

**Tailoring Inorganic Sorbents for SRS Strontium and Actinide
Separations: Optimized Monosodium Titanate
Phase II Interim Report for External Release**

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Acronyms

ARP	Actinide Removal Process
CST	Chemical Science and Technology
DF	decontamination factor
DWPF	Defense Waste Processing Facility
HLW	high-level waste
ITP	In-Tank Precipitation
K_d	batch distribution constant
MST	monosodium titanate
SNL	Sandia National Laboratories
SRS	Savannah River Site
SRNL	Savannah River National Laboratory
SWPF	Salt Waste Processing Facility
TIP	titanium(IV) isopropoxide or tetraisopropoxytitanium(IV)

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1.0 Executive Summary

This document provides an interim summary report of Phase II testing activities for the development of a modified monosodium titanate (MST) that exhibits improved strontium and actinide removal characteristics compared to the baseline MST materials. The activities included determining the key synthesis conditions for preparation of the modified MST, preparation of the modified MST at a larger laboratory scale, demonstration of the strontium and actinide removal characteristics with actual tank waste supernate and characterization of the modified MST. Key findings and conclusions include the following.

- Samples of the modified MST prepared by Method 2 and Method 3 exhibited the best combination of strontium and actinide removal.
- We selected Method 3 to scale up and test performance with actual waste solution.
- We successfully prepared three batches of the modified MST using the Method 3 procedure at a 25-gram scale.
- Performance tests indicated successful scale-up to the 25-gram scale with excellent performance and reproducibility among each of the three batches. For example, the plutonium decontamination factors (6-hour contact time) for the modified MST samples averaged 13 times higher than that of the baseline MST sample at half the sorbent concentration (0.2 g L^{-1} for modified MST versus 0.4 g L^{-1} for baseline MST).
- Performance tests with actual waste supernate demonstrated that the modified MST exhibited better strontium and plutonium removal performance than that of the baseline MST. For example, the decontamination factors for the modified MST measured 2.6 times higher for strontium and between 5.2 to 11 times higher for plutonium compared to the baseline MST sample. The modified MST did not exhibit improved neptunium removal performance over that of the baseline MST.
- Two strikes of the modified MST provided increased removal of strontium and actinides from actual waste compared to a single strike. The improved performance exhibited by the modified MST indicates that fewer strikes of the modified MST would be needed to successfully treat waste that contain very high activities of ^{90}Sr and alpha-emitting radionuclides compared to the baseline MST.
- Reuse tests with actual waste confirmed that partially loaded MST exhibits reduced removal of strontium and actinides when contacted with fresh waste.
- Samples of modified MST prepared by Method 3 and the baseline MST exhibited very similar particle size distributions.
- Dead-end filtration tests showed that the modified MST samples exhibited similar filtration characteristics as the baseline MST sample.
- Performance testing indicated no change in strontium and neptunium removal after storing the modified MST for 6-months at ambient temperature. The results suggested that plutonium removal performance may be decreased slightly after 6-months of storage. However, the change in plutonium removal is not statistically significant at the 95% confidence limit.

Based on these findings we recommend continued development of the modified MST as a replacement for the baseline MST for waste treatment facilities at the Savannah River Site.

2.0 Introduction

MST is an inorganic sorbent material that exhibits high selectivity for strontium and actinide elements in the presence of strongly alkaline and high sodium containing salt solutions.^{1,2} The Savannah River Site (SRS) selected this material for strontium and plutonium removal from high-level waste solutions in the early 1980s as part of the In-Tank Precipitation (ITP) process.³ In 2001, the Department of Energy (DOE) selected MST for the strontium/actinide separation step within the Salt Waste Processing Facility (SWPF).⁴ Subsequently, Salt Processing Program Engineering selected MST for use in the Actinide Removal Process (ARP) to treat waste solutions low in cesium activity in a treatment facility located in 512-S.⁵

Original development of MST at Sandia National Laboratory (SNL) produced a dried powder. Unpublished studies conducted by L. L. Kilpatrick and D. T. Hobbs during the 1980s indicated that air drying of the MST at elevated temperature (≥ 100 °C) adversely impacted strontium removal performance. Principally due to the poorer sorption characteristics of MST dried at elevated temperature, procurement of MST at SRS for the ITP process specified that the vendor prepare and isolate the material without drying and deliver the MST as an aqueous solution containing 10 – 20 wt% MST solids.⁶

The proposed SWPF and existing ARP facilities have significantly different reactor configurations and process cycle times than that in the abandoned ITP operation. In particular, contact times between the MST and the alkaline waste solutions in the SWPF and ARP will be less than 12 hours versus approximately 2 weeks in the ITP process. Increased waste characterization data indicates that alpha removal characteristics (and principally plutonium removal) represent a greater challenge than that for ⁹⁰Sr removal. Based on recent testing at SRNL, the performance of MST to efficiently and rapidly remove alpha-emitting radionuclides serves as the limiting factor in sizing the equipment and operational throughput. Even higher alpha activities are projected for the SWPF and ARP operations as a result of initiatives to accelerate the disposal of HLW at the SRS. Due to the limited solubility of titanium in HLW borosilicate glass, there are limits on the amount of MST that can be used in SWPF and ARP facilities.⁷ Consequently, the need exists for an improved Sr/alpha removal material that exhibits increased actinide capacity and removal kinetics.

In 2003 the DOE Office of Cleanup Technologies (EM-21) funded a project to develop improved sorbent materials for strontium/actinide separations at SRS.⁸ This work identified a methodology for modifying the synthesis of MST that produced materials that exhibited significantly improved performance for strontium and actinide removal. Improved performance included both increased capacity and sorption kinetics. Preliminary evaluation of the use of the modified MST materials for use in the treatment of SRS high-level waste (HLW) in the ARP facility suggested that the throughput could be increased by as much as a factor of three. With these promising results, the DOE

Office of Cleanup Technologies funded SRNL to continue development of the modified MST materials under a Phase II project. This interim report provides a summary of work completed to date on Tasks 1 – 4 as specified in the Task Technical and Quality Assurance Plan for Phase II testing.^{9,10}

3.0 Experimental

3.1. Preparation of Modified MST Samples

We prepared modified MST samples by one of three methods identified as Method 1, Method 2 and Method 3. These methods are variations on the synthesis of MST by the published sol-gel technique.^{1,2,11} Specific details about the syntheses are not reported in this document as this technology is currently in patent prosecution.

3.2. Performance Testing with Simulated Waste Solution

Testing of combined strontium and actinide removal performance occurred at the Savannah River National Laboratory (SRNL) using the simulated waste solution composition as shown in Table 1 including plutonium, uranium, and neptunium in addition to ⁸⁵Sr. Strontium and actinide removal testing occurred at 25 ± 2 °C at a MST solids concentration of 0.4 g/L. Sampling of the test bottles occurred at 4, 24 and 168 hours of contact. We filtered the samples through 0.45-micron syringe filters (nylon membrane) to remove MST solids. Gamma spectroscopy measured the ⁸⁵Sr and neptunium content while alpha spectroscopy measured the total alpha activity. We measured the ^{238,239,240}Pu content by radiochemical separation of the plutonium followed by alpha counting of the extracted plutonium.

Table 1. Composition of Simulated Waste Solution

<u>Component</u>	<u>Target Concentration</u>	<u>Measured Concentration</u>	<u>Analytical Uncertainty</u>
NaNO ₃	2.60 M	2.44 M	0.24 M
NaOH	1.33 M	1.36 M	0.14 M
Na ₂ SO ₄	0.521 M	0.551 M	0.055 M
NaAl(OH) ₄	0.429 M	0.503 M	0.050 M
NaNO ₂	0.134 M	0.116 M	0.012 M
Na ₂ CO ₃	0.0260 M	0.016 M	0.010 M
Total Na	5.6 M	5.2 M	0.52 M
Total Sr	0.6 mg L ⁻¹	0.484 mg L ⁻¹	0.032 mg L ⁻¹
⁸⁵ Sr *	≥1000 dpm mL ⁻¹	1.65E+05 dpm mL ⁻¹	3.22E+03 dpm mL ⁻¹
Total Pu	0.2 mg L ⁻¹	0.218 mg L ⁻¹	0.013mg L ⁻¹
²³⁷ Np	0.5 mg L ⁻¹	0.461 mg L ⁻¹	0.090 mg L ⁻¹
Total U	10 mg L ⁻¹	9.55 mg L ⁻¹	0.33 mg L ⁻¹

* Value at time solution was first prepared. ⁸⁵Sr has a 64.8 day half-life and, therefore, the ⁸⁵Sr activity in the solution is continuously decreasing.

3.3. Performance Testing with Actual Waste

Closure Business Unit (CBU) supplied SRNL with a three liter sample of supernate taken in May 2005 (Sample ID: HTF-049). Upon receipt, the sample was vented and allowed to stand undisturbed overnight. In preparation for characterization, we pumped the supernate sample from the 3-L sampler into a 4-L carboy using a Masterflex[®] peristaltic pump and Tygon[®] tubing. The total volume of supernate measured 3.18 L.

Visual inspection of the supernate composite showed the material to be clear and light in color with no observable solids. We determined the density of the as-received material at $1.265 \pm 0.0071 \text{ g mL}^{-1}$ using 10 mL glass Class A micro-volumetric flasks. We determined chemical composition of the material by diluting an aliquot of the as-received waste sample in either 2 M nitric acid or deionized water.

The as-received material measured $5.48 \pm 0.37 \text{ M}$ in sodium, which is consistent with measured density and within the desired range for performance testing in Task 3 (see Table 2). We then measured the $^{238,239,240}\text{Pu}$ content by radiochemical separation of the plutonium followed by alpha counting of the extracted plutonium. ^{90}Sr was determined by chemical separation and beta counting of the extracted strontium. ^{137}Cs was determined by gamma spectroscopy. Stable strontium and actinide isotopes were determined by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS). Free and total base, other base excluding carbonate, carbonate, nitrate, nitrite, sulfate, phosphate, halides, and oxalate were determined by titration and ion chromatography. Table 2 provides the measured composition of the as-received waste sample.

Table 2. Composition of the As-Received Waste Supernate Sample

Analyte	Unit	Concentration	Standard Deviation
Na	M	5.48E+00	3.67E-01
OH ⁻	M	2.10E+00	7.00E-02
NO ₃ ⁻	M	2.47E+00	1.51E-02
NO ₂ ⁻	M	6.48E-01	6.33E-03
Al(OH) ₄ ⁻	M	4.23E-01	6.79E-03
CO ₃ ²⁻	M	5.66E-01	7.85E-03
SO ₄ ²⁻	M	5.18E-02	1.01E-03
PO ₄ ³⁻	M	2.17E-03	4.23E-05
F ⁻	M	bdl	-
Cl ⁻	M	bdl	-
^{137}Cs	pCi/mL	2.26E+08	0
^{90}Sr	pCi/mL	3.37E+05	3.64E+04
Total Pu	pCi/mL	8.25E+04	7.43E+02

bdl = below method detection level

Analytical results revealed that the tank waste sample was much lower in ^{90}Sr and alpha activity than expected (see Table 2). Therefore, we adjusted the composition of the tank waste solution to increase plutonium and strontium content to provide a more challenging matrix for evaluating the MST performance characteristics.

Plutonium and strontium adjustment proceeded by adding three different solutions: (1) ^{238}Pu , (2) $^{239/240}\text{Pu}$ and (3) natural abundance strontium into 2.5 L of the waste solution. A concentrated, nitric acid solution of ^{238}Pu (3.3 mL of $1.3 \text{ E}+09 \text{ dpm/mL}$ in 15 M HNO_3) from the Actinide Technology Section of SRNL served as the source of ^{238}Pu . After analyzing the ^{238}Pu solution for purity we added 25 mL of $0.05 \text{ M Na}_2\text{CO}_3$ solution slowly while stirring to obtain a slightly alkaline solution. We prepared a slightly alkaline solution of weapons-grade plutonium by slowly adding 10 mL of a $0.019 \text{ M Na}_2\text{CO}_3$ solution to a concentrated, nitric acid solution of weapons-grade $^{239/240}\text{Pu}$ ($1.05 \text{ mg Pu mL}^{-1}$ in $1.3 \text{ mL of } 0.5 \text{ M HNO}_3$). We prepared the natural abundance strontium solution by dissolving the desired quantity of strontium nitrate (0.066 g) in distilled, deionized water (10 mL). All solution volumes were minimized to reduce the level of dilution of the waste solution.

We added the spike solutions in the following order with indicated hold times (1) strontium solution followed by stirring for 2 hours, (2) ^{238}Pu solution followed by stirring for 1 hour and (3) $^{239/240}\text{Pu}$ solution followed by stirring for 1 hour. We continued mixing for 13 days at ambient temperature. We collected aliquots and analyzed aliquots of the spiked solution after 6 and 13 days. Results, shown in Table 3 below, indicate that all analytes except strontium were at or near equilibrium. Prior to use we filtered the spiked waste sample through a cartridge filter (Whatman PolycapTM 75 TF unit with $0.1 \mu\text{m}$ PTFE membrane). We analyzed additional aliquots at the start of testing and included a test bottle without added MST to serve as control samples to account for the decreasing strontium concentration and the lead time between these analytical results and the initiation of the performance tests.

Table 3. Analysis of Waste Supernate after Spiking with Strontium and Plutonium

Analyte	Method	Units	6 day Sample	13 day Sample
^{90}Sr	Radchem	pCi/mL	$3.73\text{E}+04 \pm 3.8\text{E}+03$	$2.75\text{E}+04 \pm 2.8\text{E}+03$
Total Sr	ICP-MS	$\mu\text{g/L}$	$2.14\text{E}+03 \pm 4.3\text{E}+02$	$1.58\text{E}+03 \pm 3.2\text{E}+02$
Total Pu	PuTTA	$\mu\text{g/L}$	$2.54\text{E}+02 \pm 1.3\text{E}+01$	$2.54\text{E}+02 \pm 1.4\text{E}+01$
Total Pu	ICP-MS	$\mu\text{g/L}$	$1.97\text{E}+02 \pm 3.9\text{E}+01$	$2.06\text{E}+02 \pm 4.1\text{E}+01$
^{237}Np	ICP-MS	$\mu\text{g/L}$	$1.49\text{E}+02 \pm 3.0\text{E}+01$	$1.29\text{E}+02 \pm 2.6\text{E}+01$
Total U	ICP-MS	$\mu\text{g/L}$	$9.99\text{E}+03 \pm 2.00\text{E}+03$	$1.02\text{E}+04 \pm 2.05\text{E}+03$

Table 4 provides a listing of the first set of seven tests carried out to evaluate the performance of the modified MST (Test Set #1). Testing included a single control test (no added MST), a single test with the baseline MST sample (Optima Chemical Company, Inc. Lot #00-QAB-417) at 0.2 g L^{-1} , duplicate tests with modified MST at 0.1 and 0.2 g L^{-1} MST and a final test to determine if the presence of the solvent mixture planned for use in the Caustic Side Solvent Extraction (CSSX) process influences the performance of the modified MST. For this test we mixed 0.02 grams (0.2 g L^{-1}) of modified MST with 0.050 mL (500 ppm) of the CSSX solvent.

Researchers poured 100-mL of the filtered and spiked waste into test bottles equipped with magnetic stirring bars. The test bottles were placed into a temperature controlled waterbath ($25 \pm 3 \text{ }^{\circ}\text{C}$) and incubated overnight. Individually, we removed each bottle from the waterbath, added a preweighed amount of the appropriate MST, and returned the test bottle to the waterbath.

We sampled each test bottle after 2, 4, 6, 12, 24 and 168-hours of contact. At the desired sampling time, we removed a 3-mL aliquot from each test bottle. We filtered the aliquot through a syringe filter (Millex[®] VV 0.1- μm PVDF membrane) and collected the filtrate in a clean sample bottle. Personnel then pipetted 1-mL portions of the filtered sample into a second set of sample bottles containing 19 mL of 2 M nitric acid. The acidified samples were shaken for approximately 15 seconds and then allowed to equilibrate with periodic shaking for a minimum of 2 hours prior to submittal for analysis. The acidified samples were analyzed to determine the following, $^{238/239/240}\text{Pu}$, ^{90}Sr , stable strontium and actinides.

Table 4. Test Conditions for First Set of Actual Waste Performance Tests

<u>Test #</u>	<u>MST ID</u>	<u>[MST], g/L</u>	<u>Contact Time, hours</u>
1	modified	0.0	0, 2, 4, 6, 12, 24 and 168
2	modified	0.1	0, 2, 4, 6, 12, 24 and 168
3	modified	0.1	0, 2, 4, 6, 12, 24 and 168
4	modified	0.2	0, 2, 4, 6, 12, 24 and 168
5	modified	0.2	0, 2, 4, 6, 12, 24 and 168
6	baseline	0.2	0, 2, 4, 6, 12, 24 and 168
7	modified	0.2*	0, 2, 4, 6, 12, 24 and 168

*with 500 ppm CSSX solvent

We conducted a supplemental set of actual waste tests evaluating the efficiency of modified MST in both multiple strike and reuse configurations using the same equipment as described for Test Set #1. The supplemental tests consisted of seven tests, which we performed in two stages. Tests S-2, S-3, and S-1C preceded Tests S-4, S-5, S-1A, and S-1B by two weeks. Tests S-1A, -1B, and -1C served as control tests (i.e., no modified MST were added). Tests S-2 and S-3 served as multiple strike tests which were

facilitated by filtering the test solutions approximately 6 hours after first contacting with modified MST and then recharging the filtrate with fresh modified MST to simulate a second strike. Samples were obtained at approximately 2, 6, 8, 12, 30, and 174 h after the addition of the first strike with MST. Tests S-2 and S-3 were contacted with 0.1 g L^{-1} and 0.2 g L^{-1} MST, respectively, in each strike. Samples were prepared and analyzed as described previously.

