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**Thermal and Chemical Stability of  
Crystalline Silicotitanate Sorbent**

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## ABSTRACT

The Savannah River Site (SRS) is evaluating technologies for removing radioactive cesium ( $^{137}\text{Cs}$ ) from the supernate solutions stored in the high-level waste tanks at the site. Crystalline silicotitanate sorbent (IONSIV IE-911,<sup>®</sup> UOP LLC, Des Plaines, IL), which is very effective at removing cesium from high-salt solution, is one of three technologies currently being tested. Because of the extremely high inventory of  $^{137}\text{Cs}$  expected for the large columns of crystalline silicotitanate (CST) that would be used for treating the SRS supernate, any loss of flow or cooling to the columns could result in high temperatures from radiolytic heating. Also, even for normal operation, the CST would be exposed to the supernates for up to a year before being removed. Small-scale tests using simulant solutions were used to determine the long-term stability of the CST to the solutions at various temperatures.

In the tests performed in this study, the cesium capacity of the CST decreased significantly (76%) as the temperature of the simulant and CST during loading was increased from 23 to 80°C. CST exposed to recirculating SRS average simulant solution at room temperature in a column test showed a slow decrease in cesium loading capacity (measured at 23°C), with a drop of 30% for CST from the top of the bed and 13% for CST from the bottom of the bed after a 12-month period of exposure. A similar column test using a high-pH salt solution did not show any change in the cesium capacity of the CST. An increase was noted in pressure drop through the column using average simulant, but no change was observed for the column using high-pH salt solution.

In batch tests, no change in cesium capacity was noted for CST samples stored at 23°C in average, high-hydroxide, or high-nitrate simulant solutions for up to 12 months. Samples of granular CST (IONSIV IE-911) stored in the simulants at 80°C showed a drop in cesium capacity of about 23% after 1 month of storage when the loading tests were performed at room temperature. There was no further change in the cesium capacity for storage times up to 12 months at 80°C. CST samples stored at lower temperatures for up to 12 months showed drops in cesium capacity of 9, 11, and 15% for storage temperatures of 30, 35, and 50°C, respectively. A sample of CST powder (IONSIV IE-910) stored in average simulant at 80°C showed a continuing decrease in cesium capacity during the test, losing 75% after 12 months.

CST in both the batch and the column tests had a tendency to become agglomerated with cancrinite (a sodium aluminosilicate), formed when aluminum, silicon, and sodium from the simulant solutions, and possibly additional silicon leached from the CST, reacted together. The ratio of cancrinite to CST in the samples could be calculated from measured concentrations of aluminum and titanium. This ratio was used to eliminate the cancrinite weight from the calculation of the cesium loading for the CST samples from the leaching tests, and to calculate an adjusted cesium capacity for only the CST in the samples. Such calculations show that the cancrinite accounts for the decrease in cesium capacity for most of the CST samples from the batch and column leaching tests. Two sets of samples indicated more loss in cesium capacity than can be accounted for by the cancrinite precipitate: IE-911 samples from the top of the column leaching test, using average simulant after 8 to 12 months; and IE-910 from the batch test in average simulant at 80°C. In each of these tests, an additional, unknown mechanism was involved in the reduced cesium capacity of the CST.

CST clumps were visible in batch samples stored at 30, 35, 50, and 80°C after 1 month, and in room-temperature samples after 2 months. These clumps have the potential (1) to cause fluid channeling within a column and (2) to make the CST more difficult to sluice from a column. The

CST used in these tests was pretreated at a lower sodium hydroxide concentration (0.1 *M*) than was present in the simulants (1.2 to 3 *M*), which may affect the performance of the CST.

New batch and column leaching tests are in progress using samples of baseline (9090-76) and improved (MH-9098-9) CST from UOP. The baseline CST is similar to the earlier samples, while the improved CST was treated at UOP to reduce the amount of niobium and silicon that leaches from the CST. The baseline CST was pretreated with 3 *M* NaOH prior to being used; the improved CST was used as received. Both samples have shown less tendency to clump than the earlier CST, which may be related to the lower NaOH concentration used to pretreat the earlier samples. Each of the new CST samples formed clumps after storage in simulant for 1 month at 80°C, and the improved sample formed clumps after 2 months at 50°C. All of the remaining batch samples were free-flowing after 3 months. The column test using baseline CST contained clumps of CST after exposure to recirculating simulant for 2 months.

The cesium capacities of the new samples from the batch and column tests at room temperature were similar to those of comparable samples from the original tests. A slower decrease in cesium capacity has been observed for the new samples stored at higher temperatures in the batch leaching tests. The samples stored at 50 and 80°C showed drops of 4 and 14%, respectively, after 2 months, whereas the samples stored at lower temperatures have exhibited no decrease in cesium capacity to date.



## 1. INTRODUCTION

The Savannah River Site (SRS) is evaluating technologies for removing cesium-137 ( $^{137}\text{Cs}$ ) from the supernate solutions stored in high-level waste tanks at the site. IONSIV IE-911,<sup>®</sup> an inorganic ion-exchange material with a high affinity for cesium, is the key material in one of the processes being tested. Results obtained from work conducted at ORNL in 1999 showed that higher temperatures could cause desorption of previously loaded cesium from this material, a crystalline silicotitanate (CST), and that the CST would not reload the cesium when the temperature was reduced following 60 days at the higher temperatures.<sup>1</sup> Because of the extremely high inventory of  $^{137}\text{Cs}$  that would be expected for the large columns of CST that would be used for treating the SRS supernate, any loss of flow or cooling to the columns could result in high temperatures within the column from radiolytic heating.

Long-term batch and flow-through leaching tests were conducted to determine any detrimental effect from long-term exposure of the CST to supernate simulant solutions under various conditions. Simulant solutions that represent the average and extremes of composition expected for the SRS supernates during treatment were used, along with a simplified simulant, called high-pH salt solution, that contains only sodium nitrate and sodium hydroxide.

## 2. MATERIALS AND METHODS

Most of the CST used in the tests reported here was the commercial, granular form of the sorbent IONSIV IE-911<sup>®</sup> (UOP LLC, Des Plaines, IL), which consists of silicotitanate crystals (IONSIV IE-910<sup>®</sup>) and a zirconium-based binder. Lot 999098810005 CST (98-5), which was manufactured in 1998 using a chloride-based process, was used for all of the test conditions. A few of the test conditions were duplicated using Lot 999099810007 (99-7) CST, which was manufactured in 1999 using a newer, nitrate-based process, and the IE-910 that was used to make the 1998 batches of IE-911. The as-received granular CST was contacted with recirculating, dilute sodium hydroxide for 24 h to stabilize the pH at ~13 and then backwashed to remove fines prior to use. The IE-910 was used as received. A standardized procedure for pretreating CST before contact with the SRS simulants was developed after these tests were started. This procedure uses once-through NaOH at the same (or higher) concentration as the simulant.<sup>2</sup> Two new granular CST samples and the CST powder used to prepare them were received from UOP in November 2000 and January 2001. The first IE-911 sample is a baseline material (labeled as 9090-76) that was prepared using the same nitrate-based process as the 99-7 CST, but with a higher level of documentation and quality control. The second IE-911 sample (designated as MH-9098-9) was treated at UOP to remove most of the leachable material (mainly niobium) from the CST. The IE-910 powder sample is designated as 30950-48. Batch and column leaching tests have been started with these new samples, and early results are included in this report for comparison. The 9090-76 CST was pretreated with once-through 3 M NaOH for 24 h (10 L NaOH for 400 g CST) and then backwashed prior to use. The MH-9098-9 and 30950-48 samples were used as received for the batch tests. The MH-9098-9 for the column tests was pretreated with recirculating 1.9 M NaOH for 24 h to ensure that the alkalinity of the CST was stabilized prior to starting the tests.

## 2.1 Loading Tests

Batch loading tests were performed at temperatures of 23, 30, 35, 40, 45, 50, 60, 70, and 80°C, using 0.1 g of CST and 100 mL of supernate simulant, which contained 50 mg/L cesium and 0.1  $\mu\text{Ci/L}$   $^{137}\text{Cs}$ . These tests were conducted to determine the short-term effect of temperature on the CST cesium loading capacity. The CST and simulant were mixed for 7 days, inside capped Teflon flasks, at 120 rpm in Model 3527-5 Environ Shakers (Lab-Line Instrument, Inc., Melrose Park, IL). The temperature of each sample was maintained within  $\pm 0.2^\circ\text{C}$  by a built-in, forced-air convection heater with a temperature controller. The temperatures were checked using a digital thermometer with NIST traceable calibration.

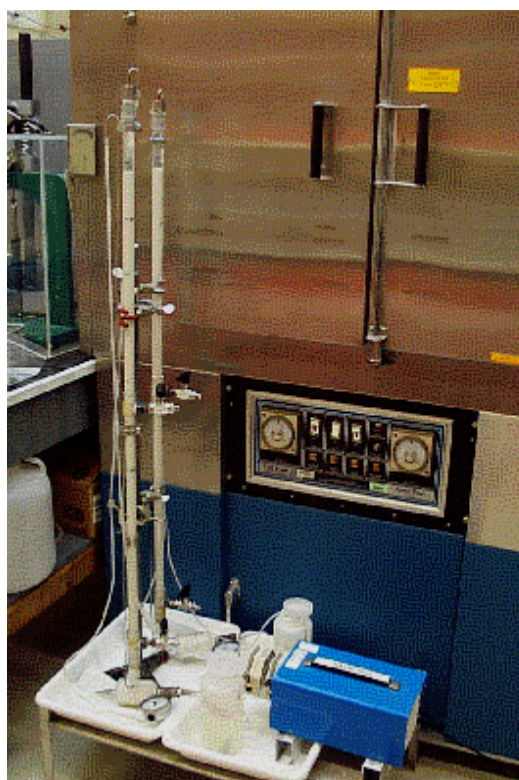
After 7 days, two samples of the supernate from each flask were filtered through 0.2- $\mu\text{m}$ -pore nylon syringe filters. A 2.00-mL portion of each filtered sample was pipetted into a polyethylene counting tube, capped, centrifuged for about 2 min to ensure that no droplets were left on the sides of the tubes, and then gamma counted for 1000 s. The samples were counted at an energy level of 664.7 keV, using a Canberra Industries, Inc. (Meriden, CT) series 90 gamma spectrometer, with a shielded germanium detector, to determine the concentration of  $^{137}\text{Cs}$ . Three samples of the starting supernate simulant and three background rates were counted in each case.

## 2.2 Column Leaching Tests

Cesium-free average supernate simulant and high-pH salt solution were recirculated through small polyvinyl chloride columns (1.6 cm ID by 120 cm tall) containing pretreated 98-5 CST at room temperature. The columns were initially filled with 235 g (220  $\text{cm}^3$ ) of the CST, which gave a bed height of 110 cm. The flow rate of each solution was 6 mL/min. The solutions were pumped through 0.45- $\mu\text{m}$ -pore filter cartridges before entering each column, using a dual-head peristaltic pump and silicon rubber tubing. Figure 1 shows a photograph of the columns, pump, and filters. Pressure gauges on the inlet of the filters were used to monitor column plugging. Samples of the solutions were analyzed periodically for dissolved metals in order to measure CST leaching and precipitation of simulant components. The solutions were replaced when their concentrations changed significantly. Samples of the CST were removed from the top, middle, and bottom of the column each month and tested for cesium sorption and elemental composition. Selected samples were analyzed for porosity, surface area, and particle size. The tests, which were started on January 31, 2000, ran for 12 months.

