.00	.04264	-11.89	20.08	.03729
$XOH\text{-}NpO_2OH^o$	-3.697	19.91	.03663	-3.365	18.49	.03456	-3.209	19.51	.03632
$XOH_2\text{-}NpO_2OH^+$	3.984	16.11	.03030	2.815	23.63	.02260	5.482	18.81	.03562
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-7.645	23.93	.04247	-5.739	22.85	.09116	-9.651	45.83	.02496

A:\BINDING.N3

6/17/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

~~Solid:~~ hematite A_{sp}: 40 m^2/g Data Source: Kohler Concentration: [Np(V)] = 1.2e-7 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.0e-10	Ionic Strength (electrolyte): 0.005 M $NaClO_4$ N_s = 2.31 sites/nm^2 1g/L

Fig 2.1 0.005 M	DLM			CCM			TLM		
	Log K_+ = 8.43 Log K_- = -10.93			Log K_+ = 11.45 Log K_- = -6.70			Log K_+ = 4.97 Log K_- = -10.87 Log K_{Cu} = -5.35 Log K_{As} = 12.14		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-3.485	20.16	.02933	-.9289	22.86	.02947	-5.534	73.65	.06462
$XOH\text{-}NpO_2^+$	5.210	38.53	.03160	***			2.950	74.84	.07138
$XO\text{-}NpO_2OH^-$	-12.31	9.281	.03106	*			-10.79	21.38	.02966
$XOH\text{-}NpO_2OH^o$	-3.485	20.16	.02933	-.9289	22.86	.02947	-2.222	23.55	.02952
$XOH_2\text{-}NpO_2OH^+$	5.210	38.53	.03160	7.911	26.54	.02981	6.341	26.60	.03005
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NPO_2^-$	-8.355	8.160	.03200	**			-8.734	7.166	.05676

* Z matrix is singular Error #2
** ITER = 31. GT. IPMAX=30 Error #1
*** 'Converged' BUT V= ??????????
σ = "

.N4

PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: hematite A_{sp}: 40 m^2/g Data Source: Kohler Concentration: [Np(V)] = 1.2e-7 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.0e-10	Ionic Strength (electrolyte): 0.005 M $NaClO_4$ N_s = 2.31 sites/nm^2 1g/L

Fig 2.2 0.01 M	DLM			CCM			TLM		
	Log K_+ = 8.41 Log K_- = -10.91			Log K_+ = 11.43 Log K_- = -6.68			Log K_+ = 4.95 Log K_- = -10.85 Log K_{Cu} = -5.35 Log K_{As} = 12.10		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-3.411	17.83	.03085	-.9546	24.26	.03125	-5.412	77.61	.08477
$XOH\text{-}NpO_2^+$	5.039	47.49	.03495	7.874	29.77	.03177	3.089	78.12	.09112
$XO\text{-}NpO_2OH^-$	-12.05	5.898	.03508	-9.744	19.47	.03115	-10.50	22.10	.03142
$XOH\text{-}NpO_2OH^o$	-3.411	17.83	.03085	-.9307	24.29	.03125	-1.956	25.37	.03134
$XOH_2\text{-}NpO_2OH^+$	5.039	47.49	.03495	7.874	29.77	.03177	.6499	9.318	.04396
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-8.037	5.062	.03705	-3.188	17.57	.03118	-8.361	76.75	.07669

BINDING.N5

6/20/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: hematite A_{sp}: 40 m^2/g Data Source: Kohler Concentration: [Np(V)] = 1.2e-7 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.0e-10	Ionic Strength (electrolyte): 0.05 M $NaClO_4$ N_s = 2.31 sites/nm^2 1g/L

Fig 2.3 0.05M	DLM			CCM			TLM		
	Log K_+ = 8.37 Log K_- = -10.87			Log K_+ = 11.39 Log K_- = -6.64			Log K_+ = 4.91 Log K_- = -10.81 Log K_{Cu} = -5.35 Log K_{As} = 12.02		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-3.046	14.15	.03187	-6.030	13.62	.02887	**		
$XOH\text{-}NpO_2^+$	5.360	21.30	.02561	8.214	14.46	.02780	**		
$XO\text{-}NpO_2OH^-$	-11.55	17.38	.04063	*			-9.507	13.41	.02961
$XOH\text{-}NpO_2OH^o$	-3.046	14.15	.03187	-6.030	13.62	.02887	-.9642	13.68	.02865
$XOH_2\text{-}NpO_2OH^+$	5.360	21.30	.02561	*			**		
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-7.504	19.57	.04459	*			**		

.N6

* 'Converged' but program found no V or σ values
** use were reported as '??????????' in the output.

See page 2-3
DM 6/2/94

~~Ruthenium~~

DM XX: these had the same problem as the files marked '*'. The FIT2 file was changed to the FIT3DM file + FITEQL re run, with the files previously resulting in? values now giving the arithmetic overflow error.

DM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: hematite A_{sp}: 40 m^2/g Data Source: Kohler Concentration: [Np(V)] = 1.2e-7 M				Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-10			Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 1 g/L		
Fig 2.4 0.1M	DLM			CCM			TLM		
	Log K_+ = 8.35 Log K_- = -10.85			Log K_+ = 11.37 Log K_- = -6.62			Log K_+ = 4.89 Log K_- = -10.79 Log K_{Cat} = -5.35 Log K_{An} = 12.20		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^+$	-2.827	.03814	.04746	-.5261	2.294	.04232	*		
$XOH\text{-}NpO_2^+$	5.325	12.36	.03635	8.254	4.048	.04084	*		
$XO\text{-}NpO_2OH^-$	-11.12	3.644	.06245	-9.309	1.155	.04381	*		
$XOH\text{-}NpO_2OH^0$	-2.827	.03814	.04746	-.5261	2.294	.04232	*		
$XOH_2\text{-}NpO_2OH^+$	5.325	12.36	.03636	8.254	4.048	.04084	*		
$XOH_2\text{-}NpO_2CO_3^0$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-7.074	7.354	.07215	-2.767	.8200	.04442	*		

.N7

None of the TLM files converged. The * indicates one of 3 errors:

DM
1)

```
VALUES OF ADJUSTABLE PARAMETERS AT EACH ITERATION: LOG K, T, LOG X
  I   V(Y): SOS/DF        3202

  0                  -5.000E+00
NDP error - arithmetic overflow (See Section 4.2.1 in Lahey Programmer's
 Reference) in surfx
Called by solvex
Called by fiteql2
```


This is seen in the XOHNP02.024 output file.

DM
2)

```
VALUES OF ADJUSTABLE PARAMETERS AT EACH ITERATION: LOG K, T, LOG X
  I   V(Y): SOS/DF        3206

  0                  6.500E+00          6.928E+01
  1                  7.243E+00
NDP error - divide by zero (See Section 4.2.1 in Lahey Programmer's Reference)
 in simq
Called by solvex
Called by fiteql2
```

This is seen in the XOH2NO21.024 output file.

DM
3)

```
VALUES OF ADJUSTABLE PARAMETERS AT EACH ITERATION: LOG K, T, LOG X
  I   V(Y): SOS/DF        3203

  0                  -5.000E+00
Invalid argument value for LOG10 function (See Section 11.2 in Lahey Language
 Reference) in solvex
Called by fiteql2
```

This is seen in the 2XONPO2.024 output file.

6/21/94 DM

DM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: gamma (γ) alumina A_{sp}: 120 m^2/g Data Source: Righetto et al Concentration: [Np(V)] = 1e-14 M				Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-17			Ionic Strength (electrolyte): .01 M $NaClO_4$ N_s = 2.31 sites/nm^2		
fig 1.3 C_s = .2 g/L	DLM			CCM			TLM		
	Log K_+ = 6.78 Log K_- = -9.12			Log K_+ = 6.92 Log K_- = -9.00			Log K_+ = 6.29 Log K_- = -10.29 Log K_{Cat} = -7.81 Log K_{An} = 8.11		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^+$	-3.540	.009068	.1388	-3.564	.05874	.1381	x		
$XOH\text{-}NpO_2^+$	*			4.489	.0049988	.2996	x		
$XO\text{-}NpO_2OH$	*			x			x		
$XOH\text{-}NpO_2OH^0$	-3.540	.009076	.1388	-3.564	005874	.1381	x		
$XOH_2\text{-}NpO_2OH^+$	*			4.489	.004988	.2993	x		
$XOH_2\text{-}NpO_2CO_3^0$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$(XO)_2NpO_2$	*			x			x		
	*: no	convergence		x: no	convergence				
	abs err =	1e-16		abs err =	1e-15				

.N8

6/22/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: natl. goethite A_{sp}: 50 m^2/g Data Source: Nakayama & Sakamoto 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 6e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.1 mass/volume ratio 1g/L	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{+a} = -7.64 Log K_{Na} = 8.56		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-3.082	13.85	.02637	-3.139	12.80	.02576	-5.314	78.62	.02906
$XOH\text{-}NpO_2^+$	4.525	.01041	.6925	4.157	2.943	.02074	2.315	84.16	.03808
$XO\text{-}NpO_2OH^-$	-10.90	20.52	.03151	-10.46	19.98	.03134	-10.67	9.472	.02887
$XOH\text{-}NpO_2OH^o$	-3.082	13.85	.02637	-3.139	12.80	.02576	-2.984	12.43	.02548
$XOH_2\text{-}NpO_2OH^+$	4.661	5.012	.02141	4.157	2.943	.02074	5.093	6.789	.02694
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-7.042	21.23	.03284	-6.657	20.29	.03229	-8.901	66.06	.02399

.N9

6/23/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: nat. hematite A_{sp}: 2.0 m^2/g Data Source: Nakayama & Sakamoto Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.2 mass to volume ratio 1 g/L	DLM			CCM			TLM		
	Log K_+ = 8.35 Log K_- = -10.85			Log K_+ = 11.37 Log K_- = -6.62			Log K_+ = 4.89 Log K_- = -10.79 Log K_{+a} = 5.35 Log K_{Na} = 11.98		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-4.174	55.54	.02015	*NC			*NC		
$XOH\text{-}NpO_2^+$	5.734	41.27	.02243	"			"		
$XO\text{-}NpO_2OH$	-13.31	56.87	.03371	"			"		
$XOH\text{-}NpO_2OH^o$	-4.174	55.54	.02015	"			"		
$XOH_2\text{-}NpO_2OH^+$	5.734	41.27	.02243	"			"		
$XOH_2\ NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-7.830	55.63	.04486	"			"		

* NC: no convergence, same errors as seen in .N7 *'d files

.N10

See pgs 126-127 of this notebook for the .N7 table + errors.

JM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: nat. magnetite A_{sp}: 5 m^2/g Data Source: Nakayama & Sakamoto 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.3 mass to volume ratio 1 g/L	DLM			CCM			TLM		
	Log K_+ = 6.61 Log K_- = -6.26			Log K_+ = 6.26 Log K_- = -7.32			Log K_+ = 4.59 Log K_- = -8.59 Log K_{+a} = -5.47 Log K_{Na} = 7.73		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	-3.503	53.02	.02037	-4.065	49.09	.02017	-2.302	40.24	.01448
$XOH\text{-}NpO_2^+$	3.387	55.46	.02230	3.297	43.18	.02215	4.905	37.61	.02815
$XO\text{-}NpO_2OH^-$	-10.52	53.36	.02163	-11.25	52.37	.02255	-8.926	33.66	.03258
$XOH\text{-}NpO_2OH^o$	-3.503	53.02	.02037	-4.065	49.09	.02017	-3.742	49.46	.02022
$XOH_2\text{-}NpO_2OH^+$	3.387	55.46	.02230	3.297	43.18	.02215	4.363	31.85	.03114
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-4.778	52.79	.02400	-6.140	50.40	.02478	-2.859	34.46	.02995

BINDING.N11

6/24/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: nat biotite $(K(Mg,Fe)_3AlSi_3O_{10}(OH)_2)$ A_{sp}: 8 m^2/g Data Source: Nakayama & Sakamoto 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.4 C_s = 1 g/L		DLM (α-Al_2O_3)			CCM			TLM		
		Log K_+ = 8.22 Log K_- = -9.62			Log K_+ = 9.08 Log K_- = -8.32			Log K_+ = 6.69 Log K_- = -10.69 Log K_{+a} = -7.73 Log K_{Na} = 9.90		
		Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}NpO_2^o$	SiOH	-4.252	39.72	.03627	-4.210	40.32	.04220	-3.109	40.09	.04907
	AlOH	-3.695		.04393	-2.887		.07324	-.2984		.05358
$XOH\text{-}NpO_2^+$	SiOH	2.861	44.42	.06008	3.097	36.99	.04052	1.113	41.67	.04387
	AlOH	4.148		.03401	4.791		.04147	4.250		.03143
$XO\text{-}NpO_2OH$	SiOH	-11.58	43.67	.02549	*NC			*NC		
	AlOH	-12.39		.1662						
$XOH\text{-}NpO_2OH^o$	SiOH	-4.252	39.72	.03627	-4.210	40.32	.04219	-4.243	41.37	.05159
	AlOH	-3.695		.04393	-2.888		.07325	-2.446		.1004
$XOH_2\text{-}NpO_2OH^+$	SiOH	2.861	44.42	.06008	3.097	36.99	.04052	.4619	40.10	.05492
	AlOH	4.148		.03401	4.791		.04147	2.578		.04964
$XOH_2\text{-}NpO_2CO_3^o$										
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$										
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$										
OTHER SPECIES:										
$2(XO)Np_2^-$	SiOH	-6.339	39.50	.02742	-6.288	40.90	.03257	-2.522	34.08	.04561
	AlOH	-6.001		.09952	-3.973		.1680	3.002		.1027

*NC no convergence
Abs error = 6.0e-7 !

.N12 pg1

.N12 pg2

Sorption on natural biotite (results seen in table .N12) was modelled using 2 sites in each file, SiOH sites + AlOH sites. Log K_+, K_-, K_{cat} + K_{an} values are α-Al_2O_3 values.

PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: syn hematite A_{sp}: 5.9 m^2/g (FROM THE ARTICLE) Data Source: Nakayama & Sakamoto 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.5	DLM			CCM			TLM		
	Log K_+ = 8.35 Log K_- = -10.85			Log K_+ = 11.37 Log K_- = -6.62			Log K_+ = 4.89 Log K_- = -10.79 Log K_{cat} = -5.35 Log K_{An} = 11.98		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}NpO_2^o$	-4.753	38.62	.02581	-2.110	37.01	.02541	-3.051	54.59	.01585
$XOH\text{-}NpO_2^+$	4.772	26.36	.01870	6.941	34.44	.02428	5.545	59.92	.02081
$XO\text{-}NpO_2OH^-$	-14.27	46.78	.03346	-11.14	39.09	.02678	NC*		
$XOH\text{-}NpO_2OH^o$	-4.753	38.62	.02581	-2.110	37.01	.02541	-2.168	36.83	.02536
$XOH_2\text{-}NpO_2OH^+$	4.772	26.36	.01870	6.941	34.44	.02428	NC**		
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-10.39	46.85	.03431	-4.618	38.95	.02730	-4.917	49.17	.02040

* NDP error - ~~with~~ log10 } NC no convergence .N13
**NDP error - ÷ 0

PM 6/24/94

6/27/94 PM

PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: syn magnetite A_{sp}: 5.0 m^2/g Data Source: Nakayama et al 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide) 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.6	DLM			CCM			TLM		
	Log K_+ = 6.61 Log K_- = -6.26			Log K_+ = 6.26 Log K_- = -7.32			Log K_+ = 4.59 Log K_- = -8.59 Log K_{cat} = -5.47 Log K_{An} = 7.73		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}NpO_2^o$	-3.371	47.68	.01964	-3.924	47.93	.02034	-2.271	33.70	.01393
$XOH\text{-}NpO_2^+$	3.497	44.43	.02006	3.448	35.16	.02056	4.808	42.30	.02842
$XO\text{-}NpO_2OH^-$	-10.38	56.06	.02299	-11.09	54.42	.02370	-8.732	23.03	.03145
$XOH\text{-}NpO_2OH^o$	-3.371	47.68	.01964	-3.924	47.93	.02034	-3.599	48.94	.02055
$XOH_2\text{-}NpO_2OH^+$	3.497	44.43	.02006	3.448	35.16	.02056	4.547	21.27	.03036
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-4.602	48.60	.02352	-5.947	50.19	.02530	-2.861	31.36	.02935

BINDING.N14

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: lepidocrocite (gamma-AlOOH) A_{sp}: 175 m^2/g Data Source: Nakayama et al 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 4.1 mass to volume ratio 1 g/L NOTE: lep's formula is taken from the article graph, NOT from the text where it is listed as γ-FeOOH	DLM (α-Al_2O_3 values)			CCM			TLM		
	Log K_+ = 8.22 Log K_- = -9.62			Log K_+ = 9.08 Log K_- = -8.32			Log K_+ = 6.69 Log K_- = -10.69 Log K_{cat} = -7.73 Log K_{An} = 9.90		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}NpO_2^o$	-3.176	19.33	.05548	-2.663	14.94	.04656	-5.426	53.75	.02467
$XOH\text{-}NpO_2^+$	5.043	9.108	.03759	5.829	12.76	.04341	3.213	59.42	.02736
$XO\text{-}NpO_2OH^-$	-11.58	26.89	.07386	-11.15	16.98	.04967	-11.46	13.95	.04860
$XOH\text{-}NpO_2OH^o$	-3.176	19.33	.05548	-2.663	14.94	.04656	-2.896	14.76	.04626
$XOH_2\text{-}NpO_2OH^+$	5.043	9.108	.03759	5.829	12.76	.04341	5.812	11.53	.04463
$XOH_2\text{-}NpO_2CO_3^o$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-8.241	28.70	.08228	-7.173	17.57	.05100	-10.21	46.63	.02675

BINDING.N15

6/28/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: boehmite (γ FeOOH) A_{sp}: 36 m^2/g Data Source: Nakayama et al 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 4.2 mass to volume ratio 1g/L NOTE: Chemical formula of boeh. taken from graph, not text, which uses γ-AlOOH	DLM (ferrihydrite values) Log K_+ = 7.18 Log K_- = -8.82			CCM (goethite values) Log K_+ = 6.47 Log K_- = -9.03			TLM (goethite values) Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}NpO_2^+$	-3.516	34.67	.03589	-3.585	34.44	.03576	-4.394	40.41	.02170
$XOH\text{-}NpO_2^+$	4.327	30.58	.03067	3.936	32.63	.02809	3.598	38.99	.02610
$XO\text{-}NpO_2OH^-$	-11.30	37.78	.03999	-11.00	38.70	.04175	-11.14	32.42	.04216
$XOH\text{-}NpO_2OH^0$	-3.519	34.68	.03589	-3.585	34.44	.03576	-3.426	34.31	.03559
$XOH_2\text{-}NpO_2OH^+$	4.331	30.55	.03062	3.936	32.63	.02809	4.698	30.90	.03950
$XOH_2\text{-}NpO_2CO_3^0$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-6.948	36.66	.03920	-7.062	38.76	.04287	-7.820	40.18	.02891

BINDING.N16

6/29/94 PM

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: α-Al_2O_3 A_{sp}: 12 m^2/g Data Source: Nakayama et al 91 Concentration: [Np(V)] = 6e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 4.3 mass to volume ratio 1g/L	DLM Log K_+ = 8.22 Log K_- = -9.62			CCM Log K_+ = 9.08 Log K_- = -8.32			TLM Log K_+ = 6.69 Log K_- = -10.69 Log K_{Cat} = -7.73 Log K_{An} = 9.90		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}NpO_2^0$	-4.974	42.86	.02171	-4.360	42.33	.02165	-4.703	36.99	.01965
$XOH\text{-}NpO_2^+$	4.108	38.09	.02139	4.442	39.50	.02147	4.349	34.95	.03059
$XO\text{-}NpO_2OH^-$	-14.09	49.43	.02447	-13.15	45.17	.02269	-12.78	32.99	.03522
$XOH\text{-}NpO_2OH^0$	-4.974	42.86	.02171	-4.360	42.33	.02165	-4.589	42.32	.02166
$XOH_2\text{-}NpO_2OH^+$	4.108	38.09	.02139	4.442	39.50	.02147	4.784	30.01	.03234
$XOH_2\text{-}NpO_2CO_3^0$									
$XOH_2\text{-}NpO_2(CO_3)_2^{2-}$									
$XOH_2\text{-}NpO_2(CO_3)_3^{4-}$									
OTHER SPECIES:									
$2(XO)NpO_2^-$	-9.537	48.49	.02522	-7.996	44.61	.02361	-8.084	29.92	.03186

BINDING.N17

Neptunium (V) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: silica A_{sp}: 175 m^2/g Data Source: Righetto 91 Concentration: $[Np^{+5}]$=1e-14	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.0e-15	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 1.202 g/L, 0.020 mol/L, 1200 ppm SiO_2, [XOH]=8.07e-4

fig. 5 no CO_2 FIT3 used for batch run file XOHNPO21.c51 resulted in error #1 and didn't converge; all other files ran for 90 iterations and did not converge.	DLM Log K_+ = - Log K_- = -7.10			CCM Log K_+ = - Log K_- = -7.05 C = 1.0 F/m^2			TLM Log K_+ = 0.79 Log K_- = -4.79 Log K_{Cat} = -6.22 Log K_{An} = 4.44		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$

BNDING.N18

1988 paper
PM 11/15/94

Thorium (IV) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: gamma (γ) alumina A_{sp}: 120 m^2/g Data Source: Righetto et al Concentration: [Th(IV)] = 1e-11	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 8.0e-15	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2

fig 1.1 C_s = .01 g/L	DLM (γ Al_2O_3 values) Log K_+ = 6.78 Log K_- = -9.12			CCM Log K_+ = 6.92 Log K_- = -9.00			TLM Log K_+ = 6.29 Log K_- = -10.29 Log K_{Cat} = -7.81 Log K_{An} = 8.11		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}Th^{3+}$	12.73	31.44	.04253	16.67	48.24	.06943	.3606	19.26	.02043
$XO\text{-}ThOH^{2+}$	8.013	44.33	.06125	10.11	50.15	.07500	-2.242	19.49	.03190
$XO\text{-}Th(OH)_2^+$	1.980*	2.420	.2555	3.543	51.91	.08132	-5.046	36.45	.04881
$XOH\text{-}Th(OH)_2^{2+}$	8.013	44.33	.06125	10.11	50.11	.07518	2.562	34.53	.04615
$XO\text{-}Th(OH)_3^0$	-1.614	56.76	.1037	-3.031	53.40	.08625	-7.860	47.02	.06704
$XOH\text{-}Th(OH)_3^+$	*NC			3.543	51.91	.08132	-.2547	45.72	.06457
$XO\text{-}Th(OH)_4^-$	-8.179	61.84	.1525	-9.616	54.70	.09207	-10.76	53.80	.08804
$XOH\text{-}Th(OH)_4^{2-}$	-1.614	56.76	.1037	-3.031	53.40	.08625	**		
$XOH_2\text{-}Th(OH)_4^+$	1.981*	2.420	.2557	2.317*	2.398	.2543	4.492	52.26	.08197
OTHER SPECIES:									
	*: Abs	error =	1e-12						
	NC: No	converge	nce						
	**: The	file had	no out-	put					

BNDING.T1

**: The file had no runtime errors, but also no output. I couldn't find any missing/extra spaces or lines in the input file, which can also cause such a result.

6/30/94 PM

Thorium (IV) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: silica A_{sp}: 175 m^2/g Data Source: Righetto 91 Concentration: $[Th^{+4}]$ = 1e-11	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1e-12	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 0.060 g/L, 0.001 mol/L, 60 ppm SiO_2, [XOH]=6.72e-7

no CO_2 fig. 5.1	DLM (I=0.1 M) Log K_+ = - Log K_- = -7.10			CCM (I=0.1 M) Log K_+ = - Log K_- = -7.05 C = 1.0 F/nm^2			TLM (I=0.1 M) Log K_+ = 0.79 Log K_- = -4.79 Log K_{Cat} = -6.22 Log K_{An} = 4.44		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
XO-Th^{3+}	5.282	1.148	.1317	5.282	1.148	.1317	7.917	1.333	.1824
XO-$ThOH^{2+}$	4.732	1.336	.1826	4.732	1.336	.1826	7.362	1.623	.2269
XO-$Th(OH)_2^+$	4.178	1.624	.2270	4.178	1.624	.2270	6.811	1.843	.2593
XOH-$Th(OH)_2^{2+}$	4.732	1.336	.1826	4.732	1.336	.1826	7.492	1.623	.2269
XO-$Th(OH)_3^0$	3.628	1.844	.2594	3.628	1.844	.2594	6.255	1.984	.2790
XOH-$Th(OH)_3^+$	4.178	1.624	.2270	4.178	1.624	.2270	6.941	1.844	.2594
XO-$Th(OH)_4^-$	-9.783	13.71	.4541	-9.783	1.371	.4541	-9.542	1.373	.4327
XOH-$Th(OH)_4^0$	3.628	1.844	.2594	3.628	1.844	.2594	6.385	1.985	.2790
XOH_2-$Th(OH)_4^+$	4.178	1.624	.2270	4.178	1.624	.2270	7.072	1.844	.2595
OTHER SPECIES:									

BINDING.T2

Note that all Abs error values = 1e-12.

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: Goethite A_{sp}: 50 m^2/g Data Source: Kohler 92 Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 1 g/L

fig. 4.1 pCO_2 = 0	DLM Log K_+ = 7.24 Log K_- = -9.06			CCM Log K_+ = 6.47 Log K_- = -9.03			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
XO-UO_2^+	3.083	9.283	.03464	2.642	10.10	.03407	-3.417	30.19	.02483
XOH-UO_2^{2+}	9.475	11.57	.02758	9.210	13.88	.03299	4.387	32.42	.02878
XO-UO_2OH^-	-3.393	13.37	.04411	-4.000	9.736	.03737	-7.487	12.76	.02840
XOH-UO_2OH^+	3.083	9.283	.03464	2.642	10.10	.03407	.2629	13.19	.02836
XO-$UO_2(OH)_2^-$	-9.875	16.79	.05344	-10.60	10.95	.03983	-11.55	9.583	.03892
XOH-$UO_2(OH)_2^0$	-3.393	13.37	.04411	-4.000	9.736	.03737	-3.908	9.579	.03710
XOH_2-$UO_2(OH)_2^+$	3.083	9.283	.03464	2.642	10.10	.03407	3.847	8.513	.03697
XOH-$UO_2(OH)_3^-$	-9.875	16.79	.05344	-10.60	10.95	.03983	-8.004	14.15	.04841
XOH_2-$UO_2(OH)_3^0$	-3.393	13.37	.04411	-4.000	9.736	.03737	-.3538	14.24	.04669
XOH-$UO_2(OH)_4^{2-}$	-16.36	20.48	.06308	-17.19	12.96	.04245	-12.02	17.76	.06154
XOH_2-$UO_2(OH)_4^-$	-9.875	16.79	.05344	-10.60	10.95	.03983	-4.482	18.77	.05872
XOH-$UO_2CO_3^0$									
XOH_2-$UO_2(CO_3)_2^-$									
XOH_2-$UO_2(CO_3)_3^{3-}$									
XOH_2-$(UO_2)_2CO_3(OH)_3^0$									

BINDING.U1

7/1/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite A_{sp}: 50 m^2/g Data Source: Kohler 92 Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2

fig. 4.2 pCO_2 = 3e-4 atm	DLM Log K_+ = 7.24 Log K_- = -9.06			CCM Log K_+ = 6.47 Log K_- = -9.03			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
XO-UO_2^+	1.029	50.64	.04264	.7203	49.55	.04666	-5.907	84.18	.2175
XOH-UO_2^{2+}	7.213	82.20	.1200	7.112	75.34	.07827	1.793	84.19	.2188
XO-UO_2OH^0	-7.341	66.85	.04853	-7.356	65.49	.04719	-9.811	82.70	.1320
XOH-UO_2OH^+	1.029	50.64	.04264	.7203	49.55	.04666	-2.156	83.17	.1472
XO-$UO_2(OH)_2^-$	-15.86	74.53	.06137	-15.84	75.92	.06566	-15.33	69.60	.05217
XOH-$UO_2(OH)_2^0$	-7.341	66.85	.04853	-7.356	65.49	.04719	-7.233	66.11	.04777
XOH_2-$UO_2(OH)_2^+$	1.029	50.64	.04265	.7203	49.55	.04666	.8535	62.06	.04530
XOH-$UO_2(OH)_3^-$	-15.86	74.53	.06138	-15.84	75.92	.06566	-16.55	78.12	.07588
XOH_2-$UO_2(OH)_3^0$	-7.341	66.85	.04853	-7.356	65.49	.04719	-8.486	78.04	.07510
XOH-$UO_2(OH)_4^{2-}$	-24.33	76.75	.06876	-24.22	77.70	.07304	-25.67	78.66	.08441
XOH_2-$UO_2(OH)_4^-$	-15.86	74.53	.06137	-15.84	75.92	.06566	-17.62	78.65	.08326
XOH-$UO_2CO_3^0$ *	-2.032	18.66	.03886	-2.381	27.77	.03804	-2.275	29.22	.03765
XOH_2-$UO_2(CO_3)_2^-$ **	-5.040	12.70	.04976	-5.598	21.61	.05292	-6.590	61.19	.03722
XOH_2-$UO_2(CO_3)_3^{3-}$ ***	-16.28	8.976	.05418	-16.03	9.113	.05139	-19.59	70.27	.05240
XOH_2-$(UO_2)_2CO_3(OH)_3^0$ ****	-5.758	16.00	.04886	-5.779	15.58	.04863	-6.620	45.01	.03549