Tests S-4 and S-5 evaluated the reuse of the modified MST sample. In these tests, we contacted the modified MST with 200-mL of the filtered, composited, residual waste solution from Test Set #1. After approximately 6 hours of contact, we separated the liquid and MST solids and then added the recovered MST solids to fresh, spiked waste solution. Samples were obtained at approximately 2, 6, 8, 12, 30, and 174 h after the first MST contact. Control tests included S-1A (fresh spiked waste) and S-1B (filtered, composited, residual waste solution remaining from Test Set #1). Tests S-4 and S-5 featured MST concentrations of 0.1 g L^{-1} and 0.2 g L^{-1} , respectively. Samples from these tests were prepared and analyzed as described previously.

3.4 Filtration Testing

We performed filtration tests in a stirred cell filtration apparatus (see Figure 1) with a simulated waste solution having the chemical composition as reported in Table 1 without the radioactive components. We added the appropriate MST sample to the solution to provide a concentration of solids of 0.55 g L^{-1} . We poured approximately 60 mL of the feed suspension into the stirred cell, agitated the cell contents, pressurized the cell to 30 psi, and measured the filtrate volume as function of time. Tests evaluated the filtration characteristics of the MST samples with the following filter media: $0.1 \mu\text{m}$ TruMem[®] ceramic (typical of the SpinTek rotary filter), $0.1 \mu\text{m}$ Mott sintered SS (as used in the ARP design), $0.1 \mu\text{m}$ Pall sintered SS (typical of the SWPF design), and $0.5 \mu\text{m}$ Pall sintered SS (as contained in the SRNL modified rotary microfilter). The Mott and Pall pore sizes are nominal. We performed two sets of tests with the $0.1 \mu\text{m}$ Mott and $0.1 \mu\text{m}$ Pall media using a fresh feed suspension for the 2nd set of tests.



Figure 1. Stirred Cell Test Apparatus

4.0 Results and Discussion

4.1 Task 1 – Identify Key Synthesis Conditions

The objective of this task was to identify the synthesis conditions that provide a modified MST material with the best strontium and actinide removal characteristics. Performance testing emphasized plutonium removal characteristics since the isotopes of this element serve as the limiting radionuclides in defining processing conditions and equipment sizing in the ARP and SWPF.

Previous Phase 1 testing identified several conditions that produced samples that exhibited improved strontium, plutonium and neptunium removal characteristics.¹² We prepared modified MST samples by three general routes identified as Method 1, Method 2 and Method 3. Within each method, we investigated a range of conditions appropriate to the particular method.

Figure 2 provides a plot of the 4-hour normalized decontamination factors (DF) for strontium, plutonium and neptunium, respectively, as measured upon contact with 0.4 g/L of the modified MST samples produced in Tests 1 – 9 using Method 1. Normalized DF factors are calculated by dividing the measured DF value of the modified MST sample by that of the baseline MST sample at the same sorbent concentration and sampling time. Attachment 8.1 provides a listing of the 4-hour normalized DF values and the uncertainties. Normalized strontium DF values ranged from a low of 0.85 to a high of 1.85. Normalized plutonium DF values measured over the range 0.60 – 3.05 and those for neptunium from 1.14 to 4.46.

The results indicate that this synthesis method produces samples with much improved neptunium and plutonium removal and modest improvement in strontium removal. These results are consistent with findings from Phase I testing. The sample with the best combination of strontium and actinide removal performance was that produced in Test #2,

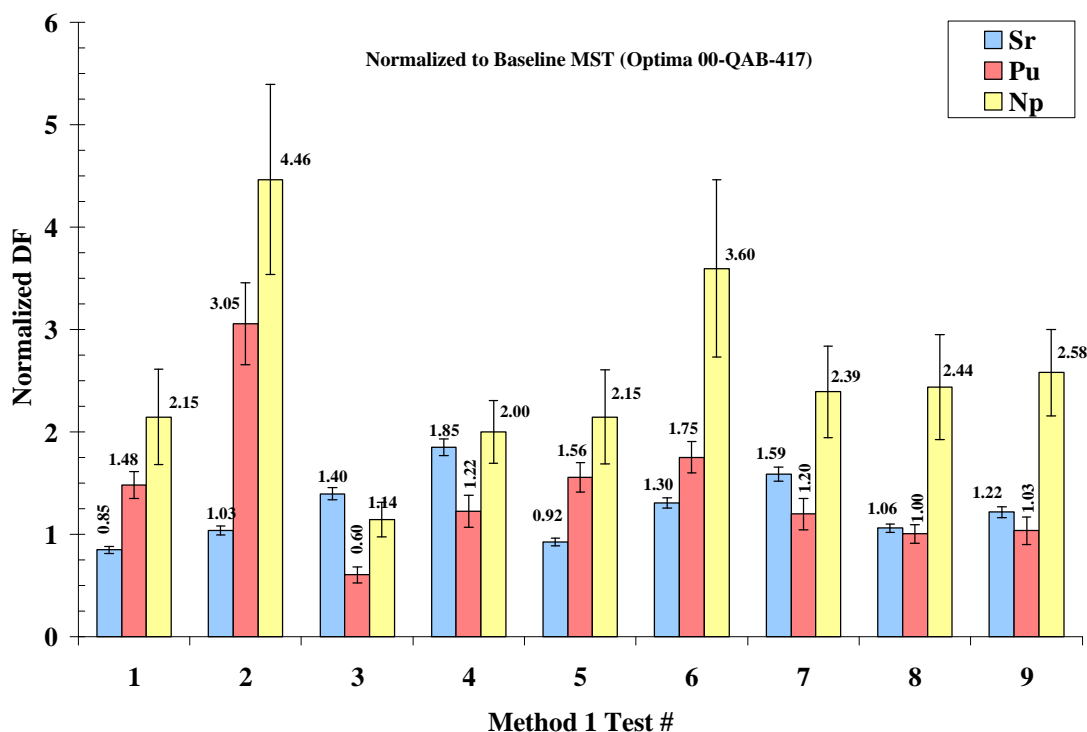


Figure 2. Normalized Decontamination Factors (DF) for Modified MST Sample Produced by Method 1

Figure 3 provides a plot of the 4-hour normalized decontamination factors (DF) for strontium, plutonium and neptunium, respectively, as measured upon contact with 0.4 g L^{-1} of the modified MST samples produced in Tests 1A – 9A using Method 2. Attachment 8.2 provides a listing of the 4-hour normalized DF values and the uncertainties. In general this synthetic route produced samples that exhibited improved sorbate removal, particularly for strontium, compared to the baseline MST sample. Compared to the samples prepared by Method 1, these samples featured better strontium removal and, in general, better plutonium, but poorer neptunium removal. Note that the sample prepared in Test 7A exhibited the highest normalized neptunium DF value (5.19) of all of the samples prepared by Methods 1 and 2.

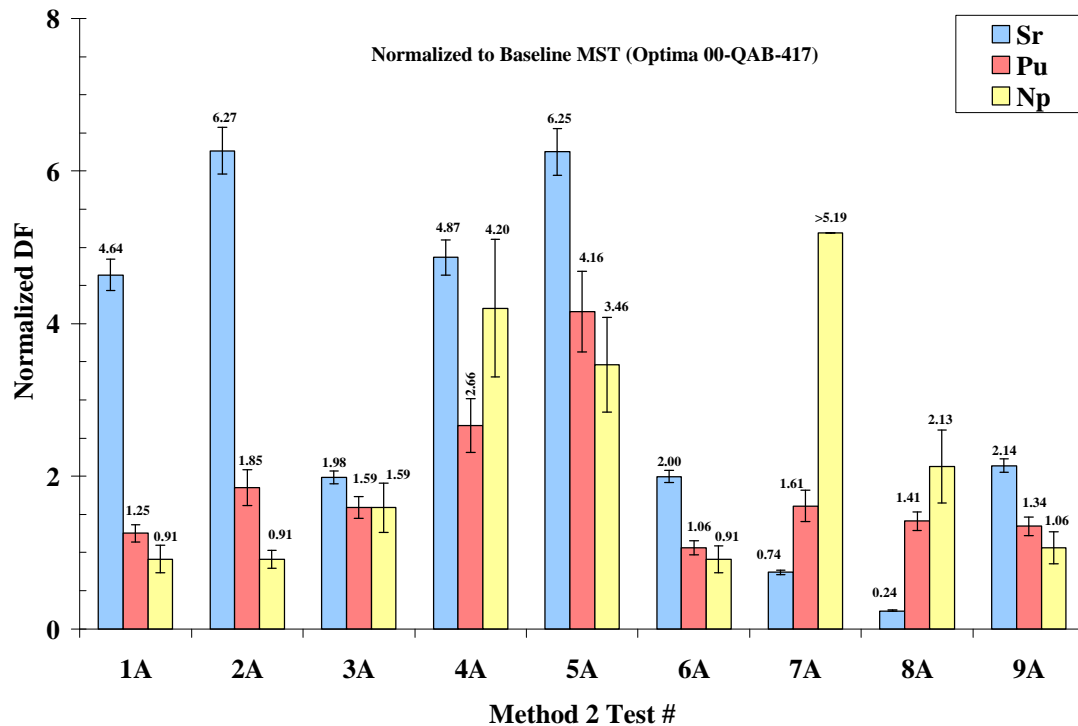


Figure 3. Normalized Decontamination Factors (DF) for Modified MST Sample Produced by Method 2

Figures 4 –6 provide plots of the measured DF values for strontium, plutonium and neptunium, respectively, produced by Method 3. Attachment 8.3 provides a listing of the DF values and the uncertainties for each of the samples prepared by Method 3.

Samples prepared by this method exhibited an increase in the DF for strontium of about 2 – 5 times (see Figure 4), for plutonium of about 6 – 31 times (see Figure 5) and for neptunium of about 1.2 – 5 times (see Figure 6) compared to the baseline MST sample. These increases in strontium and actinide removal are consistent with those measured for samples prepared by this method during Phase I testing.

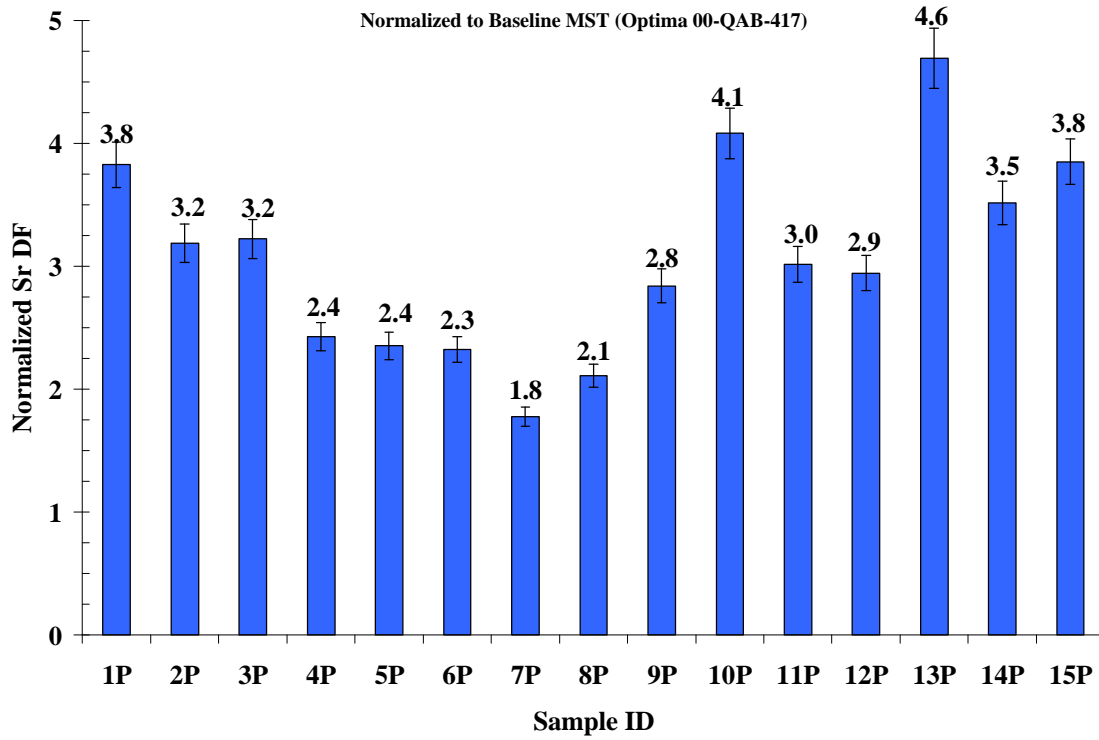


Figure 4. Normalized Strontium Decontamination Factors (DF) for Modified MST Samples Produced by Method 3

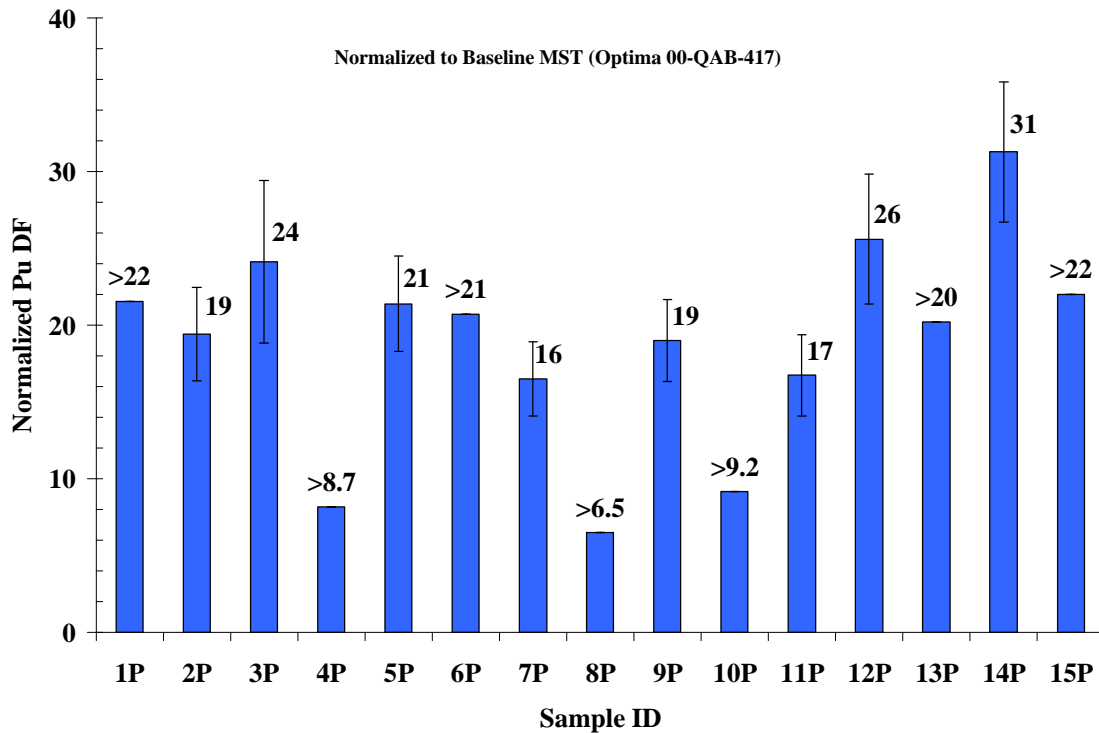


Figure 5. Normalized Plutonium Decontamination Factors (DF) for Modified MST Samples Produced by Method 3

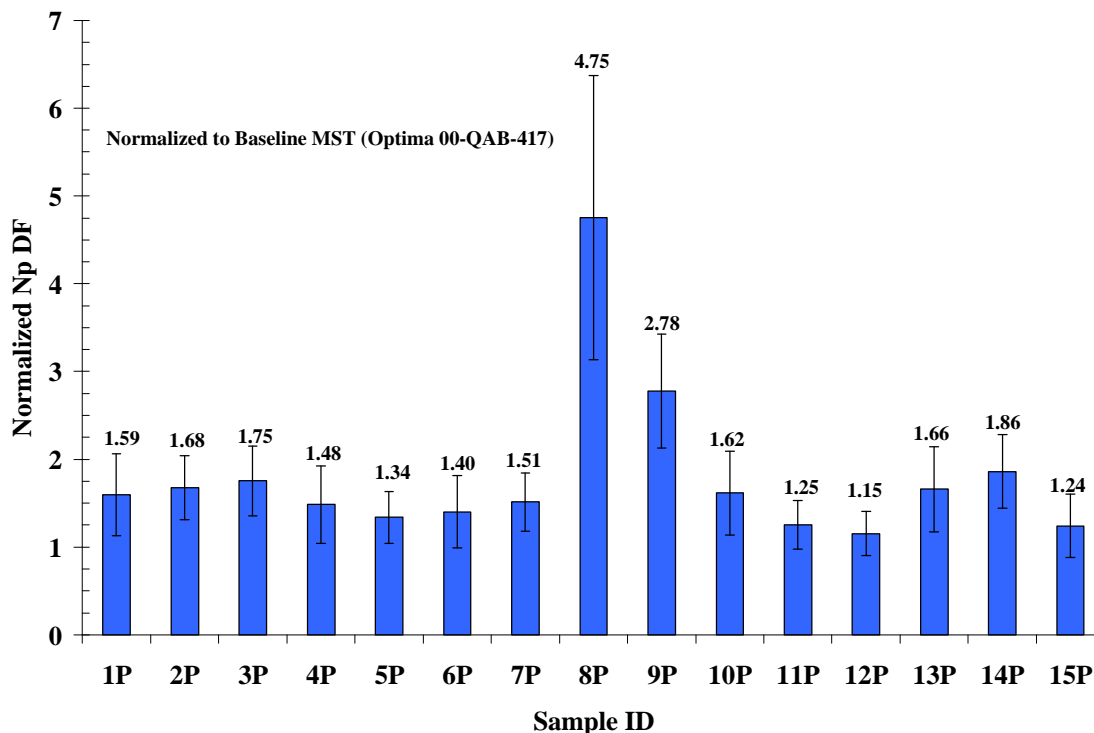


Figure 6. Normalized Neptunium Decontamination Factors (DF) for Modified MST Samples Produced by Method 3

After further review of the results from Task 1 including the findings from samples prepared by Method 3, we conducted additional preparations of modified MST samples using Method 2, but with a change in the isolation method of the precipitated solids that omitted drying of the solids. Previously, we isolated the precipitated solids by filtration, washed with deionized, distilled water and air dried at room temperature. Note that thermogravimetric analysis of the solids prepared by Method 2 and air dried contained 15 – 30 wt% water.