The new baseline (9090-76) and UOP-pretreated (MH-9098-9) CST samples are being tested using the same type of equipment and procedures as described above. Average simulant is being recirculated through each column at a flow rate of 6 mL/min. The 9090-76 column was started on December 5, 2000, and the MH-9098-9 column was started on January 18, 2001. The expected duration of these tests is approximately 6 months.

A slightly taller column (CST bed height = 115 cm) of the same diameter is being tested using MH-9098-9 CST and average simulant in a once-through flow mode. The purpose of this test is to determine if continuous exposure of the CST to fresh simulant results in the collection of higher amounts of precipitated aluminosilicate in the column. The flow rate for this column is 2.0 mL/min, giving a fluid residence time within the CST bed of about 1 h (which is about the same as that planned for a full-scale SRS column). Because the leaching and precipitation reactions have very slow kinetics, it was important to match the residence time for the full-scale columns, even though this means that the fluid velocity will be lower. This test, which was started on February 3, 2001, has an expected duration of 4 months.



**Fig. 1. Photograph of column leaching test equipment.**

### **2.3 Batch Leaching Tests**

Long-term batch leaching tests were conducted using cesium-free average, high-hydroxide, and high-nitrate supernate simulants as well as the high-pH salt solution to determine the effects of temperature and solution composition on the stability of the CST. Teflon bottles containing 16 g of CST and 240 mL of simulant were stored at room temperature and 30, 35, 50, and 80°C. Samples of the solutions were analyzed each month for dissolved metals in order to measure CST leaching and/or precipitation of simulant components. Samples of the CST were removed each month and tested for cesium sorption and elemental composition, and selected samples were analyzed for porosity, surface area, and particle size. Lot 98-5 CST was used for all of these test conditions; lot 99-7 CST and IE-910 CST powder were also tested at 23 and 80°C in average simulant. These tests, which were started on January 14, 2000, lasted for 12 months. A room-temperature leaching test using average simulant containing 50 mg/L Cs and 98-5 CST, which was started on June 15, 1999, was continued for a total of 19 months.

The new CST samples (9090-76, MH-9098-9, and 30950-48) are being tested using the same procedures as described above, except that only average simulant is being used. The 9090-76 and 30950-48 tests were started on December 5, 2000; the MH-9098-9 tests were started on January 17, 2001.

## 2.4 Analytical Methods

The CST samples from the leaching tests were rinsed three times with a small volume of deionized water and then dried at room temperature in a desiccator prior to analysis. Samples of unused, pretreated CST were contacted with each of the simulant solutions for 1 min and then rinsed and dried using the same procedure as for the batch and column leaching tests. These samples served as the baseline (zero time) for comparing all of the analytical results for the leaching test samples. Four subsamples from each group of CST samples from the leaching tests were dried to a constant weight at 105°C and then dried at 400°C for 4 h to determine the amount of moisture in the CST. The average weight loss for the CST samples was 3.84 wt % at 105°C and 13.12 wt % at 400°C. The moisture results were used to calculate cesium loadings on a dry-weight basis. The cesium loading tests used average simulant containing 50 mg/L cesium and about 1  $\mu\text{Ci/L}$   $^{137}\text{Cs}$ , and the CST and solution were contacted in Teflon flasks for 7 days at room temperature. Samples were collected and filtered in a manner similar to that used for the batch loading tests, except that they were counted on a Model 1282 Compugamma (LKB Wallac) with a sodium iodide detector. This counter has an automatic sample changer, which facilitates the analysis of a large number of samples generated from the leaching tests.

The inductively coupled plasma (ICP) analyses were performed using a model 61E Trace ICP from Thermo Jarrell Ash, following standard EPA method SW846-6010B. Standard reference samples from the National Institute of Standards and Technology (NIST) were analyzed along with the test samples to verify the accuracy of the results. Both simulant solutions and CST samples were digested using nitric and hydrofluoric acids in a microwave oven to solubilize the materials; then boric acid was added to complex the fluoride ion prior to analysis. The surface areas and pore volumes of the CST samples were measured using a Gemini III 2375 Surface Area Analyzer instrument. Selected samples of the CST were examined with a scanning electron microscope to identify any changes in the morphology and size of the particles.

## 2.5 Simulant Solutions

Average, high-nitrate, and high-hydroxide supernate simulants as well as a high-pH salt solution were prepared at ORNL using recipes supplied by SRS.<sup>3</sup> The compositions are shown in Table 1. The simulants were prepared in large batches (5 L for the high-hydroxide and high-nitrate supernate simulants, and 20 L for the average supernate simulant and high-pH salt solution) without any cesium. All of these solutions were used for the batch leaching tests, and the average simulant and high-pH salt solution were also used for the column leaching tests. A portion of each solution was spiked with 50 mg/L cesium and 1  $\mu\text{Ci/L}$   $^{137}\text{Cs}$  for use in the cesium-loading tests. Each large batch of simulant was analyzed by ICP methods for metals and by ion chromatography for anions. The spiked batches of simulant were analyzed for cesium by ICP-mass spectroscopy and for  $^{137}\text{Cs}$  by gamma counting at the ORNL Radioactive Materials Analysis Laboratory. Three large batches (Nos. 4, 5, and 6) of the average simulant were prepared and used. Only one batch of each of the remaining solutions was used. Fifteen batches of spiked (Cs and  $^{137}\text{Cs}$ ) average simulant were prepared from the three large batches, and these were used for the loading tests of the CST samples from the batch and column leaching tests. The analytical results for the spiked batches of simulant are shown in Table 2.

**Table 1. Composition of simulated waste solutions**

Component	Concentration ( <i>M</i> )			
	Average	High OH <sup>-</sup>	High NO <sub>3</sub> <sup>-</sup>	High-pH salt
Na <sup>+</sup>	5.6	5.6	5.6	5.6
K <sup>+</sup>	0.015	0.030	0.0041	
OH <sup>-</sup>	1.91	3.05	1.17	1.91
NO <sub>3</sub> <sup>-</sup>	2.14	1.10	2.84	3.69
NO <sub>2</sub> <sup>-</sup>	0.52	0.74	0.37	
AlO <sub>2</sub> <sup>-</sup>	0.31	0.27	0.32	
CO <sub>3</sub> <sup>2-</sup>	0.16	0.17	0.16	
SO <sub>4</sub> <sup>2-</sup>	0.15	0.030	0.22	
Cl <sup>-</sup>	0.025	0.010	0.040	
F <sup>-</sup>	0.032	0.010	0.050	
PO <sub>4</sub> <sup>3-</sup>	0.010	0.008	0.010	
C <sub>2</sub> O <sub>4</sub> <sup>2-</sup>	0.008	0.008	0.008	
SiO <sub>3</sub> <sup>2-</sup>	0.004	0.004	0.004	
MoO <sub>4</sub> <sup>2-</sup>	0.0002	0.0002	0.0002	

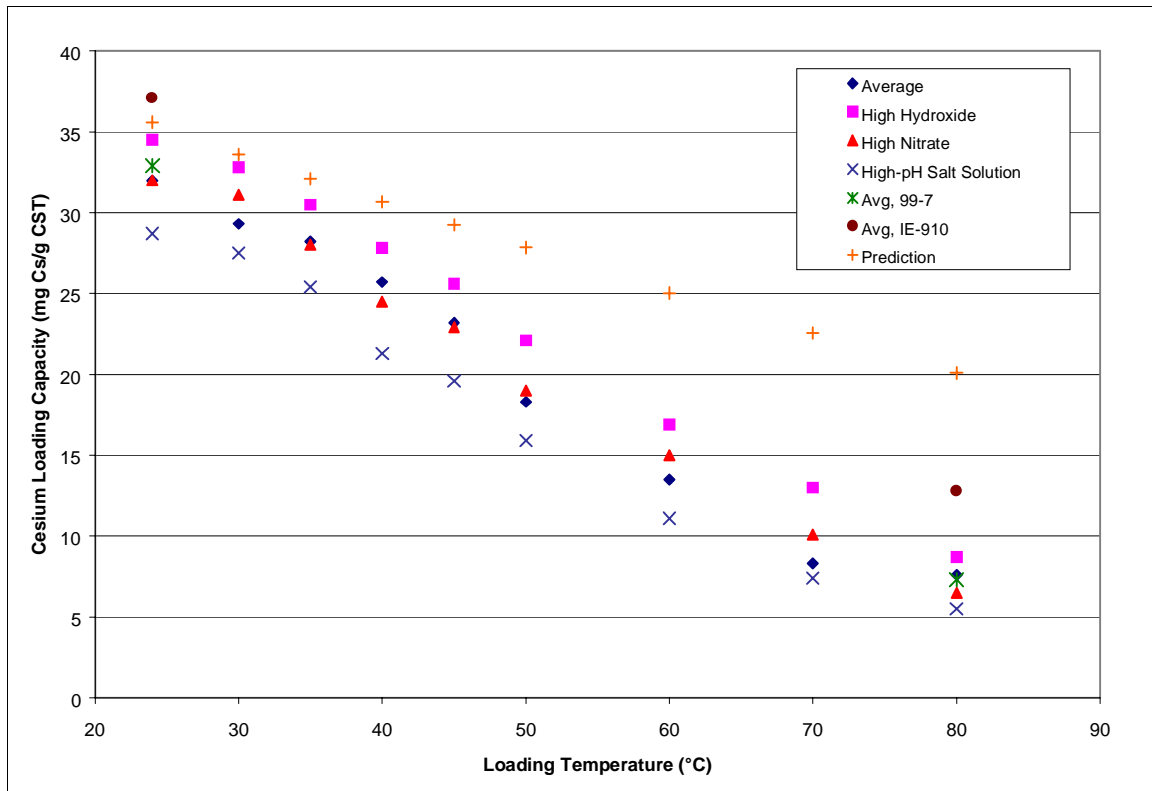
**Table 2. Analytical results for spiked batches of simulant used in loading tests**

Batch ID	Cs (mg/L)	<sup>137</sup> Cs (μCi/L)	Application
AVG 4-A	50.0	1.19	Loading tests and 1-month column
HH-1A	48.6	1.11	Loading tests
HN-1A	49.1	1.11	Loading tests
HS-1A	51.8	0.86	Loading tests
AVG 4-B	45.9	0.73	1-Month batch
AVG 4-C	42.0	1.05	2-Month batch
AVG 4-D	47.7	1.19	3-Month batch; 2- and 3-Month columns
AVG 5-A	52.4	1.22	4-Month batch and column
AVG 5-B	48.6	1.08	5-Month batch and column
AVG 5-C	57.5	1.24	6-Month batch and column
AVG 5-D	49.6	1.22	7-Month batch and column
AVG 5-E	48.3	1.16	8-Month batch and column
AVG 6-A	51.4	1.22	9-Month batch and column
AVG 6-B	50.2	1.24	10-Month batch and column
AVG 6-C	50.3	1.38	11-Month batch
AVG 6-D	56.9	1.43	11-Month column and new CST tests
AVG 6-E	54.2	1.49	12-Month batch
AVG 6-F	51.7	1.35	12-Month column and new CST tests

### 3. RESULTS AND DISCUSSION

#### 3.1 Loading Tests

The results of the cesium loading tests are summarized in Fig. 2. Temperature has a strong effect on the cesium capacity of the CST. The results are calculated on the basis of a dry weight of the CST at 400°C, which was measured using subsamples of the CST that were not included in the loading tests. The high-hydroxide simulant shows the highest capacity, followed by the average and high-nitrate simulants, and, finally, by the high-pH salt solution. The prediction of the cesium loading on the CST by the ZAM equilibrium model<sup>5</sup> (adjusted to a 400°C dry weight) for the average simulant at each temperature is shown for comparison. The predictions for temperatures of 45 to 80°C are above the validated range of the model (25 to 44°C). The high-hydroxide and average simulants have measured values about 10% lower than the predictions for temperatures up to 35°C; then the results increasingly diverge from the model as the temperature increases. For temperatures up to 35°C, the measurements for the high-pH salt solution were about 7% below the model predictions and the high-nitrate simulant results matched the predictions; however, the results for both simulants diverged at the higher temperatures. All of the solutions had measured capacities that were over a factor of 2 below the predictions at 80°C.