* Abs Error CO_3 = 1.0e-9
** Abs Error = 2.0e-9
*** Abs Error = 2.0e-9
**** Abs Error = 0.5e-9

BINDING.U2

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite A_{sp}: 50 m^2/g Data Source: Kohler Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 1 g/L

fig. 4.3 pCO_2 = 0.02 atm	DLM Log K_+ = 7.24 Log K_- = -9.06			CCM Log K_+ = 6.47 Log K_- = -9.03			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
XO-UO_2^+	3.072	39.15	.05431	2.602	39.02	.05432	-3.516	64.12	.04657
XOH-UO_2^{2+}	9.418	54.80	.04713	9.220	45.51	.05105	4.282	65.07	.05381
XO-UO_2OH^0	-3.191	19.44	.05784	-3.894	27.94	.05407	-7.562	55.71	.04877
XOH-UO_2OH^+	3.072	39.15	.05431	2.602	39.02	.05432	.1615	56.89	.05007
XO-$UO_2(OH)_2^-$	-10.62	25.90	.03932	-10.60	6.574	.03766	-11.44	24.54	.05507
XOH-$UO_2(OH)_2^0$	-3.191	19.44	.05784	-3.894	27.94	.05407	-3.806	29.03	.05419
XOH_2-$UO_2(OH)_2^+$	3.072	39.15	.05431	2.602	39.02	.05432	3.907	32.56	.05601
XOH-$UO_2(OH)_3^-$	-10.62	25.90	.03932	-10.60	6.574	.03766	-10.89	53.66	.04117
XOH_2-$UO_2(OH)_3^0$	-3.191	19.44	.05784	-3.894	27.94	.05407	-2.831	49.98	.03911
XOH-$UO_2(OH)_4^{2-}$	-18.90	46.14	.04012	-18.55	34.77	.03290	-20.66	89.57	.07853
XOH_2-$UO_2(OH)_4^-$	-10.62	25.90	.03932	-10.60	6.574	.03766	-12.58	89.37	.07500
XOH-$UO_2CO_3^0$*	-1.550	22.73	.04282	-2.209	28.89	.03931	-2.124	29.78	.03937
XOH_2-$UO_2(CO_3)_2^-$	-6.147	9.446	.05103	-7.142	21.85	.04019	-5.848	56.48	.03925
XOH_2-$UO_2(CO_3)_3^{3-}$	*			*			-19.64	80.48	.03438
XOH_2-$(UO_2)_2CO_3(OH)_3^0$*	-3.224	26.03	.06907	-3.911	31.26	.06636	71.06	21.56	.07041

*: Error #2: Z matrix is singular

.U3

7/5/94 PM

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: magnetite A_{sp}: 5 m^2/g Data Source: Venkataramani 91 Concentration: [U(VI)] = 1e-4 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 3.0e-9	Ionic Strength (electrolyte): 0.1 M $NaClO_4$ N_s = 2.31 sites/nm^2 8 g/L

Fig 2-H1 (black dots)	DLM			CCM			TLM		
	Log K_+ = 6.61 Log K_- = -6.26			Log K_+ = 6.26 Log K_- = -7.32			Log K_+ = 4.59 Log K_- = -8.59 Log K_{Cat} = -5.47 Log K_{An} = 7.73		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.318	21.34	.03353	1.185	22.10	.03496	-4.358	98.03	.05174
$XOH\text{-}UO_2^{2+}$	7.039	65.00	.03628	7.370	55.59	.04000	2.228	98.11	.06716
$XO\text{-}UO_2OH^0$	-4.744	5.164	.03627	-5.198	8.992	.03476	-7.901	87.92	.04168
$XOH\text{-}UO_2OH^+$	1.318	21.34	.03353	1.185	22.10	.03496	-1.477	92.53	.05108
$XO\text{-}UO_2(OH)_2$	-10.65	3.420	.04702	-11.41	15.44	.04259	-11.34	69.53	.05391
$XOH\text{-}UO_2(OH)_2^0$	-4.744	5.164	.03627	-5.198	8.992	.03476	-4.889	9.879	.03397
$XOH_2\text{-}UO_2(OH)_2^+$	1.318	21.34	.03353	1.185	22.10	.03496	1.495	74.52	.05418
$XOH\text{-}UO_2(OH)_3$	-10.65	3.420	.04702	-11.41	15.44	.04259	-8.257	25.23	.05935
$XOH_2\text{-}UO_2(OH)_3^0$	-4.744	5.164	.03627	-5.198	8.992	.03476	-2.576	7.362	.05137
$XOH\text{-}UO_2(OH)_4^{2-}$	-16.51	10.65	.05917	-17.67	31.34	.05032	-11.74	52.56	.09117
$XOH_2\text{-}UO_2(OH)_4$	-10.65	3.420	.04702	-11.41	15.44	.04259	-5.581	10.67	.07353
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2$									
$XOH_2\text{-}UO_2(CO_3)_3^{-3}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

BINDING.U4

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: magnetite A_{sp}: 5 m^2/g Data Source: Venkataramani 91 Concentration: [U(VI)] = 1e-4 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 4.0e-8	Ionic Strength (electrolyte): 0.1 M NaCl N_s = 2.31 sites/nm^2

Fig 2-H2 ** NOTE ** The values from data at 0.1 M and 0.01 M ionic strength were combined in this set, and need to be separated and recalculated.	DLM			CCM			TLM		
	Log K_+ = 6.61 Log K_- = -6.26			Log K_+ = 6.26 Log K_- = -7.32			Log K_+ = 4.59 Log K_- = -8.59 Log K_{Cat} = -5.47 Log K_{An} = 7.73		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$

BINDING.U5

The graph the data taken from has very poorly resolved data points; here the △ + ▽△ symbols were accidentally grouped together. This still needs to be separated + the digitized data checked so the FITEQL files can be created + run.

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: magnetite A_{sp}: 5 m^2/g Data Source: Venkataramani 91 Concentration: [U(VI)] = 1e-4 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 9.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

Fig 2-I dashed-diamond symbols .D2I file extensions	DLM			CCM			TLM		
	Log K_+ = 6.61 Log K_- = -6.26			Log K_+ = 6.26 Log K_- = -7.32			Log K_+ = 4.59 Log K_- = -8.59 Log K_{Cat} = -5.47 Log K_{An} = 7.73		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.354	3.880	.03628	1.236	5.635	.03866	-.7352	61.16	.1056
$XOH\text{-}UO_2^{2+}$	7.726	16.92	.04272	7.970	21.63	.05425	8.395	56.51	.1646
$XO\text{-}UO_2OH^0$	-4.979	1.829	.03675	-5.354	1.501	.03692	-6.773	37.84	.03812
$XOH\text{-}UO_2OH^+$	1.354	3.880	.03628	1.236	5.635	.03866	1.258	42.96	.07898
$XO\text{-}UO_2(OH)_2$	-10.98	1.651	.04989	-11.44	2.304	.05328	-9.834	13.52	.06921
$XOH\text{-}UO_2(OH)_2^0$	-4.979	1.829	.03675	-5.354	1.501	.03692	-5.026	1.373	.03595
$XOH_2\text{-}UO_2(OH)_2^+$	1.354	3.880	.03628	1.236	5.635	.03866	???????????		
$XOH\text{-}UO_2(OH)_3$	-10.98	1.651	.04989	-11.44	2.304	.05328	-7.973	2.965	.07918
$XOH_2\text{-}UO_2(OH)_3^0$	-4.979	1.829	.03675	-5.354	1.501	.03692	-3.133	9.994	.05401
$XOH\text{-}UO_2(OH)_4^{2-}$	-16.82	7.748	.07284	-17.35	11.88	.08328	-10.04	12.94	.1403
$XOH_2\text{-}UO_2(OH)_4$	-10.98	1.651	.04989	-11.44	2.304	.05328	-5.836	4.940	.1023
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2$									
$XOH_2\text{-}UO_2(CO_3)_3^{-3}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.U6

7/6/94 PM

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: magnetite A_{sp}: 5 m^2/g Data Source: Venkataramani 91 Concentration: [U(VI)] = 1e-4 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.1 Abs Error (radionuclide): 6.0e-9	Ionic Strength (electrolyte): 0.01 M $NaNO_3$ N_s = 2.31 sites/nm^2 8 g/L

Fig 2-I diamond symbols extension .d2k files	DLM			CCM			TLM		
	Log K_+ = 6.67 Log K_- = -6.32			Log K_+ = 6.32 Log K_- = -7.38			Log K_+ = 4.65 Log K_- = -8.65 Log K_{Cat} = -5.47 Log K_{An} = 7.85		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.832	13.26	.03777	1.736	14.09	.03924	-3.659	91.54	.05148
$XOH\text{-}UO_2^{2+}$	7.808	32.29	.03262	8.244	26.95	.03940	3.036	92.46	.07897
$XO\text{-}UO_2OH^0$	-4.473	25.70	.05006	-4.809	18.46	.04507	-7.155	50.39	.03371
$XOH\text{-}UO_2OH^+$	1.832	13.26	.03777	1.736	14.09	.03924	-.7435	75.68	.04877
$XO\text{-}UO_2(OH)_2$	.2071	94.10	.04376	-1.073	96.94	.04865	-10.62	26.12	.04910
$XOH\text{-}UO_2(OH)_2^0$	2.718	90.06	.04051	1.995	93.53	.04182	-4.918	18.19	.04433
$XOH_2\text{-}UO_2(OH)_2^+$	5.450	72.50	.03297	5.197	71.75	.03317	2.251	35.92	.04917
$XOH\text{-}UO_2(OH)_3$	-.2837	35.55	.02985	-.9917	55.86	.03004	-8.079	12.38	.06053
$XOH_2\text{-}UO_2(OH)_3^0$	1.832	13.26	.03777	1.736	14.09	.03924	-2.920	32.10	.07195
$XOH\text{-}UO_2(OH)_4^{2-}$	-4.473	25.70	.05006	-4.809	18.46	.04507	-11.12	26.23	.08868
$XOH_2\text{-}UO_2(OH)_4$	2.699	35.37	.07130	-2.757	33.75	.07247	-5.328	17.62	.07671
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2$									
$XOH_2\text{-}UO_2(CO_3)_3^{-3}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

BINDING.U7

BINDING.U8 is a blank table

7/7/94 PM

The BINDING.U9 - U16 tables are based on the Hsi 81 paper, which I don't have a copy of. The tables will be corrected when I have the paper to use as a reference, or corrected as far as possible if I can't get a copy.

7/8/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM 7/8/94

Solid: ferrihydrite
A_{sp}: 600 m^2/g
Data Source: Payne et al 92
Concentration: [U(VI)] = 1e-8 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0 0.10
Rel Error (radionuclide):
Abs Error (radionuclide): 1.0e-11

Ionic Strength (electrolyte): 0.1 $NaNO_3$
N_S = 2.31 sites/nm^2
10^3 M ferrihydrite

PM

fig 1.1	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{Ab} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	2.137	.7472	.05323	2.335	.7867	.05379	-3.470	19.76	.03875
$XOH\text{-}UO_2^{2+}$	8.748	2.140	.04480	9.523	.6121	.05111	4.147	22.82	.03870
$XO\text{-}UO_2OH^0$	-4.471	2.742	.06334	-4.851	1.219	.05662	-7.704	1.494	.04624
$XOH\text{-}UO_2OH^+$	2.137	.7472	.05323	2.335	.7867	.05379	-.08257	2.025	.04525
$XO\text{-}UO_2(OH)_2^-$	-11.08	5.828	.07511	-12.04	1.830	.05954	-11.92	1.799	.05941
$XOH\text{-}UO_2(OH)_2^0$	-4.471	2.742	.06334	-4.851	1.219	.05662	-4.304	1.505	.05808
$XOH_2\text{-}UO_2(OH)_2^+$	.3495	59.40	.0.054	2.335	.7867	.05379	3.315	1.237	.05682
$XOH\text{-}UO_2(OH)_3^-$	-11.08	5.828	.07511	-12.04	1.830	.05954	-8.509	5.582	.07409
$XOH\text{-}UO_2(OH)_4^{2-}$	-4.471	2.742	.06334	-4.851	1.219	.05662	-.8903	5.138	.07257
$XOH_2\text{-}UO_2(OH)_4^-$	-17.68	9.284	.08850	-19.22	2.564	.06260	-13.14	10.86	.09542
$XOH\text{-}UO_2CO_3^0$	-11.08	5.828	.07511	-12.04	1.830	.05954	-5.083	9.915	.09152
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

BINDING.U17

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM 7/8/94

Solid: ferrihydrite
A_{sp}: 600 m^2/g
Data Source: Payne et al 92
Concentration: [U(VI)] = 1e-6 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide): 0.10
Abs Error (radionuclide): 1.0e-10

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_S = 2.31 sites/nm^2
10^3 ferrihydrite

fig 1.2	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{Ab} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	1.968	13.75	.03120	2.169	13.78	.03141	-4.664	73.64	.02303
$XOH\text{-}UO_2^{2+}$	8.333	15.24	.02050	9.322	11.23	.02828	2.850	76.35	.02401
$XO\text{-}UO_2OH^0$	-4.573	23.49	.04380	-4.988	16.53	.03463	-8.004	11.55	.02254
$XOH\text{-}UO_2OH^+$	1.968	13.75	.03120	2.169	13.78	.03141	-.4080	15.60	.02163
$XO\text{-}UO_2(OH)_2^-$	-11.25	31.63	.05885	-12.15	19.22	.03798	-12.02	18.61	.03773
$XOH\text{-}UO_2(OH)_2^0$	-4.573	23.49	.04380	-4.988	16.53	.03463	-4.375	16.45	.03626
$XOH_2\text{-}UO_2(OH)_2^+$	1.968	13.75	.03120	2.169	13.78	.03141	3.293	14.01	.03497
$XOH\text{-}UO_2(OH)_3^-$	-11.25	31.63	.05885	-12.15	19.22	.03798	-9.011	30.24	.05736
$XOH\text{-}UO_2(OH)_4^{2-}$	-4.573	23.49	.04380	-4.988	16.53	.03463	-.8091	25.39	.05243
$XOH_2\text{-}UO_2(OH)_4^-$	-17.98	36.05	.07342	-19.31	21.78	.04153	-13.37	36.98	.07891
$XOH\text{-}UO_2CO_3^0$	-11.25	31.63	.05885	-12.15	19.22	.03798	-5.191	34.22	.07490
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.U18

