

**Key Words:**  
Actinide  
Removal  
High-level Waste

## **Optimized Monosodium Titanate Phase II Supplemental Testing Report – Uranium Adsorption and Shelf-Life Measurements**

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## **List of Abbreviations**

ACTS	Actinide and Chemical Technology Section
AD	Analytical Development
APT	amorphous peroxotitanate
ARP	Actinide Removal Process
DF	Decontamination Factor
HLW	High Level Waste
ICP-MS	Inductively Coupled Plasma-Mass Spectroscopy
LWO	Liquid Waste Operations
MST	monosodium titanate
mMST	modified monosodium titanate
SRNL	Savannah River National Laboratory
SWPF	Salt Waste Processing Facility

## 1.0 Executive Summary

The DOE Office of Waste Processing recently funded supplemental Phase II testing to further investigate the uranium affinity and shelf-life of modified monosodium titanate (mMST). Testing results confirmed earlier findings that the mMST exhibits much lower affinity for uranium than the baseline monosodium titanate (MST) material. The loading of uranium onto the mMST sample measured more than an order of magnitude lower than that of the MST. This finding indicates that the use of mMST provides a significant advantage over MST in that the mMST will not concentrate enriched uranium to the degree that MST does. The reduced affinity of mMST for uranium allows more operational flexibility in treating waste solutions from a nuclear criticality safety perspective.

Testing results also indicate that the mMST exhibits good shelf-life with no measurable loss in plutonium and neptunium removal upon storage of samples at ambient laboratory temperatures for up to 30-months. Testing did exhibit a change in strontium removal performance for both the mMST and MST samples at the most recent testing event. However, the decrease in strontium removal performance proved lower for the mMST than the MST sample. Given these positive findings SRNL recommends continued development of mMST as a replacement for MST in pretreatment facilities at the Savannah River Site (SRS).

## 2.0 Introduction

Phase II testing (funded by the Office of Waste Treatment) continued development of a chemically-modified monosodium titanate (MST) material that exhibited improved strontium and actinide removal characteristics compared to the baseline MST material.<sup>1</sup> The modified MST (mMST) is a member of the larger family of peroxotitanate materials referred to as amorphous peroxotitanate (APT).<sup>2</sup> The improved actinide removal performance of the mMST and APT materials offers the potential for increased throughput performance for the Actinide Removal Process (ARP), Salt Waste Processing Facility (SWPF) and Enhanced Process for Radionuclide Removal (EPRR).

An interesting finding for the mMST is a reduced affinity for uranium compared to MST. Adsorption of uranium onto a sorbent is a concentrating mechanism for uranium and, thus, represents a potential nuclear criticality safety concern if the uranium is enriched in U-235. Previous testing has measured uranium loadings as high as  $14.0 \pm 1.04$  wt % onto MST.<sup>3,4</sup> This loading is sufficiently high to require nuclear criticality control strategies when processing large volumes of waste solutions containing high concentrations of enriched uranium. Thus, development of a sorbent that exhibits reduced affinity for uranium without a decrease in strontium, plutonium and neptunium removal is an attractive characteristic for increasing throughput and flexibility for facilities that treat waste solutions containing enriched uranium. This report details supplemental testing to compare uranium adsorption by MST and mMST in the absence of other sorbates.

This information provides a more quantitative comparison of the uranium affinity of each material.<sup>5</sup>

Testing to date indicates that laboratory-prepared mMST is stable when stored at laboratory conditions for up to 12 months. Tests will extend shelf-life data for laboratory-prepared and vendor-prepared mMST samples to more than two years and one year, respectively.<sup>5</sup>

### **3.0 Experimental**

#### **3.1 Uranium Adsorption Isotherm**

The uranium isotherm (Task 1) testing consisted of contacting a simulated waste solution containing  $10 \pm 2$  mg/L of depleted uranium with varying amounts of each sorbent. Unlike previous testing, the simulated waste solution contained only uranium to limit competition between uranium and other sorbates for sites on the sorbent. The target sorbent concentrations included 0.8, 1.0, 1.6, 2, 3, 4, 5, 6, 8 and 10 g/L. Tests at 3 g/L of sorbent occurred in triplicate to determine experimental variance.

Table 1 provides the target and measured chemical composition of the simulant. The analytical results confirmed that the chemical composition of the assembled simulant met the target composition at the 95% confidence interval established by the measured value and the analytical uncertainty of the measurement. Personnel collected samples of the simulant weekly over a three week period and analyzed for uranium. The analytical results confirm that the solution did not exhibit any time dependent changes in uranium concentration over the three week time period.

Testing consisted of placing the equilibrated simulant into plastic bottles followed by the desired amount of sorbent (0.8 - 10 g/L). Bottles were continuously shaken using an Inova Model 4230 Incubator (New Brunswick Scientific) at a constant temperature of  $26 \pm 1$  °C. Personnel collected samples from the test bottles after 4, 12 and 336 hours of contact. Sampling consisted of removing a test bottle from the shaker, manually shaking to produce a homogeneous suspension, and pulling approximately 3-mL of the test mixture into a disposable 10-mL syringe. The sample mixture was filtered through a 0.10- $\mu$ m syringe filter disk (0.1-polytetrafluoroethylene (PTFE) membrane) and the filtrate collected in a clean sample bottle. Personnel pipetted 2-mL portions of the filtered sample into a second set of sample bottles containing 2-mL of 5 M nitric acid. The acidified samples were shaken for approximately 15 seconds and then allowed to equilibrate with periodic shaking overnight. The acidified samples were analyzed for uranium content by the Analytical Development (AD) organization in SRNL using inductively coupled plasma mass spectroscopy (ICP-MS).

After 336 hours, the test bottles were allowed to stand undisturbed at ambient laboratory temperature. After standing for 16 or 20 days, the remaining suspensions in the replicate test bottles containing a sorbent concentration of  $3.0 \text{ g L}^{-1}$  were filtered individually using

a Millipore 2-piece filter apparatus containing a 0.1-micron PTFE membrane disk. Each filtrate was collected and analyzed for uranium content by ICP-MS.

The sorbent solids collected on the filter disk were briefly washed with six 1-mL portions of a rinse solution comprised of sodium nitrate (4.30 M) and sodium hydroxide (1.33 M). After completing the rinsing sequence, the filter disk containing the solids was placed in a clean crystallization dish and then placed in a desiccator at ambient temperature for 4 – 5 days. The dried solids were weighed to determine a final dried weight, dissolved in sulfuric acid and the dissolved solids solution analyzed for sodium and titanium by inductively coupled plasma atomic emission spectroscopy (ICP-AES) and uranium by ICP-MS.

**Table 1. Simulated Waste Solution Composition.**

Component	Target Concentration	Measured Concentration		Units
		Value	1 $\sigma$ Uncertainty	
NaNO <sub>3</sub>	2.60	3.11	0.31	M
NaOH	1.33	1.38	0.138	M
Na <sub>2</sub> SO <sub>4</sub>	0.521	0.606	0.0909	M
NaAl(OH) <sub>4</sub>	0.429	0.461	0.0461	M
NaNO <sub>2</sub>	0.134	0.160	0.032	M
Na <sub>2</sub> CO <sub>3</sub>	0.026	0.0314	0.00471	M
Total Na <sup>+</sup>	5.60	5.10	0.51	M
Uranium	10,000	9545	294	μg/L

### **3.2 Shelf-Life Testing**

Shelf-life testing (Task 2) evaluated strontium and actinide removal after storing laboratory-prepared and vendor-prepared mMST materials as aqueous suspensions at ambient laboratory temperature. This testing featured the same simulated waste solution, SWS-12-2005, as previously described in the final report of the Phase II testing (see Table 2). Prior to use in this testing, personnel added an aliquot of <sup>85</sup>Sr to increase the concentration of radiotracer for strontium removal measurements.