**Fig. 2. Results of cesium loading tests using CST and simulant solutions, and comparison with predictions of the ZAM model for the average simulant.** All tests used 98-5 CST unless otherwise indicated.

### 3.2 Batch Leaching Tests

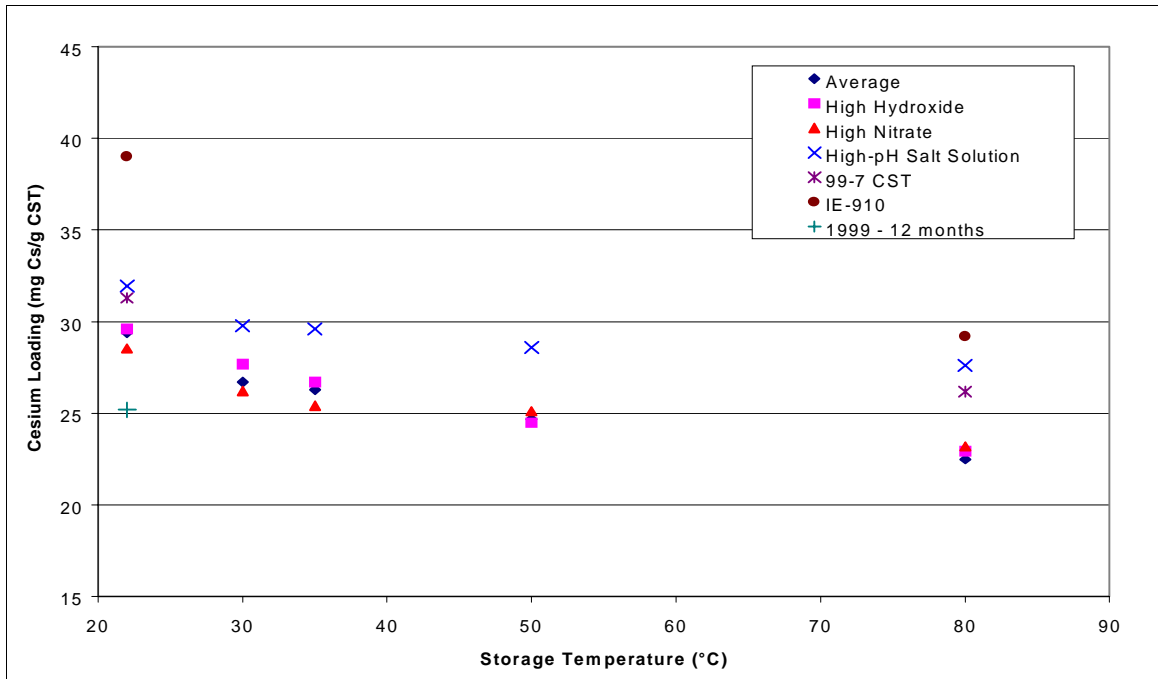
The CST stored in the average, high-hydroxide and high-nitrate simulants was weakly agglomerated after 1 month at 30, 35, 50, and 80°C. However, the samples stored at 23°C, as well as the CST in the high-pH salt solution at all temperatures, were still free flowing after 1 month. The agglomerated CST could be easily broken up by hand, using a plastic spoon. After 2 months, the samples stored at 23°C in the average, high-hydroxide, and high-nitrate simulants were also agglomerated. During subsequent samplings, the CST that had been previously broken up did not re-form into clumps, and the existing CST clumps did not become any more difficult to break apart. The CST stored in the high-pH salt solution did not form any clumps at any storage temperature during the 12 months that the tests were run. All of the simulants except the high-pH salt solution contained high concentrations of aluminum, which can form sodium aluminosilicates that in turn would cause agglomeration.

When compared with samples stored at 23°C, the cesium loading tests using granular CST (98-5 and 99-7) samples from the batch leaching tests at 80°C showed a decrease of about 19% in cesium capacity and 30% in distribution coefficient for the samples stored for 1 month or more in the average, high-hydroxide, or high-nitrate simulant solutions. This ratio did not change as the storage time was increased up to 12 months. (Note: All of the cesium loading tests were performed at 23°C using average simulant that initially contained 50 mg/L cesium.) The high-pH salt solution caused less of an effect on the cesium capacity (see Table 3). These results indicate that there was an initial degradation in the cesium sorption properties of CST as it contacted the supernate simulants at higher temperatures, sometime during the first month; however, no further change occurred thereafter. Figure 3 shows the cesium-loading results for the CST samples taken after 5 months of storage in the simulant solutions. The results for the other storage times are similar. The CST stored at intermediate temperatures also showed a drop in capacity as compared with the samples stored at 23°C—an average of 6, 7, and 11% reduction at 30, 35, and 50°C, respectively. The newer (lot 99-7) CST consistently showed a slightly higher cesium loading than the lot 98-5 material. Storage in the high-pH salt solution had the smallest effect on the cesium loading properties of the CST when compared with the results for the simulant solutions.

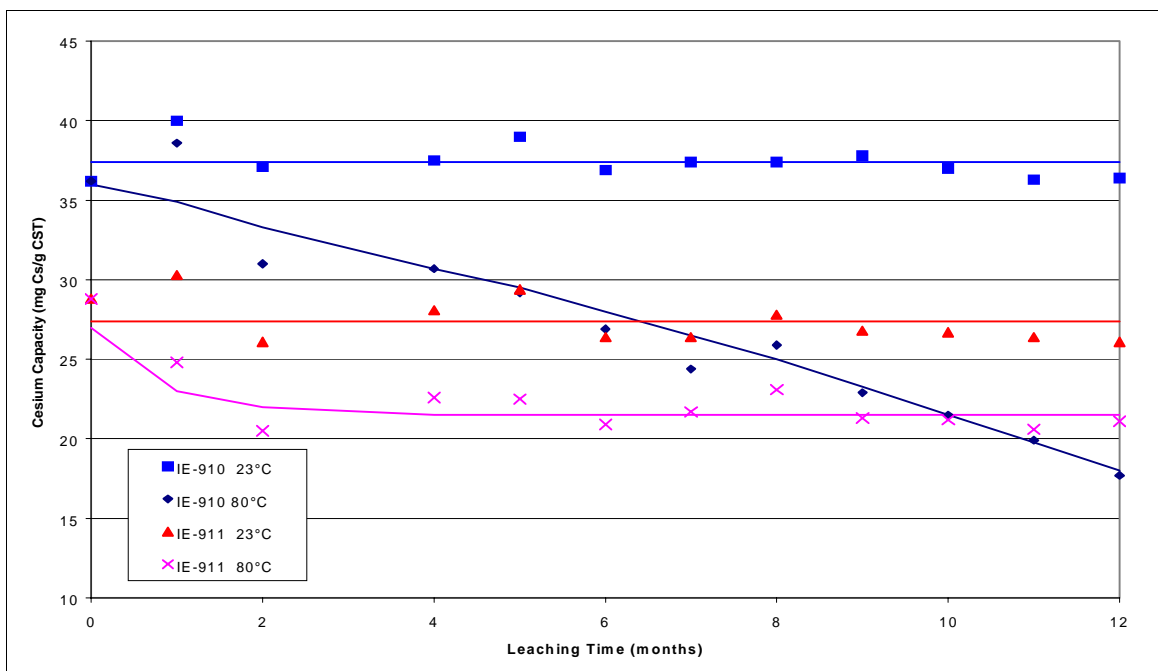
**Table 3. Average cesium loading capacities for granular CST samples stored at 23 and 80°C for 1 to 12 months**

CST	Type of simulant	Cesium capacity (mg Cs/g CST) $\pm$ S.D.		Change (%)
		Stored at 23°C	Stored at 80°C	
98-5	Average	27.6 $\pm$ 1.6	22.2 $\pm$ 1.7	19.5
98-5	High-hydroxide	28.0 $\pm$ 1.6	22.0 $\pm$ 1.5	21.5
98-5	High-nitrate	27.0 $\pm$ 1.7	22.9 $\pm$ 1.6	15.0
98-5	High-pH salt	29.1 $\pm$ 1.7	26.1 $\pm$ 1.2	10.3
99-7	Average	30.3 $\pm$ 1.5	24.8 $\pm$ 1.6	18.3

Figure 4 summarizes the results for granular CST (lot 98-5) and IE-910 CST powder stored in average simulant at 23 and 80°C for 0 to 12 months. The graph shows the initial drop in cesium loading for the samples stored at 80°C for 1 month. No further changes were observed for the granular CST for storage times up to 12 months. All of the other granular CST samples exhibited the same behavior. In contrast, the IE-910 powder showed a continuing drop in cesium loading as the storage time at 80°C increased.



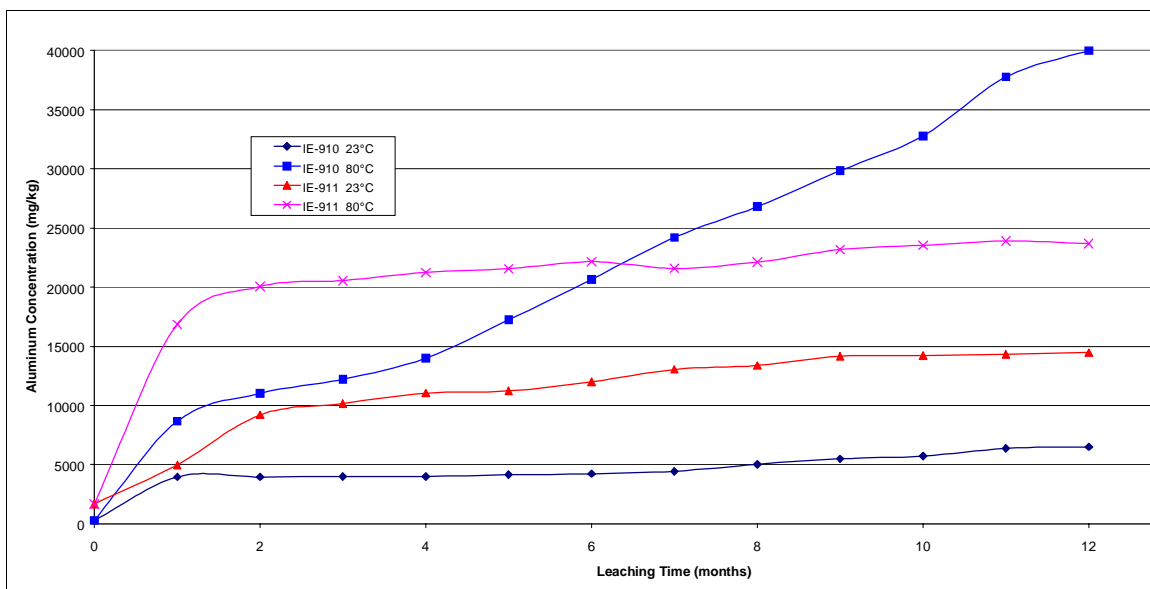
**Fig. 3. Cesium loading results for CST samples from the batch loading tests after 5 months. The 1999 sample had been exposed for 12 months. All of the loading tests were performed at room temperature. All tests used 98-5 CST unless otherwise indicated.**



**Fig. 4. Results of cesium loading tests for CST samples from batch leaching tests, using 98-5 granular CST and IE-910 powder stored in average simulant at 23 and 80°C.**



Figure 5 summarizes the aluminum concentrations for the CST samples discussed above (see Fig. 4). A decrease in cesium capacity corresponds with an increase in aluminum concentration. Work at Sandia National Laboratory<sup>4</sup> has indicated that cancrinite,  $\text{Na}_8\text{Al}_6\text{O}_{24}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ , forms on the CST particles during storage in SRS simulant solutions and that the amount increases with increasing time and temperature. The IE-911 samples initially had higher aluminum concentrations than the corresponding IE-910 samples, probably because of the difficulty in rinsing soluble aluminum from the pores of the IE-911 granules. The IE-910 sample stored at 80°C showed a continual rise in aluminum concentration, presumably from insoluble cancrinite present on the surface of the CST powder. By comparing the concentrations of aluminum and titanium in the CST samples, the ratio of CST to cancrinite can be calculated. Cancrinite is 14.8 wt % Al, IE-910 CST contains 19.9 wt % Ti, and IE-911 contains 17.8 wt % Ti. These ratios can be used to calculate an adjusted cesium loading, using only the weight of CST in each sample.