7/11/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM

Solid: ferrihydrite
A_{sp}: 600 m^2/g
Data Source: Payne et al 92
Concentration: [U(VI)] = 1e-5 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide): 0.10
Abs Error (radionuclide): 1.0e-8

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_S = 2.31 sites/nm^2
10^3 M ferrihydrite

fig 1.3	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{Ab} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	1.499	.3756	.03344	1.686	.4865	.03424	-4.090	53.51	.03169
$XOH\text{-}UO_2^{2+}$	8.207	8.344	.02706	8.933	.5014	.03222	3.670	56.66	.04173
$XO\text{-}UO_2OH^0$	-5.271	5.754	.04217	-5.547	1.733	.03683	-8.150	10.71	.03215
$XOH\text{-}UO_2OH^+$	1.499	.3756	.03344	1.686	.4865	.03424	-.3465	23.53	.04215
$XO\text{-}UO_2(OH)_2^-$	-12.09	13.51	.05226	-12.76	3.191	.03986	-12.48	.6895	.04211
$XOH\text{-}UO_2(OH)_2^0$	-5.271	5.754	.04217	-5.547	1.755	.03633	-4.534	3.293	.04891
$XOH_2\text{-}UO_2(OH)_2^+$	1.499	.3756	.03344	1.686	.4865	.03424	3.462	14.30	.06340
$XOH\text{-}UO_2(OH)_3^-$	-12.09	13.51	.05226	-12.76	3.191	.03986	-9.582	5.485	.05783
$XOH\text{-}UO_2(OH)_4^{2-}$	-5.271	5.754	.04217	-5.547	1.733	.03683	-.7285	3.093	.07046
$XOH_2\text{-}UO_2(OH)_4^-$	-18.93	19.79	.06269	-19.96	4.305	.04323	-14.13	15.36	.07445
$XOH\text{-}UO_2CO_3^0$	-12.09	13.51	.05226	-12.76	3.191	.03986	-4.997	.5273	.07959
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.U19

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM

Solid: Ferrihydrite A_{sp}: 600 m^2/g Data Source: Payne et al 92 Concentration: [U(VI) = 1e-4 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 10^{-3} ferrihydrite

fig 1.4	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.66 Log K_{An} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	.3495	59.40	.03054	.5749	55.60	.02997	-5.887	92.30	.03873
$XOH\text{-}UO_2^{2+}$	6.601	83.54	.03502	7.624	68.77	.03279	1.741	92.74	.04167
$XO\text{-}UO_2OH^0$	-6.064	15.74	.02779	-6.494	38.44	.02790	-9.796	82.07	.03684
$XOH\text{-}UO_2OH^+$	.3495	59.40	.03054	.5749	55.60	.02997	-2.195	84.92	.04085
$XO\text{-}UO_2(OH)_2^-$	-12.79	3.387	.03364	-13.67	31.56	.03007	-13.67	55.25	.03826
$XOH\text{-}UO_2(OH)_2^0$	-6.064	15.74	.02779	-6.494	38.44	.02790	-6.108	69.23	.04525
$XOH_2\text{-}UO_2(OH)_2^+$	.3495	59.40	.03054	.5749	55.60	.02997	1.505	76.72	.05195
$XOH\text{-}UO_2(OH)_3^-$	-12.79	3.387	.03364	-13.67	31.56	.03007	-10.54	39.21	.04971
$XOH_2\text{-}UO_2(OH)_3^0$	-6.064	15.74	.02779	-6.494	38.44	.02790	-2.466	65.93	.06138
$XOH\text{-}UO_2(OH)_4^{2-}$	-19.45	1.696	.04248	-20.86	32.36	.03415	-14.15	37.68	.06355
$XOH_2\text{-}UO_2(OH)_4^-$	-12.79	3.387	.03364	-13.67	31.56	.03007	-6.483	56.23	.07210
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{4-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.U20

7/12/94 PM The Hsi '81 paper was located (it's a thesis paper). Tables .U9 - .U16 can now be corrected.

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM

Solid: goethite A_{sp}: 50 m^2/g Data Source: Hsi & Langmuir 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1g/L

CO_2 (CO_3^{2-}): C_T = 0.001 Fig 64 #1	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	2.821	6.005	.04015	2.366	7.038	.04085	-2.625	68.38	.04901
$XOH\text{-}UO_2^{2+}$	9.539	29.94	.03149	9.160	18.73	.03866	6.661	60.70	.07071
$XO\text{-}UO_2OH^0$	-3.941	1.033	.05601	-4.450	.8349	.04746	-7.418	32.65	.03299
$XOH\text{-}UO_2OH^+$	2.821	6.005	.04015	2.366	7.038	.04085	.8936	34.85	.04336
$XO\text{-}UO_2(OH)_2^-$	-10.48	1.700	.06803	-11.07	.5942	.05377	-11.65	.8955	.05601
$XOH\text{-}UO_2(OH)_2^0$	-3.941	1.033	.05601	-4.450	.8349	.04746	-4.347	.8966	.04683
$XOH_2\text{-}UO_2(OH)_2^+$	2.821	6.005	.04015	2.366	7.038	.04085	3.883	2.062	.05313
$XOH\text{-}UO_2(OH)_3^-$	-10.48	1.700	.06803	-11.07	.5942	.05377	-8.326	.6513	.07019
$XOH_2\text{-}UO_2(OH)_3^0$	-3.941	1.033	.05601	-4.450	.8349	.04746	-.9556	1.372	.06058
$XOH\text{-}UO_2(OH)_4^{2-}$	-16.97	2.224	.08084	-17.61	.5583	.05968	-11.79	.4711	.1001
$XOH_2\text{-}UO_2(OH)_4^-$	-10.48	1.700	.06803	-11.07	.5942	.05377	**		
$XOH\text{-}UO_2CO_3^0$	15.29	7.681	.03612	14.72	10.50	.03110	14.82	10.92	.03076
$XOH_2\text{-}UO_2(CO_3)_2^-$	*			26.65	17.89	.02530	28.08	78.54	.02415
$XOH_2\text{-}UO_2(CO_3)_3^{5-}$	3.400	5.036	.05894	33.23	9.990	.04341	***		
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$	12.12	4.162	.05856	11.64	5.662	.05331	14.82	2.171	.06918

BINDING.U9

*: NDP ÷ 0 error
**: NDP invalid log10 fn error
***: NDP arithmetic overflow error

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM

Solid: goethite A_{sp}: 50 m^2/g Data Source: Hsi & Langmuir 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH):0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

CO_2: C_T = 0.01 Fig 64 #2 (65)	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	5.866	5.431	.04567	5.608	6.466	.04917	6.267	10.24	.07979
$XOH\text{-}UO_2^{2+}$	14.14	5.886	52.50	13.72	7.298	.05982	15.17	6.783	.09845
$XO\text{-}UO_2OH^0$	-2.330	4.264	.04139	2.377	4.403	.04158	2.180	9.237	.05517
$XOH\text{-}UO_2OH^+$	5.866	5.431	.04567	5.608	6.466	.04917	6.213	7.069	.06464
$XO\text{-}UO_2(OH)_2^-$	-10.44	1.979	.03872	-10.22	2.160	.03707	-9.807	2.625	.05291
$XOH\text{-}UO_2(OH)_2^0$	2.330	4.264	.04139	-2.377	4.403	.04158	-2.221	4.321	.04139
$XOH_2\text{-}UO_2(OH)_2^+$	5.866	5.431	.04566	5.608	6.466	.04917	6.136	3.120	.05298
$XOH\text{-}UO_2(OH)_3^-$	-10.44	1.979	.03872	-10.22	2.160	.03707	-9.471	15.21	.04103
$XOH_2\text{-}UO_2(OH)_3^0$	2.330	4.264	.04139	-2.377	4.403	.04158	-2.047	11.48	.02971
$XOH\text{-}UO_2(OH)_4^{2-}$	-22.22	2.395	.03790	-21.60	5.794	.03685	-16.03	53.80	.06908
$XOH_2\text{-}UO_2(OH)_4^-$	*Error #1			-14.00	2.227	.03711	-5.036	63.25	.2068
$XOH\text{-}UO_2CO_3^0$	17.53	12.45	.05167	-1.010	13.74	.06210	17.77	9.947	.04062
$XOH_2\text{-}UO_2(CO_3)_2^-$	29.43	12.52	.05189	-6.782	12.03	.05255	30.35	3.246	.03913
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$	33.51	12.32	.06554	-19.07	10.82	.05055	NC		
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$	16.62	10.92	.05937	-3.504	14.42	.09239	16.82	7.230	.04748

.U10

* # of iterations allowed were exceeded
NC no convergence

7/13/94 PM
BINDING.U11 table (shown on next page)
errors:
* unlisted error (output terminates w/out any iterations)
** invalid log10 fn error
*** arithmetic overflow error

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite A_{sp}: 600 m^2/g Data Source: Hsi & Langmuir 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

CO_2: C_T = 0.001 Fig 28	DLM Log K_+ = 7.18 Log K_- = -8.82			CCM Log K_+ = 7.17 Log K_- = -8.84			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.66 Log K_{An} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	4.621	.02343	1.449	4.634	.02366	1.470	7.271	.02279	1.394
$XOH\text{-}UO_2^{2+}$	12.99	.02307	1.413	13.01	.02339	1.442	15.52	.02281	1.412
$XO\text{-}UO_2OH^a$	-3.742	.02317	1.414	-3.736	.02346	1.443	-2.334	.01109	1.398
$XOH\text{-}UO_2OH^+$	4.621	.02343	1.449	4.634	.02366	1.470	5.772	.02288	1.397
$XO\text{-}UO_2(OH)_2^-$	-12.18	.01084	.8520	-12.16	.01084	1.071	-11.67	.02297	1.395
$XOH\text{-}UO_2(OH)_2^a$	-3.742	.02317	1.414	-3.736	.02346	1.443	-3.710	.02356	1.454
$XOH_2\text{-}UO_2(OH)_2^+$	4.621	.02343	1.449	4.634	.02366	1.470	4.397	.02370	1.481
$XOH\text{-}UO_2(OH)_3^-$	-12.18	.01084	.8520	-12.16	.01804	1.071	-10.68	.009100	.8360
$XOH_2\text{-}UO_2(OH)_3^a$	-3.742	.02317	1.414	-3.736	.02346	1.443	-3.056	.009275	.8346
$XOH\text{-}UO_2(OH)_4^{2-}$	-19.55	.007122	.7971	-20.08	.008844	.8190	-15.76	.007962	.8388
$XOH_2\text{-}UO_2(OH)_4$	-12.18	.01084	.8520	-12.16	.01804	1.071	-8.281	.007978	.8216
$XOH\text{-}UO_2CO_3^a$	13.59	56.14	.04082	19.08	.02191	.9745	19.10	.02191	.9745
$XOH_2\text{-}UO_2(CO_3)_2^-$	33.74	.02267	1.034	33.74	.02271	1.038	31.04	.01929	.8383
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$	*			***			***		
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^a$	**			*			15.77	.02200	1.930

BINDING.U11

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite A_{sp}: 600 m^2/g Data Source: Hsi & Langmuir 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

CO_2: C_T = 0.01 Fig 28 #2 (29)	DLM Log K_+ = 7.18 Log K_- = -8.82			CCM Log K_+ = 7.17 Log K_- = -8.84			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.66 Log K_{An} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	1.156	77.89	.06481	1.368	76.73	.06214	-7.616	87.75	2.110
$XOH\text{-}UO_2^{2+}$	6.413	87.41	.02165	8.258	81.98	.07201	-.2075	87.75	2.688
$XO\text{-}UO_2OH^a$	-3.749	20.46	.03662	-5.426	66.01	.05005	-10.05	87.09	.01603
$XOH\text{-}UO_2OH^+$	1.156	77.89	.06481	1.368	76.73	.06213	-2.648	87.38	.02107
$XO\text{-}UO_2(OH)_2^-$	-11.55	12.77	.03001	log 10			-12.91	6.029	.04559
$XOH\text{-}UO_2(OH)_2^a$	-3.749	20.46	.03662	-5.426	66.01	.05005	-5.539	68.25	.05184
$XOH_2\text{-}UO_2(OH)_2^+$	1.156	77.89	.06481	1.368	76.73	.06213	1.893	73.69	.05768
$XOH\text{-}UO_2(OH)_3^-$	-11.55	12.77	.03001	log 10			-12.51	71.01	.03968
$XOH_2\text{-}UO_2(OH)_3^a$	3.749	20.46	.03662	-5.426	66.01	.05005	-4.365	64.13	.03410
$XOH\text{-}UO_2(OH)_4^{2-}$	-19.81	28.21	.02970	-19.89	27.41	.02797	-22.38	80.19	.07390
$XOH_2\text{-}UO_2(OH)_4^-$	-11.55	12.77	.03001	log 10			-14.37	80.13	.07063
$XOH\text{-}UO_2CO_3^a$	19.08	.02192	.09753	12.42	70.19	.04424	12.33	71.14	.04471
$XOH_2\text{-}UO_2(CO_3)_2^-$	*			23.43	67.32	.04649	29.09	9.709	.02896
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$	log 10			log 10			32.61	27.22	.02418
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^a$	12.43	42.69	.05766	10.77	55.97	.06066	log 10		

* Error #2: Z matrix is singular
log 10: invalid log 10 fn error

.U12

7/14/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite A_{sp}: 50 m^2/g Data Source: Hsi & Langmuir 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