We tested the performance of these new samples in the same manner as before. Figure 7 provides a plot of the normalized DF for each of the samples along with that measured for the dried sample prepared in Test 1A. These slurry samples exhibited improved removal performance for three sorbates compared to both the baseline MST sample and the modified MST sample prepared earlier in Test 1A. Thus, we conclude that drying the modified MST reduces the performance of the MST as measured by the DF after 4-hours of contact. This result is consistent with previous findings with MST samples, which showed reduced sorption kinetics upon drying MST at elevated temperatures.¹²

The sample prepared in Test 13A proved to have the best combined performance for strontium, plutonium and neptunium. Plutonium removal matched that of the best performers prepared by Method 3. Strontium and neptunium removal performance of

this sample exceeded that of any prepared by Method 3. Thus, this preparative route should be considered as a method for preparing modified MST materials.

The particle morphology of modified MST samples prepared by Method 2 is considerably different than that by Method 3. At this time we have not determined the particle size distribution or the filtration characteristics of materials prepared by Method 2. Scanning electron micrographs (SEM) suggest that the materials prepared by Method 2 may exhibit a higher fraction of particles having small particle sizes than materials prepared by Method 3. A higher fraction of small particles may adversely impact filtration characteristics. Thus, we plan to measure the particle size distribution and filtration characteristics of samples prepared by Method 2 for comparison with those prepared by Method 3.

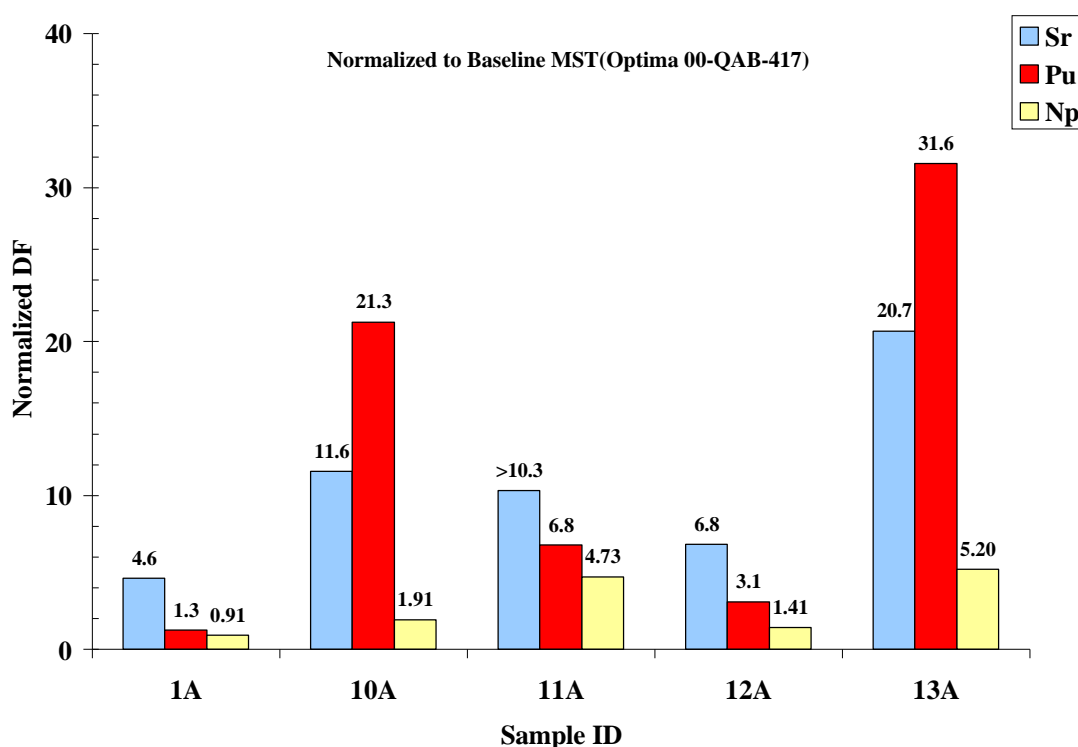


Figure 7. Normalized Decontamination Factors (DF) for Modified MST Samples Prepared by Method 2 without Drying

4.2 Task 2 – Reproduce Performance at Larger Laboratory Scale

Based on the results from Task 1, we selected Method 3 to produce samples at a larger laboratory scale. For this task we increased the previous laboratory procedure to prepare 25 grams of modified MST per batch. This represents a 25 – 100 fold increase in batch size from previous tests.

We prepared three separate batches of the modified MST material, designated as LS-1, LS-2 and LS-3 using conditions identical to Test 14P. Performance tests used the same

simulant used in evaluating samples prepared under Task 1. The performance tests added the MST samples at two different sorbent concentrations, 0.1 and 0.2 g L⁻¹. Each performance test with the modified MST samples was performed in duplicate. A single test evaluated the performance of the baseline MST sample at each sorbent concentration. We also analyzed these materials for a variety of chemical and physical properties and filtration characteristics. Results of these analyses are presented in Task 4.

Figures 8 and 9 provide plots of the normalized 4-hour DF values for strontium and plutonium, respectively, with the three modified MST samples. The normalized DF is calculated by dividing the DF of the modified MST sample by the DF of the baseline MST sample. For strontium and plutonium, the results indicate that the modified MST samples show significantly improved performance compared to the baseline MST sample. These results are consistent with those determined with modified MST samples prepared at the smaller laboratory scale. There was very good agreement among the three different batches of modified MST indicating that the synthesis is reproducible at this scale.

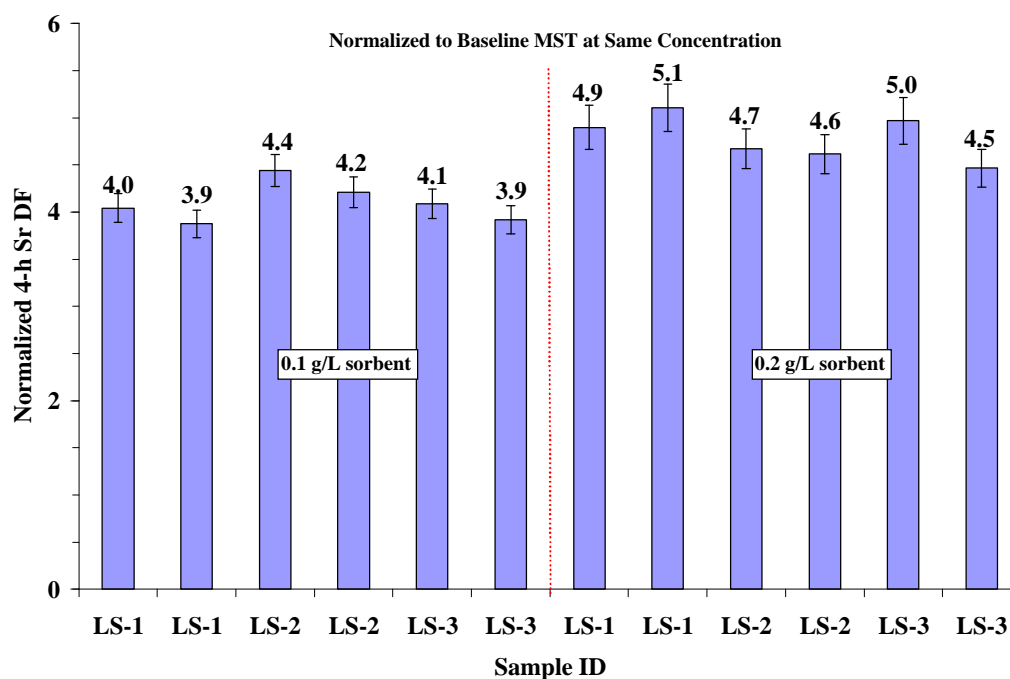


Figure 8. Normalized 4-Hour Strontium Decontamination Factors (DF) for Modified MST Samples Produced by Method 3 at the 25-gram Scale

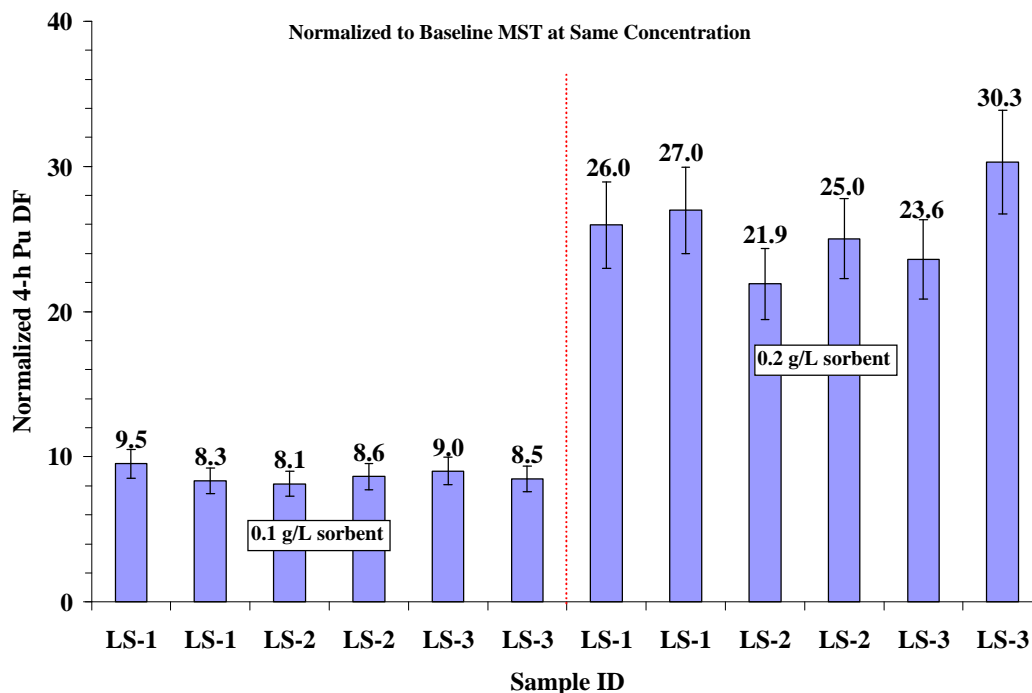


Figure 9. Normalized 4-Hour Plutonium Decontamination Factors (DF) for Modified MST Samples Produced by Method 3 at the 25-gram Scale

For neptunium, the normalized 4-hour DFs for the modified MST samples measured around 1.3 indicating better removal than the baseline material. However, considering the uncertainty in the normalized DF values (see Table 9), we cannot conclude that the modified MST samples exhibited improved neptunium removal after a contact time of 4 hours at the 95% confidence limit.

The uranium removal characteristics for the modified MST samples proved indeterminate in this test set. Given the analytical uncertainty in the uranium measurement by ICP-MS and the variance among duplicate tests, we observed no statistical difference among the uranium concentrations measured in samples from tests with and without MST. Note that we did not analyze for uranium in the samples from tests with 0.2 g/L MST at contact times other than 4 hours. At longer contact times (e.g., 168-hours), there may have been sufficient uranium sorption by the MST samples to provide a measurable difference in the uranium concentrations remaining in solution.

In addition to the 4-hour sample, we also pulled and analyzed samples after 2, 6, 12, 24 and 168 hours of contact with the MST samples to evaluate removal kinetics. Table 9 provides a summary of the average strontium, plutonium and neptunium DFs for the modified and baseline MST samples at each sampling time and each sorbent concentration. Table 10 provides the average normalized DF value for the modified MST at each sampling time and sorbent concentration.

Comparison of the baseline MST and modified MST samples at each time and concentration revealed that the modified MST consistently exhibited very high DF values for strontium and plutonium. For strontium, the DF value for the modified MST consistently measured between factors of 4 to 5 higher than that of the baseline MST (see Table 10). The modified MST exhibited exceptionally good plutonium removal with DF values of 24.0 and 49.3 upon contact with 0.1 g L^{-1} of at 6 and 12-hours, respectively at a sorbent concentration of 0.1 g L^{-1} and values of 95.7 and 172, respectively at a sorbent concentration of 0.2 g L^{-1} (see Table 9). Thus, the modified MST samples exhibited plutonium DF values between 10 and 50 times higher than those of the baseline MST sample (see Table 10).

The average neptunium DF values for the modified MST samples measured higher than the baseline MST sample at both sorbent concentrations at each sampling time. In some cases, however, the difference between the modified and baseline DF values was not statistically significant. Thus, the modified MST samples exhibited a small increase of no more than about a factor of 2 in neptunium removal compared to that of the baseline MST sample. This trend is consistent with that previously measured for samples prepared by Method 3.¹²

**Table 5. Average Strontium and Plutonium Decontamination Factors for Modified MST Samples
Produced by Method 3 at the 25-gram Scale**

Strontium								
Time (h)	Baseline MST @ 0.1 g/L		Modified MST @ 0.1 g/L		Baseline MST @ 0.2 g/L		Modified MST @ 0.2 g/L	
	Average	Std Dev	Average	Std Dev	Average	Std Dev	Average	Std Dev
2	5.42	0.13	20.6	1.18	17.8	0.45	84.6	2.57
4	6.19	0.16	25.4	1.28	21.7	0.60	104	5.26
6	8.50	0.21	27.4	1.57	21.4	0.56	113	14.0
12	6.88	0.18	30.6	1.36	24.8	0.71	128	5.80
24	8.45	0.22	37.4	1.88	29.9	0.87	147	12.0
168	11.1	0.30	47.6	2.09	37.7	1.2	200	14.8

Plutonium								
Time (h)	Baseline MST @ 0.1 g/L		Modified MST @ 0.1 g/L		Baseline MST @ 0.2 g/L		Modified MST @ 0.2 g/L	
	Average	Std Dev	Average	Std Dev	Average	Std Dev	Average	Std Dev
2	1.52	0.13	5.44	0.38	2.29	0.16	26.9	2.86
4	1.73	0.13	15.1	0.87	2.83	0.20	72.6	8.22
6	1.85	0.13	24.0	2.60	2.85	0.20	95.7	17.6
12	2.11	0.16	49.3	2.69	3.21	0.24	172	6.16
24	2.54	0.20	89.6	7.32	4.23	0.35	276	20.8
168	7.15	0.49	119	11.3	7.49	0.49	528	39.0

Neptunium								
Time (h)	Baseline MST @ 0.1 g/L		Modified MST @ 0.1 g/L		Baseline MST @ 0.2 g/L		Modified MST @ 0.2 g/L	
	Average	Std Dev	Average	Std Dev	Average	Std Dev	Average	Std Dev
2	1.13	0.067	1.43	0.056	1.22	0.070	1.60	0.114
4	1.19	0.096	1.55	0.146	1.29	0.098	1.65	0.271
6	1.30	0.270	1.64	0.170	1.05	0.063	1.77	0.252
12	0.89	0.143	1.62	0.234	1.25	0.100	1.83	0.155
24	1.30	0.265	2.06	0.102	1.47	0.118	2.46	0.338
168	1.72	0.419	3.00	0.319	2.07	0.229	5.34	1.27

Modified MST results are average and standard deviation of six trials
Baseline MST results are single determinations with reported analytical uncertainty

Table 6. Average Normalized DF Values for Modified MST Samples

Normalized DF - Strontium				
Time (h)	[MST] = 0.1 g/L		[MST] = 0.2 g/L	
	Average	Std Dev	Average	Std Dev
2	3.80	0.22	4.75	0.14
4	4.10	0.21	4.79	0.24
6	3.23	0.18	5.31	0.66
12	4.45	0.20	5.16	0.23
24	4.43	0.22	4.93	0.40
168	4.30	0.19	5.30	0.39

Normalized DF - Plutonium				
Time (h)	[MST] = 0.1 g/L		[MST] = 0.2 g/L	
	Average	Std Dev	Average	Std Dev
2	3.59	0.25	11.8	1.25
4	8.69	0.50	25.6	2.90
6	13.0	1.41	33.6	6.18
12	23.3	1.28	53.5	1.92
24	35.3	2.88	65.3	4.92
168	16.7	3.16	70.5	5.20

Normalized DF - Neptunium				
Time (h)	[MST] = 0.1 g/L		[MST] = 0.2 g/L	
	Average	Std Dev	Average	Std Dev
2	1.26	0.049	1.31	0.094
4	1.30	0.123	1.28	0.211
6	1.26	0.131	1.70	0.241
12	1.81	0.262	1.46	0.124
24	1.58	0.079	1.67	0.230
168	1.75	0.186	2.58	0.611

Normalized DF values calculated by dividing the measured DF value for the modified MST to that measured for the baseline MST sample (Optima 00-QAB-417) at the same test condition.

