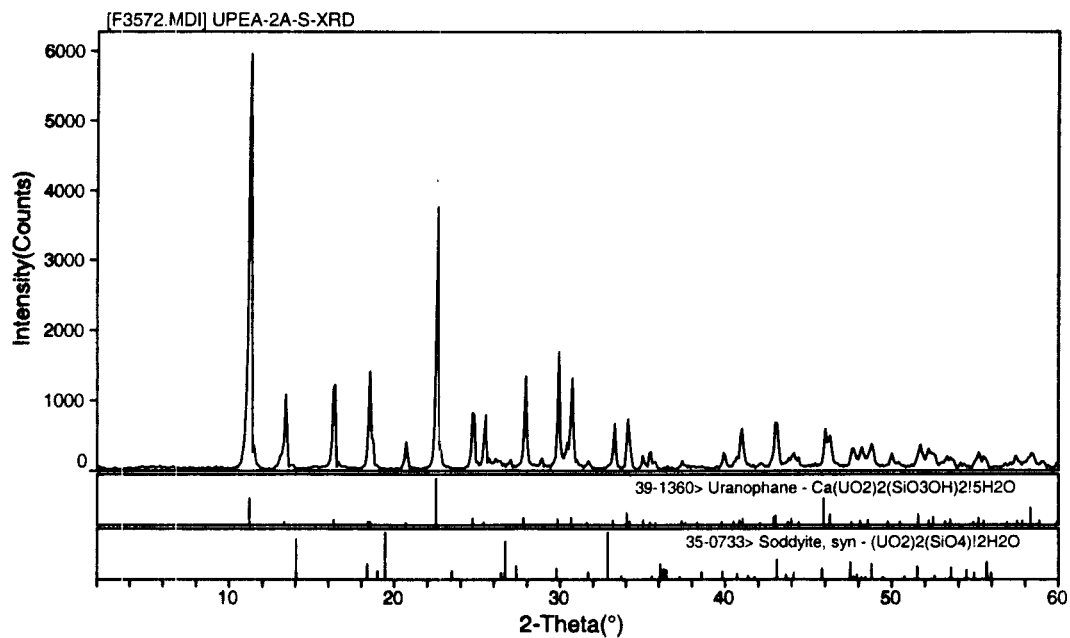
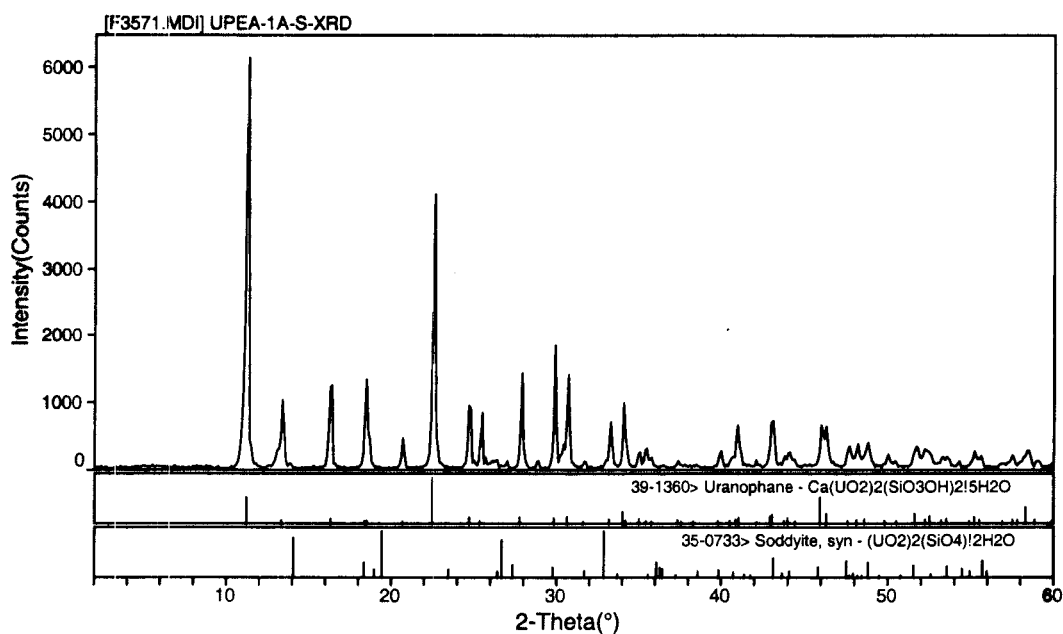
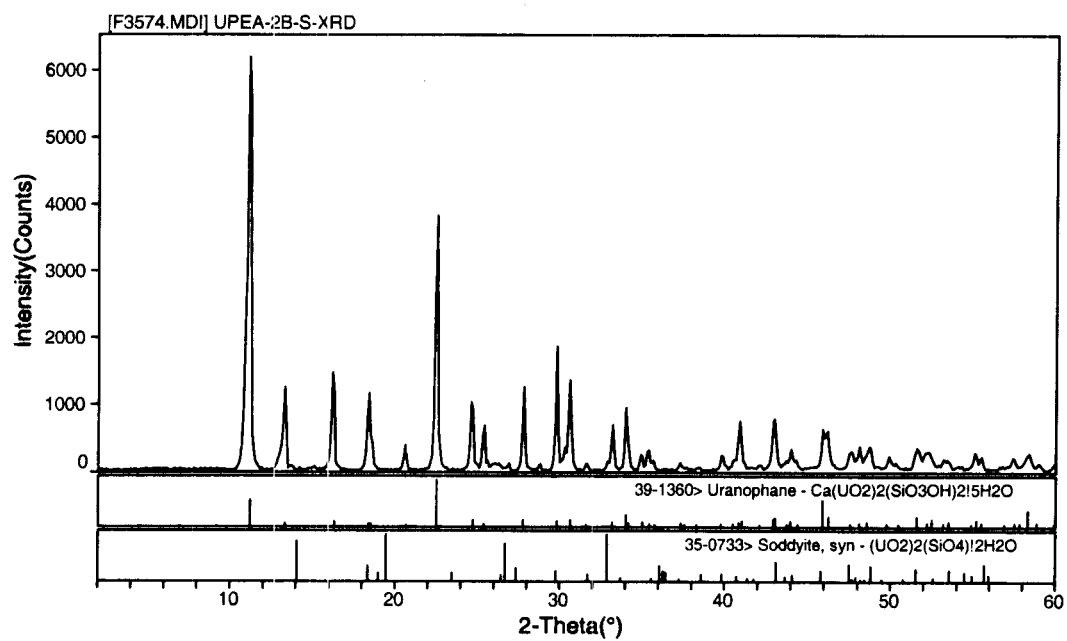
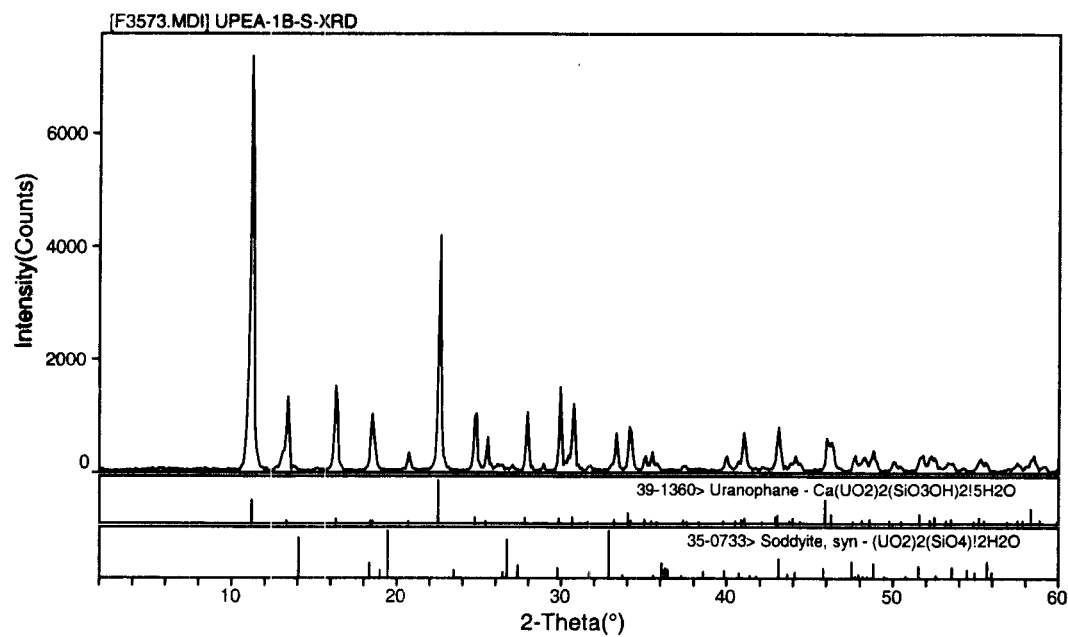