Fig. 10, extension .DGO No CO_2 Log K values (+, -, cat and an) from Pabalan & Turner 93	DLM Log K_+ = 7.24 Log K_- = -9.06			CCM Log K_+ = 6.47 Log K_- = -9.03			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	2.796	1.618	.03518	2.359	1.896	.03500	-2.840	44.97	.03995
$XOH\text{-}UO_2^{2+}$	9.416	10.82	.02779	9.115	8.260	.03381	6.125	43.25	.06379
$XO\text{-}UO_2OH^a$	-3.763	3.878	.04323	-4.304	2.036	.03772	-7.45	14.70	.02946
$XOH\text{-}UO_2OH^+$	2.796	1.618	.03518	2.359	1.896	.03500	.6401	18.34	.03845
$XO\text{-}UO_2(OH)_2^-$	-10.30	6.024	.05183	-10.88	2.351	.04109	-11.49	.7515	.04588
$XOH\text{-}UO_2(OH)_2^a$	-3.763	3.878	.04323	-4.304	2.036	.03772	-4.208	1.956	.03747
$XOH_2\text{-}UO_2(OH)_2^+$	2.796	1.618	.03518	2.359	1.896	.03500	3.963	.6632	.04580
$XOH\text{-}UO_2(OH)_3^-$	-10.30	6.024	.05183	-10.88	2.351	.04109	-8.066	2.541	.05304
$XOH_2\text{-}UO_2(OH)_3^a$	-3.763	3.878	.04323	-4.304	2.036	.03772	-.7721	4.917	.04609
$XOH\text{-}UO_2(OH)_4^{2-}$	-16.80	7.340	.06114	-17.39	2.217	.04568	-11.40	1.690	.07339
$XOH_2\text{-}UO_2(OH)_4^-$	-10.30	6.024	.05183	-10.88	2.351	.04109	-4.606	4.804	.06232
$XOH\text{-}UO_2CO_3^a$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^a$									

.U13

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite A_{sp}: 600 m^2/g Data Source: Hsi & Langmuir 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

Fig. 11 extension.DPH No CO_2 Using Log K (+, -, an, cat) from K:\turner\fiteq\table.wp	DLM Log K_+ = 7.18 Log K_- = -8.82			CCM Log K_+ = 7.17 Log K_- = -8.84			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.66 Log K_{An} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	2.178	13.81	.03138	2.373	13.78	.03123	-4.917	80.21	0.02390
$XOH\text{-}UO_2^{2+}$	8.178	25.56	.02209	9.453	15.40	.02783	2.597	82.40	.02587
$XO\text{-}UO_2OH^a$	-3.870	17.05	.04227	-4.664	14.28	.03369	-8.430	25.44	.02340
$XOH\text{-}UO_2OH^+$	2.178	13.81	.03138	2.373	13.78	.03123	-.8620	28.92	.02217
$XO\text{-}UO_2(OH)_2^-$	-9.779	21.42	.05206	-11.70	14.98	.03619	-12.25	14.19	.03449
$XOH\text{-}UO_2(OH)_2^a$	-3.870	17.05	.04227	-4.664	14.28	.03369	-4.728	14.16	.03321
$XOH_2\text{-}UO_2(OH)_2^+$	2.178	13.81	.03138	2.373	13.78	.03123	2.840	13.72	.03223
$XOH\text{-}UO_2(OH)_3^-$	-9.779	21.42	.05206	-11.70	14.98	.03619	-8.535	17.77	.04482
$XOH_2\text{-}UO_2(OH)_3^a$	-3.870	17.05	.04227	-4.664	14.28	.03369	-1.009	17.76	.04360
$XOH\text{-}UO_2(OH)_4^{2-}$	-15.56	23.47	.05752	-18.73	15.75	.03881	-12.21	20.95	.05552
$XOH_2\text{-}UO_2(OH)_4^-$	-9.779	21.42	.05206	-11.70	14.98	.03619	-4.632	21.88	.05249
$XOH\text{-}UO_2CO_3^a$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^a$									

.U14

BINDING.U15 contains K values from Hsi '81,

figure 12. There are 2 complete (all three models) sets of input files using this data.

PM 7/14/94 One ~~file~~ set ext. .D12, .C12 + .T12 uses the $UO_2(NO_3)_2$ species, which is not in the thermodynamic database I'm using. I will leave this set alone, and correct and tabulate the K values found using the other set of files. These files have .DHS, .CHS + .THS extensions.

7/15/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM

Solid: syn. hematite A_{sp}: 40 m^2/g Data Source: ~~HSI~~ 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 5.0e-6	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

Fig 12 No CO_2 ext .DHS	DLM Log K_+ = 8.35 Log K_- = -10.85			CCM Log K_+ = 11.37 Log K_- = -6.62			TLM Log K_+ = 4.89 Log K_- = -10.79 Log K_{Cat} = -5.35 Log K_{An} = 11.98		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	2.419	12.52	.02467	5.261	18.21	.01854	AO*		
$XOH\text{-}UO_2^{2+}$	9.418	46.93	.02407	Err #1*			log10*		
$XO\text{-}UO_2OH^0$	-5.413	9.273	.03629	-3.446	9.696	.02050	log10*		
$XOH\text{-}UO_2OH^+$	2.209	13.36	.02435	5.261	18.21	.01854	AO*		
$XO\text{-}UO_2(OH)_2^-$	-13.11	15.63	.05032	log10*			log10*		
$XOH\text{-}UO_2(OH)_2^0$	-5.413	9.273	.03629	-3.446	6.963	.02050	-3.553	7.209	.02603
$XOH_2\text{-}UO_2(OH)_2^+$	2.209	13.36	.02435	5.261	8.937	.02516	5.401	20.44	.03888
$XOH\text{-}UO_2(OH)_3^-$	-13.11	15.63	.05032	AO*			log10*		
$XOH_2\text{-}UO_2(OH)_3^0$	-5.413	9.273	.03629	-3.446	6.963	.02647	AO*		
$XOH\text{-}UO_2(OH)_4^{2-}$	Err #2			AO*			in surfx*		
$XOH_2\text{-}UO_2(OH)_4^-$	-13.11	15.63	.05032	log10*			log10*		
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

Err #2: Z matrix is singular
Err #1: #iterations allowed is exceeded
log 10: invalid log10 fn
AO: arithmetic overflow
in surfx: unclear error location

* Abs error = 5.0e-6 .015

8/4/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

PM

Solid: natural hematite A_{sp}: 2 m^2/g Data Source: Hsi 81 Concentration: [U(VI)] = 1e-5 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2 1 g/L

Fig. 13, ext. DHN K_+, K_- values found in MINTEQA2 input file = 7.37, -9.32	DLM Log K_+ = 8.35 Log K_- = -10.85			CCM Log K_+ = 11.37 Log K_- = -6.62			TLM Log K_+ = 4.89 Log K_- = -10.79 Log K_{Cat} = -5.35 Log K_{An} = 11.98		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.905	64.35	.04438	5.055	61.31	.03822	-2.967	69.86	.1265
$XOH\text{-}UO_2^{2+}$	9.024	68.76	.07332	in surfx			5.902	69.76	.2025
$XO\text{-}UO_2OH^0$	-5.358	12.07	.03180	-3.552	57.55	.03387	-7.664	68.81	.07420
$XOH\text{-}UO_2OH^+$	1.905	64.35	.04438	5.055	61.31	.03822	.9161	69.03	.1076
$XO\text{-}UO_2(OH)_2^-$	-12.57	28.79	.04610	in solvex			-12.14	66.19	.09698
$XOH\text{-}UO_2(OH)_2^0$	-5.092	31.20	.02952	in solvex			-3.678	58.42	.03438
$XOH_2\text{-}UO_2(OH)_2^+$	-5.092	31.20	.02952	5.055	61.31	.03822	AO		
$XOH\text{-}UO_2(OH)_3^-$	-12.57	28.79	.04610	in solvex			8.458	60.90	.09726
$XOH_2\text{-}UO_2(OH)_3^0$	-5.092	31.20	.02952	-3.552	57.55	.03387	in simq		
$XOH\text{-}UO_2(OH)_4^{2-}$	-19.97	35.16	.06373	-20.96	57.18	.04535	-12.65	64.24	.2021
$XOH_2\text{-}UO_2(OH)_4^-$	-12.57	28.79	.04610	in solvex			in simq		
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

AO: arithmetic overflow error

BINDING.U16

"in surfx" "in solvex", + "in simq" refer to where in the program the error occurred which did not allow further calculation + convergence. These messages were found in the output files of the associated species. I don't know what the actual cause of these errors is.

8/5/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite A_{SP}: 600 m^2/g Data Source: Payne et al 92 Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): .05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.10 Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.1	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	.4398	57.98	.02150	.4660	58.61	.02174	-4.590	74.91	.02643
$XOH\text{-}UO_2^{2+}$	8.384	25.76	.01772	8.787	27.02	.02070	2.922	76.42	.02811
$XO\text{-}UO_2OH^0$	-7.981	76.64	.02904	-7.974	76.33	.02869	-7.921	28.55	.02116
$XOH\text{-}UO_2OH^+$	.4398	57.98	.02150	.4660	58.61	.02174	-.3388	35.34	.02121
$XO\text{-}UO_2(OH)_2^-$	-16.29	80.25	.03405	-16.33	80.43	.03444	-16.31	80.59	.03483
$XOH\text{-}UO_2(OH)_2^0$	-7.981	76.64	.02904	-7.974	76.33	.02869	-8.293	80.01	.03397
$XOH_2\text{-}UO_2(OH)_2^+$	.4398	57.98	.02150	.4660	58.61	.02174	-.2707	79.09	.03288
$XOH\text{-}UO_2(OH)_3^-$	-16.29	80.25	.03405	-16.33	80.43	.03444	-17.58	82.26	.03948
$XOH_2\text{-}UO_2(OH)_3^0$	-7.981	76.64	.02904	-7.974	76.33	.02869	-9.552	82.11	.03923
$XOH\text{-}UO_2(OH)_4^{2-}$	-18.22	14720 *	.07672	-24.61	81.33	.03633	-27.18	84.47	.05421
$XOH_2\text{-}UO_2(OH)_4^-$	-16.29	80.25	.03405	-16.33	80.43	.03444	-19.13	84.33	.05313
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.021

*: This is the output V value, not a typo.

8/8/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrate A_{SP}: 600 m^2/g Data Source: Payne et al 92 Concentration: [U(VI)] = 1e-4 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 3.2	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 (7.28) Log K_- = -8.84 (-8.95)			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	-1.388	97.99	.04470	-1.361	98.19	.04550	-5.505	92.60	.03521
$XOH\text{-}UO_2^{2+}$	6.843	81.26	.02583	6.938	92.70	.03555	2.164	92.89	.04094
$XO\text{-}UO_2OH^0$	-9.539	98.88	.04681	-9.539	98.85	.04672	-9.397	81.76	.03003
$XOH\text{-}UO_2OH^+$	-1.388	97.99	.04470	-1.361	98.19	.04550	-1.747	85.78	.03782
$XO\text{-}UO_2(OH)_2^-$	-17.66	99.00	.04675	-17.70	99.01	.04674	-17.65	99.02	.04718
$XOH\text{-}UO_2(OH)_2^0$	-9.539	98.88	.04681	-9.539	98.85	.04672	-9.680	98.99	.04835
$XOH_2\text{-}UO_2(OH)_2^+$	-1.388	97.99	.04470	-1.361	98.19	.04550	-1.700	98.94	.05034
$XOH\text{-}UO_2(OH)_3^-$	-17.66	99.00	.04675	-17.70	99.01	.04674	-18.33	99.19	.05185
$XOH_2\text{-}UO_2(OH)_3^0$	-9.539	98.88	.04681	-9.539	98.85	.04672	-10.35	99.18	.05322
$XOH\text{-}UO_2(OH)_4^{2-}$	-25.78	99.06	.04692	-25.85	99.08	.04701	-26.97	99.28	.06263
$XOH_2\text{-}UO_2(OH)_4^-$	-17.66	99.00	.04675	-17.70	99.01	.04674	-18.99	99.28	.06323
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.022

The logK values in ( ) were used by Steve Sassman
I'm not sure how they were calculated.

8/9/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite A_{SP}: 600 m^2/g Data Source: Payne et al 92 Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 5.1 TOT Fe = 10-3 M	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	.7797	22.10	.01722	.8386	22.09	.01730	-4.899	77.31	.03080
$XOH\text{-}UO_2^{2+}$	7.608	55.61	.01983	8.380	31.55	.01972	2.700	77.93	.03203
$XO\text{-}UO_2OH^0$	-7.495	61.00	.02097	-7.489	59.68	.02044	-8.867	56.72	.02095
$XOH\text{-}UO_2OH^+$	.7797	22.10	.01722	.8386	22.09	.01730	-1.263	60.90	.02211
$XO\text{-}UO_2(OH)_2^-$	-16.14	78.85	.03421	-16.12	78.07	.03271	-16.15	79.18	.03504
$XOH\text{-}UO_2(OH)_2^0$	-7.495	61.00	.02097	-7.489	59.68	.02044	-8.045	76.67	.03089
$XOH_2\text{-}UO_2(OH)_2^+$	.7797	22.10	.01722	.8386	22.09	.01730	-.06550	72.82	.02740
$XOH\text{-}UO_2(OH)_3^-$	-16.14	78.85	.03421	-16.12	78.07	.03271	-18.45	85.47	.1064
$XOH_2\text{-}UO_2(OH)_3^0$	-7.495	61.00	.02097	-7.489	59.68	.02044	-10.36	85.36	.09893
$XOH\text{-}UO_2(OH)_4^{2-}$	-24.76	83.26	.05270	-24.68	82.36	.04633	-28.77	86.04	.2395
$XOH_2\text{-}UO_2(OH)_4^-$	-16.14	78.85	.03421	-16.12	78.07	.03271	-20.69	86.02	.2275
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.023

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: Ferrihydrite A_{SP}: 600 m^2/g Data Source: Payne et al Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): Abs Error (radionuclide): 1.0e-9	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 5.2 TOT Fe = 2.10-2M	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cu} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	-.3302	39.68	.02498	-.4039	47.81	.02602	-6.514	88.95	.06393
$XOH\text{-}UO_2^{2+}$	6.522	66.11	.02681	7.489	33.98	.02623	1.049	89.21	.06701
$XO\text{-}UO_2OH^0$	-9.161	84.67	.04336	NC*			-9.853	63.48	.02762
$XOH\text{-}UO_2OH^+$	.1854*	9.751	.03414	-.4039	47.81	.02602	-2.317	69.69	.02885
$XO\text{-}UO_2(OH)_2^-$	-17.96	90.65	.1022	-17.95	90.58	.09947	-18.01	90.81	.1108
$XOH\text{-}UO_2(OH)_2^0$	-9.161	84.67	.04336	-8.04*	29.83	.03391	-8.383*	47.92	.03269
$XOH_2\text{-}UO_2(OH)_2^+$	-.3302	39.68	.02498	-.4039	47.81	.02602	-1.695	89.07	.06663
$XOH\text{-}UO_2(OH)_3^-$	-17.96	90.65	.1021	-16.44*	51.42	.03377	-20.21	91.66	.03856
$XOH_2\text{-}UO_2(OH)_3^0$	-9.161	84.67	.04336	-9.158	84.39	.04253	-12.16	91.66	.03707
$XOH\text{-}UO_2(OH)_4^{2-}$	-26.61	91.41	.01854	-26.60	91.38	.01765	-30.04	91.70	.05175
$XOH_2\text{-}UO_2(OH)_4^-$	-16.46*	52.69	.03459	-16.44	51.42	.03377	-22.00	91.69	.5112
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.024