Figures 10 – 12 provide plots of solution concentrations of strontium, plutonium and neptunium, respectively, versus time upon contact with 0.1 and 0.2 g L⁻¹ MST. For the modified MST samples, we observed excellent agreement among the duplicate for the different preparation batches at each sample time. The concentrations plotted in Figures 10 – 12 are average concentrations of the six tests at each sorbent concentration. Error bars in these figures –shown, but not easily discernible - are the single standard deviation of the six values for the modified MST samples, the pooled single standard deviation of the six control samples taken over the entire test and the analytical uncertainty reported for each sample measurement in the baseline MST test.

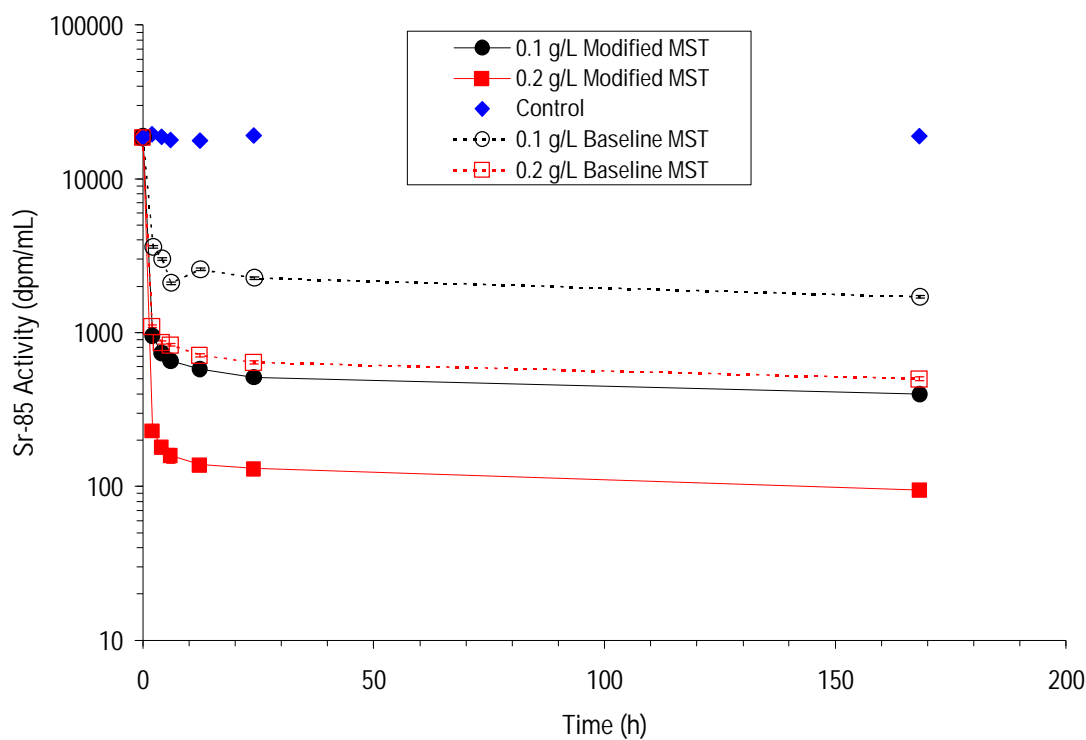


Figure 10. Strontium Removal Kinetics for Modified and Baseline MST Samples

The modified MST samples exhibited faster strontium removal kinetics than the baseline MST sample. For example, after two hours of contact the modified MST samples had removed more than 95% and 99% of the strontium at concentrations of 0.1 and 0.2 g L⁻¹, respectively, compared to 82% and 94% for the baseline MST sample. After two hours both the modified and baseline MST samples show a decrease in the rate of removal, which is typical behavior for adsorption of solution species onto solids.

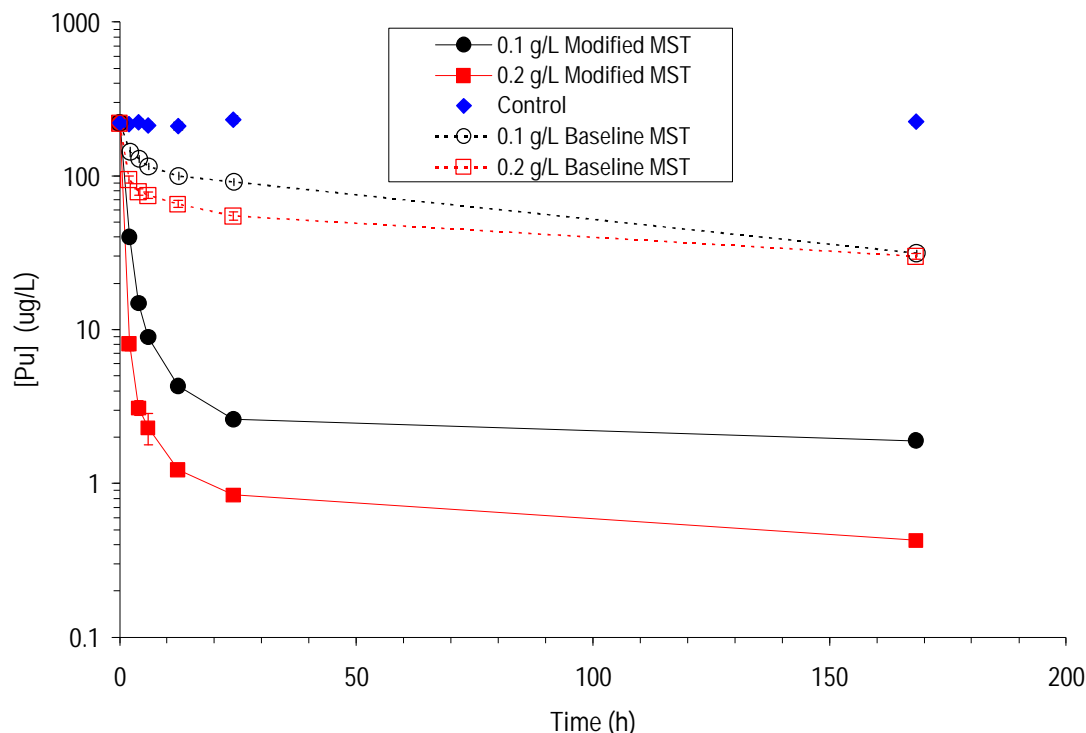


Figure 11. Plutonium Removal Kinetics for Modified and Baseline MST Samples

As with strontium, the modified MST samples exhibited much faster plutonium removal compared to the baseline MST. For example, at a MST concentration of 0.1 g L^{-1} , we observed that the modified MST samples removed on average 56% of the plutonium after 2 hours compared to 34% for the baseline MST. At a MST concentration of 0.2 g L^{-1} , the removal of plutonium increased to 96% for the modified MST samples compared to 82% for the baseline MST sample after 2 hours of contact. Note that after 12 hours of contact, the modified MST samples removed on average 98% and 99% of the plutonium, respectively, compared to 53% and 69%, respectively at MST concentrations of 0.1 g L^{-1} and 0.2 g L^{-1} .

Figure 12 provides a plot of the neptunium concentration versus time for the tests conducted at a sorbent concentration of 0.1 g L^{-1} . The modified MST samples consistently removed neptunium faster than the baseline MST sample. Furthermore, the neptunium concentrations measured for the tests with the modified MST samples measured consistently below those of the baseline MST sample. Two of the data points for the baseline MST tests (6-h at 0.2 g L^{-1} and 12-h at 0.1 g L^{-1}) measured higher than expected based on the other time samples. The corresponding data for ^{85}Sr and plutonium data at these times did not exhibit a similar increase. Thus, we have no basis for excluding these results.

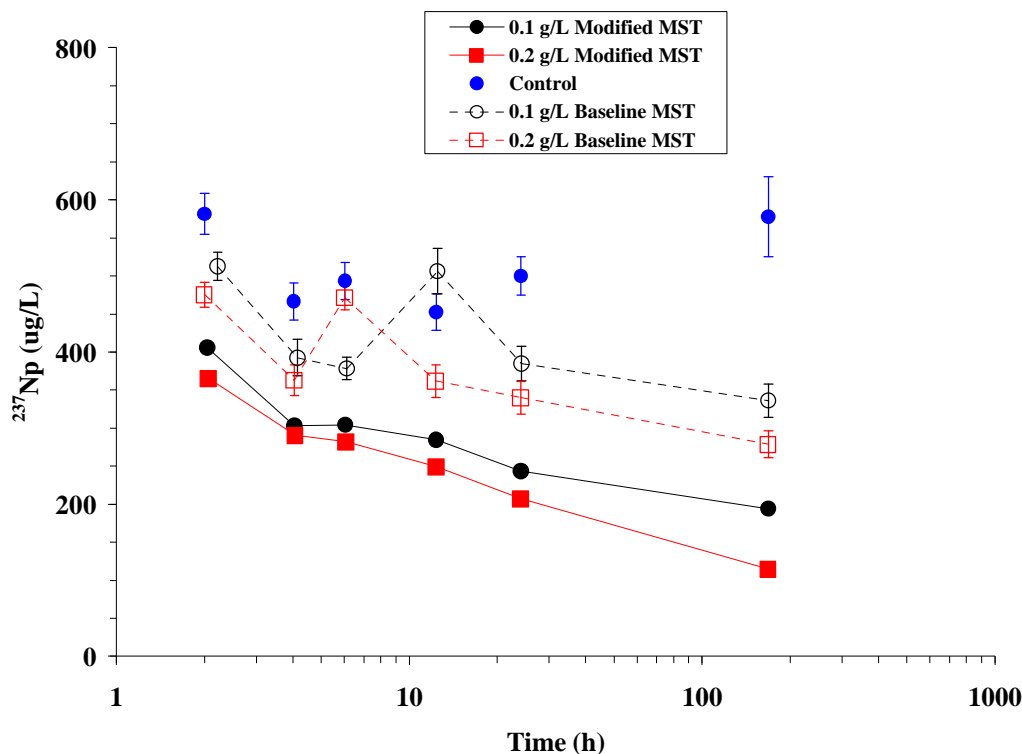


Figure 12. Neptunium Removal Kinetics for Modified and Baseline MST Samples

4.3 Task 3 – Verify Performance with Actual Waste

We carried out two sets of tests with actual waste to evaluate the performance of the modified MST material. Test Set #1 evaluated the performance at MST loadings of 0.1 and 0.2 g L⁻¹ and in the presence of the CSSX solvent. Test Set #2 (Supplemental Tests) evaluated the performance of the modified MST in multiple strike and reuse configurations. These tests used a 3-L sample of waste supernate (HTF-049) taken in May 2005.

We selected this tank waste for testing since previous samples from this tank proved high in ⁹⁰Sr and plutonium.¹³ Analysis of the supernate sample revealed that the supernate was much more dilute in salt content, ⁹⁰Sr and plutonium than the supernate sample analyzed in 2003. A review of tank transfer records revealed that most of the supernate present in the tank when sampled in 2003 had been transferred to other locations. Over the time period of 2003 – 2005 the tank received fresh canyon waste, which was much more dilute in salt content and contained much less ⁹⁰Sr and plutonium.

The sodium concentration of the supernate measured 5.48 ± 0.37 M, which is the desired concentration for testing. Therefore, the waste did not require any dilution. To increase strontium and plutonium content we added a solution of strontium nitrate and two separate solutions of plutonium(IV) nitrate to the waste sample with stirring and allowed the material to equilibrate (see Section 3.3 for details).

4.3.1 Actual Waste Test Set #1

In this set of tests we contacted the actual waste supernate with the LS-1 modified MST sample and the Optima #00-QAB-417 baseline MST sample at the conditions detailed in Table 1. The modified MST tests were conducted in duplicate at 0.1 and 0.2 g L⁻¹. The baseline MST test featured a sorbent concentration of 0.2 g L⁻¹. We also included a test in which a small amount of the solvent planned for use in Caustic Side Solvent Extraction process was added in addition to the modified MST.

Table 11 provides a summary of the measured DF values for strontium, plutonium and neptunium in the actual waste tests. Table 12 provides the normalized DF values at each sampling time. Greater than values are reported when the measured concentrations for a sorbate fell below the analytical method detection value. The values for the tests with the modified MST samples at the 0.1 and 0.2 g L⁻¹ concentrations are the average of duplicate tests. The uncertainties for tests with a single replicate are the reported analytical uncertainties.

Figure 13 provides a plot of the total strontium concentration (µg L⁻¹) versus time for the actual waste tests. For the tests with the modified MST sample, we plot the average of the duplicate trials. For these plots, the total strontium concentration derives from the ICP-MS analytical results. We also analyzed the filtrate samples for ⁹⁰Sr. However, the ⁹⁰Sr proved very scattered as the values were very close to the analytical method detection limit. Consequently the ⁹⁰Sr data provided no insight into the performance of the MST samples for strontium removal.

The graph indicates that similar strontium removal occurred when the waste was contacted with 0.1 g L⁻¹ of the modified MST or 0.2 g L⁻¹ of the baseline MST. At a modified MST concentration of 0.2 g L⁻¹, the test results indicated strontium concentrations about 2 – 3 times lower than those of the baseline MST at 0.2 g L⁻¹ or modified MST at 0.1 g L⁻¹. Unfortunately, the strontium concentration fell below the analytical method detection limit at sampling times after 4 hours. Thus, we cannot provide a quantitative value as to the performance of the modified MST at 0.2 g L⁻¹ for these times.

At the sorbent concentration of 0.2 g L⁻¹, the strontium DF values for the modified MST sample measured about 2.7 times higher than those of the baseline MST sample. Tests with the modified MST at a sorbent concentration of 0.1 g L⁻¹ resulted in strontium DF values comparable to those of the baseline MST sample. We also observed that the strontium DF values in the test with the modified MST and CSSX solvent proved very similar to those without the CSSX solvent. Thus, we conclude that the presence of the CSSX solvent did not adversely influence strontium removal by the modified MST sample.

In earlier tests with simulated waste solution, the modified MST exhibited a factor of about 5 higher strontium removal than the baseline MST sample (see Table 6). We attribute the lower increase in strontium removal performance in the actual waste tests to

the higher initial total sorbate concentration, which results in greater overall loading of the sorbent. The actual waste solution contained a total cation equivalent concentration for the four sorbates of $128 \pm 21 \mu\text{M}$, which is 32% higher than that of $97 \pm 15 \mu\text{M}$ for the simulated waste solution.