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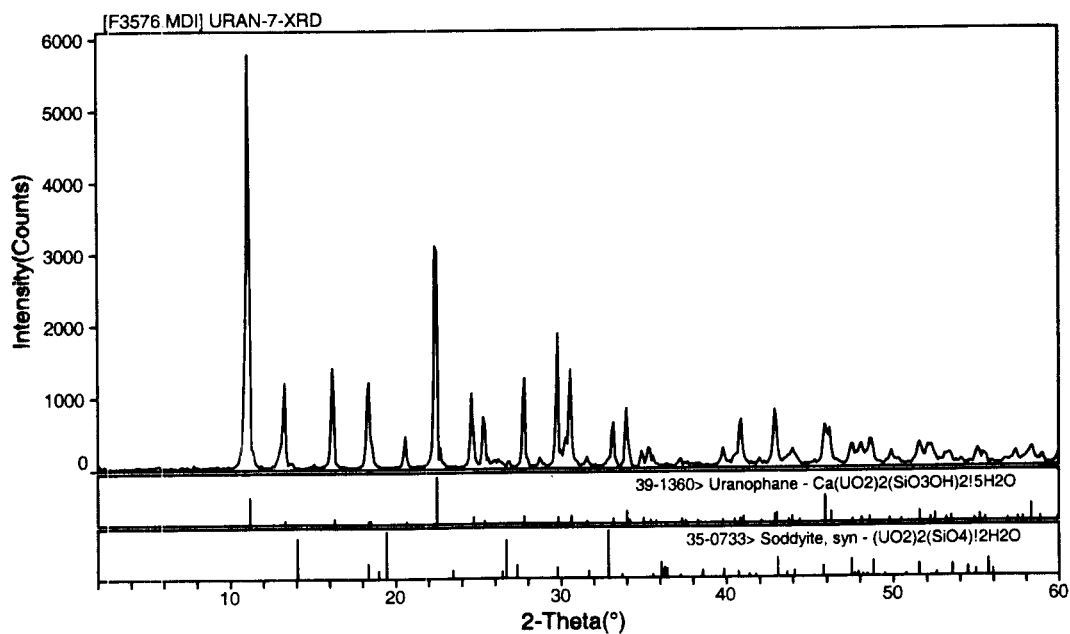
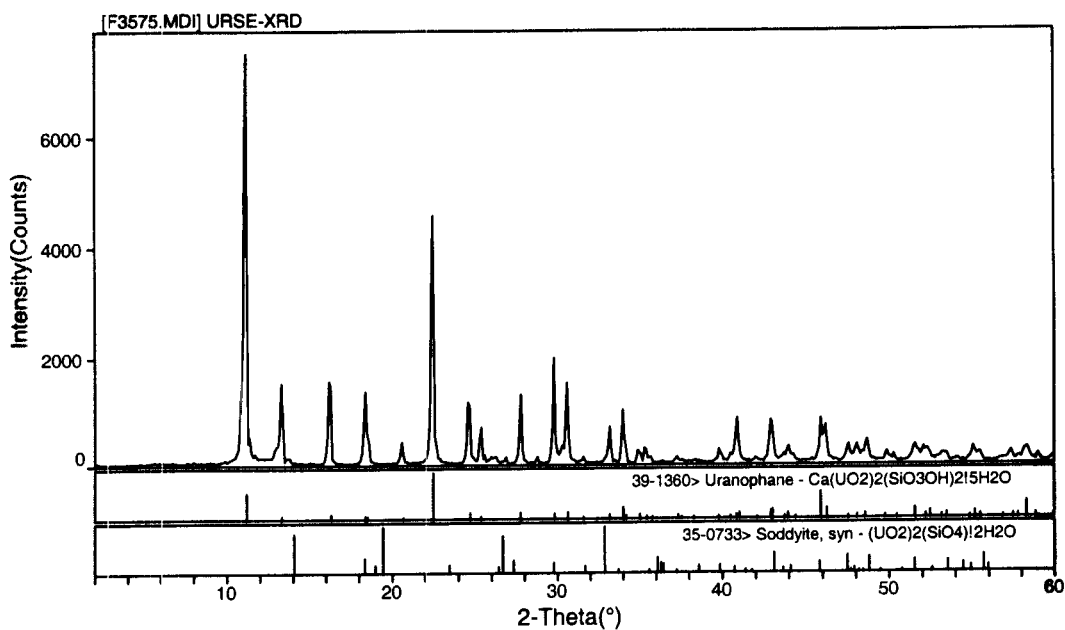
Results of xRD analyses of uranophane
sampled before and after time in
solubility experiments.



11/1/07 gr



11/1/07 JF



11/1/07 JP

Results of chemical analyses of
 solution sent to Div 01 from
 exposit NPUA and NPUB are $\rightarrow$ see p83-85
 shown of the following pages.

Hardcopies of chemical analyses for
 exposit NPUA are kept in a
 3 ring binder entitled "NPUA Results".

Hardcopies of chemical analyses for
 exposit NPUB are kept in a
 3 ring binder entitled "NPUB Results".

JP
 11/1/07

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 Man
 Moly
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 Phos
 Pota
 Seler
 Silica
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 Sodi
 Stron
 Sulfu
 Thall
 Thor
 Tin
 Titan
 Tung
 Uran
 Vana
 Yttri
 Zinc
 Zirco

3.85

[illegible]

11/1/07 JH

Results of chemical analysis of experimental solutions from Np/Uranophane coprecipitation experiment NPUB.

Sample ID	NPUB-M1	NPUB-M2	NPUB-1A	NPUB-1B	NPUB-1C
Element	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)
Aluminum	<0.500	<0.500	<0.500	<0.500	<0.500
Antimony	<0.150	<0.150	<0.150	<0.150	<0.150
Arsenic	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Barium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Beryllium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Bismuth	<0.100	<0.100	<0.100	<0.100	<0.100
Boron	<0.250	<0.250	<0.250	<0.250	<0.250
Cadmium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Calcium	498	495	495	523	515
Chromium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Cobalt	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Copper	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Iron	<1.00	<1.00	1.01	<1.00	1.30
Lanthanum	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Lead	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Lithium	<0.100	<0.100	<0.100	<0.100	<0.100
Magnesium	<0.500	<0.500	<0.500	<0.500	<0.500
Manganese	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Molybdenum	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Nickel	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Palladium	<0.250	<0.250	<0.250	<0.250	<0.250
Phosphorus	<0.200	<0.200	<0.200	<0.200	0.480
Potassium	<3.00	3.36	<3.00	3.20	14.9
Selenium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Silicon	96.0	95.9	94.9	100	96.7
Silver	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Sodium	176	176	174	186	180
Strontium	0.0793	0.0794	0.0800	0.0844	0.0844
Sulfur	<0.250	<0.250	<0.250	<0.250	<0.250
Thallium	<0.100	<0.100	<0.100	<0.100	<0.100
Thorium	<0.250	<0.250	<0.250	<0.250	<0.250
Tin	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Titanium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Tungsten	<0.100	<0.100	<0.100	<0.100	<0.100
Uranium	13.6	14.3	7.65	6.08	4.37
Vanadium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Yttrium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Zinc	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500
Zirconium	<0.0500	<0.0500	<0.0500	<0.0500	<0.0500

11/12/07 JP

Response to reviewer comments on the journal paper "The Thermodynamics of Urmophane Dissolution and Growth in CaCl_2 - SiO_2 (aq) Test Solutions" submitted to *Geochimica et Cosmochimica Acta*.

This paper was an IM for the WRC (IM. 06002.01. 222.750)

This paper details dissolution and precipitation experiments on synthetic urmophane to derive an equilibrium constant for ^{the} urmophane dissolution reaction

Data and calculations from the experiments are listed and explained in Scientific Notebook 582 pages. 142-200 and continue ^{this} in Scientific Notebook (823) in pages 4-28.

The original manuscript was submitted to *Geochimica et Cosmochimica Acta* in May, 2007. Comments from two anonymous reviewers and an associate editor were received in July, 2007.

Reviews were mixed.

The first reviewer suggested rejection of the manuscript over concerns that (1) the extensive report on urmophane thermodynamics reported by Casas et al (1994) was not considered and (2) solids were insufficiently characterized after the experiments to evaluate precipitation of phases other than urmophane. This reviewer also questioned

11/12/07

if the study did a better job at characterizing uranophane thermodynamic than previous works (e.g., Perry et al 2000).

Casas et al 1994 kinetics + thermodynamic studies of uranum minerals. Assessment of the long-term evolution of spent nuclear fuel. SKB Technical Report 94-16

Perry et al 2000. The thermodynamic + kinetics of uranophane dissolution in bicarbonate test solution. Geochimica Cosmochimica Acta 64, 4, 603-608.

The second reviewer provided a short list of review comments and suggested publication after moderate revision. This reviewer was mainly concerned with the lack of analysis of solid product phases and interpretations provided on the Perry et al 2000 data set on uranophane dissolution.

The associate editor for GSA (Eric Oelkers) also provided a list of notes + comments on the manuscript. His main concern was whether the data presented on uranophane dissolution + precipitation + mass transfer actually indicated attainment of equilibrium.

11/13/07 JP

The associate editor suggested that the manuscript be resubmitted after the following additions are made.

- 1) additional analysis of post-experiment solids.
- 2) better justification for attainment of uranophane equilibrium.
- 3) shortening manuscript by removing superfluous, repeated, or unrelated text.

11/14/07 JP

Based on reviewer comments the following revisions were made to the GCA paper on uranophane solubility.

- 1) "Thermodynamic" was removed from the title of the paper since only the equilibrium constant for uranophane was measured. The new title is "Uranophane Dissolution and Growth in CaCl_2 - SiO_2 (aq) Test Solution".
- 2) Abstract was shortened by removing detail on the initial aqueous solution compositions.

11/15/07 JF

- 3) To shorten the manuscript
 - Section 4.4 (Comparison to Other Studies) and 4.5 (Significance to Sulf Corrosion) were removed. Some of the information in these sections was condensed and added to Introduction Section.
 - Section 4.1 (Solute Chemistry + Reaction Path Characteristics) was removed. Some of the elements by element information in this section was added to the Results Section.
- 4) Section 3.3 (Thermodynamic Calculations) moved to Discussion Section.
- 5) Introduction section revised to include better justification for study and to mention Casas et al (1994) work.
- 6) Experimental Methods and Materials section is basically unchanged except that measurement uncertainties in pH and cation analysis were added.

11/19/07 JH

- 7) Data and analyses of ^{precipitates} test solution P-2A (UPEA-2A) and P-2B (UPEA-2B), which had higher initial SiO_2 contents were removed from the manuscript. Further analysis of these test solutions indicated that they were not in equilibrium with uraple when the experiments were terminated (e.g., Si was still precipitating and log Q_s was still rising). The reasons for non-equilibrium are unclear but are likely due to the high Si content. Rather than speculate about these tests and since the objective of the paper is to determine a solubility constant for uraple that tests were removed. Inclusion of these test solutions in the paper would not have changed the ultimate conclusions.

JH
11/19/07

11/20/07 JP

8) Results Section

- Table 1 removed from manuscript. data in this table is in text and can be determined from the figures.

- pH and cation concentration data were replotted in Figures 2, 3, and 4 so that trends could be more easily discerned.

In addition error bars showing analytical uncertainty in pH and cation analyses were added to the figures.

Analytical uncertainty were ± 0.03 pH units for pH, $\pm 2\%$ for Ca and Si contents, and $\pm 10\%$ for U analysis.

Figures 2, 3, and 4 are shown on the following pages.