NC: no convergence
*: Abs error (radionuclide) = $1.0e^{-8}$

8/10/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite A_{sp}: 600 m^2/g Data Source: Payne et al 92 Concentration: [U(VI)] = 1e-6 M	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): Abs Error (radionuclide): 1.0e-10	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

fig 6.1 pre-formed $Fe(OH)_3$	DLM Log K_+ = 7.18 Log K_- = -8.82			CCM Log K_+ = 7.17 Log K_- = -8.84			TLM Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.66 Log K_{An} = 8.21		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.952	11.78	.03036	2.154	11.74	.03056	-4.697	57.77	.02307
$XOH\text{-}UO_2^{2+}$	8.323	15.55	.01981	9.304	9.690	.02754	2.813	78.33	.02408
$XO\text{-}UO_2OH^0$	-4.568	20.12	.04237	-5.000	14.05	.03362	-8.016	11.71	.02174
$XOH\text{-}UO_2OH^+$	1.952	11.78	.03036	2.154	11.74	.03056	-4.185	16.59	.02087
$XO\text{-}UO_2(OH)_2^-$	-11.20	27.43	.05684	-12.15	16.33	.03681	-12.03	15.83	.03657
$XOH\text{-}UO_2(OH)_2^0$	-4.568	20.12	.04237	-5.000	14.05	.03362	-4.389	14.06	.03520
$XOH_2\text{-}UO_2(OH)_2^+$	1.952	11.78	.03036	2.154	11.74	.03056	?		
$XOH\text{-}UO_2(OH)_3^-$	-11.20	27.43	.05684	-12.15	16.33	.03681	-8.501	24.44	.05281
$XOH_2\text{-}UO_2(OH)_3^0$	-4.568	20.12	.04237	-5.000	14.05	.03362	-.8051	21.82	.05051
$XOH\text{-}UO_2(OH)_4^{2-}$	-17.89	31.61	.07123	-19.31	18.54	.04017	-12.82	31.49	.07405
$XOH_2\text{-}UO_2(OH)_4^-$	-11.20	27.43	.05684	-12.15	16.33	.03681	-5.116	29.86	.07201
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_3CO_3(OH)_3^0$									

? = output for this file
.U25

?: see this notebook, pgs 125-126 for previous ? error.

8/11/94
DRJ

Sorption Modeling for HLW Performance Assessment - Task 2

Objectives:

This scientific notebook contains the results of sorption modeling of radionuclide sorption using surface complexation modeling techniques. The objectives are to determine the model parameters necessary to reproduce the observed sorption behavior. The results are computer-based; the data used to determine the necessary model parameters are from the peer-reviewed with data sources as noted. Experimental conditions for the different data sources are noted as necessary. A basic understanding of chemical principles and DOS-based computer operation are the minimum requirements for these modeling efforts.

The computer equipment (hardware) includes:

Digitizing tablet for converting graphical data to numerical values
IBM Personal computers.

General DOS/Windows computer codes include:

Spreadsheets (Quattro, Quattro Pro, Lotus)
Graphics programs (Sigma Plot, Surfer)
Geochemical codes (EQ3/6, MINTEQA2, EQMOD)
Geochemical Parameter Estimation Code (FITEQL)

Potential sources of uncertainty and error include:

Digitization error
Experimental error/uncertainty
Conceptual model uncertainty/error
Thermodynamic data uncertainty/error

8/11/44 DRJ

Sample calculations for determining Best-estimate Log K values and 95% confidence limits. All subsequent spreadsheet calculations done in this manner

					Computation of Confidence Limits								
			TLM			DLM			CCM			TLM	
Log K	sum1/sig K	w.f.	w.f.*Log K	Log K	Sum wf*dif	Sx	conflim	Sum wf*dif	Sx	conflim	Sum wf*dif	Sx	conflim
1.70504	104.6963	0.475196	-1.90078	-4.05773	0.198545	0.37717	5.519491	0.041992	0.081124	2.559797	0.001584	0.003018	0.493694
.018119	97.95489	0.468293	1.938732	4.065561	0.020119	0.037877	1.74912	0.021113	0.039859	1.794291	0.002595	0.00488	0.627847
8.29569	86.11111	0.483871	-4.4129	-9.44	0.626754	1.117395	9.500225	0.063136	0.126025	3.190495	0.049548	0.096	2.784621
1.70504	89.21475	0.481069	-0.62539	-1.58541	0.198545	0.37717	5.519491	0.041992	0.081124	2.559797	0.039188	0.075517	2.469743
.018119	92.55697	0.47806	3.126513	6.284249	0.020119	0.037877	1.74912	0.021113	0.039859	1.794291	0.031269	0.059909	2.199774
		0.524804	-2.15695		0.178624			0.039133			0.001434		
		0.531707	2.126829		0.017758			0.018745			0.002285		
		0.516129	-5.0271		0.490641			0.062889			0.046452		
		0.518931	-0.96002		0.178624			0.039133			0.036329		
		0.52194	3.157737		0.017758			0.018745			0.02864		

nd Morel (1990)

88.2153

Best-Fit Log K = (0.4824)(-1.41) + (0.5176)(-1.98) = -1.705

-1 = [0.4824(-1.41 - (-1.705))² + 0.5176(-1.98 - (-1.705))²]/2-1 = 0.08112

4

2 values), the 95% confidence limit calculated using the students t-distribution

)(0.2014) = 2.5548 ✓

Dzombak (1986)

DRJ 8/11/94

~~8/14/94~~ DRJ

Additional Sample Calculations: ~~Enthalpies and Molecular Weights~~ Surface Charge to Total H⁺ (TOTH) 8/11/94 DRJ

type of solid: HYDROUS FERRIC OXIDE
source of data: YATES (1975)

IONIC STRENGTH (mol/L) = .01

SURFACE AREA (sq.m/g) = 257
SOLID CONC (g/L) = 1.4

DRJ

pH	SURFACE CHARGE uC/sq.cm	SURFACE CHARGE ueqv/g	SURFACE CHARGE mol/L	TOTH mol/L
4.025	30.644	816.245	1.143E-03	1.237E-03
4.308	28.038	746.831	1.046E-03	1.095E-03
4.621	24.731	658.744	9.222E-04	9.462E-04
5.041	20.889	556.407	7.790E-04	7.831E-04
5.344	18.412	490.429	6.866E-04	6.911E-04
5.736	14.903	396.962	5.557E-04	5.576E-04
6.220	10.815	288.072	4.033E-04	4.039E-04
6.726	7.465	198.841	2.784E-04	2.785E-04
7.236	4.497	119.784	1.677E-04	1.675E-04
7.830	1.165	31.031	4.344E-05	4.263E-05
8.185	-1.115	-29.700	-4.158E-05	-4.346E-05
8.419	-2.836	-75.541	-1.058E-04	-1.090E-04
8.892	-6.168	-164.293	-2.300E-04	-2.396E-04
9.695	-13.672	-364.172	-5.098E-04	-5.708E-04

← Sample Hand Calc. (see next page)

This example problem is from Dzombak (1986) and will serve as a check for the spreadsheet calculations. See next pg. (151)

8/11/94 DRL
~~8/14~~

25 Deg C; No CO2					Surface Charge		TOTH
Area(m2/g)=	257	Solid(g/l)=	1.4				
		I.S.	pH	(uC/cm2)	ueqiv/g	(mol/liter)	(mol/liter)
		0.01	4.025	30.644	816.244995	1.1427e-03	1.2371e-03
		0.01	4.308	28.038	746.830609	1.0456e-03	1.0948e-03
		0.01	4.621	24.731	658.744125	9.2224e-04	9.4617e-04
		0.01	5.041	20.889	556.407182	7.7897e-04	7.8807e-04
		0.01	5.344	18.412	490.428888	6.8660e-04	6.9113e-04
		0.01	5.736	14.903	396.961857	5.5575e-04	5.5758e-04
		0.01	6.22	10.815	288.072367	4.0330e-04	4.0388e-04
		0.01	6.726	7.465	198.84052	2.7838e-04	2.7850e-04
		0.01	7.236	4.497	119.783767	1.6770e-04	1.6754e-04
		0.01	7.83	1.165	31.0313738	4.3444e-05	4.2627e-05
		0.01	8.185	-1.115	-29.699555	-4.1579e-05	-4.3456e-05
		0.01	8.419	-2.836	-75.540752	-1.0576e-04	-1.0898e-04
		0.01	8.892	-6.168	-164.29314	-2.3001e-04	-2.3960e-04
		0.01	9.695	-13.672	-364.17248	-5.0984e-04	-5.7078e-04

Converting Surface Charge ($\mu C/cm^2$) to Total H+ (TOTH in mol/L):

$\mu C/cm^2$ to $\mu eq/g$:

(30.644 $\mu C/cm^2$)*(257 m^2/g)*(10000 cm^2/m^2)*(1/F) = 816.245 $\mu eq/g$ ✓

where the Faraday Constant, F = 96484.61 $\mu C/\mu eq$

$\mu eq/g$ to moles charge/L:

(816.245 $\mu eq/g$)*(10^{-6} moles/μeq)*(1.4 g/L) = 0.0011427 ✓moles charge/L

moles charge/L to TOTH (in mol/L):

$0.0011427 + 10^{-4.025} - 10^{-(14 - (4.025 + \log\gamma))} = 0.0012371$ ✓ mol H+/L

where log γ is the activity coefficient for the ionic strength (I.S.), calculated using the Davies Equation such that

$\log\gamma = [2(-0.508 *\{[\text{SQRT}(0.01)]/[1 + \text{SQRT}(0.01)] - 0.24*(0.01)\} = -0.09$

Sample calculation matches spreadsheet, and also matches output of Dzombak (1986) from previous page.

8/11/94 DRL

Additional Sample Calculations - Calculating ΔH_f^0 and Molecular Weight

		Americium																
				M.W.Am+3	M.W.Am+4	M.W.AmO2+1	M.W.AmO2+2	M.W.E-1	M.W.H2O	M.W.H+1	M.W.F-1	M.W.Cl-1	M.W.Br-1	M.W.I-1	M.W.CO3-2	M.W.NO3-1	M.W.PO4-3	M.W.SO4-2
				243	243	274.9988	274.9988	0	18.015	1.008	18.998	35.453	79.904	126.905	60.008	62.004	94.97	96.062
			MINTEQA2 ID #	40	41	42	43	1	2	330	270	180	130	380	140	492	580	732
	M.W.	Americium																
H.Rxn	Product	Product	H.f.prod	H.f.Am+3	H.f.Am+4	H.f.AmO2+1	H.f.AmO2+2	H.f.E-1	H.f.H2O	H.f.H+1	H.f.F-1	H.f.Cl-1	H.f.Br-	H.f.I-1	H.f.CO3-2	H.f.NO3-1	H.f.PO4-3	H.f.SO4-2
(kcal/mol)	Species	Species	(kcal/mol)	-147.424	-103.378	-192.434	-155.849	0	-68.317	0	-80.15	-39.933	-29.04	-13.6	-161.385	-49.429	-305.3	-217.4
		Solids																
147.424	243	Am (c)	0	1				3										
48.355	294.021	Am(OH)3	-304.02	1					3	-3								
352.375	294.021	Am(OH)3(am)	—	1					3	-3								
10.256	320.015	AmOHCO3	-366.87	1					1	-1					1			
		Aqueous																
2.464	363.016	Am(CO3)2-1	-467.73	1											2			
3.599	303.008	AmCO3+1	-305.21	1											1			
631.579	423.024	Am(CO3)3-3	—	1											3			
196.853	305.004	AmNO3+2	—	1												1		
246.282	367.008	Am(NO3)2+1	—	1												2		

DRL

Example Calculation:

$$Am^{3+} + 2\ CO_3^{2-} = Am(CO_3)^{2-}$$

Enthalpy of Reaction = ΣH_f (products) - ΣH_f (reactants)

H_f values:
Am^{3+} = -147.424 kcal/mol
CO_3^{2-} = -161.385 kcal/mol
$Am(CO_3)^{2-}$ = -467.73 kcal/mol

Therefore:

Enthalpy of Reaction (H. Rxn) = [-467.73] - [(-147.424) + 2(-161.385)] = 2.464 kcal/mol ✓
Molecular Weight of $Am(CO_3)^{2-}$ = (243) + 2(60.008) = 363.016 g/mol ✓

These numbers agree with those calculated for $Am(CO_3)^{2-}$ in the spreadsheet.

8/12/94 DRJ

Reference list for some of the data sources identified in this scientific notebook

Abendroth, R.P. 1970. Behavior of pyrogenic silica in aqueous electrolytes. *Journal of Colloid and Interface Science* 34:591-596.

Balistrieri, L., and J.W. Murray. 1981. The surface chemistry of goethite (α-FeOOH) in major ion sea water. *American Journal of Science* 281:788-806.

Berube, Y.G., and P.L. de Bruyn. 1968. Adsorption at the rutile-solution interface. 1. Thermodynamic and experimental study. *Journal of Colloid and Interface Science* 27:305-318.

Bolt, G.H. 1957. Determination of the charge density of silica soils. *Journal of Physical Chemistry* 61:1166-1169.

Catts, J.G., and D. Langmuir. 1986. Adsorption of Cu, Pb and Zn by δ-MnO_2: Applicability of the site binding surface complexation model. *Applied Geochemistry* 1:255-264.

Davis, J.A. 1977. *Adsorption of Trace Metals and Complexing Ligands at the Oxide/Water Interface*. Ph.D. Dissertation, Stanford University: Stanford, CA.

Davis, J.A., and J.O. Leckie. 1978. Surface ionization and complexation at the oxide/water interface 2. Surface properties of amorphous iron oxyhydroxide and adsorption of metal ions. *Journal of Colloid and Interface Science* 67:90-107.

Davis, J.A., and J.O. Leckie. 1979. Speciation of adsorbed ions at the oxide water interface. *Chemical Modeling in Aqueous Systems, Speciation, Sorption, Solubility, and Kinetics*. E A. Jenne, ed. ACS Symposium Series 93. Washington, D.C.:American Chemical Society:299-317.

Davis, J.A., R.O. James, and J.O. Leckie. 1978. Surface ionization and complexation at the oxide/water interface 1. Computation of electrical double layer properties in simple electrolytes. *Journal of Colloid and Interface Science* 63:480-499.

Davis, J.A., and D.B. Kent. 1990. Surface complexation modeling in aqueous geochemistry. *Reviews in Mineralogy: Volume 23. Mineral-Water Interface Geochemistry*. M.F. Hochella, Jr. and A.F. White, eds. Washington, DC: Mineralogical Society of America: 177-260.

