Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Wilmarth, W. R. , Mills, J. T. , Dukes, V. H. and Sullivan, R. C.(2006) 'Effects of In-Tank Precipitation of Sodium Aluminosilicate on Uranium Chemistry', Separation Science and Technology, 41: 11, 2325 — 2340
To link to this Article: DOI: 10.1080/01496390600743011
URL: http://dx.doi.org/10.1080/01496390600743011

Separation Science and Technology, 41: 2325–2340, 2006

ISSN 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390600743011

Effects of In-Tank Precipitation of Sodium Aluminosilicate on Uranium Chemistry

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Abstract: Experiments have been conducted to examine the fate of uranium during the formation of sodium aluminosilicate (NAS) when wastes containing high aluminate concentrations are mixed with wastes of high silicate concentration. Testing was conducted at varying degrees of uranium saturation. Testing examined typical tank conditions, e.g., stagnant, slightly elevated temperature (50°C). The results showed that under sub-saturated conditions uranium is not removed from solution to any large extent in both simulant testing and actual tank waste testing. This aspect was not thoroughly understood prior to this work and was necessary to avoid criticality issues when actual tank wastes were aggregated. There are data supporting a small removal due to sorption of uranium on sites in the NAS. Above the solubility limit the data are clear that a reduction in uranium concentration occurs concomitant with the formation of aluminosilicate. This uranium precipitation is fairly rapid and ceases when uranium reaches its solubility limit. At the solubility limit, it appears that uranium is not affected, but further testing might be warranted.

Keywords: Occulusion, solubility, speciation

INTRODUCTION

The Savannah River Site (SRS) stores high-level nuclear waste in 49 underground storage tanks. The wastes are to be vitrified in the defense waste processing facility (DWPF) for permanent disposal. The available tank space must be managed to ensure viability of the separation canyons to support

Received 23 October 2005, Accepted 6 March 2006
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nuclear material stabilization and continued operation of DWPF. Under normal operations, the wastes are evaporated to reduce volume. The SRS has three operational atmospheric-pressure high-level waste evaporators. One of these evaporators had not operated from October 1999 to September 2001 due to the presence of a large amount of sodium aluminosilicate scale that contained sodium diuranate (1–3). The scale is very similar to that observed in the aluminum and pulp paper industries (4–6) and was produced at SRS by reaction of the aluminate supplied by the plutonium separations facilities and the silicate from recycle water from the DWPF. The chemistry of high level waste with elevated silicon levels thermodynamically favors the formation of aluminosilicates (7).

As a result of the formation of aluminosilicates when elevated concentrations of silica are a concern, SRS changed the operational requirements for the site's high-level waste evaporators. Wastes containing high silicon concentrations, e.g., DWPF recycle, would be concentrated in a single evaporator. The criticality hazard for this evaporator was reduced by depleting the U-235 content of the waste below acceptable levels. Waste containing aluminate would be processed in the other two operational evaporators and acceptance criteria were established to monitor for the possible formation of sodium aluminosilicate (8).

Operation of the site's evaporators in a segregated mode is acceptable to prevent issues associated with sodium aluminosilicate formation. However, the segregation does generate a problem with a need exists to remove the high silicon-containing liquor from the evaporator system. With tank space at a premium, the de-inventory process has the potential to mix high silicon-containing and high aluminum-containing wastes. Rosencrance (9) examined the fate of uranium during testing involving mixing both types of wastes together. In this testing, uranium was found to precipitate in all cases. However, it should be noted that uranium was added at a concentration thought to be very near or above its solubility limit as evidenced by precipitation in the control samples.

Current plant operation has de-inventoried the high silicon-containing waste into one of the Site's underground storage tanks. A large inventory of dissolved saltcake is planned to be moved into this same storage tank. This will bring a high aluminum-containing waste into contact with the high-silicon material. Therefore, a fundamental understanding to how the uranium chemistry is affected during the expected precipitation of sodium aluminosilicate is needed.

EXPERIMENTAL DETAILS

Testing with simulated and actual waste were perform to assess the effect of sodium aluminosilicate formation would have on the dissolved uranium. The simulant solutions were prepared in a similar manner as that of Addai-Mensah

(10, 11). The simulant solutions were prepared from reagent-grade chemicals. The actual wastes were from a composite of archived wastes for the high aluminum-containing waste. It is acknowledged that there are compositional differences between the simulants and actual wastes in particular the aluminum concentrations. The actual waste compositions represented the true plant conditions while the simulant testing utilized a previously studied composition. The high silicon-containing waste was from samples taken from Tank 49H. Solutions were analyzed using a Fisons Plasmaquad PQS972 II Inductively Coupled Plasma Mass Spectrometer for uranium. Elemental analysis was performed using a JY170C Inductively Couple Plasma Emission Spectrometer. Anion analyses by ion chromatography and titration with a standard acid was used for free hydroxide. Table 1 shows the composition of these solutions. A significant effort was expended to thermodynamically model the operation of the Site's high-level waste evaporators (12, 13). In this work, a commercially available computer software called Geochemist's Workbench was used. This software package is capable of performing a number of equilibrium thermodynamic calculations simultaneously. The program is equipped with the Lawrence Livermore National Laboratory's extensive database for minerals and aqueous species. It has also been augmented with data on sodium aluminosilicate solubilities (14). This software package has undergone a thorough quality assurance validation process (15). Data on the solution compositions of the high aluminum-containing waste and the high silicon-containing waste were entered into the Geochemist Workbench program and modeled for aluminosilicate formation. In a 1:5 mixture of high silicon to high aluminum waste, the mixture was found to be subsaturated with respect to aluminosilicate precipitation.