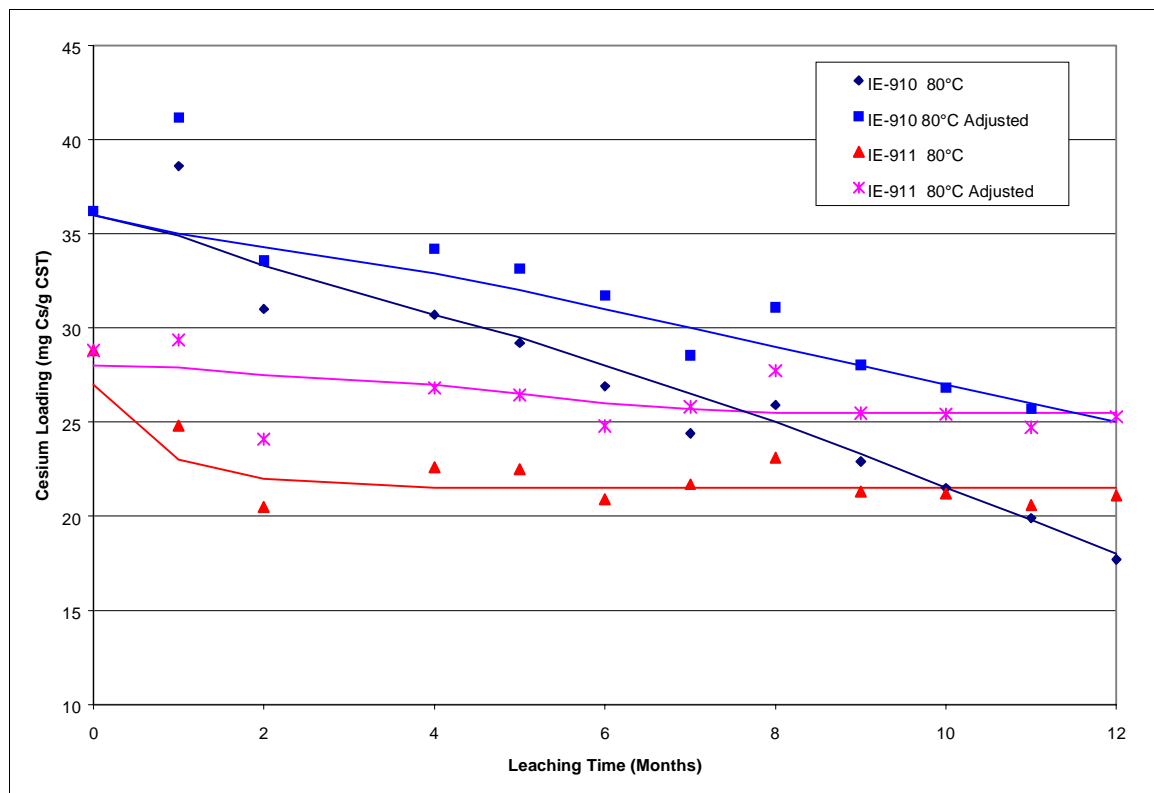


**Fig. 5. Aluminum concentrations for 98-5 granular CST and IE-910 powder stored in average simulant at 80°C.**

The results for the adjusted cesium loadings, calculated for the IE-910 and IE-911 samples stored at 80°C, are summarized in Fig. 6. The weight of cancrinite accounts for most of the loss in cesium capacity measured for the IE-911 samples, by simple dilution of the CST. The samples from the last 6 months of the test have adjusted cesium capacities that average 9% lower than for the first 6 months. The results for the IE-910 CST powder showed a significant loss in cesium capacity even after the dilution effect of the cancrinite was taken into account, suggesting that other mechanisms (i.e., blockage of the pores) are also important. The adjusted cesium capacity for the sample taken after 12 months was 30% lower than for the 1-month sample. The IE-911 samples from the batch test that was started in 1999 were stored for up to 19 months in average simulant at 23°C. These samples consistently showed slightly lower cesium capacities than those stored for 1 to 12 months; however, the aluminum concentrations in these samples were also

higher. The adjusted cesium loadings for the CST from the 1999 test were the same as those for the samples stored at 23°C in average simulant for 1 to 12 months.

The surface area, total pore volume, and micropore volume (volume of pores, <15 Å) were measured for selected samples of the CST. Samples from leaching tests conducted in 1999, which used average simulant containing 50 mg/L cesium, were initially measured. These results (see Table 4) suggested that there was a correlation between micropore volume and cesium uptake. Measurements using a larger number of CST samples (total of 52 samples) from the batch and column leaching tests this year showed wide variations in the measured micropore volume, total pore volume, and surface area, but no correlation was found between any of these measurements and the cesium loading results.

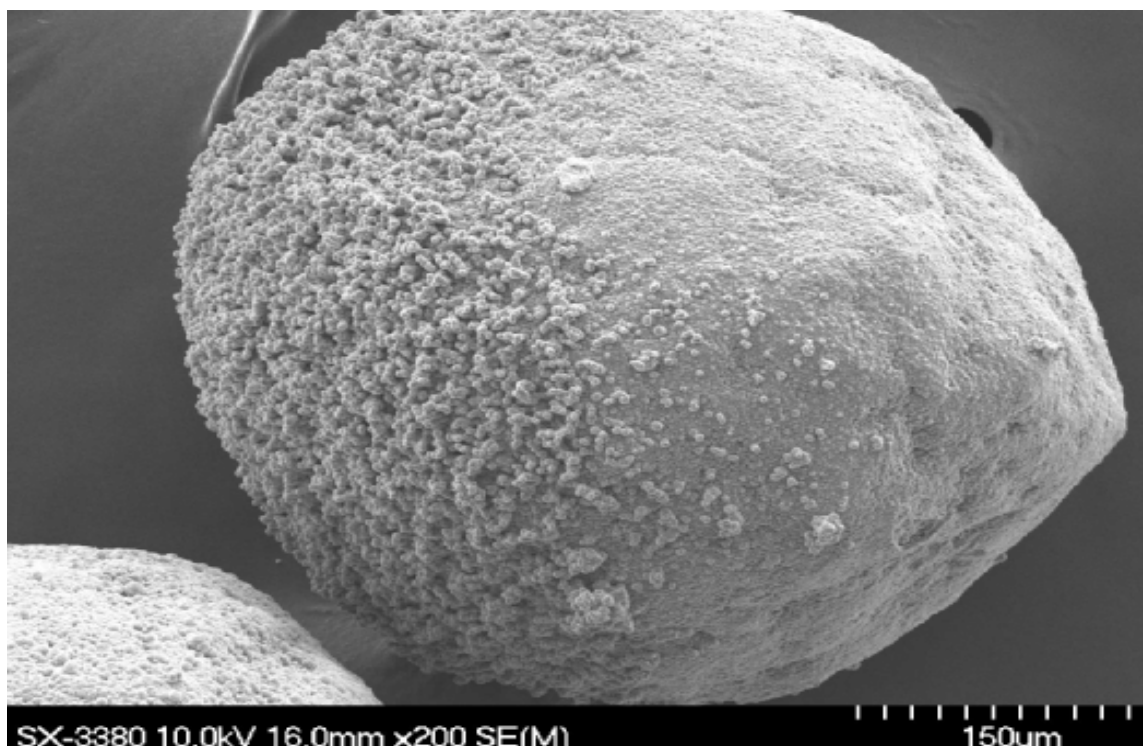


**Fig. 6. Comparison of measured and adjusted cesium capacities for IE-910 CST powder and 98-5 IE-911 CST samples stored at 80°C in average simulant.** The adjusted capacities were calculated by eliminating the weight of cancrinite in each sample.

**Table 4. Properties of 98-5 CST stored in average simulant under various conditions**

Storage conditions	Surface area (m <sup>2</sup> /g)	Total pore volume (cm <sup>3</sup> /g)	Micropore volume (cm <sup>3</sup> /g)	Cesium loading (mg Cs/g)
133 days at 23°C	30.3	0.047	0.0036	28.0
50°C for 86 days, then 23°C for 59 days	25.4	0.041	0.0035	20.8
80°C for 86 days, then 23°C for 59 days	37.3	0.057	0.0020	19.6
120°C for 86 days, then 23°C for 59 days	24.8	0.076	0.0000	2.3

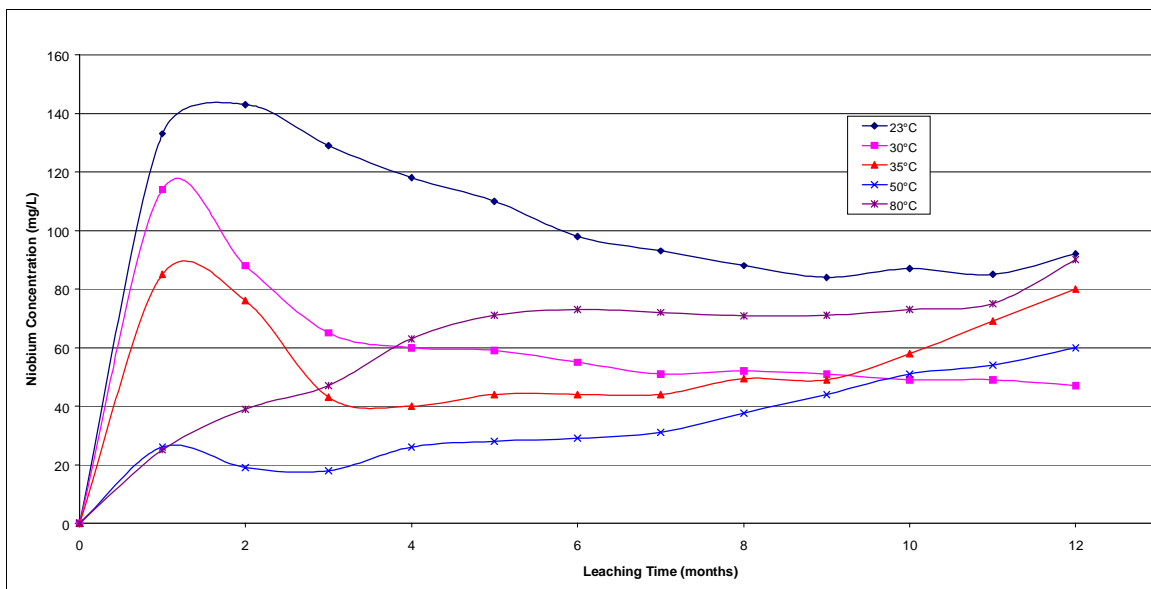
The photomicrographs of the CST samples stored in average, high-hydroxide, and high-nitrate simulant solutions, particularly those stored at higher temperatures, show precipitates on the surface of the CST particles (see Fig. 7). Energy-dispersive X-ray (EDX) spectroscopy showed that the precipitates contained Na, Si, and Al, which is consistent with cancrinite. The CST stored in the high-pH salt solution did not show any precipitates or other visible changes in the CST particles.