Testing consisted of placing 60-mL of the SWS-12-2005 in each of the test bottles and adding the desired amount of mMST (0.2 g L<sup>-1</sup>) and baseline MST (0.2 and 0.4 g L<sup>-1</sup>) in the appropriate bottles. Duplicate tests were performed with each of the following mMST samples: LS-1, LS-2, LS-3 and Optima Batch #06-QAB-0139. Samples LS-1, LS-2, LS-3 represent laboratory-prepared samples of mMST stored for more than 29 months under ambient laboratory conditions since their synthesis. Optima Batch #06-QAB-0139 represents the first vendor-prepared batch of modified MST, prepared by Optima Chemical Group LLC (Douglas, GA) and stored at ambient laboratory conditions for more than 16 months after receipt of the material at SRNL. The baseline MST

sample is from a commercial batch of material produced by Optima Chemical Group LLC and identified as Batch #00-QAB-417.

Prior to the addition of the sorbent, the simulated waste solution was equilibrated in each of the test bottles overnight in a temperature-controlled waterbath equipped with an orbital shaker operating at 175 rpm. The following day personnel removed the bottles from the waterbath, added the appropriate quantity of sorbent and returned the bottles to the waterbath. The average and single standard deviation of the recorded temperatures during the equilibration and testing measured  $26.19 \pm 0.14$  °C.

Sorption performance was determined by measuring solution concentrations after 6 and 12 hours of contact. Personnel used the same sampling protocol as described above. The  $^{85}\text{Sr}$  and  $^{237}\text{Np}$  removals were determined by gamma spectroscopy. Plutonium removal was determined by radiochemical separation of the plutonium followed by alpha counting of the extracted plutonium ( $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ). The  $^{237}\text{Np}$ ,  $^{239,240}\text{Pu}$  and uranium removals were determined by ICP-MS. Strontium and actinide removal performance in this test set is compared to that previously measured to determine any change in performance with longer storage times.

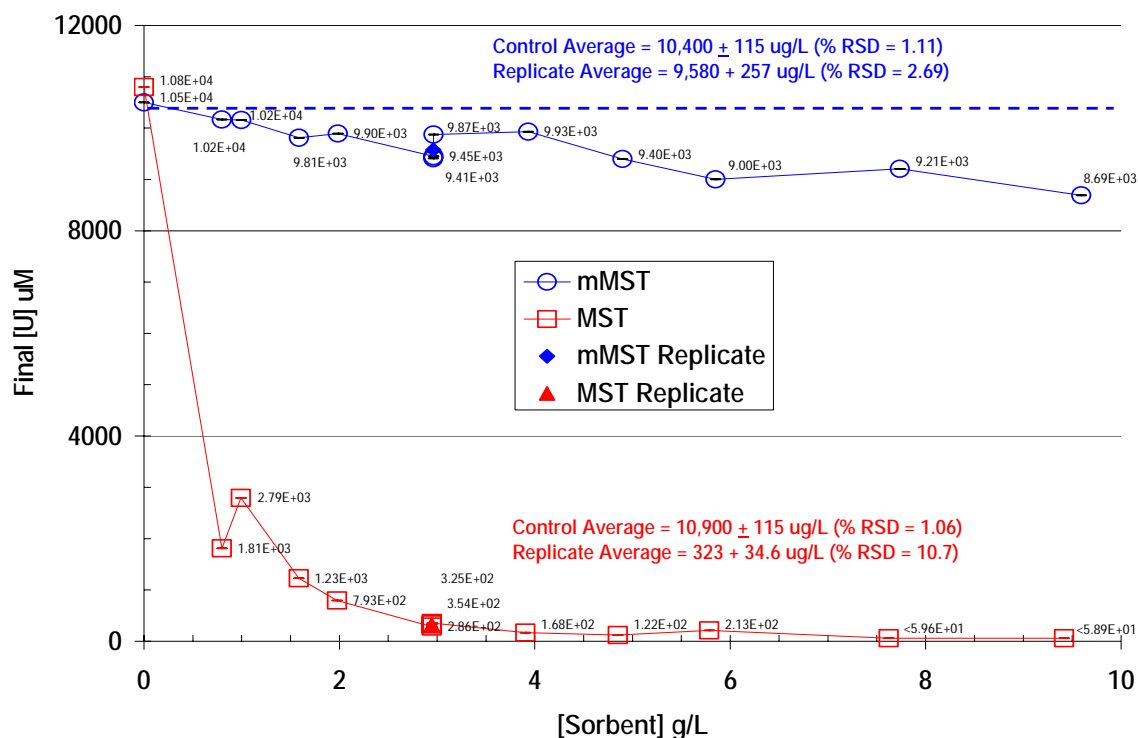
## 4.0 Results and Discussion

### 4.1 Uranium Adsorption

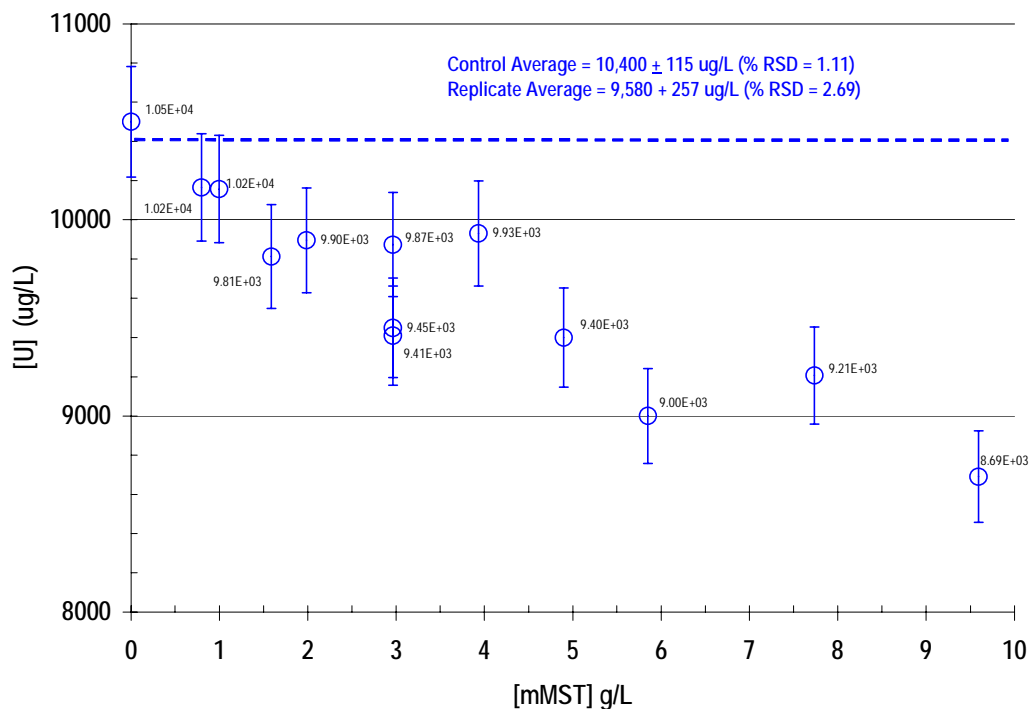
Figures 1 and 2 provide plots of the solution phase uranium concentration after 336 hours of contact versus sorbent concentration. For this test set, the target sorbent concentration ranged from 0.8 to 10.0 g L<sup>-1</sup>. Due to the dilution resulting from the liquid associated with the MST and mMST samples, the calculated sorbent concentrations ranged from 0.8 – 9.4 and 0.8 – 9.6 g L<sup>-1</sup> for tests with MST and mMST, respectively.