JP 11/20/07

11/20/07 JF

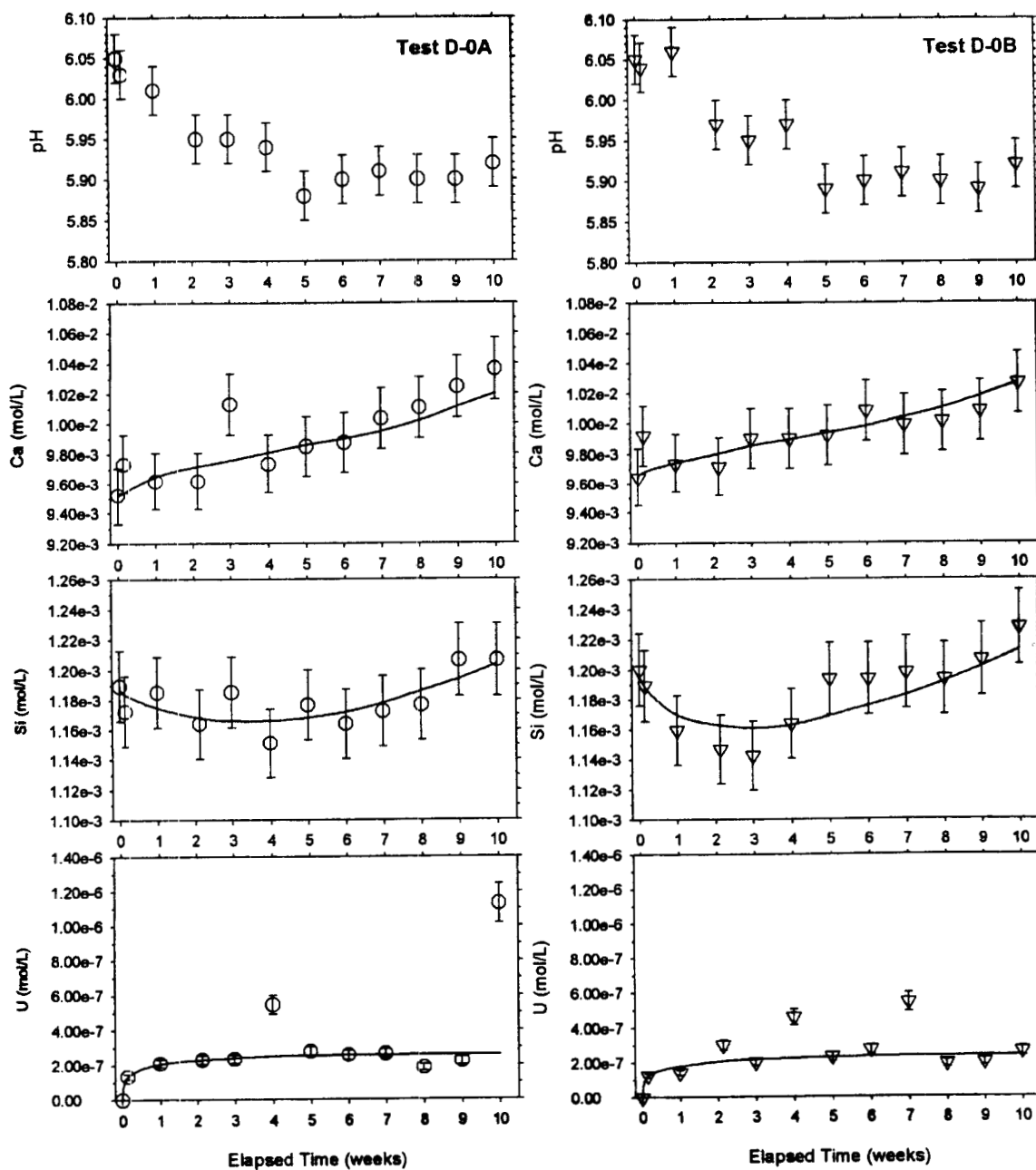


Fig 2. The pH + concentrations of Ca, Si, + U in Tests D-0A + D-0B.

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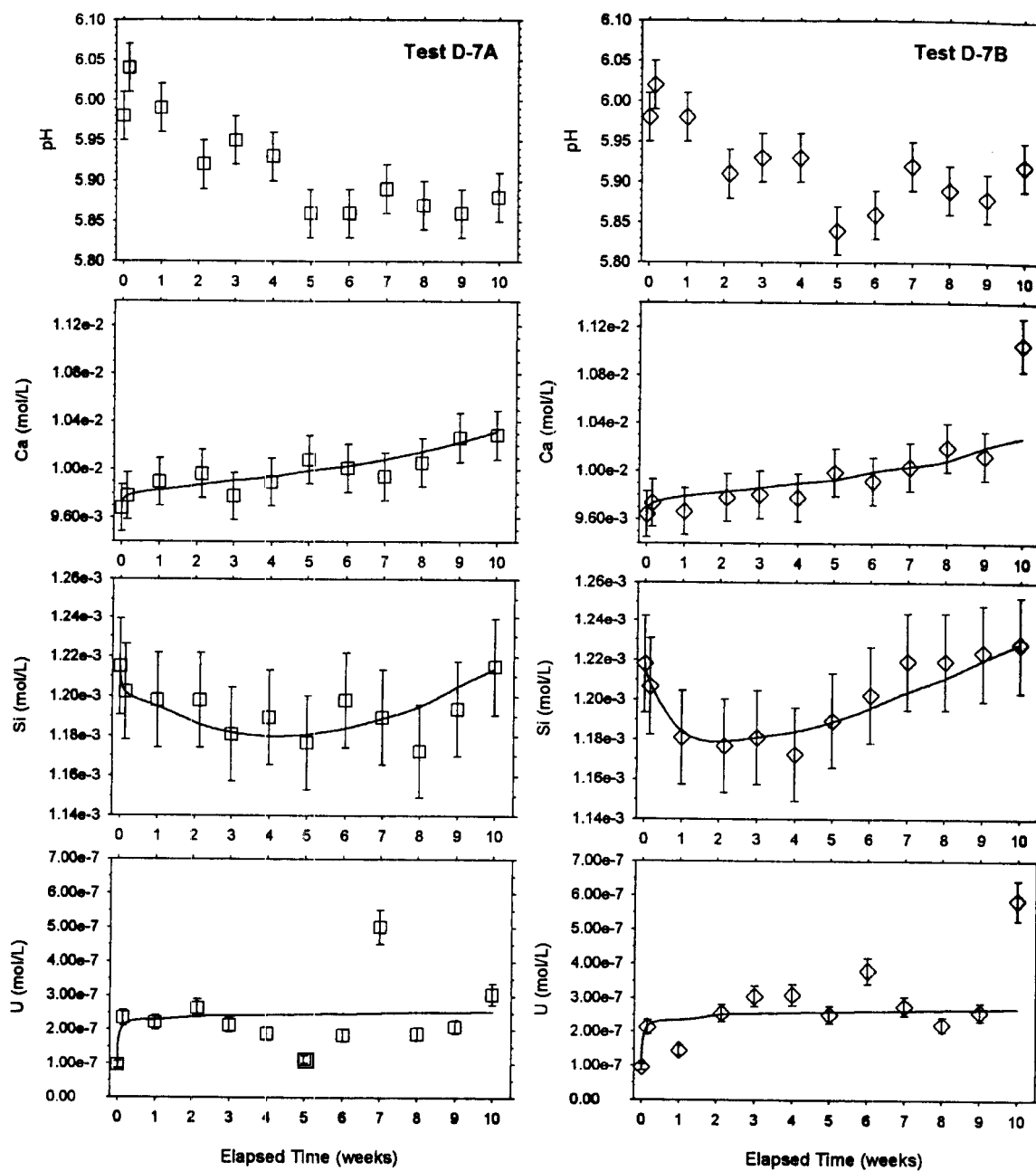


Fig 3. The pH & concentrations of Ca, Si, & U in tests D-7A & D-7B.

11/20/07 JP

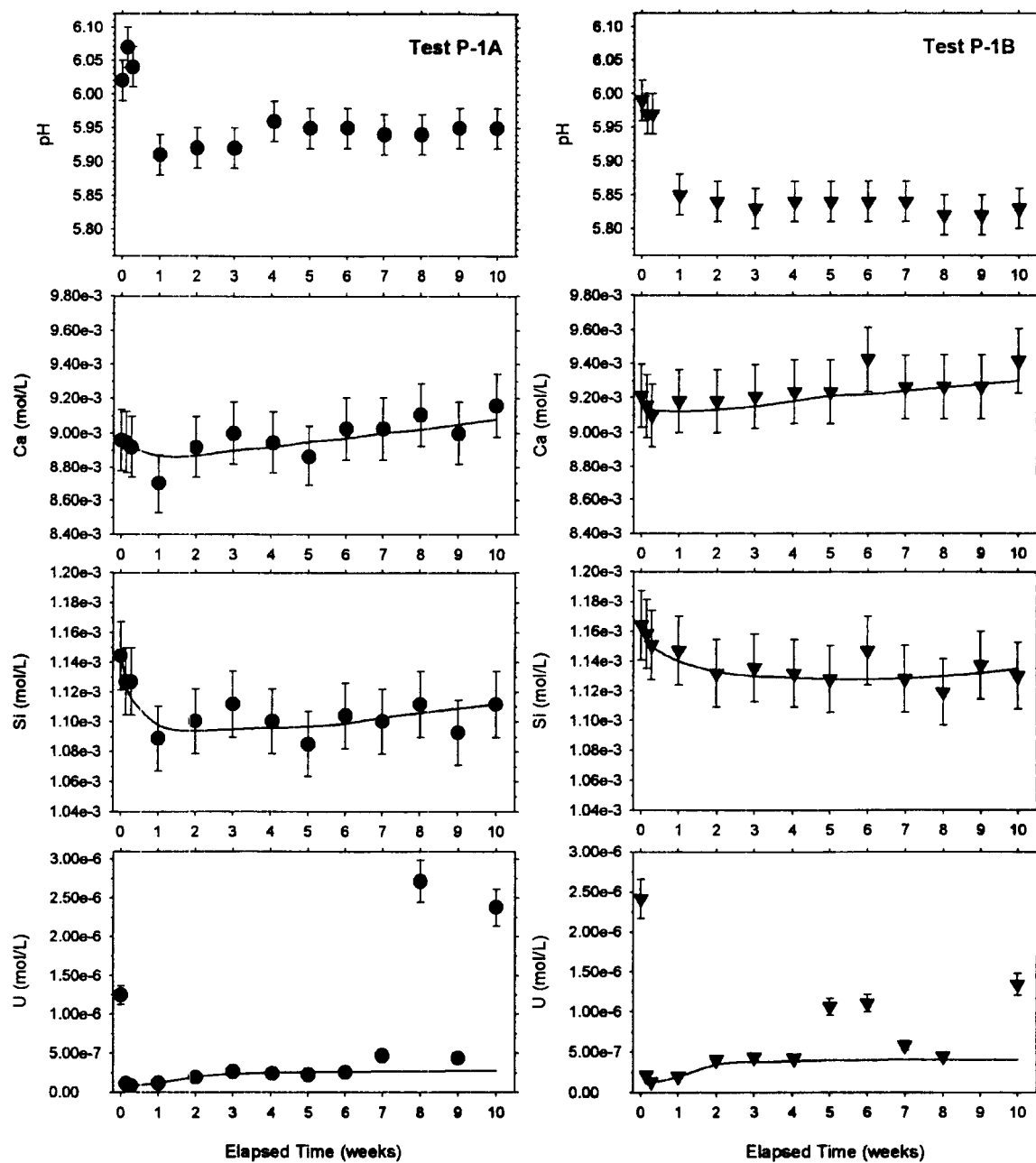


Fig 4. The pH + concentration of Ca, Si, + U
in tests P-1A + P-1B

11/21/07 JP

3) Results Section (cont).