**Fig. 7. Photomicrograph of 98-5 CST stored in high-nitrate simulant at 80°C for 4 months.**

ICP analyses of the CST samples from the batch tests revealed no significant changes in the concentrations of the major components of the IE-910 and IE-911 CST (Si, Ti, Nb and Na). In addition, no change was noted in the concentration of zirconium, which is the main component of the binder for the IE-911 samples.

ICP analyses of the simulant solutions from the batch leaching tests generally showed a slight decrease in their aluminum concentrations, which is consistent with the precipitation of cancrinite. Niobium, which is one of the components of CST, exhibited unusual behavior in the simulant solutions from the batch tests with the IE-911 CST. The results for the average simulant are summarized in Fig. 8. Similar results were measured for the high-hydroxide and high-nitrate simulants. The concentration of niobium initially increased as it was leached from the CST, particularly at the lower temperatures, but then usually decreased, presumably as it precipitated. At 80°C the niobium concentration was initially low but continued to increase over time, approaching the concentration measured in the 23°C solution after 12 months. In the high-pH salt solution, the niobium concentration increased during the first month and then leveled off and remained fairly constant at 150-180 mg/L for the 23°C samples and at about 70 mg/L for the samples stored at 80°C. The simulant from the batch tests with the IE-910 CST powder showed consistent niobium concentrations of about 400 mg/L at 23°C and about 40 mg/L at 80°C.



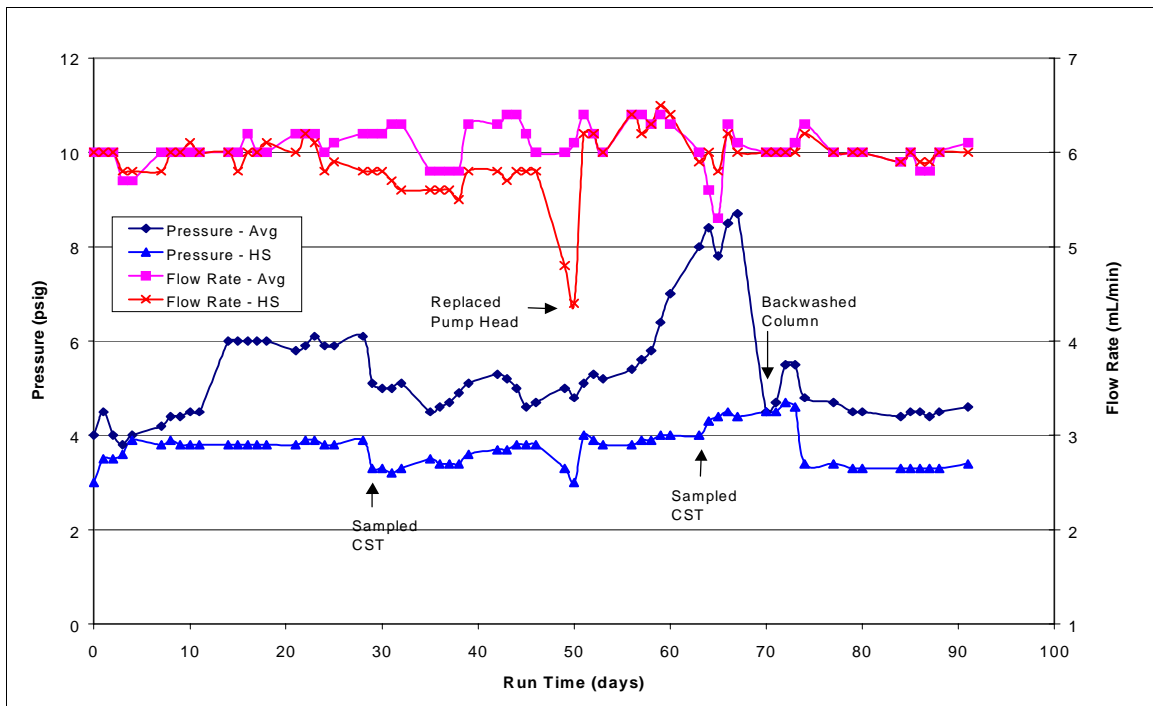
**Fig. 8. Niobium concentrations in average simulant from batch leaching tests with 98-5 CST.**

### 3.3 Column Leaching Tests

For the column with 98-5 CST and average simulant, the combined back pressure for the filter and column was 4 to 5 psig during the first 10 days; then the pressure increased to about 6.0 psig for the next 20 days. The back pressure for the high-pH salt solution column remained steady at 3 to 5 psig during the 12 months that the test was run. The pressures and flow rates for both columns during the first three months of operation are shown in Fig. 9.

The CST in the top of the average simulant column was clumped together when the first sample was taken after 1 month. The back pressure in the column decreased after sampling; however, it increased again during the second month of operation, reaching 8.0 psig when the 2-month samples were taken. This time the pressure did not decrease after sampling, continuing above

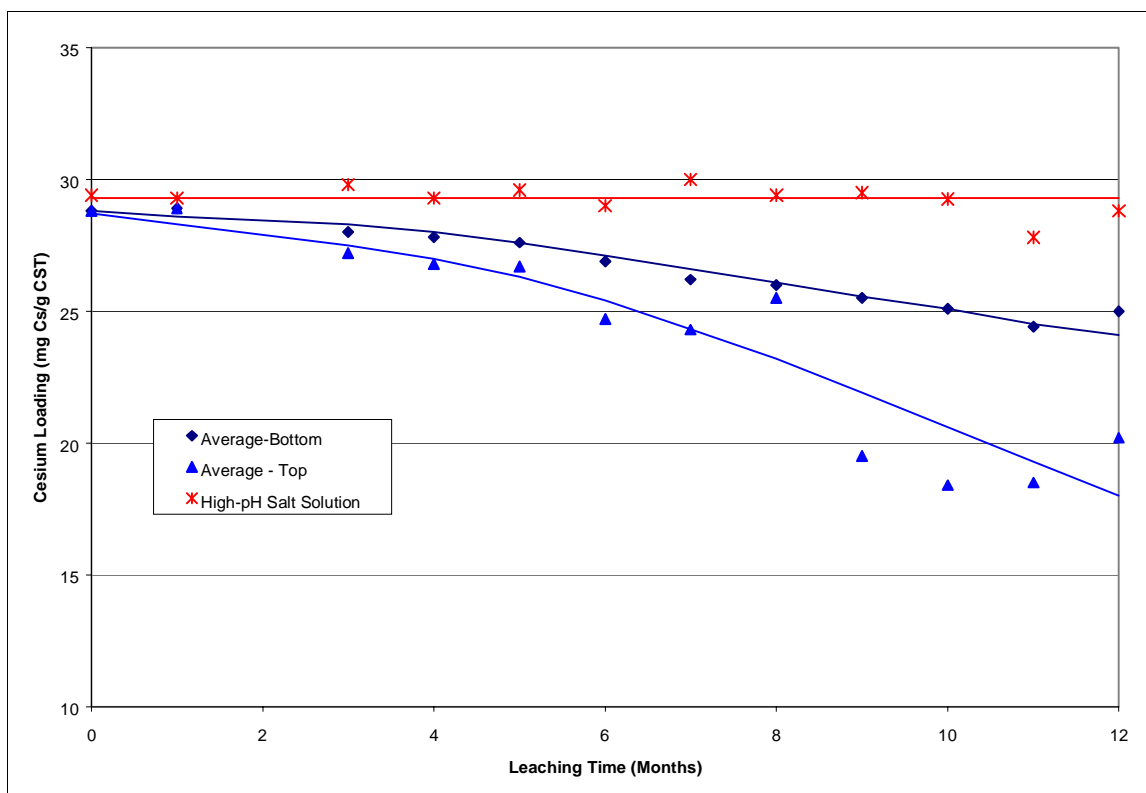
8 psig until the column was backwashed on day 67. The column was backwashed for 2 min at a flow rate of 15 mL/min, which suspended some fines above the bed of CST; however, the backwashing was not vigorous enough to remove the fines from the column or to fluidize the CST. The CST throughout the column was lightly agglomerated and tended to move up the column during backwashing. Light tapping on the column helped break up the clumps of CST sufficiently to resettle the bed. The back pressure remained below 6 psig for the next 7 months of operation. Apparently, redistributing the fines and partially breaking up the clumps of CST was sufficient to restore fluid flow through the bed of CST. The back pressure increased again after a total of 9 months of operation, staying in the range of 6 to 9 psig until the test was stopped at 12 months. The column was backwashed several times during this period, with very limited effect on the pressure drop. The height of the CST bed, which was gradually reduced as samples were removed each month, was down to 70 cm at the end of the test, as compared with the original height of 110 cm. The pressure drop for the bed of CST alone was 0.3 psi/ft at the start of the test and 1.8 psi/ft at the end.



**Fig. 9. Flow rate and pressure drop data for column leaching tests using 98-5 CST.**

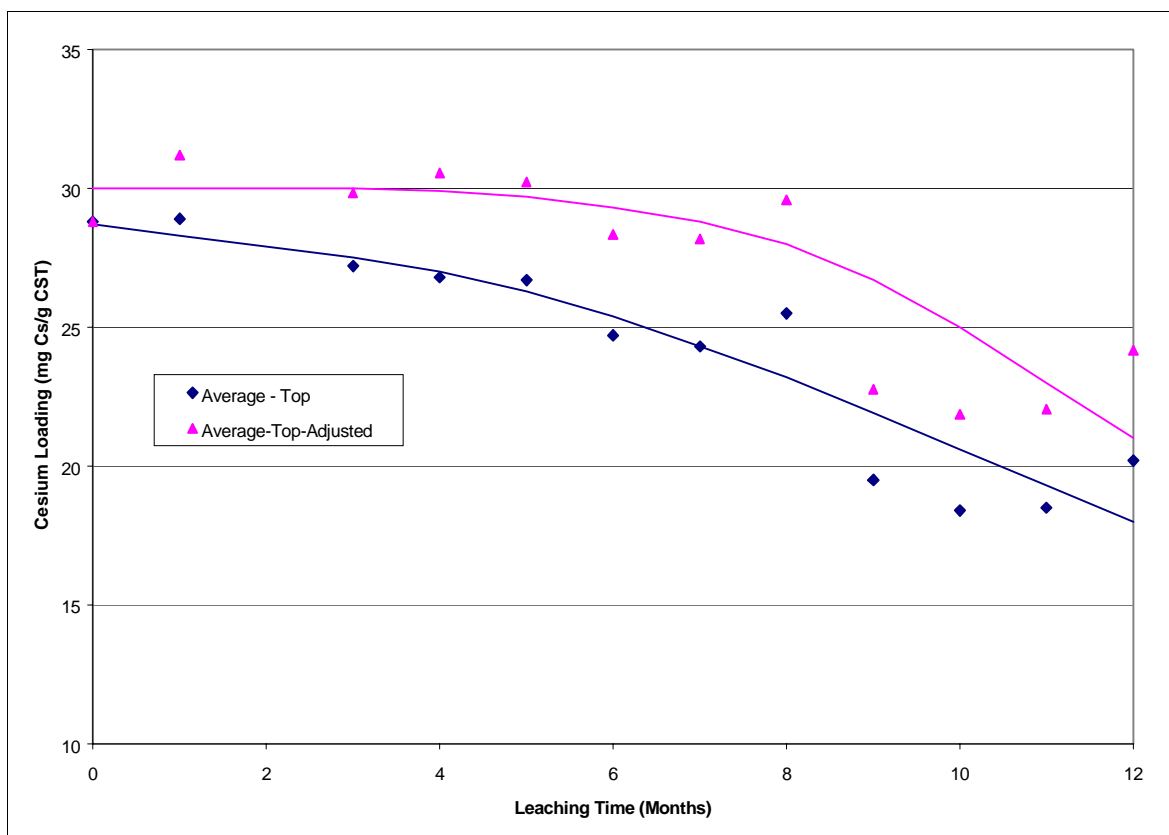
A white precipitate slowly formed in the silicone rubber tubing leading from the filter to the top of the average simulant column and from the exit of the column back to the feed tank. These pieces of tubing were replaced three times during the 12 months the test was in progress. Samples of the precipitate were analyzed by ICP, which confirmed that Na, Al, and Si were the major constituents. N. Greeley of UOP, LLC identified the precipitate as Zeolite-A, using X-ray diffraction. Apparently the surface structure of the silicon rubber tubing promotes the formation of Zeolite-A rather than cancrinite, which forms on the CST particles. There was no indication that solids collected in the filter, on the walls of the column, or in the feed tank.