Upon addition of increasing amounts of MST, the uranium concentration rapidly decreases until below the quantifiable limit at sorbent concentrations at or above 7.6 g L<sup>-1</sup>. Thus, at the higher MST concentrations (7.6 and 9.4 g L<sup>-1</sup>) more than 99.5% of the uranium was removed by sorption onto the MST.

In contrast to MST, addition of mMST resulted in much lower change in the uranium concentration (see Figure 1). From Figure 2 an addition of at least 1.5 g L<sup>-1</sup> of mMST was required before a statistically significant amount of uranium was removed from solution given the analytical and experimental uncertainty in the uranium measurements. The uranium concentration decreased approximately linearly with the mMST concentration. At the highest mMST concentration (9.6 g L<sup>-1</sup>), only 17% of the uranium was removed from solution by the simulated waste solution. Clearly, the mMST exhibited much lower affinity for uranium than that exhibited by the baseline MST.



**Figure 1. Uranium Concentration ( $\mu\text{M}$ ) versus Sorbent Concentration ( $\text{g L}^{-1}$ )**

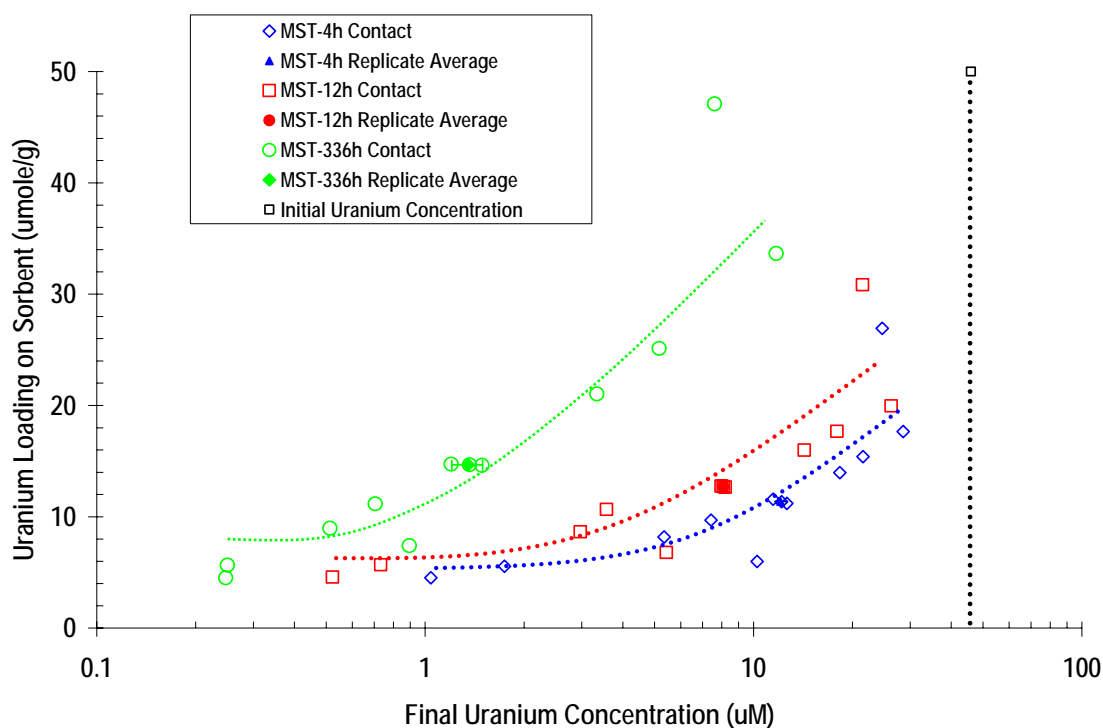


**Figure 2. Uranium Concentration ( $\mu\text{M}$ ) versus mMST Concentration ( $\text{g L}^{-1}$ ) – expanded y-axis**



The adsorption of metal ions from aqueous solutions by solid sorbates generally shows an S-shaped curve when the quantity of sorbate loaded onto the sorbent is plotted versus the equilibrium sorbate concentration. This curve, referred to as an adsorption isotherm, generally provides a useful metric for comparing the affinity of two different sorbents (e.g., MST and mMST) for a common sorbate (e.g., uranium in this study). Figure 3 provides such a plot of the uranium adsorption isotherm for MST measured at three different contact times, 4-h, 12-h and 336-h. From the graph it is evident that the adsorption of uranium increases with time. Due to the limited experimental measurements for MST near the initial uranium concentration, I did not observe the top portion of the S-shaped curve, which would provide a measure of the maximum loading of uranium onto the MST. This information could be obtained by performing additional tests at a higher phase ratio (i.e., lower MST concentration).

Replicate tests using a target sorbent concentration of  $3 \text{ g L}^{-1}$  exhibited very similar results for each sampling event. The measured uranium loading increased with contact time for tests with MST. The relative standard deviation among the replicate test measured less than 2% for the three sampling events (see Table 2).



**Figure 3. Uranium Adsorption Isotherms for MST**

**Table 2. Measured Average Loadings of Uranium onto MST and mMST at a Sorbent Concentration of 3 g L<sup>-1</sup>**

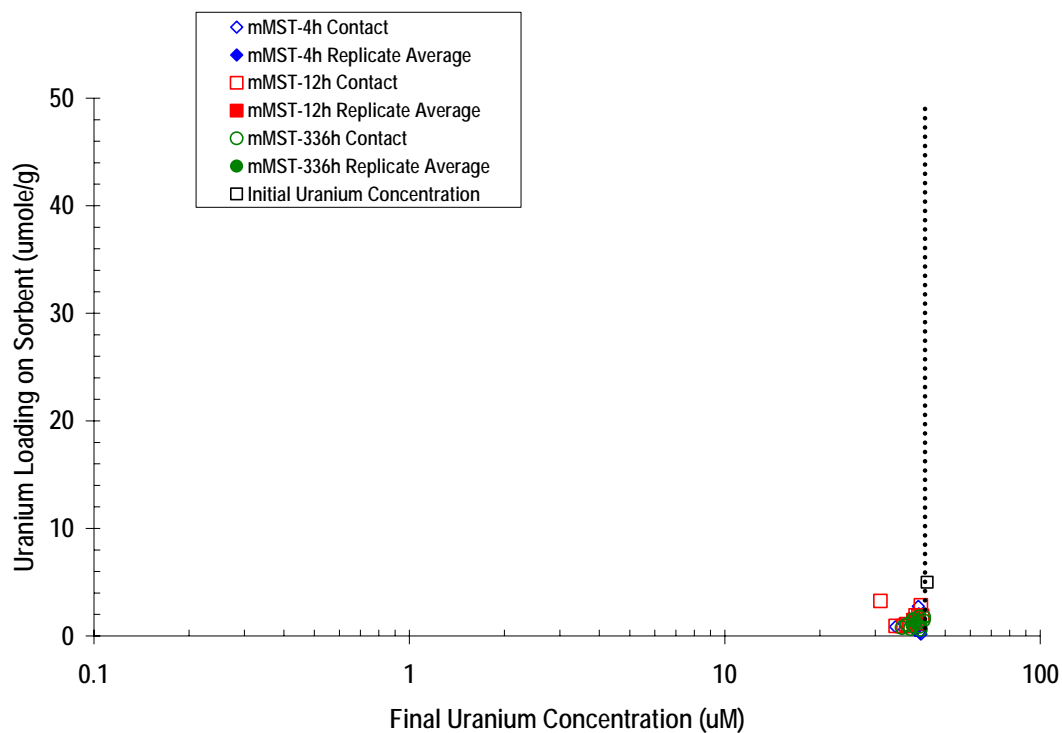
<u>Contact Time (h)</u>	<u>Uranium Loading on Sorbent (μmole/g)*</u>	
	<u>MST</u>	<u>mMST</u>
4	11.4 (0.19)	1.15 (0.215)
12	12.7 (0.042)	1.37 (0.16)
336	14.7 (0.049)	1.29 (0.36)