- To address concerns about attainment of equilibrium, regression analyses were performed on measured Ca, Si, + U concentrations data to eliminate some of the uncertainty + variation in the mass transfer data.

Regression analyses were performed using the Regression Wizard in SigmaPlot Ver 8. Before performing the regression, spurious analytical data for Ca and U were removed. For Ca these data included week 3 of test D-0A, + week 10 of D-7B. For U these data included week 4 + 10 of D-0A, week 4 + 7 of D-0B, week 7 of D-7A, and weeks 6 + 10 of D-7B, weeks 3 + 10 of P-1A, and weeks 5, 6, 9, + 10 of P-1B.

Best fit curves for the regression analyses are shown in Figs 2, 3, + 4 on the previous pages.

Based on calculated values from the regression analyses, mass transfer data (i.e., net moles of Ca, Si, + U released to solution) as a function of time were replotted for the duplicate tests in Fig 5 + for the triplicate tests in Fig 6. These figures are shown on the following pages and show good evidence for equilibrium in the latter parts of the tests.

12/13/07 JP

For results of regression analyses applied to mass transfer calculations see page 127.

11/21/07 JP

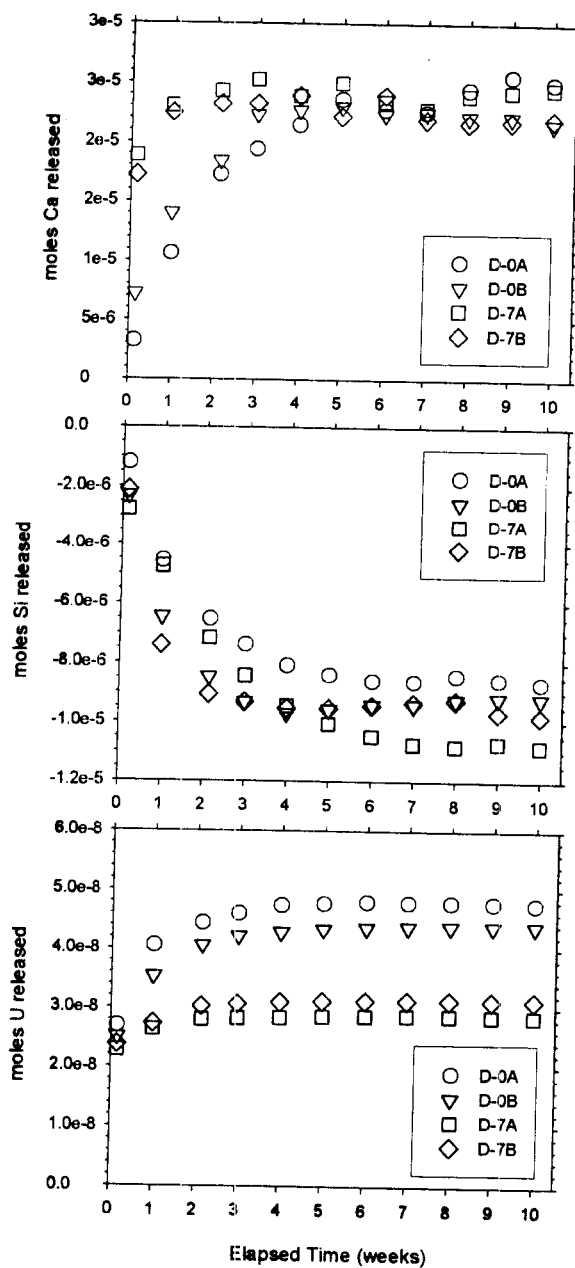


Fig 5. Net moles of Ca, Si, + U released to solution in dissolution tests.

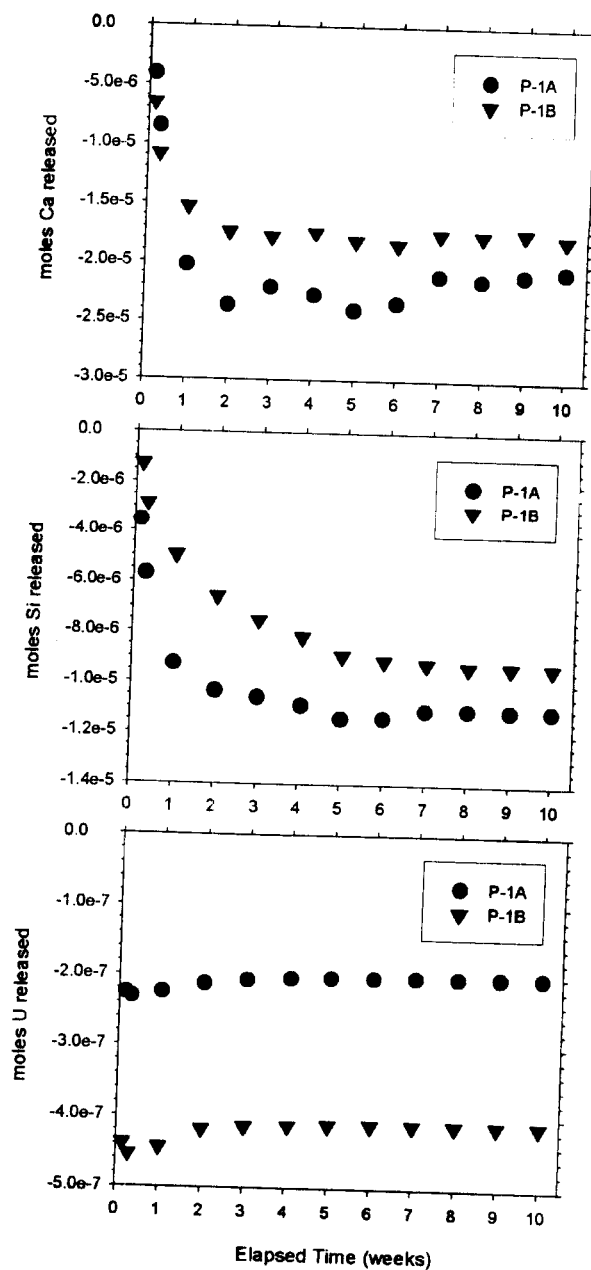


Fig 6. Net moles of Ca, Si, + U released to solution in precipitation tests.

11/28/07 JP

9) Discussion Section

- Thermodynamic Calculated section moved to Discussion Section and shortened. Table 2 removed.

Log Q_s were recalculated using measured test solution pH and calculated Ca, Si, & U concentrations based on the regression analysis. Recalculated log Q_s were replotted as Figure 7 which is shown below.

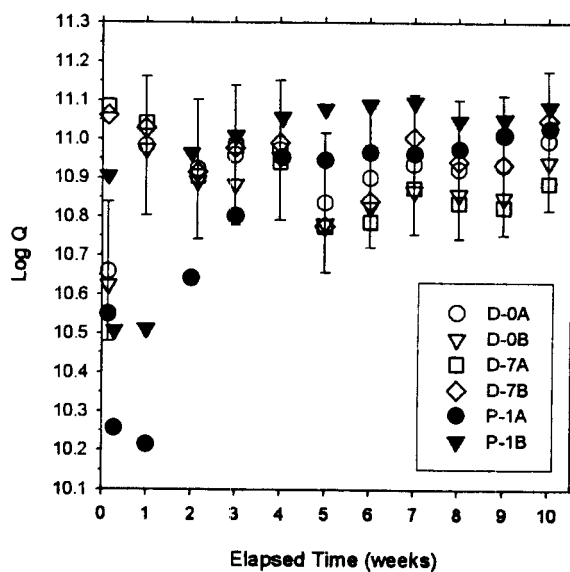


Fig 7. Calculated log Q_s plotted as
 function of time.

Error bar showing possible range in calculated log Q values resulting from error in pH measurement (± 0.03 pH units) are shown for test D-0A in Fig 7.

11/28/07 JP

The possible error range for DOA results from error in pH measurement could be expected for all test solutions.

11/29/07 JP • Evaluation of Solids Precipitation Section (4.2) was revised to include discussion of SEM + XRD characterization of recovered solids.

Results of SEM and XRD analysis of untreated samples and recovered solids from the dissolution + precipitation tests are shown in scientific notebook 582 pages 149-157 and in scientific notebook 823 pages 91-105.

Results of SEM + XRD analysis did not show evidence for strong leaching of samples treated or solids precipitated. A Figure (Figure 8) was included in the paper which detailed results of the SEM analysis. The Figure is shown on the next page.

JP 11/29/07

11/29/07 JP

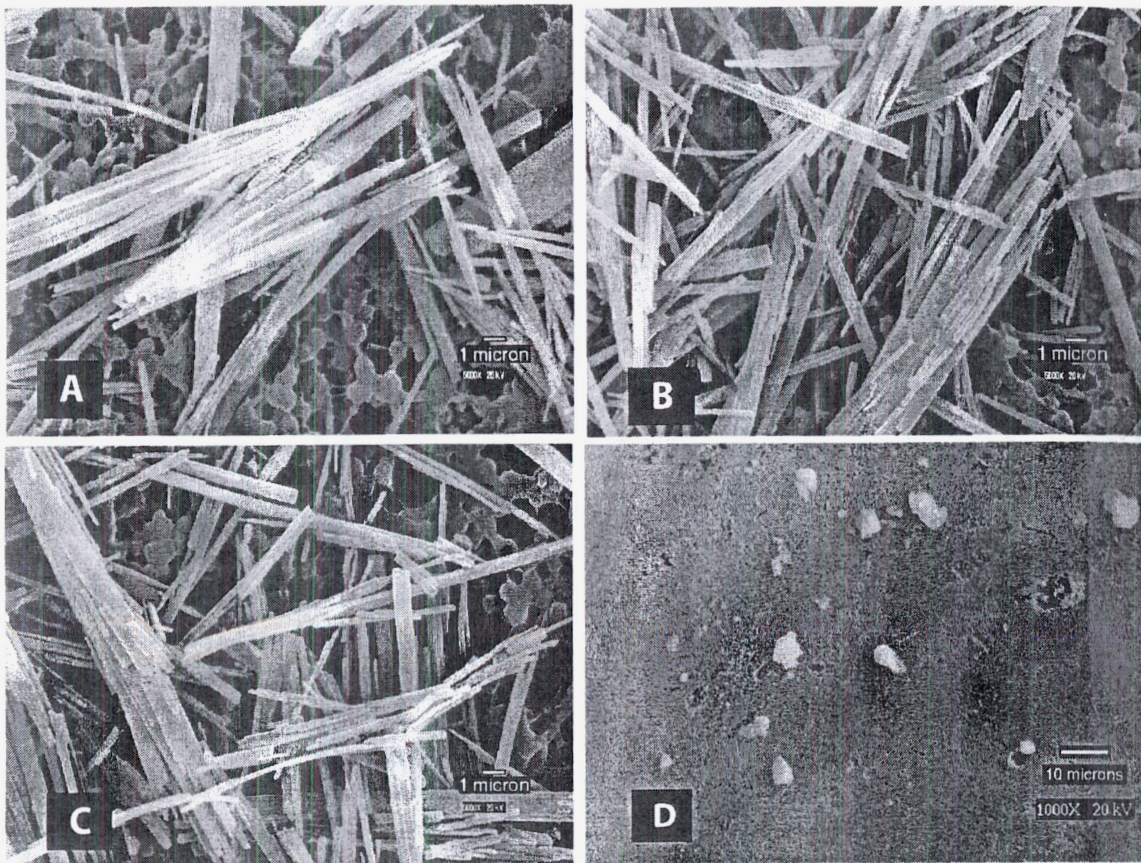
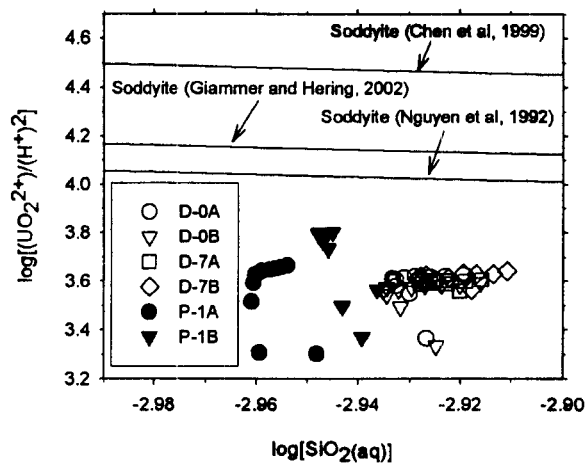