The cesium loading results for the CST from the column leaching tests are summarized in Fig. 10. The CST from the column using average simulant consistently showed lower cesium capacities for the samples from the top of the bed than for those from the bottom. There was no difference in the cesium capacities of CST samples from the top and bottom of the column using high-pH salt solution. The cesium loading results for the 2-month samples from the two columns were not plotted because they were unusually high and “out of line” with the other data.



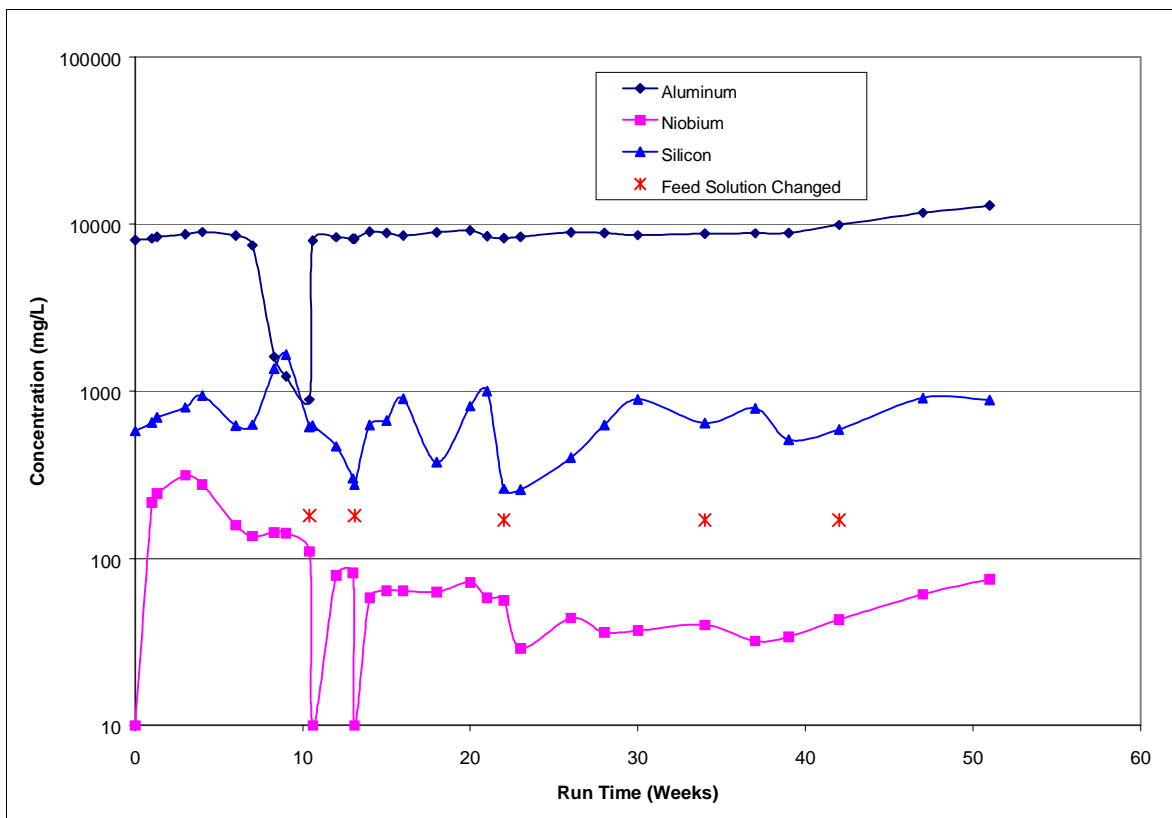
**Fig. 10. Cesium distribution coefficients for 98-5 CST samples from the column leaching tests.**

Slow increases were noted in the aluminum concentrations of the CST samples from the average simulant column, particularly for the samples from the top of the bed—indicating increased amounts of cancrinite on the CST. An adjusted cesium loading capacity was calculated for the CST in the samples by using the ratio of aluminum to titanium in each sample. For the samples from the bottom of the column, the adjusted loading showed a consistent value of about 30 mg Cs/g CST; thus, dilution of the CST by cancrinite was the cause of the drop in the measured loading for these samples. The adjusted results for the samples from the top of the bed are shown in Fig. 11. Dilution of the CST by cancrinite accounted for most of the loss in cesium capacity during the first 8 months of the test; however, even the adjusted capacity decreased significantly during the last 4 months of the test, indicating an additional mechanism for the loss in cesium capacity.



**Fig. 11. Comparison of measured and adjusted cesium capacities for 98-5 CST samples from the top of the average simulant column.** The adjusted capacities were calculated by eliminating the weight of cancrinite in each sample.

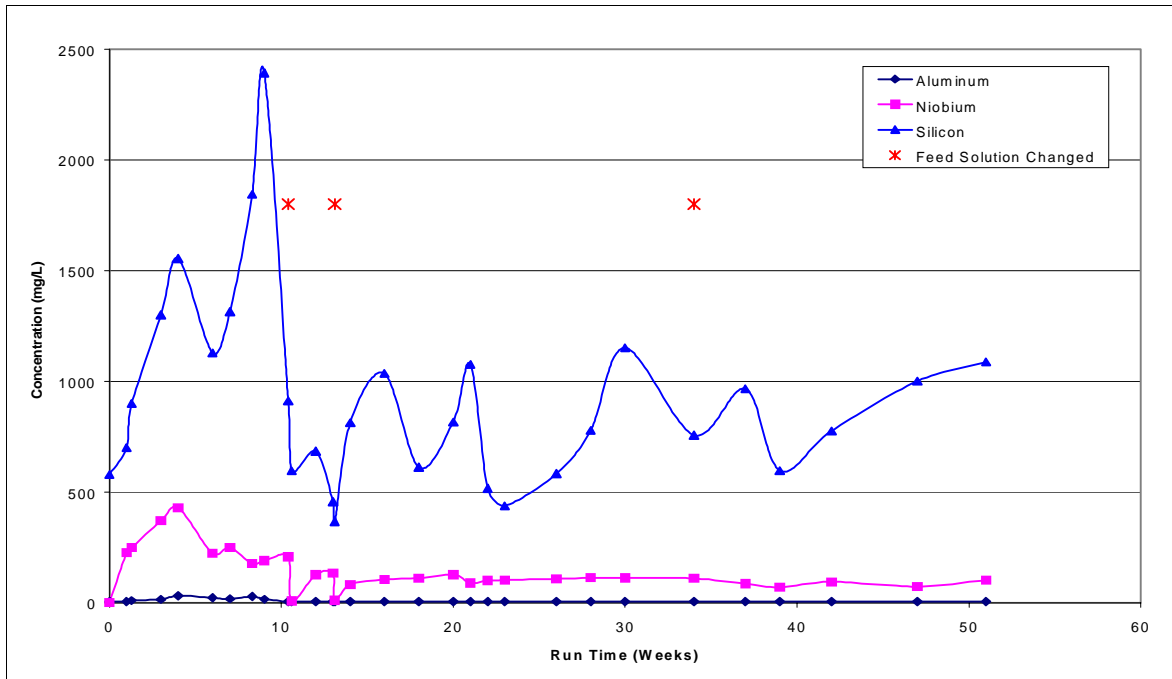
The ICP results for Al, Si, and Nb in the recirculating simulant solutions are shown in Figs. 12 and 13 (note that different y-axis scales are used). For the average simulant column, the silicon concentration slowly increased initially but then decreased along with the aluminum concentration, indicating the precipitation of cancrinite. The concentrations were restored when the feed solution was changed, and the aluminum concentration remained constant for the remainder of the 12-month test. The silicon concentration has continued to change but has not shown any consistent trend, so this is probably just analytical variation. For the column with high-pH salt solution, the silicon concentration increased during the first 10 weeks but then dropped back to the starting level when the feed solution was changed. As with the results for the average simulant column, considerable scatter was noted in the silicon concentrations. Analysis of the starting solution (0 weeks) showed 500 mg/L silicon, even though there was no silicon in the solution. This analytical variation was apparently caused by small variations in the measured silicon concentrations of blank solutions (ultrapure acid carried through the same digestion process as the samples). Because of the large dilutions required for the samples, small amounts of silicon in the reagents could cause large variations in the calculated sample concentrations. In each solution, the niobium rapidly leached from the CST—up to an apparent solubility limit—each time new feed solution was introduced.



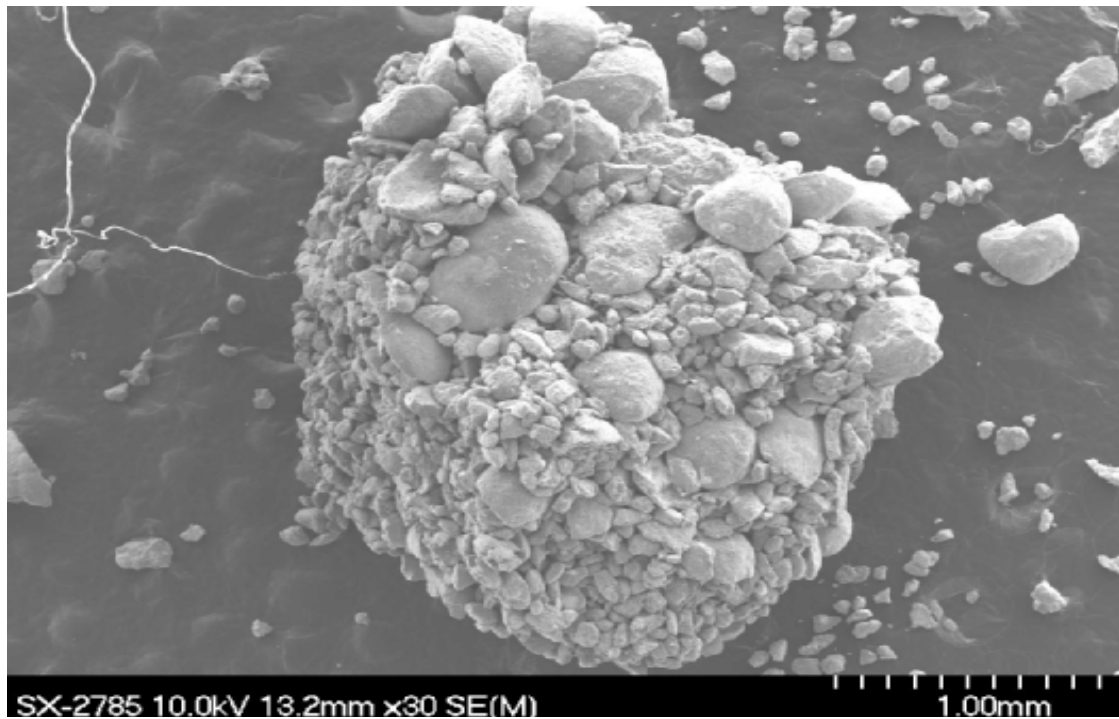
**Fig. 12. Metal concentrations in the average simulant from the column leaching test using 98-5 CST.**

Photomicrographs show that smaller CST particles and fragments tend to collect in the upper parts of the columns. The first sample from the top of the column using average simulant, taken after 1 month, showed CST particles and fines clumped together (see Fig. 14). The 1-month sample from the column using high-pH salt solution showed numerous smaller and broken CST particles, which migrated to the top of the column during initial backwashing of the CST; however, no clumping was visible (see Fig. 15).