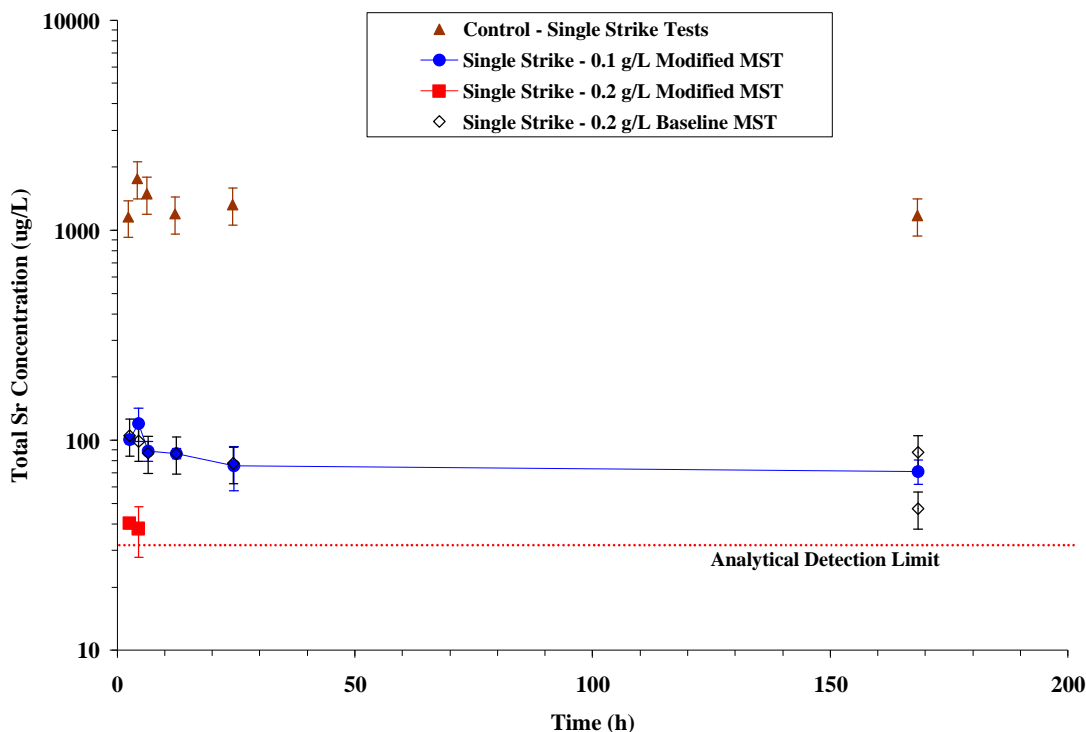


Figure 13. Total Strontium Concentration versus Time in Actual Waste Test Set #1

Figure 14 provides a plot of the total plutonium activity (pCi mL^{-1}) versus time for the actual waste tests. For the tests with the modified MST samples, the plot provides the average of the duplicate trials. As with strontium, the modified MST sample exhibited increased plutonium removal compared to the baseline MST. Within 12-hours, the plutonium activity decreased to below the current Waste Acceptance Criteria (WAC) limit of $22,500 \text{ pCi mL}^{-1}$ when the waste solution was contacted with 0.2 g L^{-1} . At this time the plutonium activity in the test with the baseline MST sample remained more than an order of magnitude higher than the WAC limit. Contact of this actual waste with 0.1 g L^{-1} MST produced plutonium concentrations above the WAC limit at all contact times, but still about 2 – 3 lower at the 6 and 12-hour sampling times than that of the baseline MST sample, which was added at 0.2 g L^{-1} .

The 168-hour result for the baseline MST tests showed a significant decrease in plutonium activity compared to the earlier sampling times. Additional analyses confirmed the low plutonium result. We have not observed this type of behavior with the baseline MST in other tests with simulants or tank wastes. Thus, we believe this result is in error.

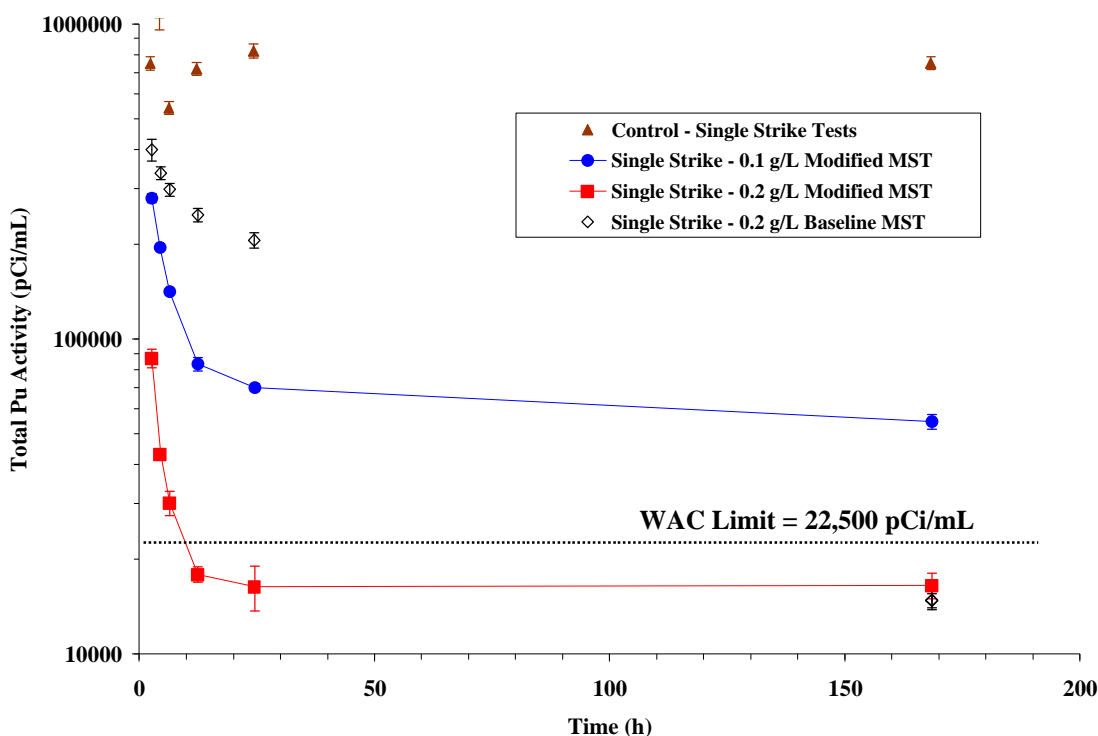


Figure 14. Total Plutonium Activity versus Time in Actual Waste Test Set #1

At a sorbent concentration of 0.2 g L^{-1} the plutonium DF values for the modified MST sample measured between 5 and 11 times higher than those of the baseline MST sample (see Tables 11 & 12). Tests with the modified MST at a sorbent concentration of 0.1 g L^{-1} resulted in plutonium DF values between 1.5 and 3.5 times higher than those of the baseline MST sample at 0.2 g L^{-1} . Given these results, we conclude that the modified MST sample clearly demonstrated improved plutonium removal performance compared to the baseline MST sample.

We also observed that the plutonium DF values in the test with the modified MST and CSSX solvent proved very similar to those without the CSSX solvent. Thus, we conclude that the presence of the CSSX solvent did not adversely influence plutonium removal by the modified MST sample.

In earlier tests with simulated waste solution, the modified MST exhibited a factor of 11 to 70 times higher in the plutonium DF value than the baseline MST sample (see Table 10). For the actual waste tests the modified MST sample exhibited increases in the DF values of between 5 and 11 times that of the baseline MST sample. As with strontium, we attribute the lower increase in plutonium removal performance in the actual waste tests to the higher initial total sorbate concentration in the actual waste. As noted earlier, the plutonium DF values for the modified MST sample at 0.1 g L^{-1} measured 1.5 – 3.5 times that of the baseline MST at the higher sorbent concentration of 0.2 g L^{-1} . In the case of strontium, the DF values were very similar for the two MST samples at the respective concentrations. The actual waste solution measured about 25% higher in

plutonium concentration than that in the simulant (275 ± 56 versus $218 \pm 13 \mu\text{g L}^{-1}$). These findings confirm that the modified MST sample clearly exhibits increased capacity for plutonium compared to the baseline MST sample.

We observed similar neptunium DF values for the modified MST sample in each of the tests. The baseline MST sample exhibited higher DF values for neptunium than the modified MST at each of the sampling times (see Table 11). In previous simulant tests we observed that the modified MST sample exhibited higher DF values than the baseline MST sample (see Table 9). The initial neptunium concentration in the actual waste solution measured more than one-third lower than that in the simulant tests (131 ± 19 versus $461 \pm 90 \mu\text{g L}^{-1}$). Given the lower neptunium concentration and the previous findings with the simulated waste solution, the results with the modified MST sample are surprising. Perhaps the higher loading of strontium and plutonium reduced the sorption of neptunium onto the modified MST.

We observed no measurable removal of uranium in any of the tests. Previous testing with simulated waste solutions at low sorbent concentrations also showed no measurable uranium removal. Given the similar initial uranium concentrations in both the actual waste ($9,550 \pm 333 \mu\text{g L}^{-1}$) and simulated waste ($10,200 \pm 2,040 \mu\text{g L}^{-1}$) solutions, the lack of measurable uranium removal is not unexpected. Higher sorbent concentrations are needed to determine uranium removal performance of the modified MST sample with actual waste solutions.

Table 7. Strontium, Plutonium and Neptunium Decontamination Factors in Actual Waste Tests Set #1

Strontium Decontamination Factors								
Time (h)	Baseline MST @ 0.2 g/L		Modified MST @ 0.2 g/L		Modified MST @ 0.2 g/L with CSSX Solvent		Modified MST @ 0.1 g/L	
	Value	Uncertainty	Average	Std Dev	Value	Uncertainty	Average	Std Dev
2.6	1.10E+01	3.10E+00	2.86E+01	1.60E+00	2.68E+01	7.57E+00	1.15E+01	5.57E-01
4.5	1.78E+01	5.03E+00	4.80E+01	1.29E+01	3.94E+01	1.11E+01	1.49E+01	2.70E+00
6.4	1.71E+01	4.85E+00	>4.54E+01		>4.52E+01		1.69E+01	1.84E+00
12.4	1.39E+01	3.92E+00	>2.30E+01		>2.33E+01		1.39E+01	7.70E-01
24.5	1.71E+01	4.83E+00	>4.02E+01		>4.07E+01		1.80E+01	4.29E+00
168.5	2.48E+01	7.02E+00	>3.56E+01		>3.57E+01		1.67E+01	2.22E+00

Plutonium Decontamination Factors								
Time (h)	Baseline MST @ 0.2 g/L		Modified MST @ 0.2 g/L		Modified MST @ 0.2 g/L with CSSX Solvent		Modified MST @ 0.1 g/L	
	Value	Uncertainty	Average	Std Dev	Value	Uncertainty	Average	Std Dev
2.6	1.84E+00	2.05E-01	9.58E+00	5.78E-01	8.69E+00	1.48E+00	2.82E+00	1.45E-01
4.5	2.99E+00	2.12E-01	1.66E+01	1.19E+00	1.47E+01	1.43E+00	4.96E+00	3.69E-02
6.4	1.86E+00	1.36E-01	1.22E+01	2.09E+00	1.08E+01	1.14E+00	3.53E+00	1.95E-01
12.4	2.35E+00	1.79E-01	2.67E+01	9.54E-01	3.32E+01	4.79E+00	8.20E+00	4.62E-01
24.5	4.46E+00	4.43E-01	3.96E+01	1.28E+01	4.01E+01	6.26E+00	7.87E+00	5.55E+00
168.5	3.37E+01	5.13E+00	2.70E+01	8.99E+00	3.22E+01	4.56E+00	7.62E+00	5.36E+00

Neptunium Decontamination Factors								
Time (h)	Baseline MST @ 0.2 g/L		Modified MST @ 0.2 g/L		Modified MST @ 0.2 g/L with CSSX Solvent		Modified MST @ 0.1 g/L	
	Value	Uncertainty	Average	Std Dev	Value	Uncertainty	Average	Std Dev
2.6	2.21E+00	6.26E-01	1.59E+00	1.57E-01	1.55E+00	4.38E-01	1.35E+00	3.97E-02
4.5	2.87E+00	8.13E-01	2.21E+00	1.03E-01	2.42E+00	6.83E-01	1.64E+00	4.25E-01
6.4	2.55E+00	7.21E-01	1.60E+00	6.56E-02	1.49E+00	4.21E-01	1.35E+00	2.05E-02
12.4	3.24E+00	9.17E-01	1.88E+00	3.76E-02	1.80E+00	5.09E-01	1.59E+00	2.80E-02
24.5	3.10E+00	8.76E-01	1.82E+00	1.66E-01	2.00E+00	5.65E-01	1.38E+00	4.35E-01
168.5	4.84E+00	9.67E-01	2.30E+00	1.74E-01	2.04E+00	4.07E-01	1.62E+00	5.13E-02

Table 8. Average Normalized Strontium and Plutonium DF Values

Time (h)	Normalized DF Value			
	Strontium		Plutonium	
	Average	Std Dev	Average	Std Dev
2.6	2.61E+00	1.45E-01	5.21E+00	3.14E-01
4.5	2.70E+00	7.29E-01	5.54E+00	3.99E-01
6.4	>2.65E+00		6.53E+00	1.12E+00
12.4	>1.66E+00		1.13E+01	4.06E-01
24.4	>2.35E+00		8.88E+00	2.87E+00
168.4	>1.44E+00		8.02E-01	2.67E-01

4.3.2 Supplemental Actual Waste Tests - Test Set #2

Supplemental actual waste tests (Test Set #2) evaluated the performance of modified MST in both double-strike and reuse configurations. Seven tests in all were conducted in two stages. Tests S-2, S-3, and S-1C occurred approximately two weeks prior to Tests S-4, S-5, S-1A, and S-1B. Tests S-1A, -1B, and -1C are control tests (i.e., no modified MST were added).

Tests S-2 and S-3 are double-strike tests conducted by filtering the test solutions approximately 6 hours after first contacting with modified MST and then adding fresh MST to the filtrate. Tests S-2 and S-3 added 0.1 g L^{-1} and 0.2 g L^{-1} MST, respectively, in each strike (see Attachment 8.4 for the measured radiochemical concentrations).

Tests S-4 and S-5 used filtered, composited, residual waste solution remaining from the Actual Waste Test Set #1 (see Attachment 8.4). These MST reuse tests filtered the decontaminated test solutions approximately 6 hours after first contacting with modified MST and then added the collected MST solids to fresh spiked waste solution. Tests S-4 and S-5 used 0.1 g L^{-1} and 0.2 g L^{-1} MST, respectively. This test design mimics the proposed operations in the Alpha Finishing Plant of the SWPF.

Personnel obtained samples from all tests except Control Test 1C at approximately 2, 6, 8, 12, 30, and 174 h after the first MST contact. Control Tests S-1A (using fresh spiked waste) and S-1B (using filtered, composited, residual spiked waste from earlier testing) were also sampled at the start of testing (time = 0 h). Control Test 1C (fresh spiked waste) was sampled at 2, 8, and 174 h. All tests were conducted at $25 \pm 3 \text{ }^{\circ}\text{C}$.

As with the previous test set, the ^{90}Sr data is scattered and provides no discernible trends in performance. The total strontium data, while limited by the method detection limit, appears well behaved. The total plutonium data from both PuTTA and ICP-MS methods show reasonable agreement, although the PuTTA method provides a lower detection limit compared to the ICP-MS method.

Figures 15 and 16 provide plots of strontium concentration and plutonium activity, respectively, versus time for the single and double strike tests. We observed good agreement between the 2 and 4-h sample results in the single-strike tests (Test Set #1) and that for the first strike of the double-strike tests (Test Set #2). Unfortunately, the first sample after the second MST addition resulted in the total strontium falling below the analytical method detection limit. Thus, we cannot quantitatively assess the magnitude of the strontium removal upon the second strike of MST.

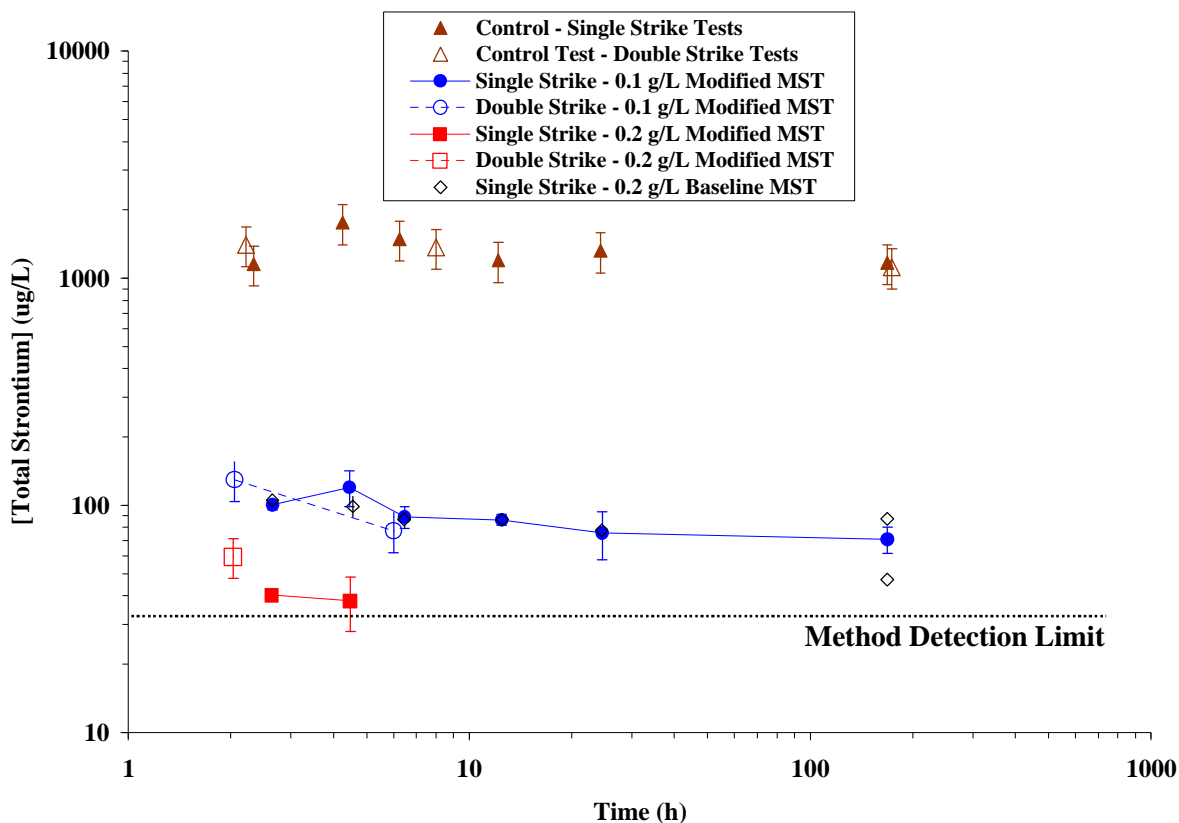


Figure 15. Total Strontium Concentration versus Time for the Single and Double Strike Tests with Actual Waste

We observe good agreement between the 2 and 4-h sample results for the single strike tests (Test Set #1) and that for the first strike of the double strike tests (Test Set #2) with respect to plutonium activity. Upon the addition of the second strike of modified MST, the plutonium activity decreased to a value well below the total alpha activity WAC limit at both MST concentrations (see Figure 16). The 30-h result for the double-strike test indicates a higher plutonium concentration than the previous two samples and the 170-h sample which is below the method detection limit. We believe this value is in error, most likely the result of cross contamination during sample handling.