Figure 8. SEM photomicrographs showing (A) unreacted synthetic uranophane, (B) uranophane recovered from test D-0B, (C) uranophane recovered from test P-1A, and (D) particles other than uranophane recovered from test P-1A.

11/29/07 JP

11/30/07 JP

The $\log$ activity diagram of $\text{UO}_2^{2+}/(\text{H}^+)^2$ versus $\text{SiO}_2(\text{aq})$ plotting the points of the test solution with respect to the solubility limit of soddyite was replotted using UO_2^{2+} , H^+ , and $\text{SiO}_2(\text{aq})$ activities derived from the regression analysis. This diagram was replotted in Figure 9 and is shown below.



11/30/07 JP

12/3/07 gp

• Extration of $\log K_{sp}$ for Unphle
Dissolution Section.

- $\log K_{sp}$ recomputed.

For last 5 sampling intervals, range of $\log Q_s$ for unphle in dissolution tests was 10.79 to 11.06 + range $\log Q_s$ in precipitation tests was 10.96 to 11.10. The $\log Q$ midpoint between these ranges is 11.01 and this value was taken as best approximation for the $\log K_{sp}$ for unphle. Standard deviation in $\log Q_s$ was 0.09.

- A table of aqueous phase $\log(VI)$ reaction used to derive the $\log K_{sp}$ was added (Table 1). Reaction & dissociation constants were extracted from the data0.com.R2 database.

- A table (Table 2) showing properties of dominant mineral species in the test solution was added. Data in the table derived from EQ3NR output of measured solution pH and calculated P_a , S_i , & U constants based on the species values.

These tables are shown on the next page.

12/3/07 JF

Table 1. Aqueous phase U(VI) reactions and associated thermodynamic stability constants used to derive $\log K_{sp}$ for uranophane.

Speciation reaction	Log K (I=0) ¹
$UO_2^{2+} + H_2O = UO_2OH^+ + H^+$	-5.21
$UO_2^{2+} + 2H_2O = UO_2(OH)_2(aq) + 2H^+$	-10.31
$UO_2^{2+} + 3H_2O = UO_2(OH)_3^- + 3H^+$	-19.22
$UO_2^{2+} + 4H_2O = UO_2(OH)_4^{2-} + 4H^+$	-33.03
$2UO_2^{2+} + H_2O = (UO_2)_2OH^{3+} + H^+$	-2.71
$2UO_2^{2+} + 2H_2O = (UO_2)_2(OH)_2^{2+} + 2H^+$	-5.63
$3UO_2^{2+} + 4H_2O = (UO_2)_3(OH)_4^{2+} + 4H^+$	-11.93
$3UO_2^{2+} + 5H_2O = (UO_2)_3(OH)_5^+ + 5H^+$	-15.59
$3UO_2^{2+} + 7H_2O = (UO_2)_3(OH)_7^- + 7H^+$	-31.05
$4UO_2^{2+} + 7H_2O = (UO_2)_4(OH)_7^+ + 7H^+$	-21.95
$UO_2^{2+} + HCO_3^- = UO_2CO_3(aq) + H^+$	-0.66
$UO_2^{2+} + 2HCO_3^- = UO_2(CO_3)_2^{2-} + 2H^+$	-3.75
$UO_2^{2+} + 3HCO_3^- = UO_2(CO_3)_3^{4-} + 3H^+$	-9.43
$3UO_2^{2+} + 6HCO_3^- = (UO_2)_3(CO_3)_6^{6-} + 6H^+$	-8.06
$2UO_2^{2+} + HCO_3^- + 3H_2O = (UO_2)_2CO_3(OH)^{3-} + 4H^+$	-11.22
$3UO_2^{2+} + HCO_3^- + 3H_2O = (UO_2)_3O(OH)_2(HCO_3)^+ + 4H^+$	-9.71
$3UO_2^{2+} + HCO_3^- + 4H_2O = (UO_2)_3(OH)_5CO_2^+ + 4H^+$	-9.62
$11UO_2^{2+} + 6HCO_3^- + 12H_2O = (UO_2)_{11}(CO_3)_6(OH)_{12}^{2-} + 18H^+$	-25.73

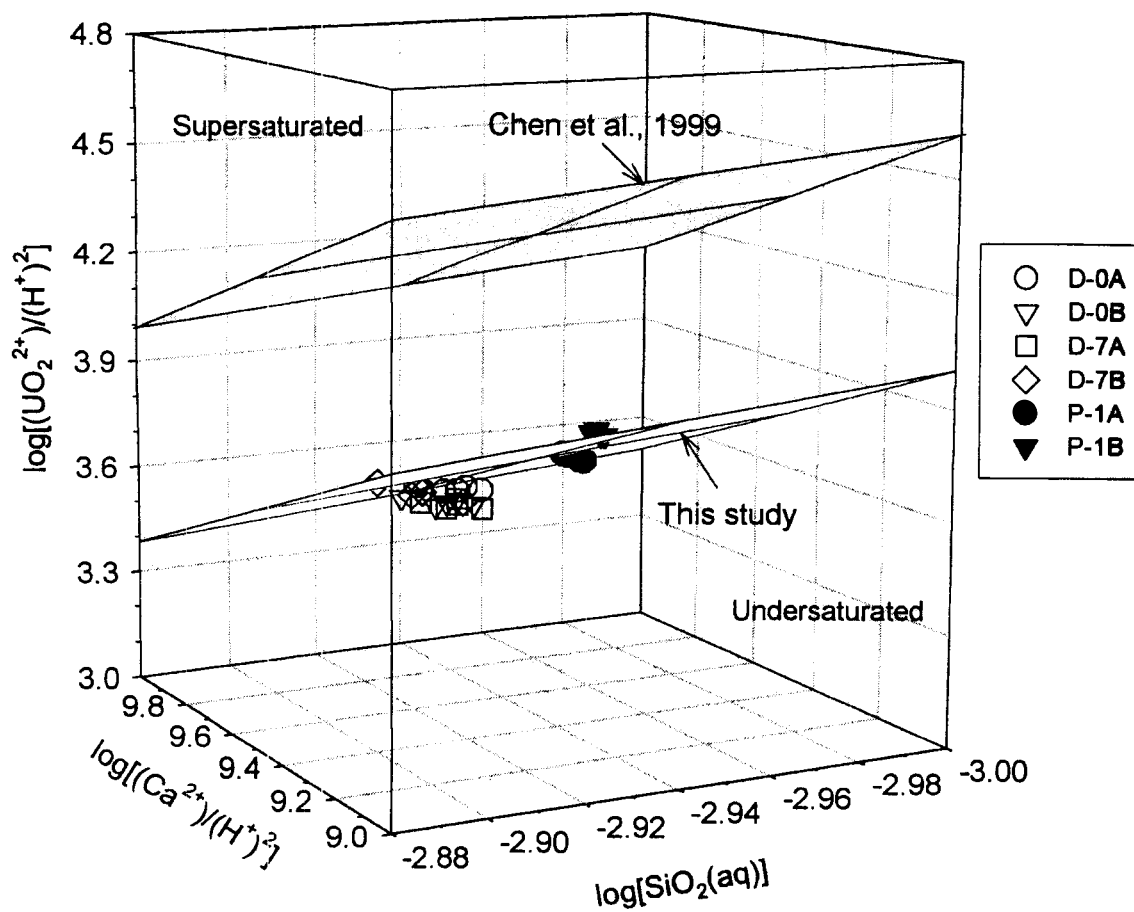
¹Source: Grenthe et al. (1992).

Table 2. Proportions of major uranyl species in dissolution and precipitation test solutions from week 6 to 10.

	$UO_2(OH)_2(aq)$	UO_2OH^+	UO_2^{2+}	$UO_2CO_3(aq)$
<i>Dissolution tests</i>				
D-0A	0.780 – 0.788	0.144 – 0.148	0.045 – 0.049	0.017
D-0B	0.775 – 0.788	0.143 – 0.151	0.045 – 0.051	0.017
D-7A	0.761 – 0.775	0.151 – 0.159	0.051 – 0.057	0.017
D-7B	0.761 – 0.788	0.143 – 0.158	0.045 – 0.057	0.017
<i>Precipitation tests</i>				
P-1A	0.798 – 0.802	0.135 – 0.137	0.039 – 0.040	0.017
P-1B	0.741 – 0.751	0.168 – 0.163	0.061 – 0.066	0.016

12/4/07 JP

- The log activity diagram of $\text{UO}_2^{2+}/(\text{H}^+)^2$ versus $\text{SiO}_2(\text{aq})$ versus $\text{Ca}^{2+}/(\text{H}^+)^2$ was replotted in Figure 10 to reflect recompiled UO_2^{2+} , $\text{SiO}_2(\text{aq})$ & Ca^{2+} activities. Figure 10 is shown below. For last 5 supply runs.



12/10/07 JP

Conclusion section of unaple solubility paper was shortened by removing repeated text.

12/12/07 JP

Revised manuscript on unaple dissolution & growth was placed into Center review.