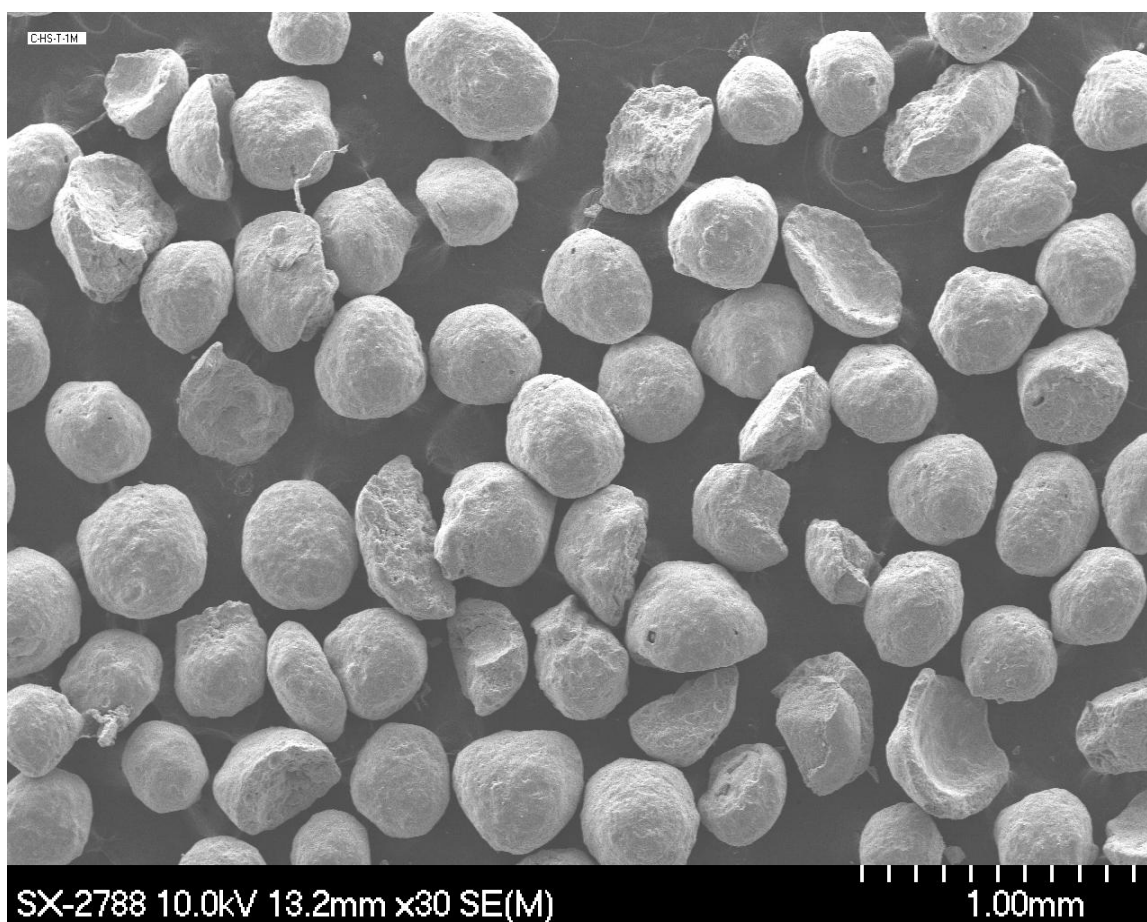
**Fig. 13. Metal concentrations in the high-pH salt solution from the column leaching test using 98-5 CST.**



**Fig. 14. SEM photomicrograph of CST clump that was formed at the top of the column using average simulant and 98-5 CST after 1 month.**

### 3.4 New Batch Leaching Tests

Batch leaching tests using the new UOP baseline IE-911 CST (9090-76) and the corresponding IE-910 powder were started on December 5, 2000. Samples were taken after leaching times of 1, 2, and 3 months. The samples of 9090-76 CST stored at 80°C showed a decrease in cesium capacity of 10% after 1 month, 14% after 2 months, and 16% after 3 months. The samples stored at the lower temperatures indicated no significant change in cesium loading, except for the sample stored at 50°C for 3 months (5.6% loss). The CST stored at 80°C was lightly clumped together after 1 month, but all of the samples at the lower temperatures were still free-flowing after 3 months. The 9090-76 CST, which was pretreated with once-through 3 M NaOH, had less tendency to clump than the earlier CST samples that were pretreated using recirculating 0.1 M NaOH. The IE-910 powder samples stored at 80°C showed decreases in cesium capacity of 8, 9, and 16% after 1, 2, and 3 months, respectively. Results for the sample stored at 50°C for 3 months indicated a drop of 4% in cesium capacity; no significant changes were noted in the other samples.



**Fig. 15. SEM photomicrograph of 98-5 CST from the top of the column using high-pH salt solution after 1 month of operation.**

The batch tests with the UOP-pretreated CST (MH-9098-9) were started on January 17, 2001. Samples were taken after leaching time of 2 weeks and again after 2 months. The samples stored

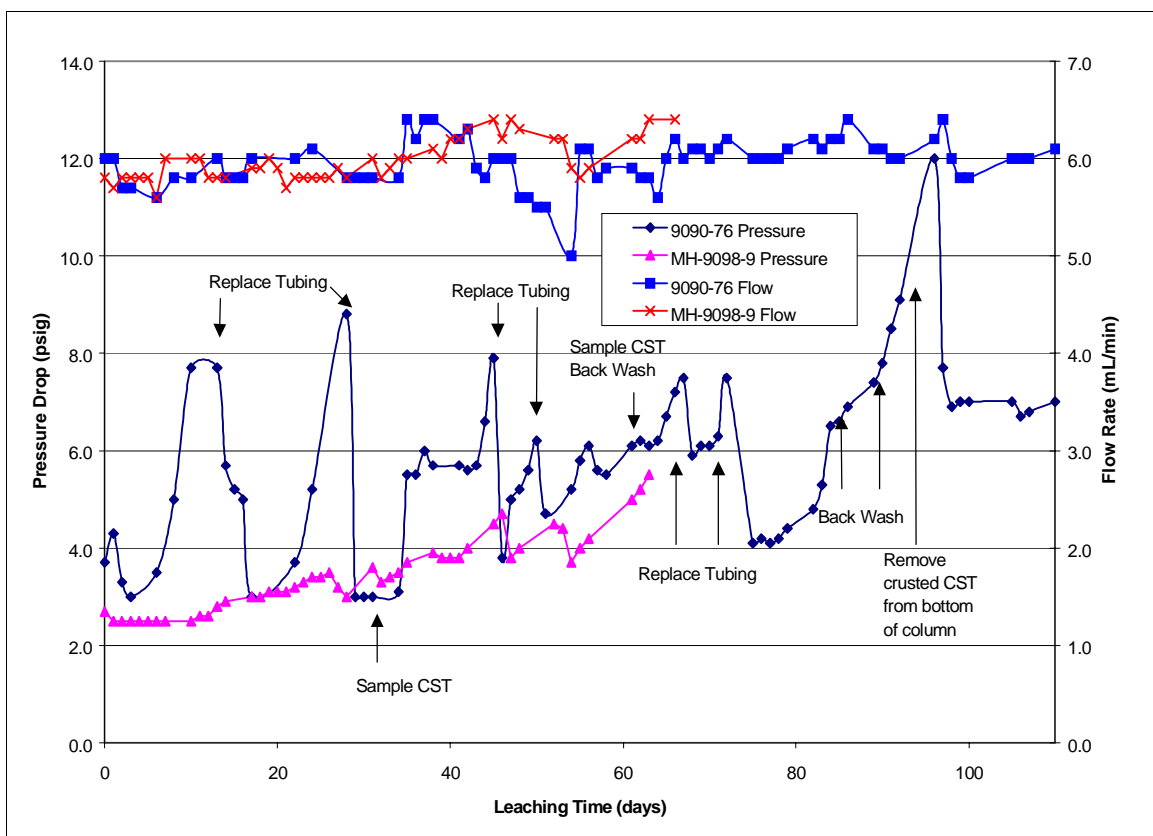
at 80°C have shown decreases in cesium capacity of 9 and 15% after 2 weeks and 2 months, respectively. The MH9098-9 CST stored at 80°C was lightly clumped after 2 weeks, as was the sample stored at 50°C for 2 months. All of the samples stored at lower temperatures were free flowing after 2 months. The cesium loading results for the new batch tests are very similar to those for the completed (12-month) tests using 98-5 and 99-7 granular CST and the corresponding CST powder. All of the new CST samples stored at 80°C showed increases in aluminum concentration in the CST, consistent with the precipitation of cancrinite. Using the aluminum-to-titanium ratio to calculate an adjusted cesium capacity indicated that the weight of cancrinite on the CST would account for most of the drop in measured cesium capacity for all of the new CST samples.

### 3.5 New Column Leaching Tests

The column test with the new baseline CST (9090-76) and recirculating average simulant was started on December 5, 2000. Samples of CST after contact times of 1, 2, and 3 months showed no change in cesium capacity, averaging 32 mg Cs/g CST, which is similar to the early results for the CST samples from the column test using 98-5 CST. The amount of aluminum measured in the CST samples was much lower than that for the column test using 98-5 CST and average simulant after the same contact time (3000 to 4000 mg/kg for 9090-76 CST and 9000 mg/kg for 98-5 CST after 2 months). A white precipitate continued to collect in the silicone rubber tubing leading to and from the column for the first 3 months. The tubing was replaced six times over a period of 3 months due to flow restrictions in the tubing. The amount of precipitate collected in the tubing was much larger than that seen in the earlier column test using 98-5 CST.