Table 1. Solution compositions of tank samples

	Units	High silicon	High aluminum	Simulant
Al	M	0.14	0.42	1.7×10^{-3}
Cr	M	0.0025	0.0047	—
K	M	0.04	0.06	—
Li	M	0.009	0.004	—
Na	M	9.53	7.85	6.0
P	M	0.01	0.01	—
Si	ppm	185	28	48
Carbonate	M	1.35	0.91	—
Formate	M	0.05	0	—
Nitrite	M	1.73	1.16	1.0
Nitrate	M	2.22	2.03	1.0
Sulfate	M	0.01	0.03	—
Free OH	M	5.13	3.49	4.0

Therefore, using the *REACT* command, 10 g of SiO_2 (aq) was added. The model showed that if one wanted to produce 20 g of NAS in 1 L of this solution, one would need to add approximately 7 g of SiO_2 (aq), or 3.25 g of elemental Si.

For the actual waste testing, each test combined amounts of the high silicon material with high aluminum material in a 1:5 ratio, respectively. An addition of ~ 6.63 g of sodium metasilicate was added to the combined waste matrix at the beginning of the test. Testing was performed at 50°C in Teflon bottles in duplicate on a 100 mL scale. Periodically during the test, the samples were removed from the oven allow to cool to ambient temperature and an aliquot was removed for analysis. A sub-sample of the aliquot was filtered through a cellulose 0.45 micrometer filter and gravimetrically diluted into 0.2 M nitric acid prior to analysis. For the simulant tests, the aluminate concentration was raised to ~ 2500 mg/L and silicon was raised to over 1000 mg/I to promote aluminosilicate formation. The test solutions were spiked to a level of 5, 10, 15, 25, and 50 mg/L uranium(VI) in the form of uranyl nitrate in a 2 M nitric acid solution.

RESULTS AND DISCUSSION

Simulant Studies Involving Uranium and NAS Formation

The past experience with the uranium incorporation into aluminosilicate phases causes concerns if the NAS formation occurs in the large waste tanks ($\sim$ 1 MGal), whereby large amount of uranium are available even though the absolute concentration is small. Previously, Rosencrance (9) had studied the fate of uranium and found under all conditions studied that uranium precipitated. Rosencrance, however, initiated the study with relatively high uranium concentrations and may have been above the solubility limit for the base simulant composition. In performing his uranium sorption experiments on the different sodium aluminosilicate phases, Addai-Mensah (16, 17) established the uranium solubility in a 6 M sodium salt solution and the effect of increasing the aluminate concentration. Figure 1 and Fig. 2 show the results of that work. Using the base simulant shown in Table 1 and adjusting the starting aluminate ion concentration, experiments were conducted at known uranium concentrations that were well below the solubility limit up to well beyond the solubility limit. A total of 5 uranium concentrations were explored from 5 mg/L to 50 mg/L.

Using the above graphs to determine that the uranium concentration in the simulated waste solutions was 14–15 mg/L, experiments were conducted at target uranium concentrations of 25 and 50 mg/L. In these experiments, the targeted uranium was spiked into the waste solution and held at temperature (50°C) for 50 h. Samples were taken very shortly after uranium addition (recorded as Time = 0 h) and at 50 h to determine if the uranium had precipitated. An aliquot of reagent grade sodium metasilicate was added ($\sim$6.26 g) to

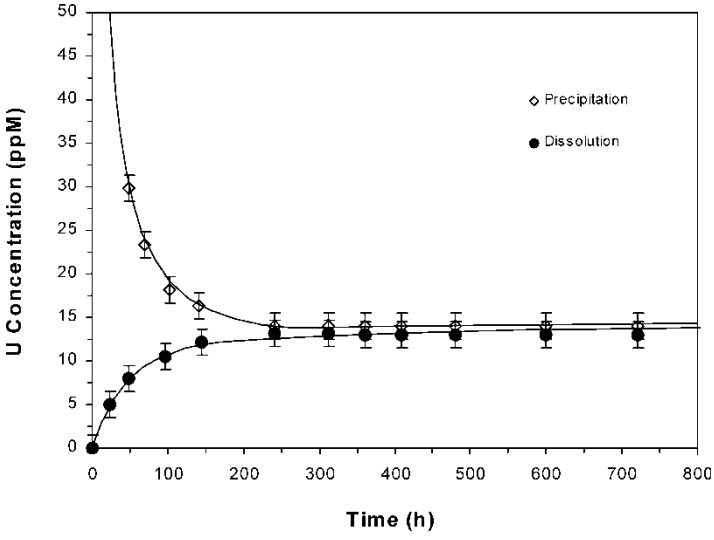

Figure 1. Uranium solubility curves in simulated waste solution.

the waste solution and a second sample was taken following the dissolution of the metasilicate. Lastly, samples were taken periodically over approximately 200 h from the reaction vessel maintained at 50°C.