12/13/07 JP

Results of mass transfer calculations for Ca, Si, and U in the unaple dissolution & precipitation tests using calculated Ca, Si, & U concentrations from regression analyses of measured data are shown on the following pages.

12/13/07 JP

Sample URSE-0A (D-C-A)

Amount of uranophane added = 0.5009 g
 Amount of 0.0 M U solution added = 200.25 g
 Total sample wt = 229.32 g
 Wt of bottle = 28.57 g

Concentrations of components in starting solutions

	mg/L	molarity	moles
U	0	0	0
Ca	381.56	0.00952	0.00190638
Si	33.41	0.001189474	0.000238192

JP 12/13/07
circulated by regression analysis**Sampling data**

Elapsed time (hrs)	Wt of bottle before sampling (g)	Wt of soln before sampling (g)	Wt of bottle after sampling (g)	Wt of soln removed (g)	pH	U (ppm)	Ca (ppm)	Si (ppm)
24.47	229.24	200.17	219.21	10.03	6.03	0.032	382.36	33.26
166.42	217.81	188.74	207.80	10.01	6.01	0.050	386.77	33.01
358.13	207.19	178.12	196.86	10.33	5.95	0.055	389.58	32.81
502.00	196.40	167.33	186.32	10.08	5.95	0.057	391.18	32.75
670.25	185.72	156.65	175.68	10.04	5.94	0.060	393.18	32.75
837.58	175.16	146.09	164.97	10.19	5.88	0.060	395.19	32.81
1007.72	164.33	135.26	154.57	9.76	5.90	0.061	396.79	32.92
1173.60	153.92	124.85	143.97	9.95	5.91	0.061	398.80	33.09
1342.02	143.36	114.29	133.50	9.86	5.90	0.061	401.60	33.31
1509.78	132.68	103.61	122.68	10.00	5.90	0.062	405.21	33.54
1677.83	121.80	92.73	111.86	9.94	5.92	0.062	408.82	33.82

Calculation of cumulative release (moles of U, Ca, Si released or precipitated as a function of time)

Defined by:

Cumulative moles released = (moles in soln at sampling time) + (moles extracted in all samples removed prior to sampling) - (initial moles in soln) where

Moles in soln at sampling time = (measured molar concentration in sample) * (wt of soln before sampling) / 1000

Moles extracted during sampling = (measured molar concentration in sample) * (wt of soln removed during sampling) / 1000

	Elapsed time	Moles in soln at sampling time	Moles extracted during sampling	Moles extracted in all samples removed prior to sampling	Cumulative moles released
U	24.47	2.70228E-08	1.35405E-09	0	2.70228E-08
	166.42	3.92577E-08	2.08208E-09	1.35405E-09	4.06118E-08
	358.13	4.09674E-08	2.3759E-09	3.43613E-09	4.44035E-08
	502.00	4.0159E-08	2.4192E-09	5.81203E-09	4.5971E-08
	670.25	3.91623E-08	2.51E-09	8.23123E-09	4.73935E-08
	837.58	3.6965E-08	2.57807E-09	1.07412E-08	4.77018E-08
	1007.72	3.46263E-08	2.49856E-09	1.33193E-08	4.79456E-08
	1173.60	3.20862E-08	2.55715E-09	1.58179E-08	4.79041E-08
	1342.02	2.94866E-08	2.54388E-09	1.8375E-08	4.78616E-08
	1509.78	2.68348E-08	2.59E-09	2.09189E-08	4.77536E-08
	1677.83	2.41096E-08	2.5844E-09	2.35089E-08	4.76185E-08
Ca	24.47	0.001909613	9.58862E-05	0	3.23321E-06
	166.42	0.001821332	9.65965E-05	9.58862E-05	1.06385E-05
	358.13	0.001731318	0.000100408	0.000192283	1.72204E-05
	502.00	0.001633132	9.83808E-05	0.00029269	1.94423E-05
	670.25	0.001536728	9.84924E-05	0.000391071	2.14188E-05
	837.58	0.001440439	0.000100473	0.000489564	2.3622E-05
	1007.72	0.001339085	9.6624E-05	0.000590037	2.2722E-05
	1173.60	0.001242249	9.90025E-05	0.000686661	2.25294E-05
	1342.02	0.001145177	9.87972E-05	0.000785663	2.44602E-05
	1509.78	0.001047488	0.0001011	0.000884461	2.55686E-05
	1677.83	0.000945837	0.000101388	0.000985561	2.50174E-05
Si	24.47	0.000237	1.18755E-05	0	-1.1919E-06
	166.42	0.000221768	1.17618E-05	1.18755E-05	-4.5482E-06
	358.13	0.000208043	1.20654E-05	2.36373E-05	-6.5117E-06
	502.00	0.000195106	1.17533E-05	3.57027E-05	-7.3837E-06
	670.25	0.000182653	1.17066E-05	4.7456E-05	-8.0833E-06
	837.58	0.000170632	1.19019E-05	5.91626E-05	-8.3974E-06
	1007.72	0.000153524	1.14387E-05	7.10646E-05	-8.6039E-06
	1173.60	0.000147072	1.17211E-05	8.25033E-05	-8.6166E-06
	1342.02	0.000135547	1.1694E-05	9.42244E-05	-8.4209E-06
	1509.78	0.000123709	0.00001194	0.000105918	-8.5645E-06
	1677.83	0.000111646	1.19678E-05	0.000117858	-8.6879E-06

12/13/07 JP

Sample URSE-08 (D-0A)

Amount of uranophane added = 0.5006 g
 Amount of 0.0 M U solution added = 200.06 g
 Total sample wt = 229.17 g
 Wt of bottle = 28.6 g

Concentrations of components in starting solutions

	mg/L	molarity	moles
U	0	0	0
Ca	386.37	0.00964	0.001928578
Si	33.708	0.0012	0.000240072

JP 12/13/07
 calculated from
 regression analysis

Sampling data

Elapsed time (hrs)	Wt of bottle before sampling (g)	Wt of soln before sampling (g)	Wt of bottle after sampling (g)	Wt of soln removed (g)	pH	U (ppm)	Ca (ppm)	Si (ppm)
24.50	229.08	199.98	218.79	10.29	6.04	0.030	387.97	33.40
166.42	218.32	189.22	208.28	10.04	6.06	0.043	390.38	32.87
358.15	207.63	178.53	197.30	10.33	5.97	0.050	392.78	32.67
502.02	196.84	167.74	186.80	10.04	5.95	0.052	394.79	32.61
670.27	186.20	157.10	176.17	10.03	5.97	0.054	396.39	32.67
837.60	175.54	146.44	165.55	9.99	5.89	0.055	398.19	32.84
1007.73	164.87	135.77	155.12	9.75	5.90	0.055	400.00	33.03
1173.62	154.38	125.28	144.44	9.94	5.91	0.056	402.40	33.23
1342.03	143.72	114.82	133.85	9.87	5.90	0.057	404.81	33.48
1509.80	133.03	103.93	123.10	9.93	5.89	0.057	408.01	33.76
1677.85	122.22	93.12	112.28	9.94	5.92	0.058	411.62	34.07

Calculation of cumulative release (moles of U, Ca, Si released or precipitated as a function of time)

Defined by:

Cumulative moles released = (moles in soln at sampling time) + (moles extracted in all samples removed prior to sampling) - (initial moles in soln) where

Moles in soln at sampling time = (measured molar concentration in sample) * (wt of soln before sampling) / 1000

Moles extracted during sampling = (measured molar concentration in sample) * (wt of soln removed during sampling) / 1000

	Elapsed time	Moles in soln at sampling time	Moles extracted during sampling	Moles extracted in all samples removed prior to sampling	Cumulative moles released
U	24.50	2.49974E-08	1.28625E-09	0	2.49974E-08
	166.42	3.40595E-08	1.8072E-09	1.28625E-09	3.53457E-08
	358.15	3.74912E-08	2.1693E-09	3.09345E-09	4.05846E-08
	502.02	3.69027E-08	2.2088E-09	5.26275E-09	4.21654E-08
	670.27	3.53474E-08	2.25675E-09	7.47155E-09	4.28189E-08
	837.60	3.36811E-08	2.2977E-09	9.7283E-09	4.34094E-08
	1007.73	3.16343E-08	2.27175E-09	1.2026E-08	4.36603E-08
	1173.62	2.95659E-08	2.34584E-09	1.42978E-08	4.38637E-08
	1342.03	2.72794E-08	2.34906E-09	1.66436E-08	4.3923E-08
	1509.80	2.49431E-08	2.3832E-09	1.89927E-08	4.39357E-08
	1677.85	2.25349E-08	2.40548E-09	2.13759E-08	4.39107E-08
Ca	24.50	0.001935801	9.96072E-05	0	7.22219E-06
	166.42	0.001842997	9.77896E-05	9.96072E-05	1.40258E-05
	358.15	0.001749588	0.000101234	0.000197397	1.84065E-05
	502.02	0.001652233	9.8894E-05	0.000298631	2.22855E-05
	670.27	0.001553713	9.91967E-05	0.000397525	2.26595E-05
	837.60	0.001454875	9.92506E-05	0.000496722	2.30185E-05
	1007.73	0.001354979	0.000097305	0.000595972	2.23724E-05
	1173.62	0.001257805	9.97976E-05	0.000693277	2.25039E-05
	1342.03	0.001157656	0.000099687	0.000793075	2.21523E-05
	1509.80	0.001058001	0.000101087	0.000892762	2.21846E-05
	1677.85	0.000956336	0.000102084	0.000993849	2.1607E-05
Si	24.50	0.000237776	1.22348E-05	0	-2.29649E-06
	166.42	0.000221387	1.17468E-05	1.22348E-05	-6.45049E-06
	358.15	0.00020763	1.20138E-05	2.39816E-05	-8.4807E-06
	502.02	0.000194745	1.16564E-05	3.59954E-05	-9.33116E-06
	670.27	0.000182707	1.16649E-05	4.76518E-05	-9.71356E-06
	837.60	0.000171188	1.16783E-05	5.93167E-05	-9.56781E-06
	1007.73	0.000159665	0.000011466	7.0995E-05	-9.41215E-06
	1173.62	0.000148206	1.1759E-05	8.2461E-05	-9.40543E-06
	1342.03	0.000136626	1.1765E-05	9.42201E-05	-9.22562E-06
	1509.80	0.000124923	1.19359E-05	0.000105985	-9.16376E-06
	1677.85	0.000112954	1.20572E-05	0.000117921	-9.19721E-06

12/13/07 JP

Sample UPEA-1A (P-1A)

Amount of uranophane added = 0.5002 g
 Amount of matrix solution added = 200.16 g
 Total sample wt = 229.21 g
 Wt of bottle = 28.54 g

Concentrations of components in starting solutions

	mg/L	molality	moles
U	0.29585916	1.24295E-06	2.48789E-07
Ca	359.02	0.008957477	0.001792929
Si	32.14	0.001144315	0.000229046