\* numbers in parenthesis are single standard deviation of triplicate measurements

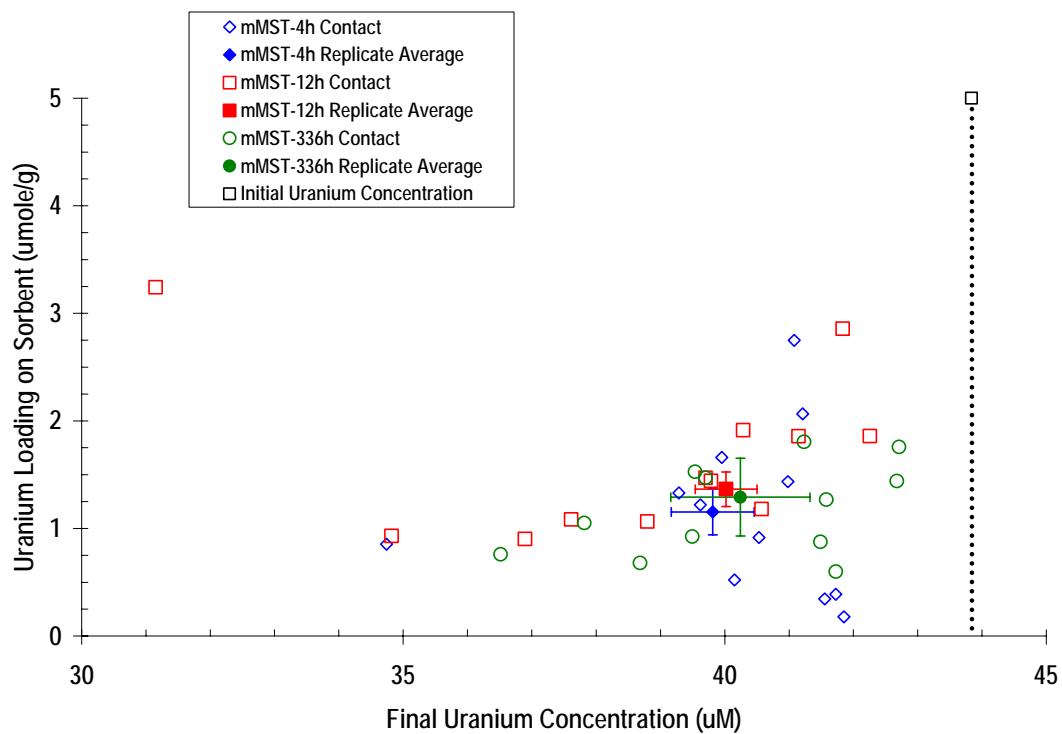
Figures 4 and 5 provide similar plots as Figure 3 as measured for the mMST material. Figure 4 provides a plot using the same scale as that shown in Figure 3. Note that all of the data points fall in the bottom right hand corner indicating much lower adsorption of uranium onto the mMST material compared to that measured with MST. Figure 5 provides an expanded scale allowing a better comparison of the loading of uranium onto mMST across the dataset.

One of the sample results (12-h result at a target sorbent concentration of 4.0 g L<sup>-1</sup>) appears inconsistent with the remainder of the dataset. The solution concentration of uranium measured 7540 μg L<sup>-1</sup>, which is considerably lower than that measured at the 4-h and 336-h sampling events (9930 μg L<sup>-1</sup>). I suspect that a pipetting error occurred resulting in the delivery of a smaller aliquot of the sample upon dilution in nitric acid for the determination of uranium by ICP-MS.

Inspection of Figures 3 – 5 clearly reveals that the affinity of mMST for uranium is much lower than that exhibited by MST. This is also apparent by comparing the measured uranium loadings for the MST and mMST materials at a sorbent concentration of 3 g L<sup>-1</sup> (see Table 2). The average uranium loadings for the mMST sample measured about an order of magnitude lower than that of the MST sample. Note also that, unlike MST, which exhibited increasing uranium loading with contact time, uranium loadings over the range of sorbent concentrations did not increase in the tests with mMST between 4 and 336 hours of contact.



**Figure 4. Uranium Adsorption Isotherms for mMST**



**Figure 5. Uranium Adsorption Isotherms for mMST (expanded scale)**

After receiving the ICP-MS results (384 hours later for the MST samples and 480 hours later for the mMST samples), personnel separated the remaining solution and sorbent solids in each of the  $3.0 \text{ g L}^{-1}$  tests by filtration. The uranium concentration was determined in the final filtrate and the uranium and titanium content in the sorbent solids. Table 3 provides the measured uranium concentrations in the final filtrate as well as those measured after 336 hours of contact. In all of the tests, the uranium concentration in the final filtrate measured below that determined earlier after 336 hours of contact. This finding indicates that adsorption of uranium continued during the time period in which the test bottles were allowed to stand unmixed at ambient laboratory temperature.

**Table 3. Uranium Concentrations in the Final Filtrate and in Solution After 336-hours of Contact\***

Sample ID	Sorbent	[U] in Final Filtrate ( $\mu\text{M}$ )	[U] after 336-h of Contact ( $\mu\text{M}$ )	% of Remaining Uranium Adsorbed
MMST-205	MST	<1.68E-01	1.20E+00	>86.0%
MMST-206	MST	9.79E-01	1.37E+00	28.3%
MMST-207	MST	9.92E-01	1.49E+00	33.4%
MMST-192	mMST	3.80E+01	3.97E+01	4.23%
MMST-193	mMST	3.73E+01	3.95E+01	5.63%
MMST-194	mMST	3.76E+01	4.15E+01	9.36%

\* Total contact time for the final filtrate samples is 720 hours for the tests with MST and 816 hours for the tests with mMST.