Figure 3 shows the results for the test conducted with 50 mg/L uranium. The initial samples showed uranium concentrations of 53 and 61 mg/L. The first sample taken at 50 h measured 64 mg/L while the second sample

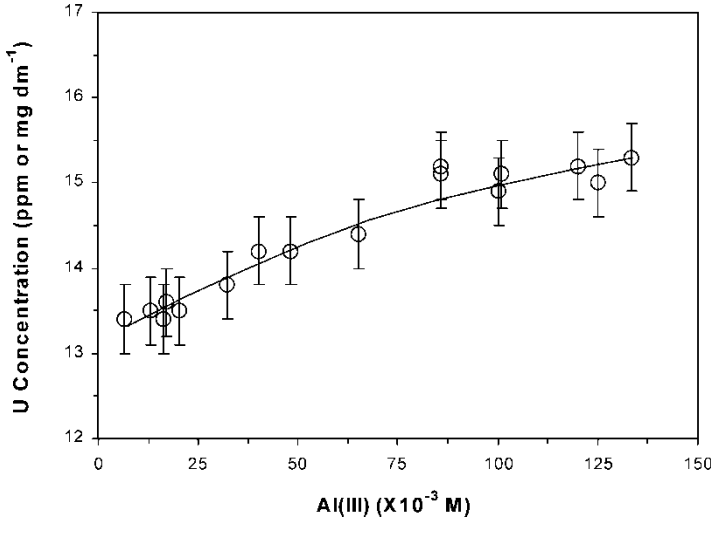

Figure 2. Effect of aluminate ion on uranium solubility.

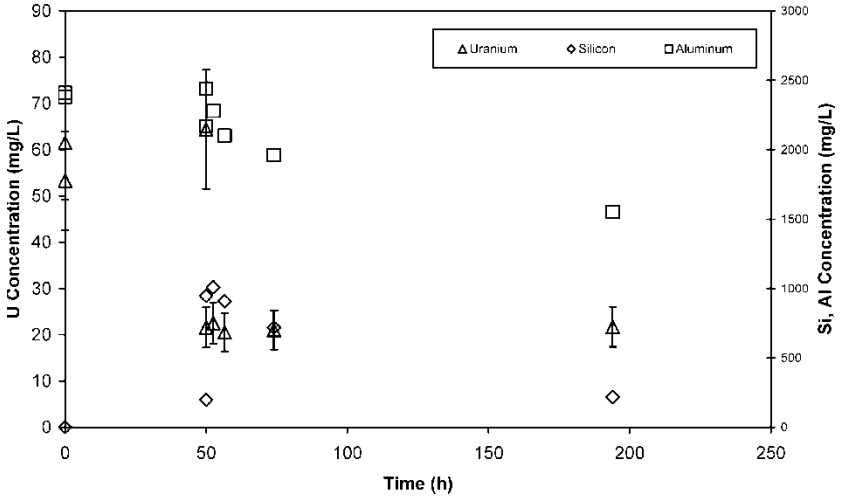

Figure 3. Results from the 50 mg/L uranium test.

taken, after the metasilicate was added, measured 21.6 mg/L. At the time the second sample was taken during the 50th h, a concomitant reduction in the aluminum concentration occurred indicating the formation of aluminosilicate had commenced. Evident from the silicon and aluminum data, aluminosilicate formation continued over the duration of the test. However, the uranium concentration plot does not indicate a continued loss of uranium from solution.

A second test was conducted at a uranium concentration of 25 mg/L with the expectation that similar solution chemistries would occur. The results from the 25 mg/L uranium test are shown in Fig. 4. In this instance the starting

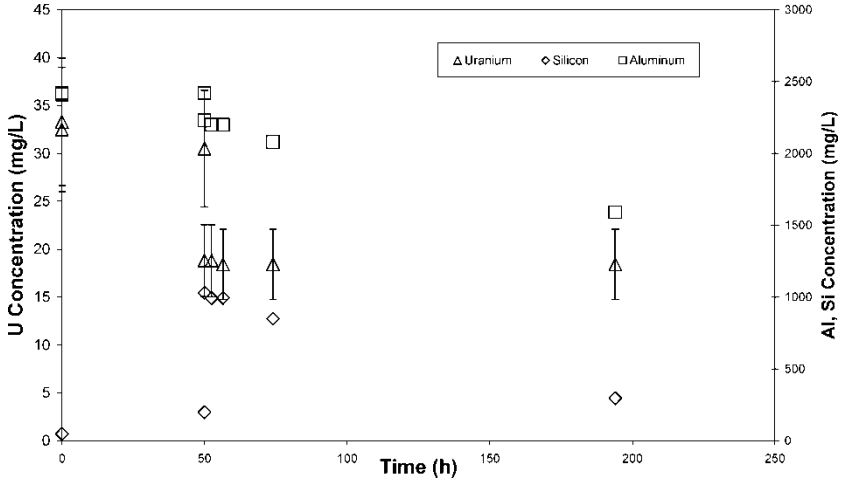

Figure 4. Results from the 25 mg/L uranium test.

uranium concentration was slightly higher than the 25 mg/L target and measured slightly above 30 mg/L. The supersaturated solution was stable at the 50 h mark. When the silicon was added, the same phenomenon as in the 50 mg/L uranium test occurred. A drop in both uranium and aluminum concentrations are observed. This is followed by further reaction to produce additional aluminosilicate without a significant removal of uranium from solution.