Table 13 provides a summary of the measured DF values for strontium, plutonium and neptunium in the double strike tests. Greater than values are given when the measured concentrations for a sorbate are below the analytical method detection limit. Uncertainties for the single replicate tests are determined from the analytical uncertainties. Strontium, plutonium, and neptunium DFs for the two double-strike tests (S-2 and S-3) exhibit the expected increase with time. The large increase in DF between the 6 and 8-hour sampling time resulted from the removal of the MST solids from the first strike and addition of fresh MST.

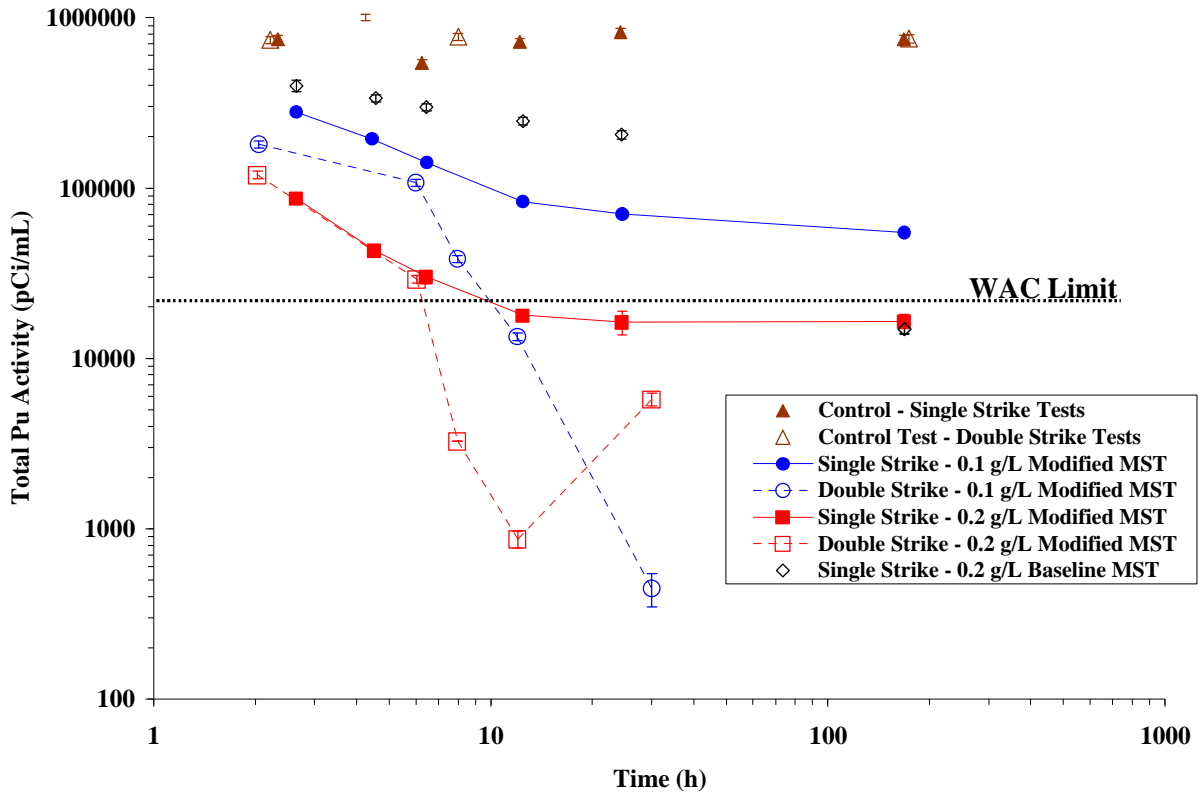


Figure 16. Total Plutonium Activity versus Time for the Single and Double Strike Tests with Actual Waste

Table 9. Strontium, Plutonium and Neptunium Decontamination Factors in Double Strike Actual Waste Tests

Decontamination Factors (DFs)							
Test Description	Time (h)	Total Sr		Total Pu*		²³⁷Np	
		Value	Uncertainty	Value	Uncertainty	Value	Uncertainty
Supplemental Test S-2	2.1	1.00E+01	2.83E+00	4.18E+00	2.78E-01	1.40E+00	3.95E-01
Double-strike test with 0.1 g/L Modified MST	6.0	1.68E+01	4.74E+00	7.02E+00	4.77E-01	1.55E+00	4.37E-01
	8.0	>2.49E+01		1.97E+01	1.36E+00	1.86E+00	5.27E-01
	12.0	>2.62E+01		5.63E+01	3.99E+00	1.96E+00	5.53E-01
	30.0	>2.66E+01		1.69E+03	3.83E+02	2.21E+00	6.24E-01
	174	>8.12E+01		>1.04E+02		2.41E+00	6.82E-01
Supplemental Test S-3	2.0	2.18E+01	6.17E+00	6.31E+00	4.48E-01	1.34E+00	3.79E-01
Double-strike test with 0.2 g/L Modified MST	6.0	>2.43E+01		2.59E+01	1.87E+00	1.50E+00	4.23E-01
	8.0	>2.64E+01		>2.31E+02		2.60E+00	7.35E-01
	12.0	2.20E+01	6.24E+00	8.71E+02	1.06E+02	2.79E+00	7.89E-01
	30.0	>2.64E+01		1.31E+02	1.34E+01	3.29E+00	9.29E-01
	174	>8.06E+01		>7.61E+02		4.62E+00	1.31E+00

DF calculated using average concentrations of Control Test 1C

*Calculated from PuTTA data for ²³⁸Pu and ^{239/240}Pu

Single tests (S-4 at 0.1 g L⁻¹ and S-5 at 0.2 g L⁻¹) evaluated the performance of the modified MST upon reuse. In these tests we first contacted the modified MST with a filtered composite of the residuals from Test Set #1, which had a reduced radioactivity compared to the initial waste. After approximately six hours, we recovered the MST solids from the waste solution, and added the solids to a fresh batch of the waste solution at the original spiked concentrations (i.e., same material as used in the Test Set #1 and Supplemental Tests S-2 and S-3). Figure 17 provides a plot of the total plutonium activity as a function of total test time for the reuse tests. Included in Figure 17 are the curves measured for plutonium removal in the single-strike tests (Test Set #1) allowing for a 6-hour offset from the beginning of the experiment.

The observed changes in plutonium activity with time during the test met expectations. Initially, the plutonium activity decreases for the first two samples. After separating the solids and contacting the recovered MST solids with fresh waste supernate, the plutonium activity increased reflecting the higher plutonium activity in the fresh waste versus that of the initial strike which had a much lower initial plutonium activity ($8.18 \pm 0.44 \text{ E}+05$ versus $3.02 \pm 0.16 \text{ E}+04 \text{ pCi mL}^{-1}$). Subsequent samples showed decreasing plutonium activity with time.

Comparison of the plutonium activity changes with time in the reuse and single strike tests indicates that less removal of plutonium occurs in the reuse test. Generally, the plutonium activity is about a factor of 10 higher in the tests with the reused MST compared to a single strike of fresh MST. The results indicate that a higher MST concentration is required to achieve the WAC limit if the material is previously used in the Alpha Finishing Plant.

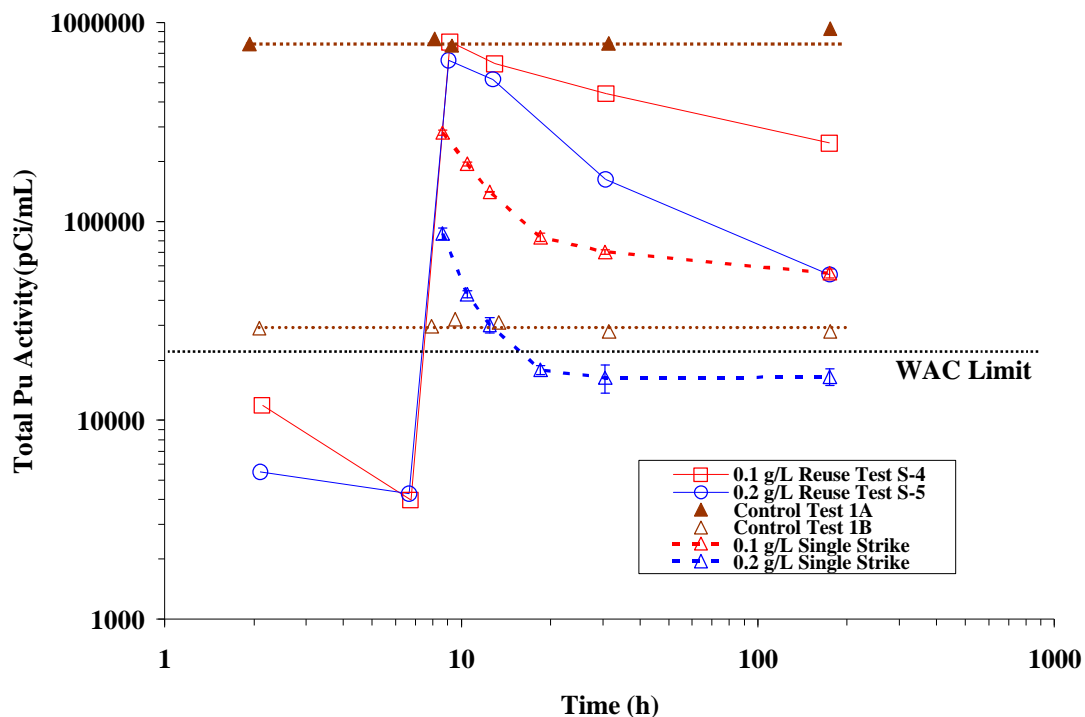


Figure 17. Total Plutonium Activity versus Time in the Modified MST Reuse Tests

Figure 18 provides a plot of the total strontium concentration versus total test time for the reuse tests. Included in Figure 18 are the data measured for strontium removal in the single-strike tests (Test Set #1) allowing for a 6-hour offset from the beginning of the experiment. Data points that are filled are the method detection values reported by the Analytical Development Section for that sample and corrected for dilution. Unfortunately, a large number of the samples fell below the detection limit. This prevents obtaining a quantitative measure of the influence of the reuse on strontium removal.

From the available data we see that the removal of strontium by the reused MST is less than that measured in the single-strike tests. This result is consistent with the trend observed with plutonium. From Figure 18 we estimate that after 2 hours of contact the reused MST lagged that of fresh MST by a factor of ten. After 6 and 24 hours of contact the reused MST lagged the fresh MST by a factor of about 2 – 3.

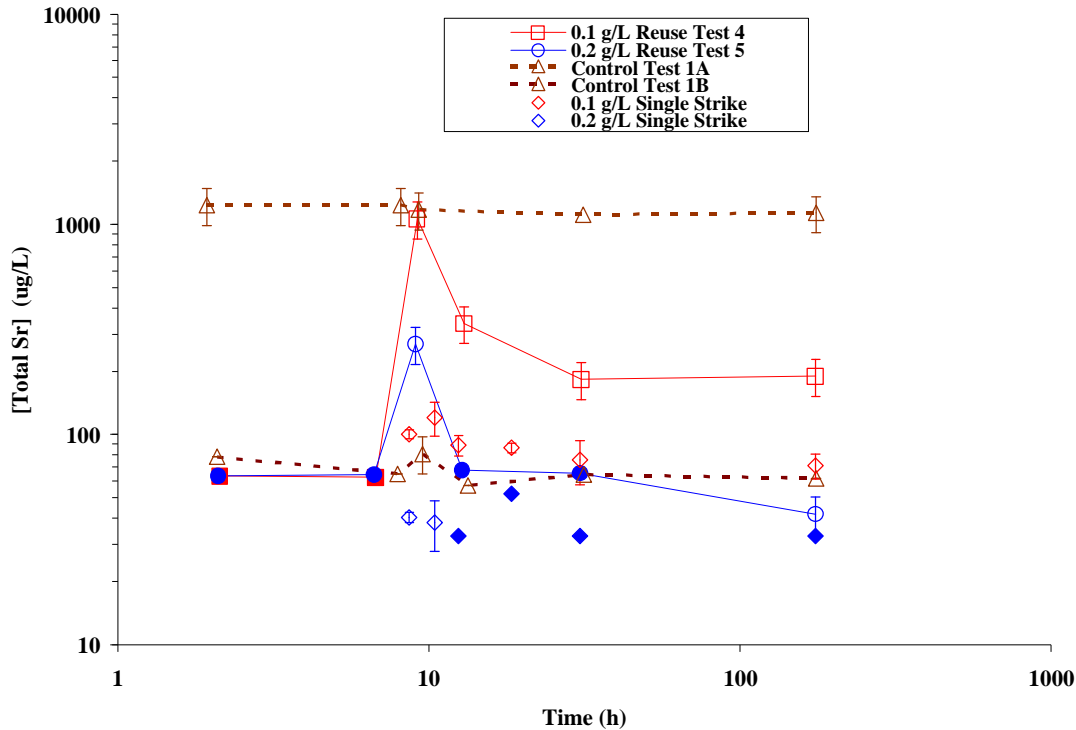


Figure 18. Total Strontium Concentration versus Time in the Modified MST Reuse Tests

4.4 Task 4 – Characterization of Modified MST Samples

4.4.1 Particle Size Distribution

We measured the particle size distribution of the three modified MST samples prepared at the 25-gram scale using a Microtrac S3000 instrument. Figure 19 provides a graph of the volume distribution data for these three samples as well as a sample of the baseline MST material (Optima Chemicals, Inc. Batch #00-QAB-417). For all materials, we diluted the sample into deionized distilled water for the particle size measurement.

The particle size data indicate very little change in the particle size distribution of the modified MST samples compared to the baseline MST sample. The modified MST samples exhibit a similar bimodal distribution of particle volumes as compared to the baseline MST. Two of the three modified MST samples (LS-1 and LS-3) exhibited a slightly smaller fraction of particles smaller than 1 micron and larger than 10 microns. This may indicate some removal of fines and larger particles during the synthesis of the modified MST samples. However, the amount of change is very small.

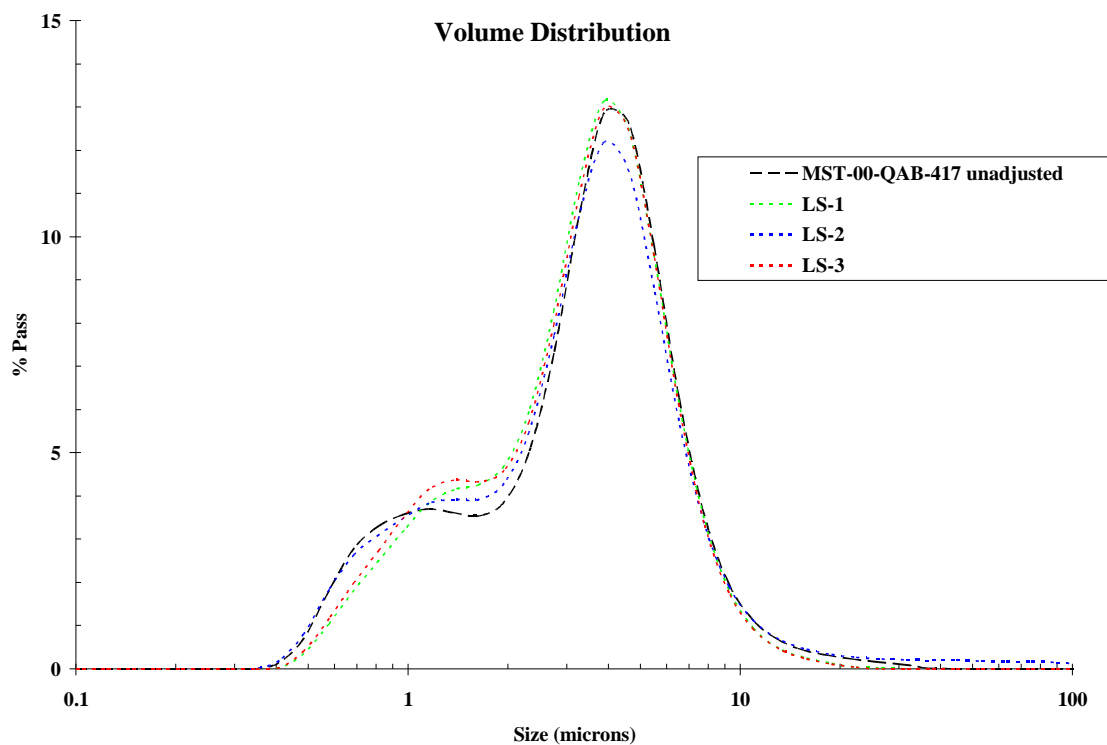


Figure 19. Particle Size Distribution of Modified and Baseline MST Samples

4.4.2. Filtration Characteristics

Previous SRNL work shows that dead end filters, such as the stirred cell, can provide a reliable qualitative comparison of the filterability of different feed slurries.^{1,2} Using the stirred cell as a screening tool allows personnel to perform many tests to be run in a short time and at much lower cost than performing all of the tests with a crossflow filter. These tests used a simulated waste solution having the same chemical composition as that provided in Table 1, but without the added sorbates.