The additional amount of uranium adsorbed ranged from 28.3% to more than 86.0% for MST and 4.23% to 9.36% for mMST based on the measured concentration of uranium in solution. The relative high percentage of additional uranium adsorption for the tests with MST reflects little uranium ( $1.2 - 1.5 \mu\text{M}$ ) remaining in solution for adsorption to occur after 336 hours of contact with the MST. The mMST material continues to show low affinity for uranium as evidenced by only between 4% and 10% of the remaining uranium ( $39.5 - 41.5 \mu\text{M}$ ) adsorbing onto the mMST solids after an additional 480 hours of unagitated contact time.

Table 4 provides the mass balance for the six tests. For the tests using MST as the sorbent, most of the uranium was found on the recovered solids. On average, the mass balance accounted for about 85% of the uranium present in the MST tests, not including filtrate discarded during sample collection events. For the tests using mMST as the sorbent, most of the uranium remained in solution. On average the mass balance accounted for about 90% of the uranium present in these tests, not including filtrate and discarded during sample collection. The 10 – 15% of unaccounted for uranium is likely due to the 3 sampling events which resulted in the removal of about 20% of the initial test suspension volume and corresponding sorbent solids.

**Table 4 Uranium Mass Balance in Adsorption Tests**

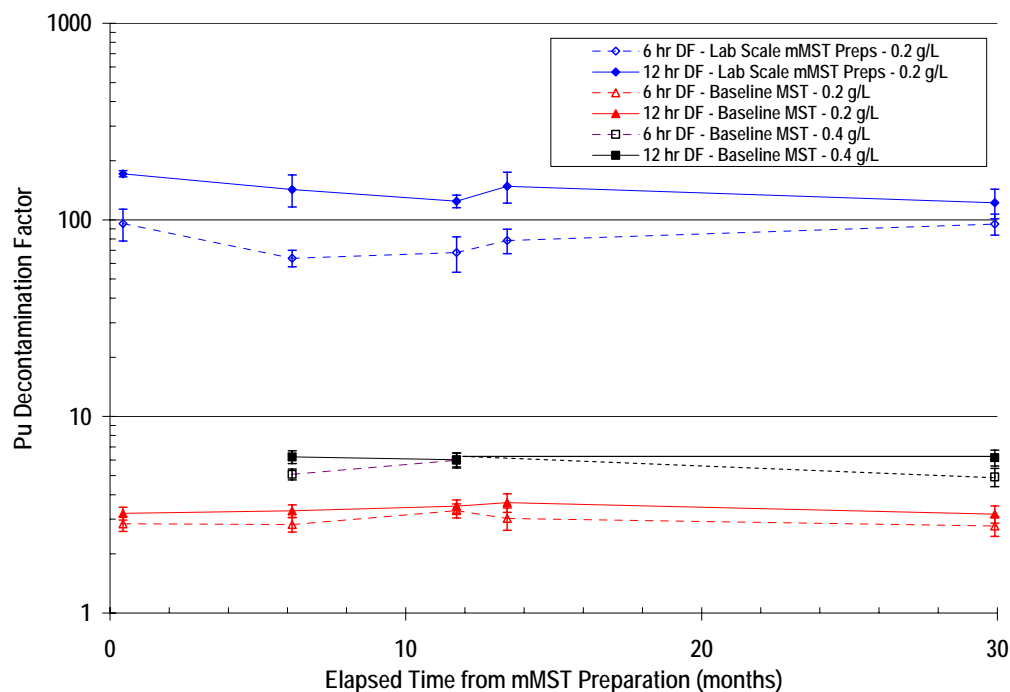
Test ID	Sorbent	Total U in Expt (μg)	Measured Uranium in Solids (μg)	Measured Uranium Remaining in Solution (μg)	Total U Accounted For (μg)	% Uranium Accounted For
MMST-205	MST	6.56E+02	5.36E+02	2.40E+00	5.39E+02	82.1%
MMST-206	MST	6.56E+02	5.45E+02	1.40E+01	5.59E+02	85.2%
MMST-207	MST	6.56E+02	5.38E+02	1.42E+01	5.52E+02	84.2%
MMST-192	mMST	6.26E+02	2.96E+01	5.43E+02	5.73E+02	91.5%
MMST-193	mMST	6.26E+02	2.74E+01	5.33E+02	5.60E+02	89.5%
MMST-194	mMST	6.26E+02	2.20E+01	5.37E+02	5.59E+02	89.3%

#### **4.2 Shelf-life Testing**

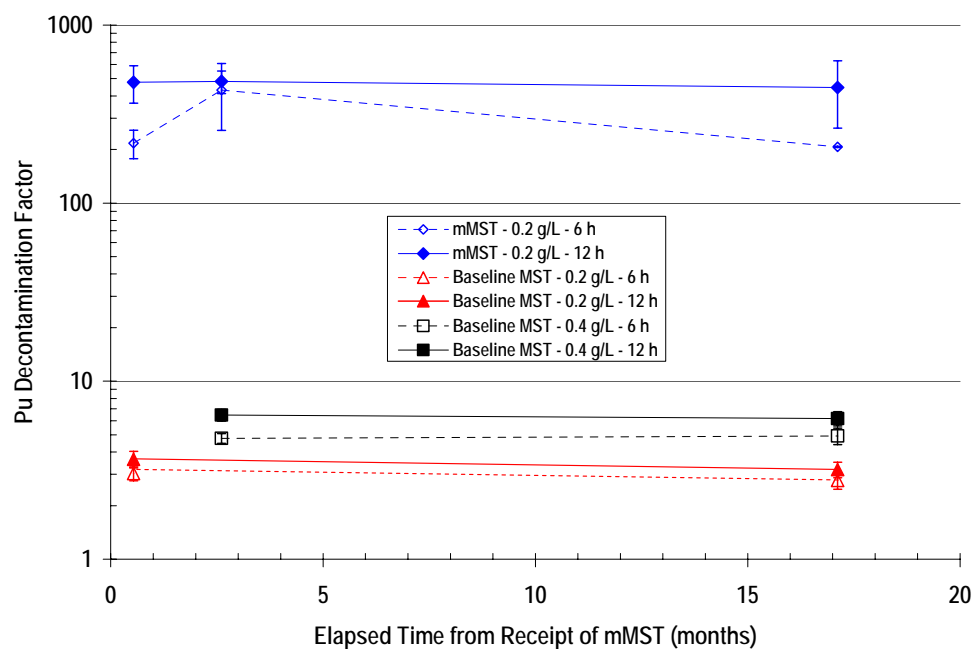
Testing evaluated the performance of three laboratory-prepared (LS-1, LS-2, LS-3) and one vendor-prepared (Optima #06-QAB-0139) mMST samples. Storage of the laboratory-prepared mMST samples reached 30 months since their preparation compared to 17-months since the receipt of the vendor-prepared batch of mMST. In all cases, the mMST and MST samples were stored as aqueous suspension at ambient laboratory temperature.

Appendices 7.1 and 7.2 provide summary tables of the 6-hour and 12-hour decontamination factors (DF) for strontium, plutonium and neptunium determined for the laboratory-prepared and vendor-prepared mMST samples at a sorbent concentration of 0.2 g L<sup>-1</sup> and the baseline MST sample at sorbent concentrations of 0.2 and 0.4 g L<sup>-1</sup>. The decontamination factors for the laboratory-prepared mMST samples represent the average of duplicate tests for each of the three mMST samples: LS-1, LS-2, and LS-3 (total of 6 tests). The reported uncertainty for the MST samples represents the analytical uncertainty in the measurement of the sorbate.