Della Mea, G., J.C. Dran, V. Moulin, J.C. Petit, J.D.F. Ramsay, and M. Theyssier. 1992. Scavenging properties of inorganic particles toward heavy elements. *Radiochimica Acta* 58/59:219-223.

Dzombak, D.A. 1986. *Toward a Uniform Model for the Sorption of Inorganic Ions on Hydrous Oxides*. Ph.D. Dissertation. Massachusetts Institute of Technology: Boston, MA.

Dzombak, D.A., and F.M.M. Morel. 1990. *Surface Complexation Modeling: Hydrous Ferric Oxide*. New York: John Wiley and Sons.

Girvin, D.C., L.L. Ames, A.P. Schwab, and J.E. McGarrah. 1991. Neptunium adsorption on synthetic amorphous iron oxyhydroxide. *Journal of Colloid and Interface Science* 141:67-78.

Hayes, K.F., G. Redden, W. Ela, and J.O. Leckie. 1990. *Application of Surface Complexation Models for Radionuclide Adsorption: Sensitivity Analysis of Model Input Parameters*. NUREG/CR-5547. U.S. Nuclear Regulatory Commission: Washington, D.C.

Hayes, K.F., G. Redden, W. Ela, and J.O. Leckie. 1991. Surface complexation models: An evaluation of model parameter estimation using FITEQL and oxide mineral titration data. *Journal of Colloid and Interface Science* 142:448-469.

Hsi, C-K.D. 1981. *Sorption of Uranium(VI) by Iron Oxides*. Ph.D. Dissertation. Golden, Colorado: Colorado School of Mines.

Hsi, C-K.D. and D. Langmuir. 1985. Adsorption of uranyl onto ferric oxyhydroxides: Application of the surface complexation site-binding model. *Geochimica et Cosmochimica Acta* 49:1931-1941.

Huang, C.P., and W. Stumm. 1972. The specific surface area of γ-Al_2O_3. *Surface Science* 32:287-296.

Hunter, K.A., D.J. Hawke, and L.K. Choo. 1988. Equilibrium adsorption of thorium by metal oxides in marine electrolytes. *Geochimica et Cosmochimica Acta* 52:627-636.

James, R.O., and G. Parks. 1982. Characterization of aqueous colloids by their electrical double-layer and intrinsic surface chemical properties. *Surface and Colloid Science* 12:119-216.

Kohler, M., E. Wieland, and J.O. Leckie. 1992. Metal-ligand-surface interactions during sorption of uranyl and neptunyl on oxides and silicates. *Proceedings of the 7th International Symposium on Water-Rock Interaction - WRI-7. Volume 1: Low Temperature Environments*. Y.K. Kharaka and A.S. Maest, eds. Rotterdam: A.A. Balkema.

Krupka, K.M., R.L. Erikson, S.V. Mattigod, J.A. Schramke, and C.E. Cowan. 1988. *Thermochemical Data Used by the FASTCHEM Package*. EPRI-EA-5872, Palo Alto, California:Electric Power Research Institute (EPRI).

LaFlamme, B.D., and J.W. Murray. 1987. Solid/Solution Interaction: The effect of carbonate alkalinity on adsorbed thorium. *Geochimica et Cosmochimica Acta* 51:243-250.

Lieser, K.H., and B. Thybusch. 1988. Sorption of uranyl ions on hydrous titanium dioxide. *Fresenius Zeitschrift für Analytisch Chemie* 332:351-357.

Mesuere, K.L.G. 1992. *Adsorption and Dissolution Reactions Between Oxalate, Chromate, and an Iron Oxide Surface: Assessment of Current Modeling Concepts*. Ph.D. Dissertation. Oregon Graduate Institute of Science and Technology: Beaverton, OR.

8/12/94 DRJ

Reference List - continued

Moulin, V., D. Stammose, and G. Ouzounian. 1992. Actinide sorption at oxide-water interfaces: Application to α alumina and amorphous silica. *Applied Geochemistry Supplemental Issue* 1:163-166.

Murray, J.W. 1974. The surface chemistry of hydrous manganese dioxide. *Journal of Colloid and Interface Science* 46:357-371.

Nakayama, S., and Y. Sakamoto. 1991. Sorption of neptunium on naturally-occurring iron-containing minerals. *Radiochimica Acta* 52/53:153-157.

Payne, T.E., and T.D. Waite. 1991. Surface complexation modelling of uranium sorption data obtained by isotope exchange techniques. *Radiochimica Acta* 52/53:487-493.

Payne, T.E., K. Sekine, J.A. Davis, and T.D. Waite. 1992. Modeling of radionuclide sorption processes in the weathered zone of the Koongarra ore body. P. Duerden, ed. *Alligator Rivers Analogue Project Annual Report, 1990-1991*. Australian Nuclear Science and Technology Organization (ANSTO):57-85.

Regazzoni, A.E., M.A. Blesa. and A.J.G. Maroto. 1983. Interfacial properties of zirconium dioxide and magnetite. *Journal of Colloid and Interface Science* 91:560-570.

Riese, A.C., *Adsorption of Radium and Thorium onto Quartz and Kaolinite: A Comparison of Solution/Surface Equilibria Models*. Ph.D. Dissertation. Colorado School of Mines: Golden, CO.

Righetto, L., G. Bidoglio, B. Marcandalli, and I.R. Bellobono. 1988, Surface interactions of actinides with alumina colloids. *Radiochimica Acta* 44/45:73-75.

Righetto, L., G. Bidoglio, G. Azimonti, and I.R. Bellobono. 1991. Competitive actinide interactions in colloidal humic acid-mineral oxide systems. *Environmental Science and Technology* 25: 1913-1919.

Sanchez, A.L., J.W. Murray, and T.H. Sibley. 1985. The adsorption of plutonium IV and V on goethite. *Geochimica et Cosmochimica Acta* 49:2297-2307.

Sprycha, R. 1984. Surface charge and adsorption of background electrolyte ions at anatase/electrolyte interface. *Journal of Colloid and Interface Science* 102:173-185.

Sprycha, R. 1989. Electrical double layer at alumina/electrolyte interface. *Journal of Colloid and Interface Science* 127:1-11.

Stammose, D., J.M. Dolo. 1990. Sorption of americium at trace levels on a clay mineral. *Radiochimica Acta* 51:189-193.

Swallow, K.C. 1978. *Adsorption of Trace Metals by Hydrous Ferric Oxide*. Ph.D. Dissertation. Massachusetts Institute of Technology: Cambridge, MA.

Tripathi, V.S. 1984. *Uranium(VI) Transport Modeling: Geochemical Data and Submodels*. Ph.D. Dissertation. Stanford, California: Stanford University.

Venkataramani, B., and A.R. Gupta. 1991. Effect of anions on the sorption of uranyl ions on hydrous oxides: Application of the surface hydrolysis model. *Colloids and Surfaces* 53:1-19.

Yates, D.A. 1975. *The Structure of the Oxide/Aqueous Electrolyte Interface*. Ph.D. Dissertation. University of Melbourne: Melbourne, Australia.

Yates, D.A., and T.W. Healy. 1975. Mechanism of anion adsorption at the ferric and chromic oxide/water interfaces. *Journal of Colloid and Interface Science* 52:222-228.

Zachara, J.M., D.C. Girvin, R.L. Schmidt, and C.T. Resch. 1987. Chromate adsorption on amorphous iron oxyhydroxide in the presence of major groundwater ions. *Environmental Science and Technology* 21:589-594.

7/12/94 PM K Tables Continued

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite
A_{sp}: 600 m²/g
Data Source: Payne et al 92
Concentration: [U(VI)] = 1e-6 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide):
Abs Error (radionuclide): 1.0e-10

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_s = 2.31 sites/nm²

fig 6.2 co-precipitated $Fe(OH)_3$	DLM			CCM			TLM		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Na} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.871	2.877	.02587	2.073	2.963	.02593	1.196	8.763	.02327
$XOH\text{-}UO_2^{2+}$	7.703	55.39	.02295	9.146	6.633	.02371	1.395	88.88	.06296
$XO\text{-}UO_2OH^0$	-4.483	6.848	.03680	-5.022	2.258	.02846	-8.407	31.67	.02086
$XOH\text{-}UO_2OH^+$	1.871	2.877	.02587	2.073	2.963	.02593	-.9267	45.11	.02185
$XO\text{-}UO_2(OH)_2^-$	-11.01	14.84	.04957	-12.13	3.198	.03130	-12.01	2.907	.03102
$XOH\text{-}UO_2(OH)_2^0$	-4.483	6.848	.03680	-5.022	2.258	.02846	-4.410	2.235	.02974
$XOH_2\text{-}UO_2(OH)_2^+$	1.871	2.877	.02587	2.073	2.963	.02593	3.196	2.463	.02861
$XOH\text{-}UO_2(OH)_3^-$	-11.01	14.84	.04957	-12.13	3.198	.03130	-8.3667	11.03	.04521
$XOH_2\text{-}UO_2(OH)_2^0$	-4.483	6.848	.03680	-5.022	2.258	.02847	-.7002	8.579	.04337
$XOH\text{-}UO_2(OH)_4^{2-}$	-17.62	20.99	.06391	-19.26	4.959	.03434	-12.52	19.96	.06460
$XOH_2\text{-}UO_2(OH)_3^-$	-11.01	14.84	.04957	-12.13	3.198	.03130	-4.812	17.53	.06160
$XOH\text{-}UO_2CO_3^0$									
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

.U26

8/16/94 PM

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite
A_{sp}: 600 m²/g
Data Source: Payne et al 92
Concentration: [U(VI)] = 1e-6 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide):
Abs Error (radionuclide): 1.0e-10

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_s = 2.31 sites/nm²

fig 4.1 C_s = 0.1777 $P(CO_2)$ = 0.03%	DLM			CCM			TLM *		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Na} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	.3492	33.03	.02042	.3805	33.14	.02049	-4.641	43.82	.02785
$XOH\text{-}UO_2^{2+}$	8.471	13.07	.01750	8.704	17.75	.02034	2.838	44.65	.02978
$XO\text{-}UO_2OH^0$	-8.065	41.93	.02542	-8.060	41.79	.02521	-7.698	14.13	.02161
$XOH\text{-}UO_2OH^+$	.3492	33.03	.02402	.3805	33.14	.02049	-.1278	18.40	.02205
$XO\text{-}UO_2(OH)_2^-$	-16.33	43.34	.02693	-16.37	43.48	.02725	-16.35	43.51	.02725
$XOH\text{-}UO_2(OH)_2^0$	-8.065	41.93	.02542	-8.060	41.79	.02521	-8.347	43.31	.02725
$XOH_2\text{-}UO_2(OH)_2^+$	.3492	33.03	.02042	.3722	33.37	.02054	-.3364	42.95	.02709
$XOH\text{-}UO_2(OH)_3^-$	-16.33	43.34	.02693	-16.37	43.48	.02725	-17.59	43.96	.02806
$XOH_2\text{-}UO_2(OH)_3^0$	-8.065	41.93	.02542	-8.057	41.78	.02521	-9.563	43.79	.02783
$XOH\text{-}UO_2(OH)_4^{3-}$	-24.56	43.41	.02656	-24.64	43.65	.02712	-27.27	46.49	.04196
$XOH_2\text{-}UO_2(OH)_4^-$	-16.33	43.34	.02693	-16.36	43.47	.02723	-19.22	46.33	.04074
1) $XOH\text{-}UO_2CO_3^0$	-.7220	62.07	.03017	-1.048	61.88	.02443	-1.088	61.99	.02397
2) $XOH_2\text{-}UO_2(CO_3)_2^-$	-3.822	76.00	.03368	-4.707	76.35	.02571	1.155	65.53	.03973
3) $XOH_2\text{-}UO_2(CO_3)_3^{3-}$	-13.89	82.63	.03547	-15.72	84.01	.02725	-3.886	64.07	.05804
4) $XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$	-3.083	41.02	.05851	-3.479	50.91	.05216	-1.548	67.93	.01996

*: TLM FILES run under FIT2DIM as FIT2 only accepts 10 components

.U27

FIT2 + FIT2DIM are FITEQL executable files.

Errors for input files numbered 1), 2), 3), + 4):

	abs error (rad)	a.e. (rad)	a.e. (rad)	a.e. (rad)
UO_2 ads	1) 1.0e-10	2) 1.0e-10	3) 1.0e-10	4) 1.0e-10
CO_3 ads	1.0e-10	2.0e-10	3.0e-10	5.0e-11
H^+	0.0	0.0	0.0	0.0

8/17/94 PM

PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: ferrihydrite
A_{sp}: 600 m²/g
Data Source: Payne et al 92
Concentration: [U(VI)] = 1e-6 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide):
Abs Error (radionuclide): 1.0e-09

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_s = 2.31 sites/nm²

fig 4.2 C_s = 0.1777 $P(CO_2)$ = 1%	DLM			CCM			TLM*		
	Log K_+ = 7.18 Log K_- = -8.82			Log K_+ = 7.17 Log K_- = -8.84			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Na} = -7.66 Log K_{As} = 8.21		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	2.105	19.97	.03913	2.302	18.73	.03917	-4.555	35.88	.04448
$XOH\text{-}UO_2^{2+}$	8.268	31.45	.03682	9.412	22.43	.03711	2.999	36.31	.04684
$XO\text{-}UO_2OH^0$	-4.791	17.83	.04085	-4.955	16.80	.03853	-9.082	35.11	.04495
$XOH\text{-}UO_2OH^+$	2.105	19.97	.03913	2.302	18.73	.03917	-1.103	31.53	.03745
$XO\text{-}UO_2(OH)_2^-$	-12.75	29.69	.04196	-12.72	24.20	.03794	-18.52	40.52	1.276
$XOH\text{-}UO_2(OH)_2^0$	-4.791	17.83	.04085	-4.955	16.80	.03853	-10.53	40.52	1.085
$XOH_2\text{-}UO_2(OH)_2^+$	2.105	19.97	.03913	2.302	18.73	.03917	-2.538	40.52	.9126
$XOH\text{-}UO_2(OH)_3^-$	-12.75	29.69	.04196	-12.72	24.20	.03794	-19.44	40.53	6.867
$XOH_2\text{-}UO_2(OH)_3^0$	-4.791	17.83	.04085	-4.955	16.80	.03853	-11.46	40.53	6.083
$XOH\text{-}UO_2(OH)_4^{2-}$	-20.89	37.99	.06339	-20.78	34.48	.04618	-28.35	40.53	36.16
$XOH_2\text{-}UO_2(OH)_4^-$	-12.75	29.69	.04196	-12.72	24.20	.03794	-20.37	40.53	32.09
1) $XOH\text{-}UO_2CO_3^0$	-3.774	29.44	.03660	-4.317	35.17	.03035	-2.816	63.26	.04069
2) $XOH_2\text{-}UO_2(CO_3)_2^-$	-9.691	25.50	.04754	-10.94	33.73	.03314	-1.625	59.45	.06672
3) $XOH_2\text{-}UO_2(CO_3)_3^{3-}$	-22.21	17.57	.05629	-24.68	29.26	.03858	-7.261	60.09	.09428
4) $XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$	-5.686	31.85	.06639	-6.237	33.43	.05778	-5.029	69.26	.04742

* FIT2DIM used

.U28

Errors for input files numbered 1), 2), 3), + 4)

	abs error (rad)	a.e. (rad)	a.e. (rad)	a.e. (rad)
UO_2 ads	1) 1.0e-9	2) 1.0e-9	3) 1.0e-9	4) 1.0e-9
CO_3 ads	1.0e-9	2.0e-9	3.0e-9	5.0e-10
H^+	0.0	0.0	0.0	0.0

Next is Dave Turner's explanation of the data used for the Tripathi input files:

Using the Data of Tripathi (1984) in FITEQL

1. As noted in Tripathi (1984), below pH ≈ 7, the sorption data are the same for both CO_2-free and atmospheric CO_2 experiments. In the presence of atmospheric CO_2, additional experiments were conducted at pH > 7. For the purposes of constructing the FITEQL input files, the data were digitized for each U(VI) concentration. For those concentrations where both CO_2-free and atmospheric CO_2 experiments were conducted, the two sets of digitization results for data points below pH ≈ 7 were averaged and entered into FITEQL. For atmospheric CO_2 experiments, data points at pH > 7 were not replicated in the CO_2-free experiments, and only one set of digitization results was used.