Figure 20 shows the results from the tests conducted in triplicate with the TruMem[®] media. We observed no difference in filtrate rate between the baseline MST and the modified MST with the TruMem[®] media.

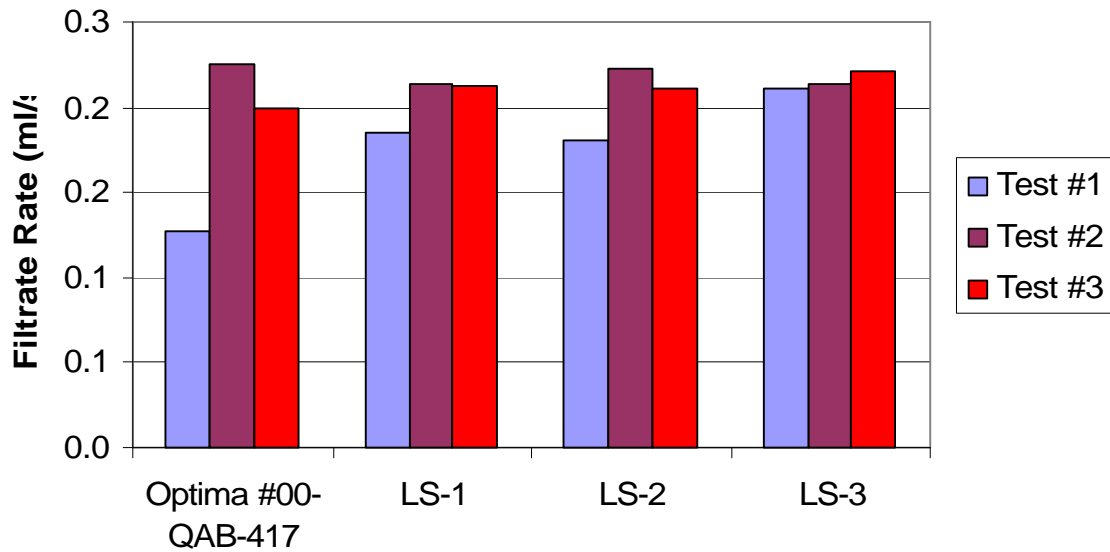


Figure 20. Filtration Rate with 0.1-mm TruMem® Media (each MST sample tested in triplicate)

Figure 21 shows the results from the tests conducted with the Mott media. We performed two sets of tests each in triplicate with this filter media. In general we observe a decrease in filtration rate in the second test set compared to the first set. However, within each test set, we observe no difference in filtrate rate between the baseline MST and the modified MST.

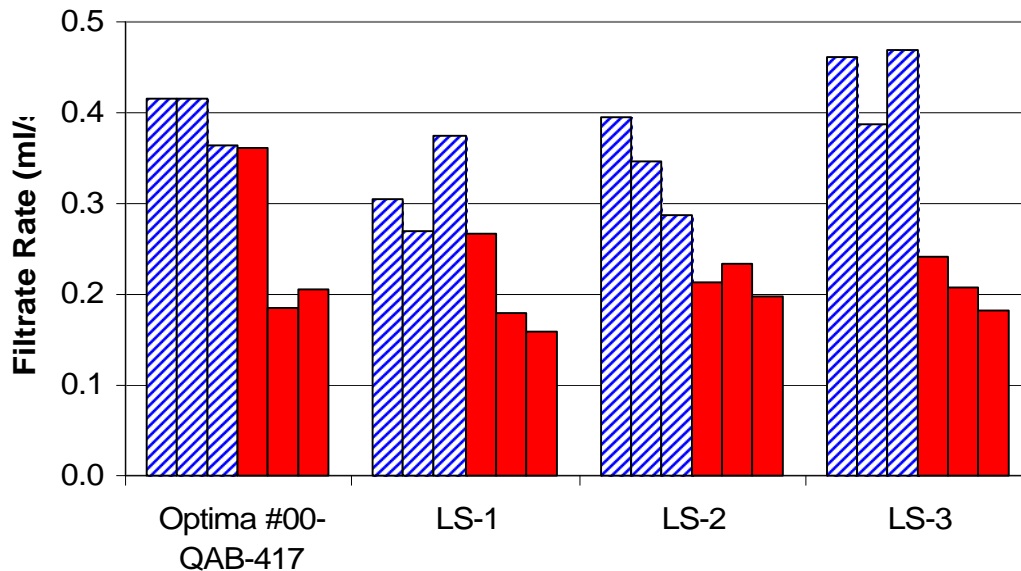


Figure 21. Filtration Rate with 0.1 micron Mott Media (each MST sample tested in triplicate in two different test sets)

Figure 22 shows the results with the 0.1 micron Pall media. The modified MST shows a lower filtration rate than the baseline MST for both sets of tests.

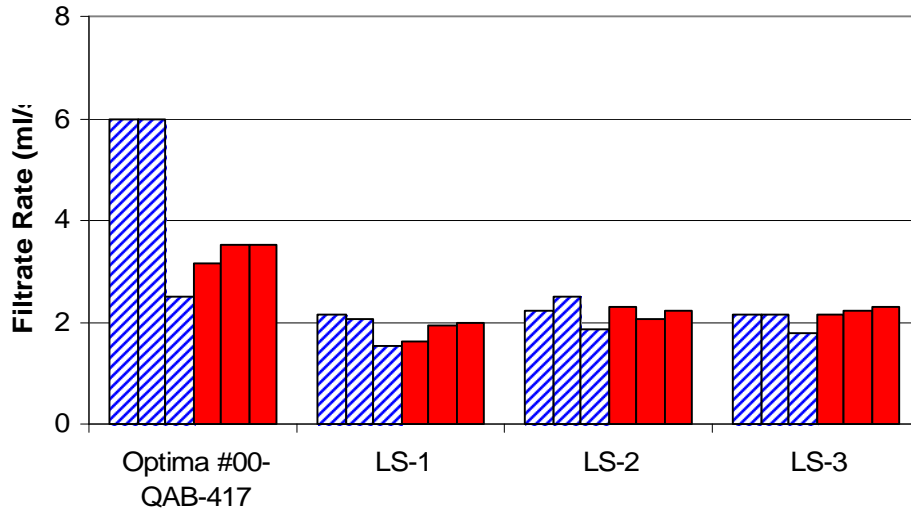


Figure 22. Filtration Rate with 0.1 micron Pall (ach MST sample tested in triplicate in two different test sets)

Figure 23 shows the results for the 0.5 micron Pall media. The modified MST shows a lower filtration rate than the Baseline MST.

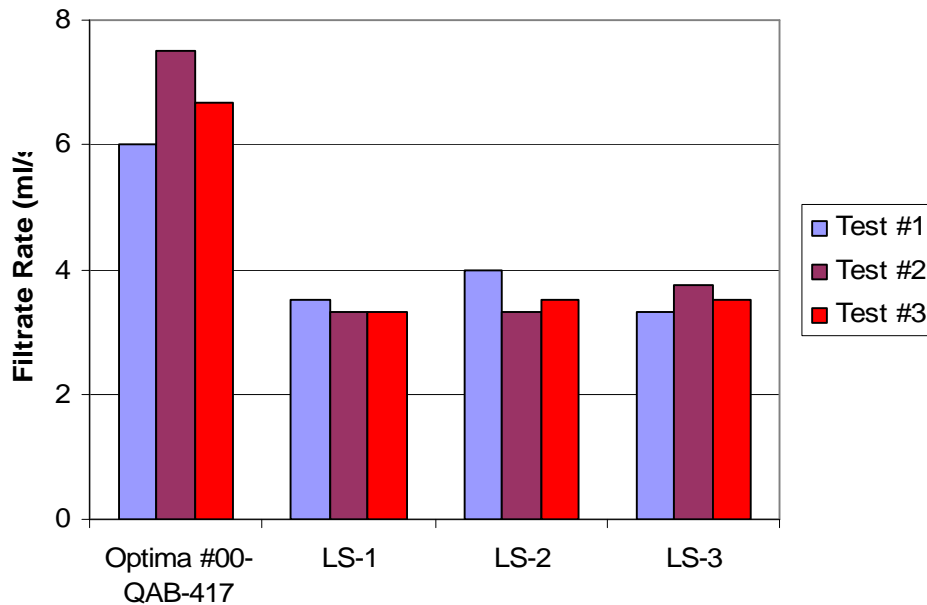


Figure 23. Filtration Rate with 0.5 micron Pall (each MST sample tested in triplicate)

The filter media with smaller absolute pore size (TruMem[®] and Mott) show no difference in the filtration rates between the baseline MST and the modified MST. The larger pore size media (Pall) show a decrease in filtrate rate with the modified MST. To determine whether there is a correlation between filter absolute pore size and filter ability of the modified MST, the authors plotted the normalized filtrate rate (optimized MST filtrate rate/baseline MST filtrate rate) as a function of absolute pore size in Figure 24.

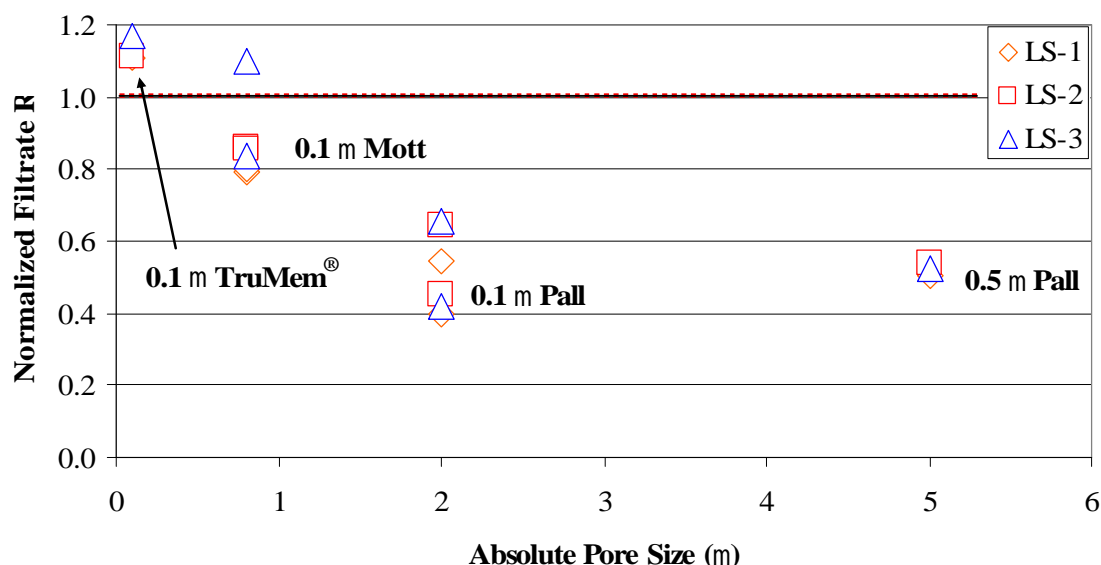


Figure 24. Normalized Filtration Rate of Modified MST as a Function of Absolute pore Size

Figure 24 shows a correlation between absolute pore size and normalized filtration rate of the optimized MST. When the absolute pore size is less than 1 μ , there is no significant difference in filtration rate. When the absolute pore size is greater than 1 μ , the modified MST filters more slowly than the baseline MST. This result suggest that the modified MST may have more fine particles than the baseline MST when dispersed in the simulated salt solution. Note that when dispersed in water for particle size measurement, the particle size data do not show a significant difference between the baseline and modified MST samples (see Figure 19).

4.4.3 Shelf-Life

This task measured the strontium and actinide removal performance of the modified MST samples, LS-1, LS-2 and LS-2, prepared by Method 3 at the 25-gram scale after storage for six months at ambient laboratory temperature (see Section 4.2 Task 2). We used the same simulant that we used when we first tested the performance of the modified MST samples. Prior to the test we added a small amount of ⁸⁵Sr radiotracer. This addition was necessary to bring the ⁸⁵Sr activity to a level similar to that when we tested the performance of the samples 6-months earlier. The addition of the ⁸⁵Sr radiotracer provides an insignificant increase in the strontium concentration of the simulant.

For these tests we limited the modified MST testing to a single sorbent concentration (0.2 g L^{-1}) in duplicate for each sample with sampling events at 6 and 12-hours. For the baseline MST sample we tested at both 0.2 and 0.4 g L^{-1} . Table 14 provides the average and standard deviation of the strontium, plutonium and neptunium DF values for the modified MST and baseline MST samples at both testing dates. Note, we did not test the performance of the baseline MST sample at 0.4 g L^{-1} at the initial time date. Figure 25 provides a plot of the plutonium concentration for the modified MST (average of 6 trials) and baseline MST (single trial) samples at both the initial test set and the set after 6-months of storage. Figure 26 provides a plot of the plutonium concentrations for each of the six trials at the 6 and 12-hour sampling times for the modified MST at both test dates.

Inspection of Table 14 indicates that the removal of strontium and neptunium was not altered upon storage of the modified MST for 6-months at ambient laboratory temperature. For strontium, we observed that the modified MST exhibited an average DF value 5 times greater than that of the baseline MST sample after 6 and 12-hours of contact at a 0.2 g L^{-1} sorbent concentration for both the initial and 6-month testing dates. Comparison of the modified MST results at 0.2 g L^{-1} with that of the baseline MST at the higher concentration of 0.4 g L^{-1} revealed that the modified MST exhibited a strontium DF value of 1.5 times that of the baseline MST.

Testing results indicated lower average plutonium DF values at the 6-hour and 12-hour sampling times after storing the modified MST samples for six months (see Table 14). At the 95% confidence level the range of plutonium DF values at the initial and 6-months dates overlap indicating the DF values are not statistically different. Thus, we cannot absolutely conclude that the plutonium removal performance decreased over the 6-month storage time. Inspection of Figure 19, which shows the individual data points for each trial, indicates that the 6-month plutonium concentrations are consistently higher than those measured immediately after synthesis with the exception of a single trial result in each data set. This trend suggests that the material has lost a small fraction of capacity. However, the plutonium removal remains quite high ($>98\%$ removal). For example, the plutonium DF values after 6-months measured between 23 and 43 times higher than the baseline MST added at 0.2 g L^{-1} and 13 to 23 times higher than the baseline MST added at 0.4 g L^{-1} . Thus, after 6-months of storage, the modified MST continues to provide excellent removal characteristics for strontium and actinides. We plan to conduct additional shelf-life tests as the approved work scope for the Phase II project includes measuring the performance of the modified MST samples after storing for 12 months.⁹

Table 10. Strontium, Plutonium and Neptunium DF Values for the Modified and Baseline MST Samples at the Initial Synthesis and 6-month Storage Times

	Strontium DF											
	Modified MST @ 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	
	<u>Average</u>	<u>Std Dev</u>	<u>Average</u>	<u>Std Dev</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>
Initial	1.13E+02	1.40E+01	1.28E+02	5.80E+00	8.50E+00	2.09E-01	6.88E+00	1.79E-01	nd	-	nd	-
6-months	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

	Plutonium DF											
	Modified MST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	<u>Average</u>	<u>Std Dev</u>	<u>Average</u>	<u>Std Dev</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>
	<u>Average</u>	<u>Std Dev</u>	<u>Average</u>	<u>Std Dev</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>
Initial	9.57E+01	1.76E+01	1.72E+02	6.16E+00	1.85E+00	1.27E-01	2.11E+00	1.63E-01	nd	-	nd	-
6-months	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

	Neptunium DF											
	Modified MST @ 0.2 g/L				Baseline MST @ 0.2 g/L				Baseline MST @ 0.4 g/L			
	<u>Average</u>	<u>Std Dev</u>	<u>Average</u>	<u>Std Dev</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>
	<u>Average</u>	<u>Std Dev</u>	<u>Average</u>	<u>Std Dev</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>	<u>Value</u>	<u>Uncertainty</u>
Initial	1.77E+00	2.52E-01	1.83E+00	1.55E-01	1.30E+00	8.21E-02	8.94E-01	7.11E-02	nd	-	nd	-
6-months	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	2.37E+00	1.80E-01

nd = not determined

Modified MST results are average and standard deviation of six trials

Baseline MST results are single determinations with reported analytical uncertainty