One experiment was conducted with a uranium concentration approximately at the uranium solubility limit. In this experiment, the targeted uranium concentration was 15 mg/L. Figure 5 shows the uranium, silicon and aluminum elemental analysis results. As observed in the tests conducted with uranium concentrations of 50 and 25 mg/L, silicon was added at the 50 h mark and reaction to form the sodium aluminosilicates occurs as evident by both aluminum and silicon concentrations declining with time. In this experiment, the starting measured uranium concentration was slightly higher than targeted and measured near 20 mg/L with an anticipated solubility limit of 15 mg/L. The uranium data suggests that at the point at which aluminosilicate starts to form that the uranium concentration is reduced. However, as indicated by the uncertainty bands, many of the measurements for uranium after the addition of metasilicate (at 50 h) many of the uranium measurements overlap the starting concentration. As in the other tests, there does not appear to a continual change with time after the addition of metasilicate.

It is evident from the results of the first three tests that the formation of sodium aluminosilicate will influence the level of supersaturated uranium

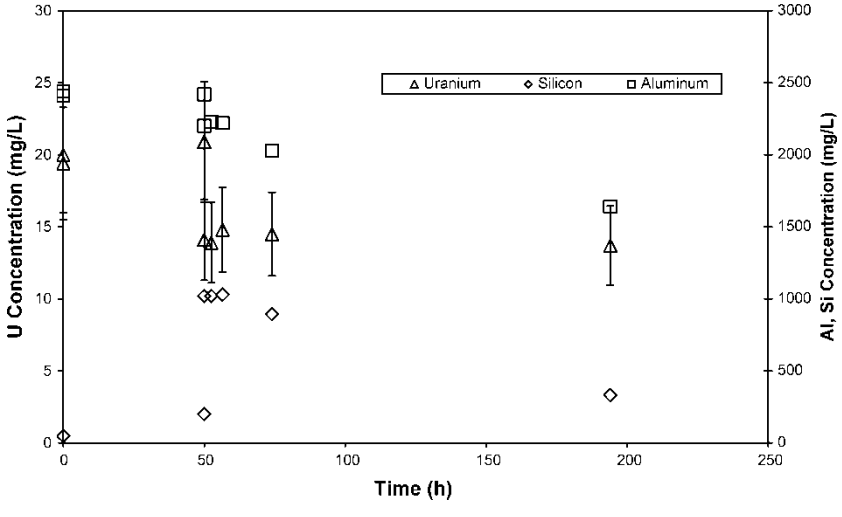

Figure 5. Results from testing at 15 mg/L uranium.

causing removal of uranium from solution. To examine the other extreme, a solution was prepared with uranium well below its solubility in this case at a concentration of 5 mg/L. The identical reaction sequence was followed including the thermal treatment of the solution prior to the silicon spike. The results of this test are shown in Fig. 6. As in the previous tests conducted at higher uranium concentrations, aluminosilicate forms immediately upon the addition of metasilicate as evidenced by the drop in aluminum concentration and a continued reaction producing aluminosilicate solids for the remainder of the test. The uranium concentration appears to be constant throughout the test within the error associated with the uranium measurement. This indicates that when below the solubility limit uranium is unaffected during the formation of aluminosilicate. This was examined at a slightly higher uranium concentration as shown in Fig. 7. In this experiment, the uranium concentration was raised to a target of 10 mg/L and measurements taken throughout the test indicate the uranium concentration is constant. The results from all five of these tests show that when the uranium concentration is at or below the solubility limit that the uranium is unaffected during aluminosilicate formation.

Characterization of Solids

Solids were collected from the reaction vessels from each of the five aluminosilicate formation tests. The samples were characterized by powder X-ray diffraction and scanning electron microscopy along with energy dispersive spectroscopy. In addition, portions of the solid samples were digested in