Figures 6 and 7 provide plots of the plutonium decontamination factors measured after 6 and 12-hours of contact with 0.2 g L<sup>-1</sup> of the laboratory-prepared and vendor-prepared mMST samples. Results with 0.2 and 0.4 g L<sup>-1</sup> of MST are also included in the graphs. Both graphs indicate that the mMST materials showed no loss in performance over the storage period with respect to the removal of plutonium from the simulated waste solution for both the laboratory-prepared mMST samples and the vendor-prepared mMST sample. Over this storage period the plutonium removal performance of the baseline MST sample also remained constant.



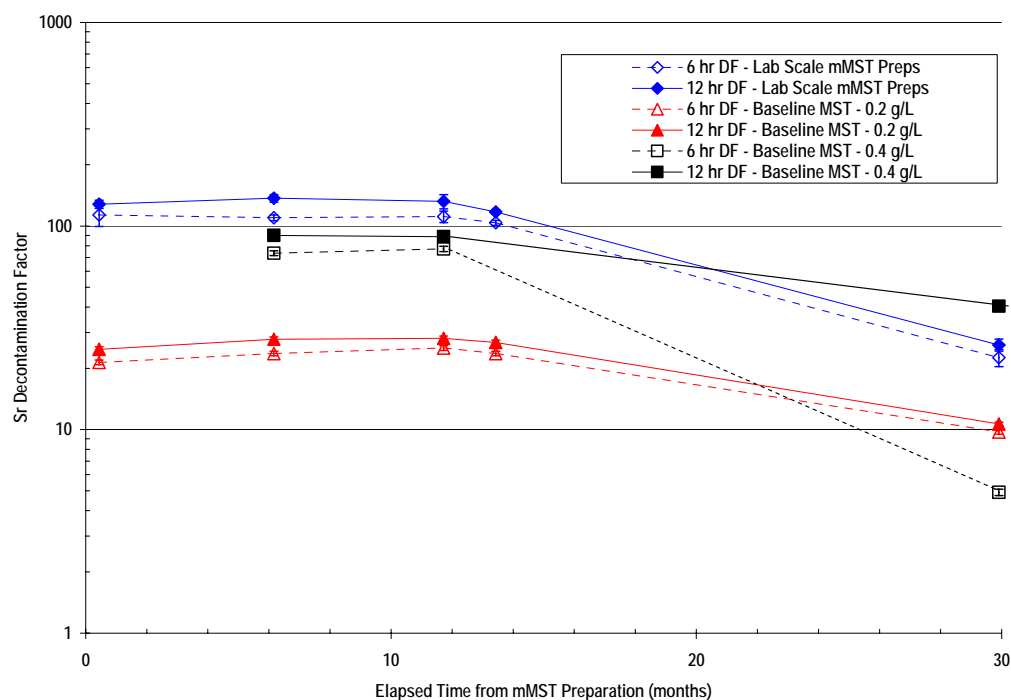
**Figure 6. Plutonium Decontamination Factors versus Elapsed Time Since Laboratory Preparations of mMST**



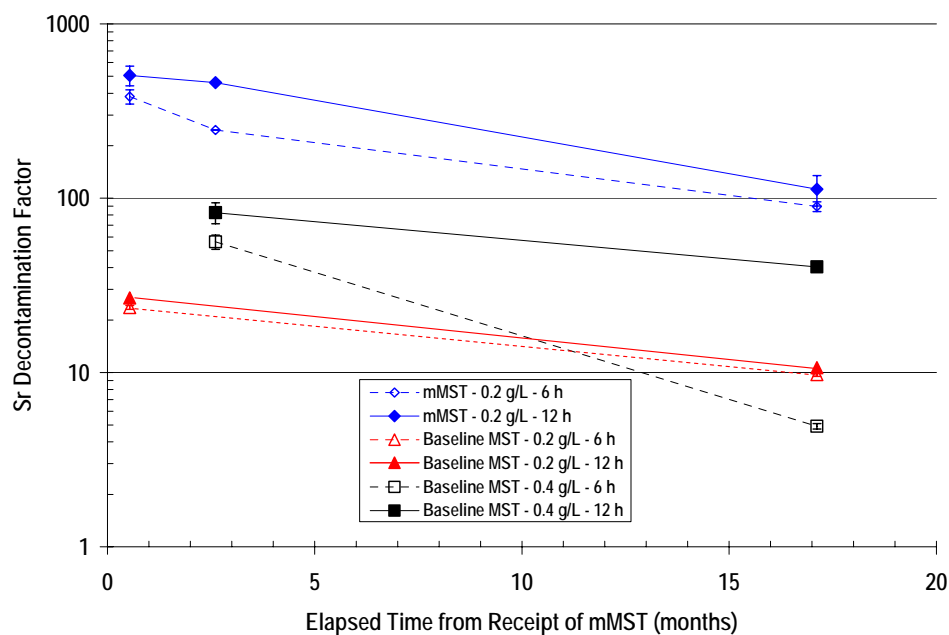
**Figure 7. Plutonium Decontamination Factors versus Elapsed Time Since Receipt of Vendor-Prepared mMST**

Figures 8 and 9 provide plots of the strontium decontamination factors in a similar manner as previously discussed for plutonium. For the laboratory-prepared mMST samples, the Sr DF values remain consistent until the testing event after 30-months of storage. The baseline MST sample also exhibited a decrease in strontium removal after this storage time (see Figure 8). The vendor-prepared mMST sample exhibited a similar decrease in strontium removal in this test set as well (see Figure 9). Comparing strontium DF values, the mMST samples exhibited a larger decrease at the most recent testing event than the baseline MST sample. However, if one compares the percent of  $^{85}\text{Sr}$  removed (see Table 5), the mMST samples exhibit a smaller decrease in mass of strontium removed than observed for the baseline MST sample. For example, the percentage  $^{85}\text{Sr}$  removed by the vendor-prepared mMST sample decreased from 99.8 to 99.1 compared to a decrease from 96.3 to 90.6 for the baseline MST after a 12-hour contact with  $0.2 \text{ g L}^{-1}$  of sorbent. Thus, the mMST samples exhibited the greater total removal of strontium mass compared to that of MST.

Initially, I attributed the observed decrease in strontium removal performance for both the mMST and MST samples to the addition of  $^{85}\text{Sr}$  radiotracer prior to the last testing event. The  $^{85}\text{Sr}$  has a short half-life of 64.84 days and, consequently, the  $^{85}\text{Sr}$  activity in the simulated waste solution had decreased significantly over the 16.5-month time period between performance testing events. Thus, I added a quantity of additional  $^{85}\text{Sr}$  to the simulant. The  $^{85}\text{Sr}$  is not carrier-free and, therefore, the addition of the radiotracer adds both  $^{85}\text{Sr}$  and stable strontium isotopes. I analyzed a sample of the simulant after the testing to determine if the stable strontium concentration may have increased significantly due to the  $^{85}\text{Sr}$  addition. Analysis indicated that the total strontium concentration measured  $598 \pm 38.9 \text{ } \mu\text{g L}^{-1}$  compared to an average of  $569 \pm 105 \text{ } \mu\text{g L}^{-1}$  (RSD = 18.4%) during the preparation of the simulant. Given this finding, I conclude that the  $^{85}\text{Sr}$  addition did not change the total strontium concentration sufficiently to result in the observed decrease in strontium removal. Currently, I do not have an explanation for the cause of the reduced strontium removal performance in the latest testing event. However, given that the decrease in strontium removal performance is smaller for the mMST samples compared to that of the baseline MST, I conclude that the mMST exhibits very good shelf-life when stored as aqueous suspension at ambient laboratory temperature.