Sampling data

Elapsed time (hrs)	Wt of bottle before sampling (g)	Wt of soln before sampling (g)	Wt of bottle after sampling (g)	Wt of soln removed (g)	pH	U (ppm)	Ca (ppm)	Si (ppm)
23.42	229.13	200.09	218.17	10.96	6.07	0.027	358.32	31.66
47.30	218.10	189.06	207.38	10.72	6.04	0.021	357.51	31.35
167.75	207.06	178.02	196.80	10.26	5.91	0.030	355.51	30.84
335.60	196.42	167.38	186.43	9.99	5.92	0.048	355.51	30.73
503.70	186.07	157.03	176.03	10.04	5.92	0.057	356.71	30.76
679.23	175.63	146.59	165.65	9.98	5.96	0.061	357.51	30.79
839.50	165.05	136.01	155.05	10.00	5.95	0.062	358.72	30.81
1007.58	154.84	125.80	144.91	9.93	5.95	0.063	359.52	30.87
1175.52	144.78	115.74	134.83	9.95	5.94	0.064	360.72	30.98
1344.10	134.56	105.52	124.58	9.98	5.94	0.065	361.52	31.07
1512.95	124.31	95.27	114.35	9.96	5.95	0.066	362.72	31.15
1680.08	114.11	85.07	104.10	10.01	5.95	0.067	363.93	31.24

Calculation of cumulative release (moles of U, Ca, Si released or precipitated as a function of time)

Defined by:

Cumulative moles released = (moles in soln at sampling time) + (moles extracted in all samples removed prior to sampling) - (initial moles in soln) where

Moles in soln at sampling time = (measured molar concentration in sample) * (wt of soln before sampling) / 1000

Moles extracted during sampling = (measured molar concentration in sample) * (wt of soln removed during sampling) / 1000

	Elapsed time	Moles in soln at sampling time	Moles extracted during sampling	Moles extracted in all samples removed prior to sampling	Cumulative moles released
U	23.42	2.30103E-08	1.2604E-09	0	-2.25778E-07
U	47.30	1.70154E-08	9.648E-10	1.2604E-09	-2.30513E-07
U	167.75	2.22525E-08	1.2825E-09	2.2252E-09	-2.24311E-07
U	335.60	3.3476E-08	1.998E-09	3.5077E-09	-2.11805E-07
U	503.70	3.76872E-08	2.4096E-09	5.5057E-09	-2.05596E-07
U	679.23	3.73804E-08	2.5449E-09	7.9153E-09	-2.03493E-07
U	839.50	3.53625E-08	2.6E-09	1.04602E-08	-2.02966E-07
U	1007.58	3.33369E-08	2.63145E-09	1.30602E-08	-2.02392E-07
U	1175.52	3.12497E-08	2.6865E-09	1.56917E-08	-2.01847E-07
U	1344.10	2.88069E-08	2.72454E-09	1.83782E-08	-2.01604E-07
U	1512.95	2.62945E-08	2.74896E-09	2.11027E-08	-2.01392E-07
U	1680.08	2.38195E-08	2.8028E-09	2.38517E-08	-2.01117E-07
Ca	23.42	0.001788803	9.79824E-05	0	-4.12572E-06
Ca	47.30	0.001686413	9.56224E-05	9.79824E-05	-8.53271E-06
Ca	167.75	0.001579036	9.10062E-05	0.000193605	-2.02881E-05
Ca	335.60	0.001484659	8.86113E-05	0.000284611	-2.36587E-05
Ca	503.70	0.001397565	8.9356E-05	0.000373222	-2.2141E-05
Ca	679.23	0.001307581	8.90216E-05	0.000462578	-2.27692E-05
Ca	839.50	0.001217288	0.0000895	0.0005516	-2.40409E-05
Ca	1007.58	0.001128424	8.90721E-05	0.0006411	-2.34044E-05
Ca	1175.52	0.001041658	8.955E-05	0.000730172	-2.10983E-05
Ca	1344.10	0.000951789	9.00196E-05	0.000819722	-2.14179E-05
Ca	1512.95	0.000862192	9.0138E-05	0.000909742	-2.09952E-05
Ca	1680.08	0.000772434	9.08908E-05	0.00099988	-2.06151E-05
Si	23.42	0.000225501	1.23519E-05	0	-3.54483E-06
Si	47.30	0.000210991	1.19635E-05	1.23519E-05	-5.70338E-06
Si	167.75	0.000195466	1.12655E-05	2.43154E-05	-9.26485E-06
Si	335.60	0.000183114	1.09291E-05	3.55809E-05	-1.03516E-05
Si	503.70	0.000171948	1.09938E-05	4.651E-05	-1.05884E-05
Si	679.23	0.000160662	1.09381E-05	5.75038E-05	-1.08798E-05
Si	839.50	0.000149203	0.00001097	6.84419E-05	-1.14014E-05
Si	1007.58	0.000138254	1.09131E-05	7.94119E-05	-1.13802E-05
Si	1175.52	0.000127661	1.09749E-05	9.03249E-05	-1.10601E-05
Si	1344.10	0.000116705	1.10379E-05	0.0001013	-1.10414E-05
Si	1512.95	0.000105654	1.10456E-05	0.000112338	-1.10542E-05
Si	1680.08	9.45976E-05	1.11311E-05	0.000123383	-1.10651E-05

12/13/07 JP

Sample UPEA-1B

P-15

Amount of uranophane added = 0.5004 g
 Amount of 0.0 M U solution added = 200.16 g
 Total sample wt = 229.27 g
 Wt of bottle = 28.59 g

Concentrations of components in
 starting solutions

	mg/L	molality	moles
U	0.574434443	2.41329E-06	4.83043E-07
Ca	367.93	0.00918	0.001837469
Si	32.69676	0.001164	0.000232986

Sampling data

Elapsed time (hrs)	Wt of bottle before sampling (g)	Wt of soln before sampling (g)	Wt of bottle after sampling (g)	Wt of soln removed (g)	pH	U (ppm)	Ca (ppm)	Si (ppm)
23.43	229.19	200.10	218.20	10.99	5.97	0.052	366.73	32.53
47.27	218.14	189.05	207.43	10.71	5.97	0.033	365.93	32.30
167.72	207.14	178.05	197.09	10.05	5.85	0.048	365.53	32.02
335.55	196.68	167.59	186.66	10.02	5.84	0.083	365.93	31.83
503.72	186.27	157.18	176.23	10.04	5.83	0.090	366.73	31.74
679.20	175.79	146.70	165.79	10.00	5.84	0.093	367.93	31.71
839.48	165.27	136.18	155.23	10.04	5.84	0.095	369.14	31.69
1007.52	155.06	125.97	145.12	9.94	5.84	0.096	369.54	31.69
1175.53	144.97	115.88	135.05	9.92	5.84	0.097	370.34	31.70
1344.15	134.81	105.72	124.57	10.24	5.82	0.097	371.14	31.74
1512.90	124.39	95.30	114.43	9.96	5.82	0.098	371.94	31.80
1680.10	114.19	85.10	104.24	9.95	5.83	0.098	372.74	31.88

Calculation of cumulative release (moles of U, Ca, Si released or precipitated as a function of time)

Defined by:

Cumulative moles released = (moles in soln at sampling time) + (moles extracted in all samples removed prior to sampling) - (initial moles in soln) where

Moles in soln at sampling time = (measured molar concentration in sample) * (wt of soln before sampling) / 1000

Moles extracted during sampling = (measured molar concentration in sample) * (wt of soln removed during sampling) / 1000

	Elapsed time	Moles in soln at sampling time	Moles extracted during sampling	Moles extracted in all samples removed prior to sampling	Cumulative moles released
U	23.43	4.40219E-08	2.4178E-09	0	-4.39021E-07
U	47.27	2.64669E-08	1.4994E-09	2.4178E-09	-4.54159E-07
U	167.72	3.56099E-08	2.01E-09	3.9172E-09	-4.43516E-07
U	335.55	5.86564E-08	3.507E-09	5.9272E-09	-4.1846E-07
U	503.72	5.97282E-08	3.8152E-09	9.4342E-09	-4.13881E-07
U	679.20	5.72128E-08	3.9E-09	1.32494E-08	-4.12581E-07
U	839.48	5.44718E-08	4.016E-09	1.71494E-08	-4.11422E-07
U	1007.52	5.10177E-08	4.0257E-09	2.11654E-08	-4.1086E-07
U	1175.53	4.72209E-08	4.0424E-09	2.51911E-08	-4.10631E-07
U	1344.15	4.32393E-08	4.18816E-09	2.92335E-08	-4.1057E-07
U	1512.90	3.90728E-08	4.0836E-09	3.34217E-08	-4.10549E-07
U	1680.10	3.49759E-08	4.08945E-09	3.75053E-08	-4.10562E-07
Ca	23.43	0.001830911	0.000100559	0	-6.55746E-06
Ca	47.27	0.001726023	9.77823E-05	0.000100559	-1.08875E-05
Ca	167.72	0.001623812	9.1656E-05	0.000198341	-1.53156E-05
Ca	335.55	0.001530093	9.14826E-05	0.000289997	-1.7379E-05
Ca	503.72	0.001438193	9.1866E-05	0.000381479	-1.77961E-05
Ca	679.20	0.001346702	0.0000918	0.000473345	-1.74211E-05
Ca	839.48	0.001254214	9.24684E-05	0.000565145	-1.81093E-05
Ca	1007.52	0.00116144	9.16468E-05	0.000657614	-1.84153E-05
Ca	1175.53	0.001070728	9.16608E-05	0.000749261	-1.74807E-05
Ca	1344.15	0.000978963	9.48224E-05	0.000840921	-1.75839E-05
Ca	1512.90	0.00088436	9.24288E-05	0.000935744	-1.73447E-05
Ca	1680.10	0.000791426	0.00092535	0.001028173	-1.78699E-05
Si	23.43	0.000231715	1.27264E-05	0	-1.2709E-06
Si	47.27	0.000217407	1.23165E-05	1.27264E-05	-2.85278E-06
Si	167.72	0.000202977	0.000011457	2.50429E-05	-4.96678E-06
Si	335.55	0.000189879	1.13527E-05	3.64999E-05	-6.6073E-06
Si	503.72	0.000177613	1.13452E-05	4.78526E-05	-7.52071E-06
Si	679.20	0.000165624	0.00001129	5.91978E-05	-8.16461E-06
Si	839.48	0.000153611	1.13251E-05	7.04878E-05	-8.88787E-06
Si	1007.52	0.000142094	1.12123E-05	8.18129E-05	-9.07963E-06
Si	1175.53	0.00013077	1.11947E-05	9.30252E-05	-9.19089E-06
Si	1344.15	0.000119463	1.15712E-05	0.00010422	-9.30315E-06
Si	1512.90	0.000107879	1.12747E-05	0.000115791	-9.31595E-06
Si	1680.10	9.6588E-05	1.12933E-05	0.000127066	-9.33233E-06