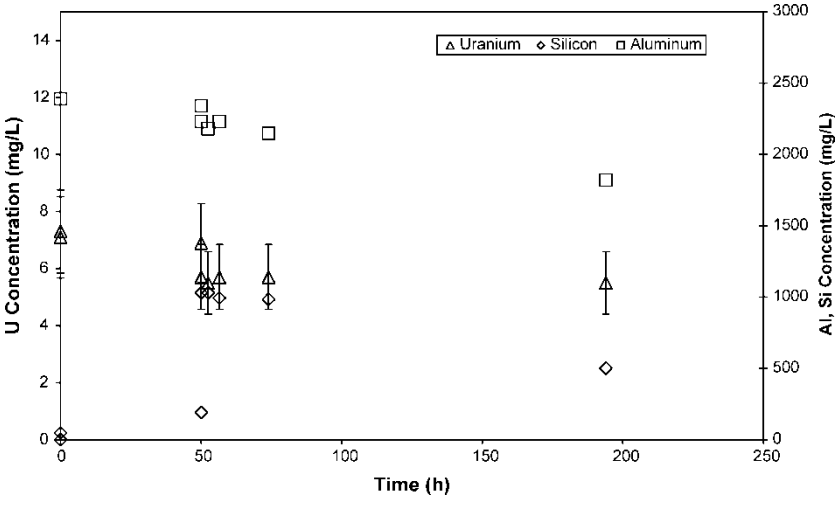

Figure 6. Results from the 5 mg/L uranium test.

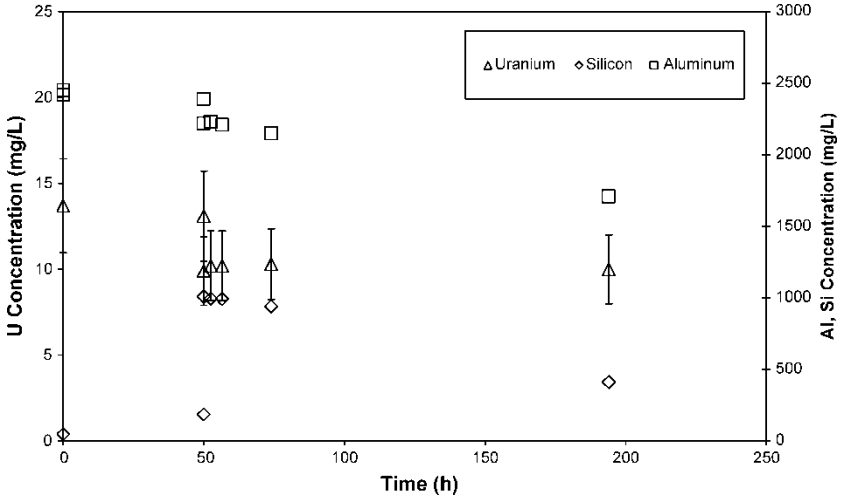

Figure 7. Results from 10 mg/L uranium test.

nitric acid and submitted for uranium analysis. The solids were white and flaky in color.

The results from the X-ray diffraction showed powder patterns typical of that shown in Fig. 8. The pattern matches the reference pattern for a nitrated cancrinite typical of that observed in the high-silicon evaporator. The pattern does show amorphous character as evidenced by the broad band in the 15–40 degree 2θ. Even in the test involving 50 mg/L, there was no indication of a

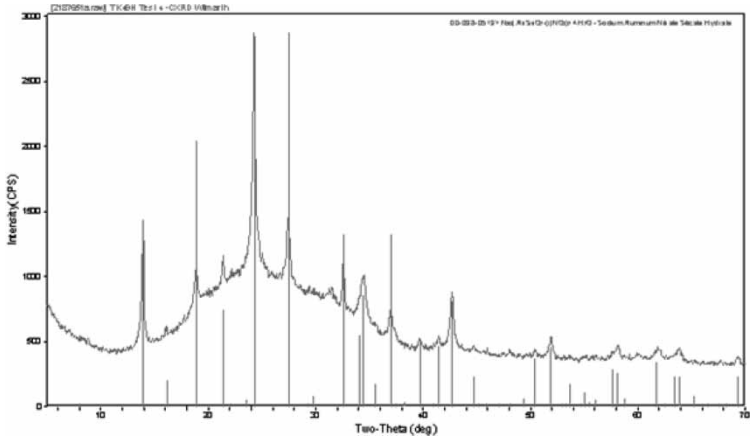

Figure 8. Typical powder diffraction spectrum from solids produced during aluminosilicate formation.

crystallographic phase of a uranium compound. This is not unexpected due to the small amount of uranium present in each test.

Several scanning electron micrographs are shown in Fig. 9. For all of the tests, the basic structure of the material is the zeolytic ball structure. The material is cancrinite based on the X-ray powder pattern and the scanning electron images agree with that published by Addai-Mensah (18). This "yarn-ball" effect observed in the micrograph in the upper right of Fig. 9 can be seen in higher magnification in all portions of the samples. The energy dispersive spectrum of the materials show peaks only for sodium, aluminum, silicon and oxygen.

In the test that was conducted with 50 mg/L uranium in the starting solution, a different image is observed and is shown in Fig. 10. The backscatter image shows a few small bright images that contain uranium. This test is where uranium was well above the solubility limit and is expected to precipitate and precipitation was observed when aluminosilicate formed. The occlusion of these small uranium compounds into the aluminosilicate solids is similar to that observed in the 2H Evaporator scale (3). The energy dispersive spectrum of the solids is shown in Fig. 11. The spectrum contains peaks characteristic of uranium at about 3 keV. These uranium solids were not observed in the solids from the tests with starting uranium concentrations of 5 to 25 mg/L.