**Figure 8. Strontium Decontamination Factors versus Elapsed Time Since Laboratory Preparations of mMST**



**Figure 9. Strontium Decontamination Factors versus Elapsed Time Since Receipt of Vendor-Prepared mMST**

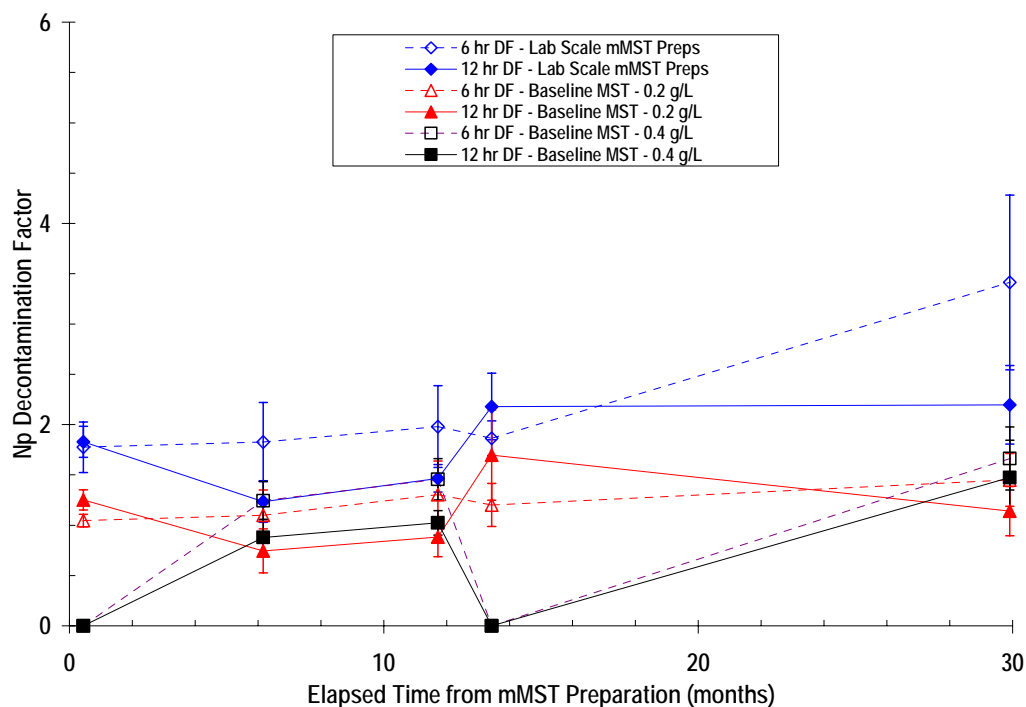


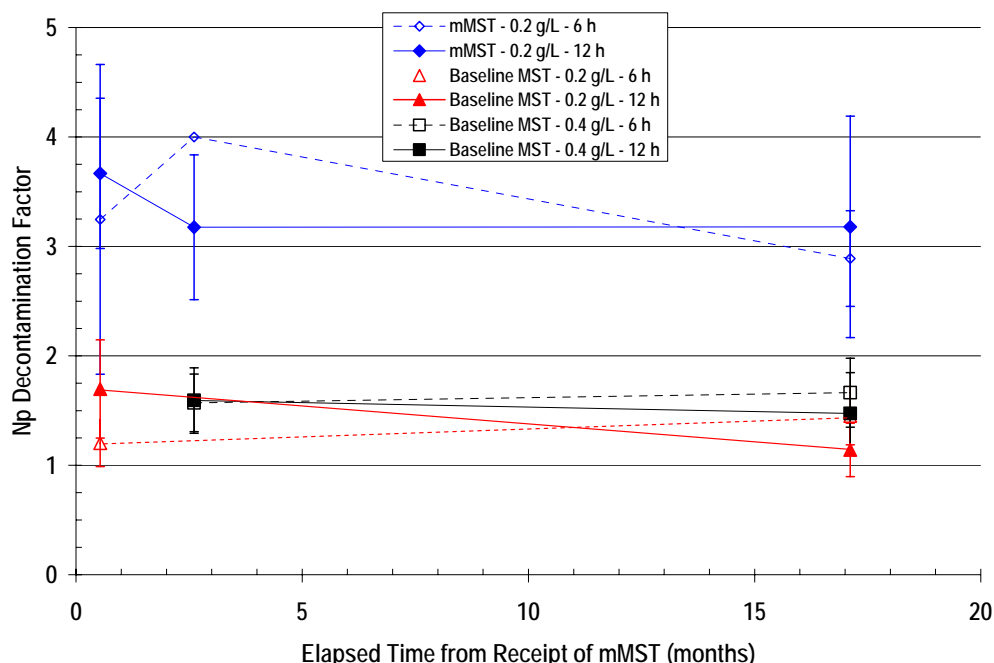
**Table 5 Percent Strontium Removed upon Contact of Simulated Waste Solution with mMST and MST Samples**

Elapsed Time from Prep to Performance Test (months)	% Strontium Removed											
	Laboratory-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hours		12-hours		6-hours		12-hours		6-hours		12-hours	
	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
0.44	9.91E+01	1.22E+01	9.92E+01	4.50E+00	9.53E+01	2.51E+00	9.60E+01	2.75E+00	nd	-	nd	-
6.2	9.91E+01	3.16E+00	9.93E+01	4.16E+00	9.58E+01	2.42E+00	9.64E+01	2.59E+00	9.86E+01	2.71E+00	9.89E+01	3.78E+00
11.7	9.91E+01	6.38E+00	9.92E+01	7.84E+00	9.60E+01	2.51E+00	9.64E+01	2.56E+00	9.87E+01	2.95E+00	9.89E+01	3.20E+00
13.4	9.90E+01	2.77E+00	9.91E+01	2.71E+00	9.58E+01	2.40E+00	9.63E+01	2.36E+00	nd	-	nd	-
29.9	9.56E+01	9.12E+00	9.62E+01	6.58E+00	8.97E+01	2.25E+00	9.06E+01	2.06E+00	7.97E+01	2.87E+00	9.75E+01	2.50E+00

Elapsed Time from Prep to Performance Test (months)	Vendor-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hours		12-hours		6-hours		12-hours		6-hours		12-hours	
	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
0.54	9.97E+01	9.34E+00	9.98E+01	1.30E+01	9.58E+01	2.40E+00	9.63E+01	2.36E+00	nd	-	nd	-
2.6	>9.96E+01	-	>9.98E+01	-	nd	-	nd	-	9.82E+01	9.05E+00	9.88E+01	1.35E+01
17.1	9.89E+01	6.45E+00	9.91E+01	1.95E+01	8.97E+01	2.25E+00	9.06E+01	2.06E+00	7.97E+01	2.87E+00	9.75E+01	2.50E+00

Figures 10 and 11 provide plots of the neptunium decontamination factors in a similar manner as that described previously for plutonium. There is considerable scatter in the data, particularly for the baseline MST sample, across the datasets. However, the DF values for both the laboratory and vendor-prepared mMST batches of material are reasonably similar over the entire dataset. Given the similar neptunium DF values and lower variance compared to the baseline MST sample, I conclude that there is no statistically significant loss in sorption performance for neptunium by either the laboratory-prepared or the vendor-prepared mMST samples.