12/13/07 JP

Sample URSE-7A D-7A

Amount of uranophane added = 0.5004 g
 Amount of 0.0 M U solution added 200.04 g
 Total sample wt = 229.15 g
 Wt of bottle = 28.62 g

Concentrations of components in starting solutions

	mg/L	molality	moles
U	0.022886139	9.61481E-08	1.92335E-08
Ca	387.97	0.00968	0.001936387
Si	34.13086134	0.001215054	0.000243059

Sampling data

Elapsed time (hrs)	Wt of bottle before sampling (g)	Wt of soln before sampling (g)	Wt of bottle after sampling (g)	Wt of soln removed (g)	pH	U (ppm)	Ca (ppm)	Si (ppm)
24.75	229.04	199.92	219.07	9.97	6.04	0.050	391.98	33.78
166.62	218.53	189.41	208.53	10.00	5.99	0.055	393.99	33.57
358.35	207.84	178.72	197.83	10.01	5.92	0.057	395.79	33.31
502.18	197.33	168.21	187.32	10.01	5.95	0.058	397.19	33.20
670.48	186.71	157.59	176.69	10.02	5.93	0.058	398.40	33.15
837.78	176.06	146.94	166.03	10.03	5.86	0.059	400.40	33.17
1007.97	165.33	136.21	155.57	9.76	5.86	0.059	402.00	33.28
1173.83	154.82	125.70	144.89	9.93	5.89	0.060	404.21	33.40
1342.25	144.26	115.14	134.39	9.87	5.87	0.060	406.81	33.57
1510.07	133.60	104.48	123.68	9.92	5.86	0.060	410.02	33.85
1678.15	122.88	93.76	112.90	9.98	5.88	0.060	413.63	34.10

Calculation of cumulative release (moles of U, Ca, Si released or precipitated as a function of time)

Defined by:

Cumulative moles released = (moles in soln at sampling time) + (moles extracted in all samples removed prior to sampling) - (initial moles in soln) where

Moles in soln at sampling time = (measured molar concentration in sample) * (wt of soln before sampling) / 1000

Moles extracted during sampling = (measured molar concentration in sample) * (wt of soln removed during sampling) / 1000

	Elapsed time	Moles in soln at sampling time	Moles extracted during sampling	Moles extracted in all samples removed prior to sampling	Cumulative moles released
U	24.75	4.19831E-08	2.0937E-09	0	2.27496E-08
U	166.62	4.35642E-08	2.3E-09	2.0937E-09	2.64244E-08
U	358.35	4.28927E-08	2.4024E-09	4.3937E-09	2.80529E-08
U	502.18	4.07067E-08	2.42242E-09	6.7961E-09	2.82694E-08
U	670.48	3.84519E-08	2.44488E-09	9.21852E-09	2.84369E-08
U	837.78	3.61471E-08	2.46738E-09	1.16634E-08	2.85771E-08
U	1007.97	3.378E-08	2.42048E-09	1.41308E-08	2.86773E-08
U	1173.83	3.14249E-08	2.4825E-09	1.65513E-08	2.87427E-08
U	1342.25	2.90152E-08	2.48724E-09	1.90338E-08	2.88155E-08
U	1510.07	2.64333E-08	2.50976E-09	2.1521E-08	2.87209E-08
U	1678.15	2.38149E-08	2.53492E-09	2.40308E-08	2.86122E-08
Ca	24.75	0.001955214	9.75066E-05	0	1.88265E-05
Ca	166.62	0.001861896	0.0000983	9.75066E-05	2.30158E-05
Ca	358.35	0.001764856	9.88487E-05	0.000195807	2.42755E-05
Ca	502.18	0.001666957	9.91991E-05	0.000294655	2.52253E-05
Ca	670.48	0.001566441	9.95988E-05	0.000393854	2.59079E-05
Ca	837.78	0.001467927	0.0001002	0.000493453	2.49927E-05
Ca	1007.97	0.001366182	9.78928E-05	0.000593653	2.3448E-05
Ca	1173.83	0.00126768	0.000100144	0.000691546	2.2839E-05
Ca	1342.25	0.001168667	0.000100181	0.00079169	2.39695E-05
Ca	1510.07	0.001068826	0.000101482	0.00089187	2.43094E-05
Ca	1678.15	0.000967599	0.000102994	0.000993352	2.45638E-05
Si	24.75	0.000240303	1.19839E-05	0	-2.756E-06
Si	166.62	0.000226344	0.00001195	1.19839E-05	-4.73095E-06
Si	358.35	0.000211961	1.18719E-05	2.39339E-05	-7.16398E-06
Si	502.18	0.000198824	1.18318E-05	3.58058E-05	-8.42982E-06
Si	670.48	0.000185956	1.18236E-05	4.76376E-05	-9.46601E-06
Si	837.78	0.000173536	1.18454E-05	5.94612E-05	-1.00625E-05
Si	1007.97	0.000161272	1.15558E-05	7.13067E-05	-1.04805E-05
Si	1173.83	0.000149457	1.18068E-05	8.28625E-05	-1.074E-05
Si	1342.25	0.000137592	1.17947E-05	9.46693E-05	-1.07983E-05
Si	1510.07	0.000125898	1.19536E-05	0.000106464	-1.06975E-05
Si	1678.15	0.000113824	1.21157E-05	0.000118418	-1.08177E-05

12/13/07 JJB

Sample URSE-7B (D-7B)

Amount of uranophane 0.5008 g
 Amount of 0.0 M U's 200.02 g
 Total sample wt = 229.11 g
 Wt of bottle = 28.58 g

Concentrations of components in
 starting solutions

	mg/L	molarity	moles
U	0.022886139	9.61481E-08	1.92315E-08
Ca	386.37	0.00964	0.001928193
Si	34.21362	0.001218	0.000243624

Sampling data	Elapsed time (hrs)	Wt of bottle before sampling (g)	Wt of soln before sampling (g)	Wt of bottle after sampling (g)	Wt of soln removed (g)	pH	U (ppm)	Ca (ppm)	Si (ppm)
	24.77	229.02	199.94	219.05	9.97	6.02	0.051	389.98	33.93
	166.62	218.42	189.34	208.12	10.30	5.98	0.056	392.38	33.26
	358.37	207.47	178.39	197.47	10.00	5.91	0.060	393.99	33.12
	502.20	196.96	167.88	186.95	10.01	5.93	0.061	395.19	33.17
	670.50	186.38	157.30	176.37	10.01	5.93	0.061	396.79	33.26
	837.80	175.75	146.67	165.74	10.01	5.84	0.062	397.99	33.40
	1007.98	165.02	135.94	155.26	9.76	5.86	0.062	400.64	33.60
	1173.85	154.50	125.42	144.61	9.89	5.92	0.063	402.40	33.82
	1342.27	144.01	114.93	134.15	9.86	5.89	0.063	404.41	34.02
	1510.08	133.03	103.95	123.12	9.91	5.88	0.064	408.82	34.27
	1678.17	122.41	93.33	112.45	9.96	5.92	0.064	412.02	34.49

Calculation of cumulative release (moles of U, Ca, Si released or precipitated as a function of time)

Defined by:

Cumulative moles released = (moles in soln at sampling time) + (moles extracted in all samples removed prior to sampling) - (initial moles in soln)

where

Moles in soln at sampling time = (measured molar concentration in sample) * (wt of soln before sampling) / 1000

Moles extracted during sampling = (measured molar concentration in sample) * (wt of soln removed during sampling) / 1000

	Elapsed time	Moles in soln at sampling time	Moles extracted during sampling	Moles extracted in all samples removed prior to sampling	Cumulative moles released
U					
	24.77	4.29869E-08	2.14355E-09	0	2.3755E-08
	166.62	4.44947E-08	2.4205E-09	2.14355E-09	2.7407E-08
	358.37	4.49541E-08	2.52E-09	4.56405E-09	3.0287E-08
	502.20	4.28092E-08	2.55255E-09	7.08405E-09	3.0662E-08
	670.50	4.05832E-08	2.58258E-09	9.6366E-09	3.0988E-08
	837.80	3.8134E-08	2.6026E-09	1.22192E-08	3.1122E-08
	1007.98	3.56161E-08	2.55712E-09	1.48218E-08	3.1206E-08
	1173.85	3.31107E-08	2.61096E-09	1.73789E-08	3.1258E-08
	1342.27	3.05712E-08	2.62276E-09	1.99899E-08	3.1329E-08
	1510.08	2.78584E-08	2.65588E-09	2.26126E-08	3.1239E-08
	1678.17	2.51989E-08	2.6892E-09	2.52685E-08	3.1236E-08
Ca					
	24.77	0.001945408	9.70081E-05	0	1.7216E-05
	166.62	0.001853631	0.000100837	9.70081E-05	2.2446E-05
	358.37	0.001753566	0.0000983	0.000197845	2.3218E-05
	502.20	0.001655289	9.86986E-05	0.000296145	2.3241E-05
	670.50	0.001557262	9.9099E-05	0.000394844	2.3913E-05
	837.80	0.001456425	9.93993E-05	0.000493943	2.275E-05
	1007.98	0.001358848	9.7561E-05	0.000593342	2.3997E-05
	1173.85	0.001259209	9.92956E-05	0.000690903	2.1919E-05
	1342.27	0.001159636	9.94874E-05	0.000790199	2.1641E-05
	1510.08	0.001060282	0.000101082	0.000889686	2.1775E-05
	1678.17	0.000959424	0.000102389	0.000990768	2.1999E-05
Si					
	24.77	0.000241527	1.20438E-05	0	-2.0978E-06
	166.62	0.000224178	1.21952E-05	1.20438E-05	-7.403E-06
	358.37	0.000210321	0.0001179	2.4239E-05	-9.0645E-06
	502.20	0.000198265	1.18218E-05	3.6029E-05	-9.3301E-06
	670.50	0.000186242	1.18518E-05	4.78508E-05	-9.5313E-06
	837.80	0.00017439	1.19019E-05	5.97026E-05	-9.5321E-06
	1007.98	0.000162583	1.1673E-05	7.16045E-05	-9.4366E-06
	1173.85	0.000151005	1.19076E-05	8.32775E-05	-9.3422E-06
	1342.27	0.000139179	1.19405E-05	9.5185E-05	-9.2601E-06
	1510.08	0.000126818	1.20502E-05	0.000107125	-9.6809E-06
	1678.17	0.000114608	1.22309E-05	0.000119216	-9.8004E-06

No further entries

E.C. Par

9/10/2009