2. In the presence of CO_2, only surface complexes using the uranium-carbonate species $UO_2CO_3^o$, $UO_2(CO_3)_2^{2-}$, $UO_2(CO_3)_3^{4-}$, and $(UO_2)_2CO_3(OH)_3^+$ were considered. In addition, the surface complex $XOH\text{-}UO_2(OH)_4^{2-}$ that best reproduced the observed sorption behavior in the CO_2-free experiments was also considered in the FITEQL runs.

8/22/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite
A_{sp}: 50 m²/g
Data Source: Tripathi 84
Concentration: [U(VI)] = 1e-5.4 M (1000 ppb)

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide): 0.08
Abs Error (radionuclide): 3.4e-7

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_s = 2.31 sites/nm²

log pCO_2 = -3.5

	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.988	1.196	.08361	1.491	1.245	.08575	.4329	2.154	.1216
$XOH\text{-}UO_2^{2+}$	9.918	1.489	.09248	9.069	1.536	.1015	9.647	1.829	.1527
$XO\text{-}UO_2OH^o$	-5.831	.9227	.08064	-5.893	.9362	.08065	-6.933	1.701	.08813
$XOH\text{-}UO_2OH^+$	1.988	1.196	.08361	1.491	1.245	.08575	1.562	1.549	.1100
$XO\text{-}UO_2(OH)_2^-$	-13.54	.7207	.08382	-13.10	.7142	.08517	-12.98	.9217	.1056
$XOH\text{-}UO_2(OH)_2^o$	-5.831	.9227	.08064	-5.893	.9362	.08065	-5.740	.9487	.08071
$XOH_2\text{-}UO_2(OH)_2^+$	1.988	1.196	.08361	1.491	1.245	.08575	2.795	.9756	.1049
$XOH\text{-}UO_2(OH)_3^-$	-13.54	.7207	.08382	-13.10	.7142	.08517	-11.70	.5069	.1055
$XOH_2\text{-}UO_2(OH)_3^o$	-5.831	.9227	.08064	-5.893	.9362	.08065	-4.423	.4321	.08123
$XOH\text{-}UO_2(OH)_4^{2-}$	-21.13	.5996	.09298	-20.13	.5949	.09760	-16.88	.3600	.1439
$XOH_2\text{-}UO_2(OH)_4^-$	-13.54	.7207	.08382	-13.10	.7142	.08517	-10.31	.2569	.1115
$XOH\text{-}UO_2CO_3^o$	-2.238	1.107	.04487	-2.298	1.111	.04503	-2.143	1.120	.04517
$XOH_2\text{-}UO_2(CO_3)_2^-$	-6.373	.6654	.05479	-5.983	.8358	.05273	-3.254	5.286	.08160
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$	-18.00	.8324	.07335	-16.54	1.809	.07877	-9.126	5.199	.1721
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^o$	-7.086	1.447	.06192	-7.144	1.448	.06209	AO	AO	AO

1) C1
2) C2
3) C3
4) CC

AO: arithmetic overflow error .U29 pg 1

$XOH\text{-}UO_2(OH)_4^{-2}$ wC	-22.28	4.696	.07158	-20.23	5.806	.08141	-16.86	6.725	.1370

U29 pg 2

Errors For Input Files Numbered 1), 2), 3), + 4)

	abs err (radion.)	abs error (r)	abs error (r)	abs error (r)
UO_2 ads	1) 3.4 e-7	2) 3.4 e-7	3) 3.4 e-7	4) 3.4 e-7
CO_3 ads	3.4 e-7	6.8 e-7	1.0 e-6	1.7 e-7
H^+	0 0	0.0	0.0	0.0

8/30/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite
A_{sp}: 50 m²/g
Data Source: Tripathi 94
Concentration: [U(VI)] = 1e-5.8 M (400 ppb)

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide): 0.08
Abs Error (radionuclide): 1.3e-7

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_s = 2.31 sites/nm²

	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.984	1.414	.08951	1.461	1.452	.09046	-.03675	2.884	.1257
$XOH\text{-}UO_2^{2+}$	9.802	1.801	.09556	8.849	1.839	.09879	8.560	2.544	.1453
$XO\text{-}UO_2OH^o$	-5.781	1.062	.08668	-5.845	1.087	.08679	-6.991	2.067	.09666
$XOH\text{-}UO_2OH^+$	1.984	1.414	.08951	1.461	1.452	.09046	1.179	2.005	.1084
$XO\text{-}UO_2(OH)_2^-$	-13.49	.7771	.08704	-13.07	.7944	.08769	-13.28	1.056	.1008
$XOH\text{-}UO_2(OH)_2^o$	-5.781	1.062	.08668	-5.845	1.087	.08679	-5.694	1.104	.08688
$XOH_2\text{-}UO_2(OH)_2^+$	1.984	1.414	.08951	1.461	1.452	.09046	2.502	1.158	.1010
$XOH\text{-}UO_2(OH)_3^-$	-13.49	.7771	.08704	-13.07	.7944	.08769	-11.89	.4729	.09984
$XOH_2\text{-}UO_2(OH)_3^o$	-5.781	1.062	.08668	-5.845	1.087	.08679	-4.290	.4350	.08587
$XOH\text{-}UO_2(OH)_4^{2-}$	-21.16	.5696	.09035	-20.22	.5914	.09270	-17.56	.2717	.1339
$XOH_2\text{-}UO_2(OH)_4^-$	-13.49	.7771	.08704	-13.07	.7944	.08769	-10.43	.1709	.1041
$XOH\text{-}UO_2CO_3^o$	-2.244	1.173	0.4579	-2.305	1.183	.04598	-2.150	1.193	.04613
$XOH_2\text{-}UO_2(CO_3)_2^-$	-6.411	.6642	.05373	-6.040	.8606	.05194	-3.362	4.323	.08046
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$	-18.25	.7823	.06232	-16.88	1.936	.06641	-10.74	3.950	.1574
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^o$	-6.716	1.627	.06431	-6.776	1.629	.06444	-5.399	3.948	.06852

1)
2)
3)
4)

.U30 pg 1

$XOH\text{-}UO_2(OH)_4^{-2}$ wC	-21.29	4.361	.07402	-20.30	5.103	.08190	-17.56	5.565	.1306

.U30 pg 2

Errors For Input Files Numbered 1), 2), 3), + 4)

	abs err (radion.)	abs error (r)	abs error (r)	abs error (r)
UO_2 ads	1) 1.3 e-7	2) 1.3 e-7	3) 1.3 e-7	4) 1.3 e-7
CO_3 ads	1.3 e-7	2.6 e-7	3.9 e-7	6.5 e-8
H^+	0 0	0.0	0.0	0.0

8/31/94 PM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite
A_{sp}: 50 m²/g
Data Source: Tripathi 84
Concentration: [U(VI)] = 1.05e-6 M

Rel Error (pH): 0.05
Abs Error (pH): 0.0
Rel Error (radionuclide): 0.08
Abs Error (radionuclide): 8.0e-8

Ionic Strength (electrolyte): 0.1 M $NaNO_3$
N_s = 2.31 sites/nm²

	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Cat} = -7.64 Log K_{An} = 8.56		
	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$	Log K	V_Y	$\sigma_{Log K}$
$XO\text{-}UO_2^+$	1.306	8.014	.04185	.7619	8.331	.04248	-2.420	17.95	.06439
$XOH\text{-}UO_2^{2+}$	8.767	11.35	.04441	7.821	10.61	.04506	5.889	16.32	.07565
$XO\text{-}UO_2OH^o$	-6.187	4.163	.04152	-6.282	4.589	.04126	-8.178	12.71	.04546
$XOH\text{-}UO_2OH^+$	1.3061	8.014	.04185	.7619	8.331	.04248	-.2238	12.72	.05119
$XO\text{-}UO_2(OH)_2^-$	-13.76	1.354	.04466	-13.41	1.444	.04447	-13.87	3.974	.04699
$XOH\text{-}UO_2(OH)_2^o$	-6.187	4.163	.04152	-6.282	4.589	.04126	-6.142	4.691	.04117
$XOH_2\text{-}UO_2(OH)_2^+$	1.306	8.014	.04185	.7619	8.331	.04248	1.827	5.495	.04692
$XOH\text{-}UO_2(OH)_3^-$	-13.76	1.354	.04466	-13.41	1.444	.04447	-12.11	.4397	.05425
$XOH_2\text{-}UO_2(OH)_3^o$	-6.187	4.163	.04152	-6.282	4.589	.04126	-4.370	.3827	.04856
$XOH\text{-}UO_2(OH)_4^{2-}$	-21.32	.5097	.04967	-20.49	.7417	.04890	-17.81	.2176	.07596
$XOH_2\text{-}UO_2(OH)_4^-$	-13.76	1.354	.04466	-13.41	1.444	.04447	-10.30	.2157	.06189
$XOH\text{-}UO_2CO_3^o$	-2.502	3.497	.02215	-2.581	3.890	.02220	-2.434	4.085	.02227
$XOH_2\text{-}UO_2(CO_3)_2^-$	-6.598	1.351	.02583	-6.269	1.484	.02519	-5.010	2.200	.03970
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$	-18.42	.7639	.03261	-17.26	2.084	.03458	-15.65	9.624	.08676
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^o$	-6.888	2.098	.04488	-6.963	2.222	.04489	-5.620	2.316	.05157

1)
2)
3)
4)

Continued on next page 8/31/94 PM

.U31 pg 1

$XOH\text{-}UO_2(OH)_4^{2-}$ wC	-21.69	13.57	.04123	-20.74	14.45	.04500	-17.88	15.45	.07311

,U31/pg 2

Errors For Input Files Numbered 1), 2), 3), + 4)

	abs err (rad.an.)	abs error (r)	abs error (r)	abs error (r)
UO_2 abs	1) 8.0e-8	2) 8.0e-8	3) 8.0e-8	4) 8.0e-8
CO_3 abs	8.0e-8	1.6e-7	2.4e-7	4.0e-7
H^+	0.0	0.0	0.0	0.0

9/8/94 RM

Uranium (VI) Sorption Binding Constants
Monodentate, mononuclear compounds

Solid: goethite A_{sp}: 50 m^2/g Data Source: Tripathi 94 Concentration: [U(VI)] = 1e-6.4 M (100ppb)	Rel Error (pH): 0.05 Abs Error (pH): 0.0 Rel Error (radionuclide): 0.08 Abs Error (radionuclide): 3.4e-8	Ionic Strength (electrolyte): 0.1 M $NaNO_3$ N_s = 2.31 sites/nm^2

	DLM			CCM			TLM		
	Log K_+ = 7.24 Log K_- = -9.06			Log K_+ = 6.47 Log K_- = -9.03			Log K_+ = 5.89 Log K_- = -9.89 Log K_{Ca} = -7.64 Log K_{Ab} = 8.56		
	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$	Log K	V_Y	$\sigma_{Log\ K}$
$XO\text{-}UO_2^+$	1.460	3.065	.06906	.9269	2.895	.06930	-1.638	11.63	.08711
$XOH\text{-}UO_2^{2+}$	8.999	5.341	.07039	8.026	4.392	.07042	6.327	11.01	.09730
$XO\text{-}UO_2OH^0$	-6.065	1.489	.07102	-6.159	1.742	.07034	-7.890	6.321	.07116
$XOH\text{-}UO_2OH^+$	1.460	3.065	.06906	.9269	2.895	.06930	.01630	6.564	.07534
$XO\text{-}UO_2(OH)_2^-$	-13.59	.6353	.07549	-13.24	.9652	.07325	-13.81	1.665	.07465
$XOH\text{-}UO_2(OH)_2^0$	-6.065	1.489	.07102	-6.159	1.742	.07034	-6.020	1.816	.07019
$XOH_2\text{-}UO_2(OH)_2^+$	1.460	3.065	.06906	.9269	2.895	.06930	1.885	2.114	.07378
$XOH\text{-}UO_2(OH)_3$	-13.59	.6353	.07549	-13.24	.9652	.07325	-11.94	.2638	.0839
$XOH_2\text{-}UO_2(OH)_3^0$	-6.065	1.489	.07102	-6.159	1.742	.07034	-4.147	.2689	.07974
$XOH\text{-}UO_2(OH)_4^{2-}$	-21.12	.2964	.08123	-20.31	.5076	.07760	-17.80	.2489	.1057
$XOH_2\text{-}UO_2(OH)_4^-$	-13.59	.6353	.07549	-13.24	.9652	.07325	-10.12	.3139	.09534
$XOH\text{-}UO_2CO_3^0$	NO CO_2 at	this $U(VI)_T$	----------	----------	----------	----------	----------	----------	----------
$XOH_2\text{-}UO_2(CO_3)_2^-$									
$XOH_2\text{-}UO_2(CO_3)_3^{3-}$									
$XOH_2\text{-}(UO_2)_2CO_3(OH)_3^0$									

,U32

9/9/94 RM

RM

this notebook's data will be continued in scientific notebook #123.