**Figure 10. Neptunium Decontamination Factors versus Elapsed Time Since Laboratory Preparations of mMST**



**Figure 11. Neptunium Decontamination Factors versus Elapsed Time Since Receipt of Vendor-Prepared mMST**

## 5.0 Summary and Recommendation

Testing results confirmed earlier findings that the mMST exhibits much lower affinity for uranium than the baseline monosodium titanate (MST) material. The loading of uranium onto the mMST sample measured more than an order of magnitude lower than observed for MST. This finding indicates that the use of mMST provides a significant advantage over MST in that the mMST will not concentrate enriched uranium to the degree that MST does. The reduced affinity of mMST for uranium allows more operational flexibility in treating waste solutions from a nuclear criticality safety perspective.

Testing results also indicate that the mMST exhibits good shelf-life with no measurable loss in plutonium and neptunium removal upon storage of samples at ambient laboratory temperatures for up to 30-months. Testing did exhibit a change in strontium removal performance for both the mMST and MST samples at the most recent testing event. However, the decrease in strontium removal performance proved lower for the mMST than the MST sample. Given these positive findings SRNL recommends continued development of mMST as a replacement for MST in pretreatment facilities at the Savannah River Site (SRS).

## **6.0 Reviews and Approvals**

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### **Design Review**

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### **Customer Concurrence**

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R. L. Salizzoni, HTF Chief Engineering	Date
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## 7.0 Appendices

### 7.1 Summary of Decontamination Factors for Laboratory-Prepared mMST Samples (LS-1, LS-2 and LS-3) and Baseline MST Sample (Optima #00-QAB-417) at Various Storage Times

Elapsed Time from Prep to Performance Test	Strontium DF											
	Laboratory-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time	
	months	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value
0.44	1.13E+02	1.40E+01	1.28E+02	5.80E+00	2.14E+01	5.63E-01	2.48E+01	7.12E-01	nd	-	nd	-
6.2	1.10E+02	3.50E+00	1.37E+02	5.74E+00	2.36E+01	5.97E-01	2.78E+01	7.47E-01	7.35E+01	2.02E+00	9.00E+01	3.44E+00
11.7	1.11E+02	7.16E+00	1.32E+02	1.04E+01	2.52E+01	6.59E-01	2.81E+01	7.45E-01	7.72E+01	2.31E+00	8.87E+01	2.87E+00
13.4	1.04E+02	2.90E+00	1.17E+02	3.20E+00	2.36E+01	5.93E-01	2.68E+01	6.59E-01	nd	-	nd	-
29.9	2.26E+01	2.15E+00	2.61E+01	1.78E+00	9.74E+00	2.45E-01	1.07E+01	2.43E-01	4.92E+00	1.77E-01	4.04E+01	1.03E+00

Elapsed Time from Prep to Performance Test	Plutonium DF											
	Laboratory-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time	
	months	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value
0.44	9.57E+01	1.76E+01	1.72E+02	6.16E+00	2.85E+00	1.97E-01	3.21E+00	2.41E-01	nd	-	nd	-
6.2	6.38E+01	6.14E+00	1.43E+02	2.66E+01	2.82E+00	1.82E-01	3.31E+00	2.43E-01	5.08E+00	3.23E-01	6.22E+00	4.66E-01
11.7	6.82E+01	1.39E+01	1.24E+02	9.25E+00	3.32E+00	3.11E-01	3.49E+00	2.72E-01	5.99E+00	5.28E-01	6.02E+00	4.90E-01
13.4	7.85E+01	1.12E+01	1.48E+02	2.66E+01	3.03E+00	2.71E-01	3.65E+00	3.93E-01	nd	-	nd	-
29.9	9.52E+01	1.17E+01	1.22E+02	2.09E+01	2.77E+00	2.96E-01	3.19E+00	3.16E-01	4.92E+00	5.24E-01	6.17E+00	5.71E-01

Elapsed Time from Prep to Performance Test	Neptunium DF											
	Laboratory-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time	
	months	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value
0.44	1.77E+00	2.52E-01	1.83E+00	1.55E-01	1.05E+00	6.27E-02	1.25E+00	9.99E-02	nd	-	nd	-
6.2	1.83E+00	3.93E-01	1.24E+00	2.07E-01	1.10E+00	2.52E-01	7.44E-01	2.19E-01	1.24E+00	1.90E-01	8.79E-01	1.80E-01
11.7	1.98E+00	4.07E-01	1.47E+00	1.37E-01	1.30E+00	3.33E-01	8.82E-01	1.95E-01	1.46E+00	2.06E-01	1.02E+00	1.21E-01
13.4	1.87E+00	1.72E-01	2.18E+00	3.30E-01	1.20E+00	2.14E-01	1.70E+00	4.49E-01	nd	-	nd	-
29.9	3.41E+00	8.68E-01	2.20E+00	3.91E-01	1.45E+00	2.64E-01	1.14E+00	2.45E-01	1.66E+00	3.14E-01	1.47E+00	3.73E-01

## 7.2 Summary of Decontamination Factors for Vendor-Prepared mMST Sample (Optima #06-QAB-0139) and Baseline MST Sample (Optima #00-QAB-417) at Various Storage Times

Elapsed Time from Prep to Performance Test	Strontium DF											
	Vendor-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time	
	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
months												
0.54	3.82E+02	3.58E+01	5.07E+02	6.58E+01	2.36E+01	5.93E-01	2.68E+01	6.59E-01				
2.6	>2.46E+02	-	>4.61E+02	-					5.62E+01	5.17E+00	8.27E+01	1.13E+01
17.1	8.96E+01	5.85E+00	1.12E+02	2.21E+01	9.74E+00	2.45E-01	1.07E+01	2.43E-01	4.92E+00	1.77E-01	4.04E+01	1.03E+00

Elapsed Time from Prep to Performance Test	Plutonium DF											
	Vendor-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time	
	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
months												
0.54	2.17E+02	3.97E+01	4.77E+02	1.13E+02	3.03E+00	2.71E-01	3.65E+00	3.93E-01				
2.6	4.32E+02	1.76E+02	4.83E+02	6.99E+01					4.78E+00	3.02E-01	6.45E+00	4.80E-01
17.1	2.07E+02	1.77E+00	4.47E+02	1.83E+02	2.77E+00	2.96E-01	3.19E+00	3.16E-01	4.92E+00	5.24E-01	6.17E+00	5.71E-01

Elapsed Time from Prep to Performance Test	Neptunium DF											
	Vendor-Prepared mMST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time		6-hour Contact Time		12-hour Contact Time	
	Average	Std Dev	Average	Std Dev	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
months												
0.54	3.25E+00	1.42E+00	3.67E+00	6.86E-01	1.20E+00	2.14E-01	1.70E+00	4.49E-01				
2.6	>4.00E+00		3.18E+00	6.61E-01					1.57E+00	2.63E-01	1.59E+00	2.99E-01
17.1	2.89E+00	4.37E-01	3.18E+00	1.01E+00	1.45E+00	2.64E-01	1.14E+00	2.45E-01	1.66E+00	3.14E-01	1.47E+00	3.73E-01

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