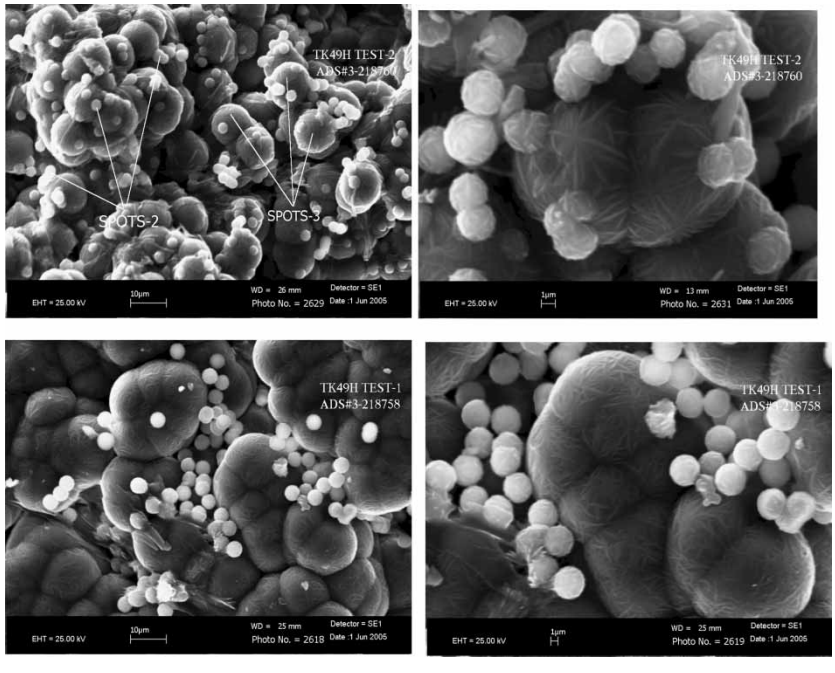

Figure 9. SEM images of solids from simulant studies.

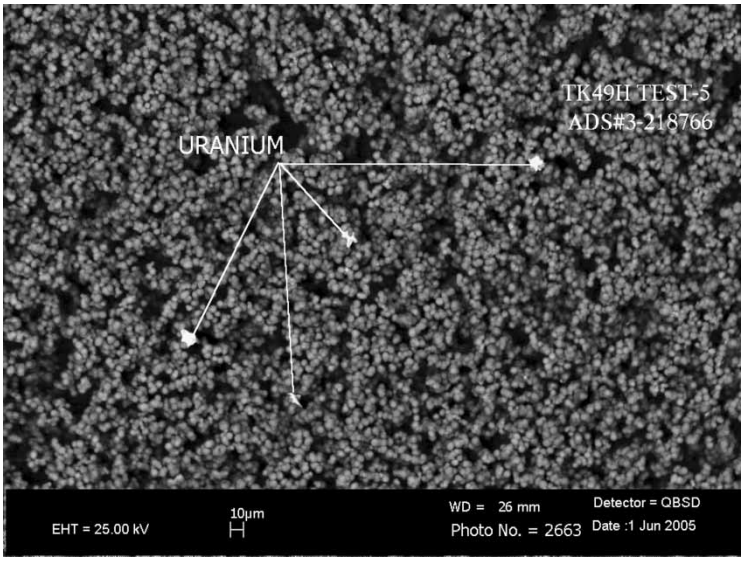

Figure 10. SEM image from simulant test with 50 mg/L uranium.

The solids from each of the simulant studies with starting uranium concentrations of 5 to 50 mg/L were collected and washed with inhibited water (0.01 M NaOH) three times. The washed solids were digested in nitric acid and submitted for uranium analysis. Table 2 displays the measured uranium concentration in the aluminosilicate solids. The uranium content of the solids increased with an increase in the starting uranium concentration. A distinctive jump in the uranium content was observed for the solids from

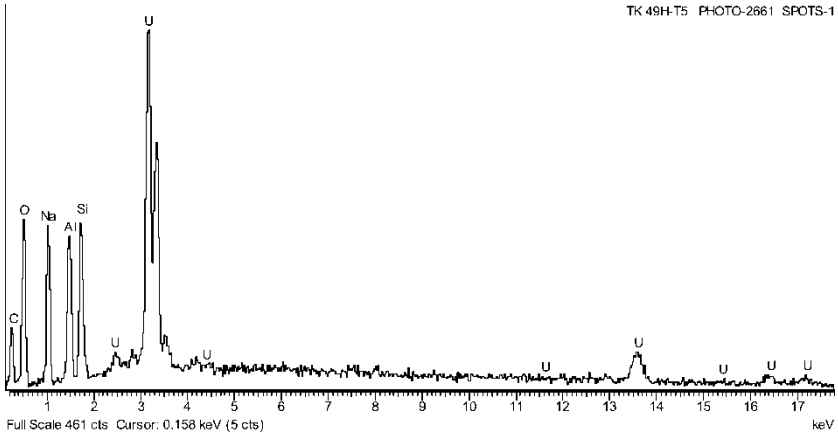

Figure 11. EDS spectrum from uranium-containing solid.