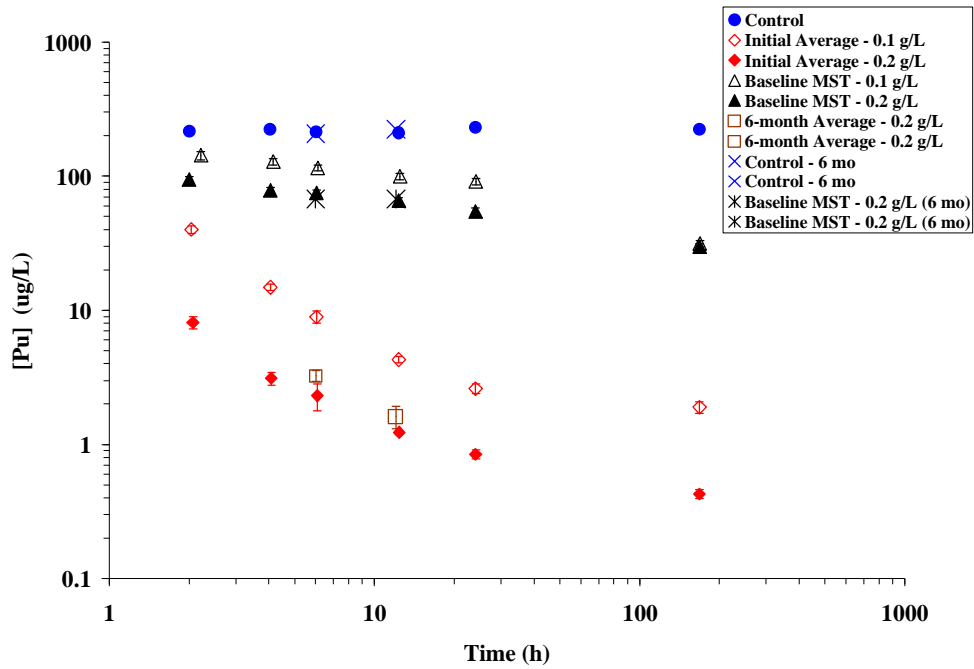


Figure 25. Plot of Average Plutonium Concentration versus Time for Tests with Modified and Baseline MST Samples at the Initial Synthesis and 6-month Storage Times

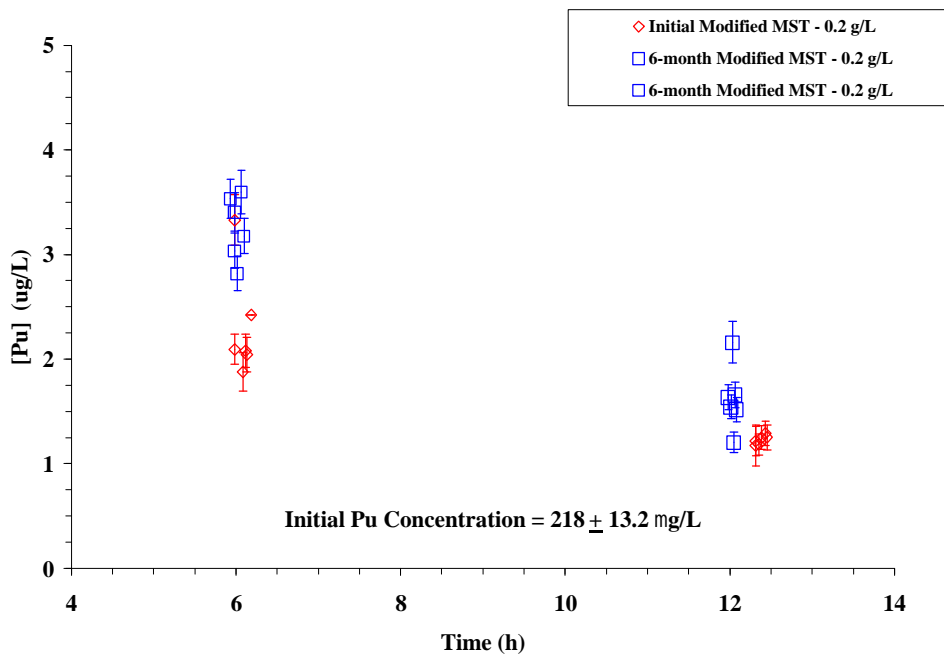


Figure 26. Plot of Individual Plutonium Concentration at the 6 and 12 Hours Sampling Times for Tests with Modified and Baseline MST Samples at the Initial Synthesis and 6-month Storage Times

5.0 Summary of Finding and Recommendations

This document provides an interim summary report of Phase II testing activities for the development of a modified MST that exhibits improved strontium and actinide removal characteristics compared to the baseline MST materials. The activities included determining the key synthesis conditions for preparation of the modified MST, preparation of the modified MST at a larger laboratory scale, demonstration of the strontium and actinide removal characteristics with actual tank waste supernate and characterization of the modified MST. Key findings and decisions include the following.

- Samples of the modified MST prepared by Method 2 and Method 3 exhibited the best combination of strontium and actinide removal.
- We selected Method 3 to scale up and test performance with actual waste solution.
- We successfully prepared three batches of the modified MST using the Method 3 procedure at a 25-gram scale.
- Performance tests indicated successful scale-up to the 25-gram scale with excellent performance and reproducibility among each of the three batches. For example, the plutonium decontamination factors (6-hour contact time) for the modified MST samples averaged 13 times higher than that of the baseline MST sample at half the sorbent concentration (0.2 g L^{-1} for modified MST versus 0.4 g L^{-1} for baseline MST).
- Performance tests with actual waste supernate demonstrated that the modified MST exhibited better strontium and plutonium removal performance than that of the baseline MST. For example, the decontamination factors for the modified MST measured 2.6 times higher for strontium and between 5.2 to 11 times higher for plutonium compared to the baseline MST sample. The modified MST did not exhibit improved neptunium removal performance over that of the baseline MST.
- Two strikes of the modified MST provided increased removal of strontium and actinides from actual waste compared to a single strike. The improved performance exhibited by the modified MST indicates that fewer strikes of the modified MST would be needed to successfully treat waste that contain very high activities of ^{90}Sr and alpha-emitting radionuclides compared to the baseline MST.
- Reuse tests with actual waste confirmed that partially loaded MST exhibits reduced removal of strontium and actinides when contacted with fresh waste.
- Samples of modified MST prepared by Method 3 and the baseline MST exhibited very similar particle size distributions.
- Dead-end filtration tests showed that the modified MST samples exhibited similar filtration characteristics as the baseline MST sample.
- Performance testing indicated no change in strontium and neptunium removal after storing the modified MST for 6-months at ambient temperature. The results suggested that plutonium removal performance may be decreased slightly after 6-months of storage. However, the change in plutonium removal is not statistically significant at the 95% confidence limit.

Based on these findings we recommend continued development of the modified MST as a replacement for the baseline MST for waste treatment facilities at the Savannah River Site.

6.0 Acknowledgements

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7.0 Reviews and Approvals

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8.0 Attachments

8.1 Decontamination Factors and Uncertainties for Modified MST Samples

Prepared by Method 1

<u>Test ID</u>	Strontium	Uncertainty Strontium	Plutonium	Uncertainty Plutonium	Neptunium	Uncertainty Neptunium
	<u>DF</u>	<u>DF</u>	<u>DF</u>	<u>DF</u>	<u>DF</u>	<u>DF</u>
1	0.85	0.032	1.48	0.13	2.15	0.46
2	1.03	0.044	3.05	0.40	4.46	0.93
3	1.40	0.060	0.60	0.078	1.14	0.17
4	1.85	0.081	1.22	0.16	2.00	0.31
5	0.92	0.036	1.56	0.15	2.15	0.46
6	1.30	0.051	1.75	0.15	3.60	0.87
7	1.59	0.069	1.20	0.15	2.39	0.45
8	1.06	0.041	1.00	0.091	2.44	0.51
9	1.22	0.052	1.03	0.13	2.58	0.42

8.2 Decontamination Factors and Uncertainties for Modified MST Samples

Prepared by Method 2

<u>Test ID</u>	Strontium	Uncertainty Strontium	Plutonium	Uncertainty Plutonium	Neptunium	Uncertainty Neptunium
	<u>DF</u>	<u>DF</u>	<u>DF</u>	<u>DF</u>	<u>DF</u>	<u>DF</u>
1A	4.64	0.204	1.25	0.11	0.91	0.18
2A	6.27	0.305	1.85	0.23	0.91	0.12
3A	1.98	0.082	1.59	0.143	1.59	0.32
4A	4.87	0.230	2.66	0.35	4.20	0.90
5A	6.25	0.305	4.16	0.53	3.46	0.62
6A	2.00	0.081	1.06	0.09	0.91	0.18
7A	0.74	0.031	1.61	0.20	>5.19	
8A	0.24	0.009	1.41	0.123	2.13	0.48
9A	2.14	0.086	1.34	0.12	1.06	0.21

8.3 Decontamination Factors and Uncertainties for Modified MST Samples Prepared by Method 3

Test ID	Strontium	Uncertainty Strontium	Plutonium	Uncertainty Plutonium	Neptunium	Uncertainty Neptunium
	DF	DF	DF	DF	DF	DF
1P	3.83	0.18	>21.5		1.59	0.46
2P	3.19	0.16	19.4	3.03	1.68	0.36
3P	3.22	0.16	24.1	5.29	1.75	0.39
4P	2.43	0.11	>8.2		1.48	0.44
5P	2.35	0.11	21.4	3.12	1.34	0.30
6P	2.32	0.10	>20.7		1.40	0.41
7P	1.78	0.080	16.5	2.42	1.51	0.33
8P	2.11	0.094	>6.5		4.75	1.62
9P	2.84	0.14	19.0	2.67	2.78	0.65
10P	4.08	0.21	>9.2		1.62	0.48
11P	3.01	0.15	16.7	2.64	1.25	0.27
12P	2.94	0.14	25.6	4.22	1.15	0.25
13P	4.69	0.24	>20.2		1.66	0.48
14P	3.52	0.18	31.3	4.57	1.86	0.42
15P	3.85	0.19	>22.0		1.24	0.36

Attachment 8.4

⁹⁰Sr, Pu and ²³⁷Np Concentrations Measured for Supplemental Actual Waste Tests

Test	Time (h)	Concentration (pCi/mL)							
		⁹⁰ Sr		Total Pu*		Total Pu**		²³⁷ Np	
Test S-2 (Restrike Test w/ 0.1 g/L MST)	2.1	3.20E+03	3.20E+02	1.80E+05	8.18E+03	3.21E+03	6.41E+02	6.50E+01	1.30E+01
	6.0	1.24E+03	1.73E+02	1.07E+05	5.10E+03	1.66E+03	3.32E+02	5.87E+01	1.17E+01
	8.0	7.84E+02	1.62E+02	3.83E+04	1.89E+03	< 1.76E+03	idl	4.87E+01	9.74E+00
	12.0	1.94E+03	2.85E+02	1.34E+04	6.93E+02	< 1.67E+03	idl	4.64E+01	9.28E+00
	30.0	3.29E+03	3.91E+02	4.47E+02	9.92E+01	< 1.65E+03	idl	4.11E+01	8.22E+00
	174	< 2.37E+02	mda	< 7.26E+03	mda	< 8.11E+02	idl	3.76E+01	7.52E+00
Test S-3 (Restrike Test w/ 0.2 g/L MST)	2.0	1.70E+03	2.04E+02	1.19E+05	6.19E+03	2.13E+03	4.27E+02	6.77E+01	1.35E+01
	6.0	5.57E+02	1.11E+02	2.92E+04	1.57E+03	< 1.81E+03	idl	6.06E+01	1.21E+01
	8.0	1.67E+03	2.80E+02	< 3.26E+03	mda	< 1.66E+03	idl	3.49E+01	6.98E+00
	12.0	1.15E+03	2.14E+02	8.65E+02	9.64E+01	< 1.72E+03	idl	3.25E+01	6.50E+00
	30.0	1.09E+03	1.90E+02	5.75E+03	5.15E+02	< 1.66E+03	idl	2.76E+01	5.52E+00
	174	2.06E+03	2.39E+02	< 9.91E+02	mda	< 8.17E+02	idl	1.96E+01	3.92E+00
Test S-1C (Control for Tests S-2 & S-3)	2.2	1.89E+04	1.89E+03	7.39E+05	3.41E+04	1.24E+04	2.48E+03	8.54E+01	1.71E+01
	8.0	2.06E+04	2.06E+03	7.72E+05	3.64E+04	1.27E+04	2.55E+03	9.41E+01	1.88E+01
	174	1.72E+04	1.74E+03	7.51E+05	3.92E+04	1.19E+04	2.38E+03	9.26E+01	1.85E+01
	Average	1.89E+04	1.89E+03	7.54E+05	3.66E+04	1.24E+04	2.47E+03	9.07E+01	1.81E+01
Test S-1A (Control for Tests S-4 & S-5)	1.9	2.24E+04	2.28E+03	7.82E+05	3.68E+04	1.23E+04	2.45E+03	9.17E+01	1.83E+01
	8.1	2.68E+04	3.48E+03	8.28E+05	4.65E+04	1.25E+04	2.49E+03	8.57E+01	1.71E+01
	9.3	2.54E+04	2.90E+03	7.65E+05	3.60E+04	1.31E+04	2.62E+03	8.92E+01	1.78E+01
	13.3	bad sample data		bad sample data		bad sample data		bad sample data	
	31.4	2.34E+04	2.67E+03	7.83E+05	3.77E+04	1.20E+04	2.40E+03	8.84E+01	1.77E+01
	175	2.15E+04	2.15E+03	9.32E+05	6.09E+04	1.11E+04	2.21E+03	8.30E+01	1.66E+01
	Average	2.39E+04	2.70E+03	8.18E+05	4.36E+04	1.22E+04	2.43E+03	8.76E+01	1.75E+01
Test S-1B (Control for Tests S-4 & S-5)	0.0	1.72E+03	2.72E+02	3.35E+04	1.96E+03	< 1.68E+03	idl	3.44E+01	6.88E+00
	2.1	9.08E+03	1.09E+03	2.90E+04	1.64E+03	< 1.98E+03	idl	4.14E+01	8.28E+00
	7.9	1.93E+03	2.50E+02	2.97E+04	1.50E+03	< 1.65E+03	idl	3.93E+01	7.86E+00
	9.5	3.98E+03	4.78E+02	3.22E+04	1.70E+03	< 1.69E+03	idl	4.01E+01	8.02E+00
	13.4	3.36E+04	3.57E+03	3.11E+04	1.66E+03	< 1.65E+03	idl	3.63E+01	7.27E+00
	31.4	1.59E+03	2.10E+02	2.80E+04	1.41E+03	< 1.63E+03	idl	4.32E+01	8.64E+00
	175	1.42E+03	1.98E+02	2.80E+04	1.39E+03	< 1.69E+03	idl	3.50E+01	7.01E+00
	Average	7.62E+03	8.66E+02	3.02E+04	1.61E+03	< 1.63E+03	idl	3.85E+01	7.71E+00
Test S-4 (Reuse Test w/ 0.1 g/L MST)	2.1	4.66E+02	9.31E+01	1.19E+04	6.90E+02	< 1.61E+03	idl	4.02E+01	8.04E+00
	6.8	1.56E+03	2.19E+02	3.98E+03	2.71E+02	< 1.59E+03	idl	2.73E+01	5.47E+00
	9.2	2.37E+04	2.49E+03	8.00E+05	4.17E+04	1.16E+04	2.32E+03	9.13E+01	1.83E+01
	13.0	6.64E+03	7.43E+02	6.24E+05	2.88E+04	9.22E+03	1.84E+03	7.41E+01	1.48E+01
	30.8	5.06E+03	5.82E+02	4.40E+05	2.17E+04	7.41E+03	1.48E+03	7.05E+01	1.41E+01
	175	1.09E+04	1.09E+03	2.48E+05	1.29E+04	3.18E+03	6.36E+02	5.98E+01	1.20E+01
Test S-5 (Reuse Test w/ 0.2 g/L MST)	2.1	8.83E+02	1.55E+02	< 5.48E+03	mda	< 1.61E+03	idl	2.81E+01	5.62E+00
	6.7	3.34E+03	4.54E+02	4.27E+03	2.72E+02	< 1.64E+03	idl	3.04E+01	6.08E+00
	9.0	5.82E+03	6.17E+02	6.46E+05	3.04E+04	1.05E+04	2.11E+03	8.33E+01	1.67E+01
	12.8	3.67E+03	4.15E+02	5.18E+05	2.50E+04	8.43E+03	1.69E+03	7.93E+01	1.59E+01
	30.6	2.20E+03	2.49E+02	1.63E+05	9.23E+03	2.86E+03	5.72E+02	6.09E+01	1.22E+01
	175	7.99E+02	1.28E+02	5.40E+04	2.81E+03	< 1.69E+03	idl	4.84E+01	9.68E+00

*Sum of PuTTA data for ²³⁸Pu and ^{239/240}Pu**ICP-MS data for ²³⁹Pu only. The mass of ²⁴⁰Pu was below the detection limit for all samples.

mda signifies the minimum detectable analysis of a isotope

idl indicates the mass was below the instrument detection limit.

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