Clumps of CST were visible throughout the column during backwashing, after 2 months of operation. The pressure and flow rate data are included in Fig. 16. After the third month, the accumulation of solids in the tubing greatly diminished, but the pressure drop through the bed of CST started increasing. Backwashing the column had little effect on the pressure drop. The column was inverted, and the bottom connection was removed to determine the cause of the high pressure drop. A layer of crusty CST, about 0.5 cm thick, which was stuck to the bottom screen of the column, was removed by scraping. Removal of the CST from the screen reduced the pressure drop through the bed from 12 psig to 7 psig. The aluminum concentration in the CST removed from the plug was 3800 mg/kg, compared with concentrations of 4300 and 2300 mg/kg for CST samples removed from the top and bottom, respectively, of the column after exposure times of 3 months. The concentration of aluminum in the plug was much higher than for the CST that was sampled from the valve near the bottom of the column, just above the plug. Apparently the CST in the plug contained more sodium aluminosilicate precipitate than the surrounding CST; however, the reason for this difference is not currently known.

The column test using the UOP-pretreated CST (MH-9098-9) and recirculating average simulant was started on January 18, 2001. Samples taken after 2 weeks and after 2 months showed cesium loadings of 32 to 33 mg Cs/g CST, which are the same as those measured for as-received MH-9098-9 CST that was contacted with simulant for only 10 min and then rinsed with water. The aluminum concentrations in the 2-month CST bottom and top samples were 2100 and 5600 mg/kg, respectively, which are similar to the results for the baseline CST column test. No precipitate was visible in the tubing for this column for the first month, but small amounts started to collect during the second month of operation. The amount of precipitate collected in the tubing was much less than that for the column using 9090-76 CST. The pressure drop for this column stayed quite low, although there was a slow increase during the 2 months of operation (see Fig. 16).



**Fig. 16. Operational data for new column leaching tests.**

### 3.6 Once-Through Column Test

The column using the UOP-pretreated CST (MH-9098-9) with average simulant pumped through the column in once-through fashion (no recirculation of simulant) was started on February 3, 2001. No increase in pressure drop through the column was observed, and no precipitates formed in the Teflon tubing leading to and from the column. CST samples were taken from the top, middle, and bottom of the bed after 1 month of operation. The aluminum concentrations of the CST samples were 1400, 890, and 840 mg/kg for samples from the top, middle, and bottom of the bed, respectively. These results are similar to those for samples taken from the column test using MH-9098-9 CST and recirculating simulant after 2 weeks, and lower than the 2-month samples. The cesium capacities of the CST samples from the once-through column were the same as those measured for as-received MH-9098-9 CST that was contacted with simulant for only 10 min and then rinsed.

#### 4. CONCLUSIONS

Experimental determination of the chemical stability of CST in long-term experiments that were initiated in FY 1999 and FY 2000 at ORNL has been completed. The work was performed to evaluate the long-term chemical stability of CST when exposed to several different waste compositions using a range of possible operating conditions and temperatures.

The cesium-loading capacity of CST decreased rapidly when measured at increasingly higher temperatures; a drop of 76% was noted as the loading temperature was increased from 23 to 80°C, due to a shift in equilibrium at the higher temperatures. Granular CST (UOP IONSIV® IE-911) that had been stored in cesium-free simulant solutions for 1 to 12 months at 80°C and then cooled to 23°C for the loading test showed a 23% drop in cesium capacity as compared with CST stored at 23°C for the same time periods. This indicates that the decrease in CST loading capacity is not entirely reversible for extended-duration exposure at elevated temperatures. It should be noted however, that these conditions are not expected under normal operation of CST columns. Most of the decrease in cesium capacity was caused by the added mass of cancrinite (a sodium aluminosilicate) that precipitated on the CST from the simulant solutions.

CST exposed to recirculating simulant solution at room temperature in a flow-through column test showed a slow decrease in cesium-loading capacity, with a drop of 30% after 12 months for samples from the top of the column and a drop of 13% for samples from the bottom. Precipitation of cancrinite on the CST accounted for the loss of cesium capacity for the samples from the bottom of the column and most (but not all) of the loss for samples from the top of the column. Long-term exposure to flowing, but not recirculating, supernate solution would be encountered in the baseline design for the SRS, where three 16-ft-tall by 5-ft-diam columns are aligned in series.

CST in both the batch and the column tests showed a tendency to become agglomerated with cancrinite, which was formed by the reaction of Al, Si, and Na from the simulant solutions with the additional Si leached from the CST. These clumps could potentially cause fluid channeling within a column and make removal of the CST more difficult. The cancrinite also causes an apparent reduction in the cesium-loading capacity by adding inert mass to the CST.

New batch and column leaching tests are in progress using samples of both baseline (9090-76) and improved (MH-9098-9) CST from UOP. The baseline CST is similar to the earlier samples; the improved sample was treated at UOP to reduce the amounts of niobium and silicon that leaches from the CST. The baseline CST was pretreated with 3 M NaOH prior to being used, and the improved CST was used as received. Both samples have shown less tendency to clump than the earlier CST, which may be related to the lower NaOH concentration used to pretreat the earlier samples. Both of the new CST samples formed clumps after storage in simulant for 1 month at 80°C, while the improved sample formed clumps after 2 months at 50°C. All of the remaining batch samples were free flowing after 3 months. The column test using baseline CST contained clumps of CST after 2 months of exposure to recirculating simulant. The cesium capacities of the new samples from the batch and column tests at room temperature are similar to those of the comparable samples from the original tests. The decrease in cesium capacity has been slower for the new samples stored at higher temperatures in the batch leaching tests. The samples stored at 50 and 80°C showed drops of 4 and 14%, respectively, after 2 months, while the samples stored at lower temperatures have shown no decrease in cesium capacity to date.

## 5. REFERENCES

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4. M. Nyman, T. Headley, and T. M. Nenoff, *Characterizations of As-Received, Pre-Treated and Simulant-Treated IE-911 (UOP batches 98-5, 99-7, 99-9) Interim Report*. Sandia National Laboratories, Albuquerque, N.M., October 2000.
5. Z. Zheng, D. Gu, and R. G. Anthony, "Estimation of Cesium Ion Exchange Distribution Coefficients for Concentrated Electrolyte Solutions When Using Crystalline Silicotitanates," *Ind. Eng. Chem. Res.* **34**, 2142-47 (1995).

## **6. APPENDIXES**

## Appendix A. Results of Cesium Loading Tests at Various Temperatures

(Results are based on CST dry weight at 400°C;  
CST is IE-911, Lot 98-5 unless otherwise indicated.)

Loading temp. (°C)	Cesium loading (mg Cs/g CST) in each simulant solution					
	Average	High- hydroxide	High- nitrate	High-pH salt solution	Avg, 99-7	Avg, IE-910
24	32.0	34.5	32.0	28.7	32.9	37.1
30	29.3	32.8	31.1	27.5		
35	28.2	30.5	28.0	25.4		
40	23.4	27.8	24.5	21.3		
45	23.2	25.6	22.9	19.5		
50	18.3	22.1	19.0	15.9		
60	13.5	16.9	15.0	11.1		
70	8.3	13.0	10.1	7.4		
80	7.6	8.7	6.5	5.5	7.3	12.8

Loading temp. (°C)	Distribution coefficients ( $K_d$ ) values (L/kg) in each simulant solution					
	Average	High- hydroxide	High- nitrate	High-pH salt solution	Avg, 99-7	Avg, IE-910
24	1407	1674	1406	1128	1487	1962
30	1171	1485	1329	1038		
35	1090	1270	1071	893		
40	779	1059	841	672		
45	767	906	750	590		
50	533	706	561	438		
60	350	474	443	273		
70	195	381	243	170		
80	174	204	146	122	132	323



**Appendix B. Cesium Loading Results for 98-5 CST Samples  
from the Column Leaching Tests**

Time (months)	Cesium loading (mg Cs/g CST)		
	Average simulant		High-pH salt
	Bottom	Top	All samples
0	28.8	28.8	28.8
1	28.9	29.2	29.3
2	32.0	32.1	33.0
3	28.0	27.2	29.8
4	27.8	26.8	29.3
5	27.6	26.7	29.6
6	26.9	24.7	29.0
7	26.2	24.3	30.0
8	26.0	25.5	29.4
9	25.5	19.5	29.5
10	25.1	18.4	29.3
11	24.4	18.5	27.8
12	25.0	20.2	28.8

### Appendix C. Cesium Loading Results (mg Cs/g CST) for CST Samples from Batch Leaching Tests

Sample ID	Leaching time (months)											
	1	2	3	4	5	6	7	8	9	10	11	12
AVG-23	30.3	26.1	30.5	28.1	29.4	26.4	26.4	27.8	26.8	26.7	26.4	26.1
AVG-30	28.3	24.5	29.2	26.0	26.7	23.7	23.3	25.2	24.6	24.9	24.1	24.8
AVG-35	26.9	25.2	28.1	25.4	26.3	24.1	23.5	24.8	24.3	24.0	23.2	22.0
AVG-50	27.0	22.3	27.0	23.7	24.7	22.7	22.2	23.5	23.0	22.5	22.2	23.0
AVG-80	24.8	20.5	26.1	22.6	22.5	20.9	21.7	23.1	21.3	21.2	20.6	21.1
HH-23	30.2	26.2	30.3	28.7	29.6	25.4	27.9	29.2	27.7	26.8	27.0	27.3
HH-30	29.7	24.5	29.5	26.5	27.7	25.0	26.2	26.9	25.8	26.3	24.5	26.1
HH-35	28.7	25.7	29.0	26.0	26.7	24.4	25.3	27.0	26.1	25.8	25.5	26.3
HH-50	27.0	24.6	27.1	23.9	24.5	22.7	24.0	25.9	24.6	24.0	23.6	24.2
HH-80	24.2	21.6	25.1	22.2	22.9	19.8	21.0	22.1	22.0	21.0	20.8	21.2
HN-23	29.2	27.3	30.5	27.1	28.5	24.2	25.4	27.1	25.9	26.2	26.2	26.1
HN-30	28.8	24.5	28.4	24.9	26.2	22.0	24.0	23.6	24.4	24.6	24.3	24.6
HN-35	27.6	22.5	27.6	24.2	25.4	22.5	23.8	24.6	23.8	24.3	23.7	24.5
HN-50	25.9	23.6	27.4	23.8	25.1	21.8	23.1	23.9	22.8	23.5	22.9	23.6
HN-80	25.3	22.5	26.8	21.8	23.2	21.5	22.7	23.5	21.6	22.0	21.7	22.7
HS-23	29.7	26.5	32.1	27.1	31.9	28.3	29.4	29.9	28.8	28.7	28.2	28.3
HS-30	21.4	-	32.0	27.4	29.8	27.4	27.9	27.5	27.4	27.4	27.0	28.0
HS-35	28.9	25.8	30.2	27.1	29.6	26.7	27.8	28.5	27.2	27.6	27.0	27.3
HS-50	28.1	24.1	29.5	26.4	28.6	26.0	26.4	27.3	26.4	26.0	25.2	26.6
HS-80	27.1	25.2	28.2	26.2	27.6	24.1	24.5	26.6	25.4	26.0	25.7	26.1
910-AVG-23	40.0	37.1	53.8	37.5	39.0	36.9	37.4	37.4	37.8	37.0	36.3	36.4
910-AVG-80	38.6	31.0	34.3	30.7	29.2	26.9	24.4	25.9	22.9	21.5	19.9	17.7
99-7-AVG-23	31.1	26.7	33.5	29.9	31.3	29.8	29.7	31.3	30.3	30.4	29.8	30.3
99-7-AVG-80	25.7	24.7	28.5	25.7	26.2	23.1	23.8	25.6	23.7	23.4	23.0	24.0
1999-AVG-23	23.6	25.8	27.4	24.6	25.2	22.3	22.4	24.4	23.8	23.2	23.0	24.1

AVG = average simulant; HH = high hydroxide; HN = high nitrate; HS = high-pH salt solution. Stored at temperatures of 23, 30, 35, 50, and 80°C.

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