Table 2. Uranium content of simulant solids

Test condition	Uranium (μg/g)	Uranium Wt%
5 mg/L	22.5 ± 4	0.0023 ± 0.0004
10 mg/L	110 ± 16	0.011 ± 0.002
15 mg/L	370 ± 36	0.04 ± 0.004
25 mg/L	1100 ± 1	0.11 ± 0.0001
50 mg/L	3140 ± 1200	0.31 ± 0.12

the test with a starting uranium concentration of 25 mg/L. The uranium content of the solids increased from 370 μg/g of NAS solids at 15 mg/L U to 1100 μg/g for the solids from the 25 mg/L U. This is expected due to the presence of uranium above the solubility limit. The uranium content of the 15 mg/L solids was also high and may have resulted from precipitation of uranium due to the uranium spike was high for that test. The starting uranium concentration was above 20 mg/L with a target of 15 mg/L. The uranium content from the sub-saturated tests was 23 and 110 μg/g for the tests starting with 5 and 10 mg/L uranium, respectively. These values are in the range of that observed from sorption tests (10). The measured sorption values for cancrinite were lower and were less than 10 μg/g. These elevated levels may result from inefficient washing of the test solution from the solids or small amounts of precipitation or uranium from solution.

Actual Waste Testing

In order to confirm that the formation of sodium aluminosilicate would not influence the uranium concentration if the uranium concentration was at or below the uranium solubility, a test was conducted in duplicate with a 1:5 mixture of actual high silicon-containing waste and some previously sampled high aluminum-containing waste. Table 1 shows the chemical composition of the wastes used in the testing. As mentioned, the tests were conducted in duplicate. The testing was similar to the simulant testing; however, the heat pretreatment was not performed. Silicon in the form of sodium metasilicate was added to the waste mixtures and the solutions were heated to 50°C. Figure 12 shows the results for aluminum, silicon, and uranium from the first of two actual waste tests. The data from the second test are very similar to these results. With the addition of elevated levels of silicon to the waste, an immediate reaction is observed with the measured silicon and aluminum concentrations declining throughout the 500 h of the test. The uranium concentration, on the other hand, remains constant at about 6–7 mg/L. This is the expected result based on the simulant studies.

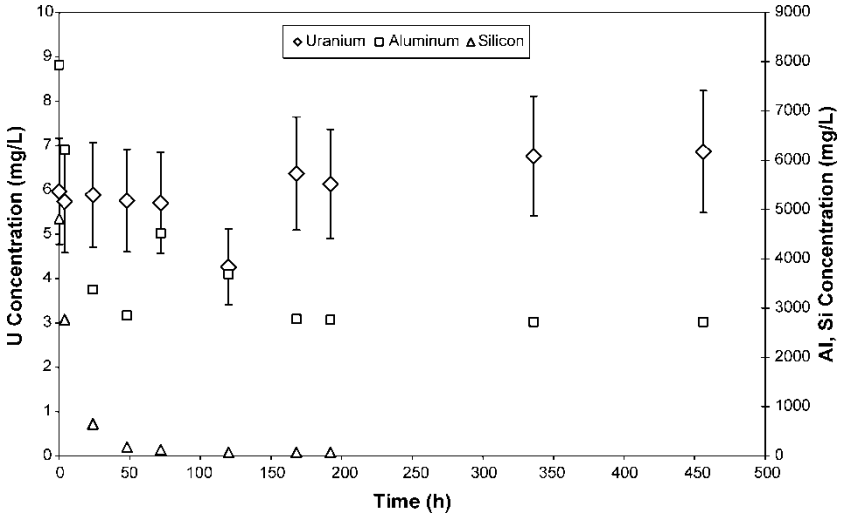

Figure 12. Uranium concentrations in the actual waste test.

The solids produced during the testing were collected and thoroughly washed with distilled, de-ionized water to remove the interstitial supernate. The samples were examined for crystal structure, imaging and uranium content. Figure 13 shows the X-ray powder pattern from the first of two samples. The result from the second sample agreed with the first. The powder pattern is that of cancrinite identical to the pattern obtained from the simulant studies. No other forms of the sodium aluminosilicate are

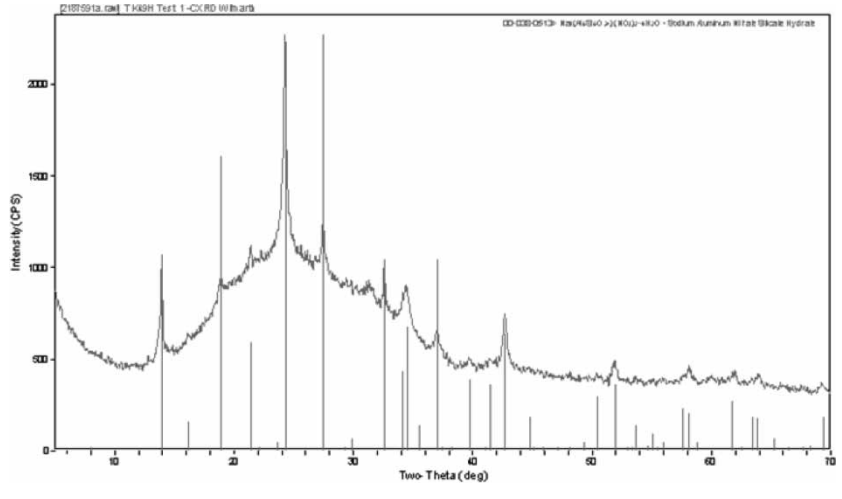

Figure 13. X-Ray diffraction pattern from actual waste test.

observed. This is reasonable since the solids were heat treated for several hundred hours at 50°C. Armstrong and Dann (19) had shown in Bayer solutions with high nitrate concentrations that cancrinite formation was not greatly influenced by reaction temperature. There is, however, a large band typically associated with amorphous material observed in the powder pattern.

Figure 14 shows a typical scanning electron micrograph of the solids collected from the second actual waste test. The morphology of these samples is less defined that the simulant studies. The "yarn ball" structure is not observed in the actual waste solids indicating that the material consists primarily of the amorphous form of the NAS which is not detected by the X-ray diffraction measurement. Both sets of solids from the actual waste have the same appearance. The energy dispersive spectra from the solids show sodium, silicon, aluminum and oxygen from the sodium aluminosilicate. Some spots within the samples showed evidence of iron and manganese typical of sludge solids. Others showed evidence of barium, calcium, and other elements which are likely contaminants of the sample from sample handling in highly radioactive environment. The uranium content of the solids was determined by digesting an aliquot of the sample and measuring the uranium using ICP-MS. The measured uranium concentrations in the actual waste samples were 5.2 and 6.0 $\mu g/g$ of NAS solid. This indicates that employing a better washing procedure removed the interstitial supernate. The magnitude is in better agreement with sorption data for cancrinite (10).

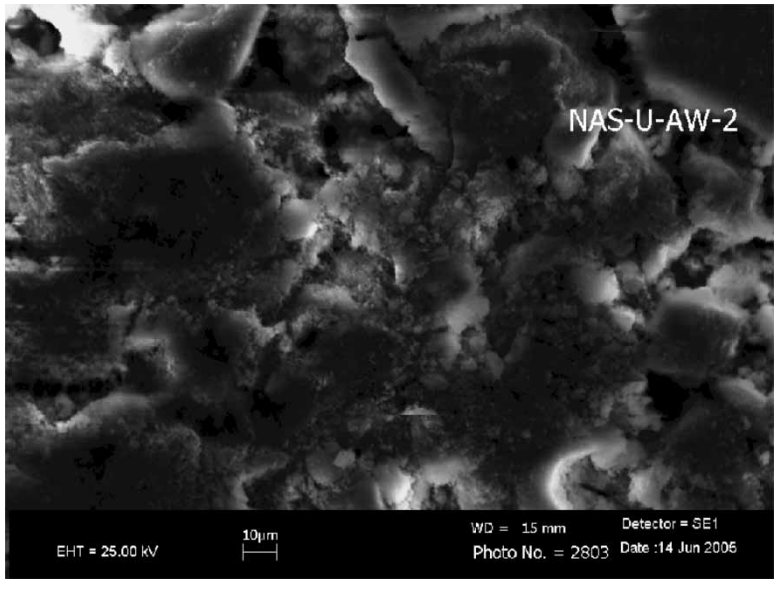

Figure 14. SEM of solids from actual waste test.

CONCLUSIONS

The focus of this work was to determine if the formation of sodium aluminosilicate occurring when wastes containing high aluminate concentrations are mixed with wastes containing high silicate concentrations could cause the precipitation of uranium-containing phase. If uranium precipitated from solution, then criticality issues would need to be addressed. The testing has shown that if the uranium concentration is above the solubility limit, i.e., the solution is supersaturated with uranium, and then the formation of sodium aluminosilicate can act as a means to reduce the uranium degree of supersaturation. The exact mechanism for the removal of uranium from solution, however, was not examined. The data from simulant studies showed a concomitant reduction in uranium, silicon and aluminum. Furthermore, testing at the highest degree of uranium supersaturation demonstrated that a separate uranium phase can be formed in the aluminosilicate matrix.

Testing with waste simulants and actual tank wastes have shown that if the solution is sub-saturated with uranium, then the formation of aluminosilicates does not influence the uranium concentrations. Analysis of the uranium content in these solids show levels typically observed during sorption experiments.

ACKNOWLEDGEMENT

This report was prepared by Westinghouse Savannah River Company (WSRC) for the United States Department of Energy under Contract No. DE-AC09-96SR18500 and is an account of work performed under that contract. Neither the United States Department of Energy, nor WSRC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, or product or process disclosed herein or represents that its use will not infringe privately owned rights. Reference herein to any specific commercial product, process or service by trademark, name, manufacturer or otherwise does not necessarily constitute or imply endorsement, recommendation, or favoring of same by WSRC or the United States Government or any agency thereof. The views and opinions of the authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

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