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In Situ Removal of Actinides and Strontium from High Level Waste Tanks: Tea Bag versus Adsorption Column

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Summary

Initiatives are underway at the Savannah River Site (SRS) to accelerate the disposition of the supernate and salt portions of the waste in the SRS High Level Waste (HLW) tank farm system. Significant savings in processing time and overall cost could be achieved by in situ treatment of waste supernate or dissolved salt inside a tank farm waste tank. For treatment of actinides and strontium in waste, the baseline method is sorption onto monosodium titanate (MST), an engineered powder with mean particle size of ~10 microns. In a separate study at the Savannah River National Laboratory (SRNL), engineered forms of MST were developed and compared on a small (250-mL) scale in batch tests. In the current study, a promising form of engineered MST was tested under two conditions: a traditional ion exchange (or adsorption) column and a porous, flow-through device called a "tea bag", immersed in solution. Both tests used the same amount of engineered MST to treat 10 L of simulated waste solution containing plutonium (108 μg/L) and strontium (710 μg/L).

In the column test, engineered MST succeeded in treating 2900 bed volumes (BV) of simulated salt waste solution. There was no significant strontium (Sr) breakthrough and only 7% plutonium (Pu) breakthrough at the end of the run which operated at 5.3 BV/hour and lasted 23 days. Stated another way, the column of engineered MST achieved an average decontamination factor (DF) of 70 for Sr and a Pu DF ranging from 140 initially to 15. In the tea bag test, activity levels for Sr and Pu were reduced by 82% and 80%, respectively, in four weeks, though most of the Sr removal occurred during the first week. The tea bag DFs were 5.5 for Sr and 4.9 for Pu.

Estimates are available for the DFs required to meet Z-Area limits. For the average waste concentration, a Sr DF of 4.5 and a Pu DF of 12 are needed. Based on the performance of the engineered MST in the column tests, this treatment approach would be sufficient to treat much of the SRS waste. The tea bag method, however, would be limited to wastes that have Pu activities below the average composition or used in tandem with other treatment methods for wastes with the average Pu waste composition or higher.

Though tea bag and column equipment could be deployed in a large tank, the column approach is more attractive for several reasons: ability to meet decontamination requirements at higher alpha activity, no observed fines generation, and relative maturity of column-related technology. Another important advantage to the column approach is that it enhances loading because the adsorbent is in equilibrium with feed adsorbate levels which are higher than product levels. Enhanced loading yields more efficient use of MST, reducing the amount of MST solids that transfer downstream for vitrification.

We scaled the results of bench-scale tests for deployment in a million-gallon waste tank. Using a 70-gal column operating in once-through mode, we would expect a DF of 70 for Sr and about 30 for Pu for the treatment of the initial 200,000 gal in a period of 23 days. We recommend further studies with engineered MST to establish both its adsorption capacity and kinetics. Kinetic data may confirm initial work showing that higher flow rates are feasible, and capacity data would allow projections of the amount of sorbent needed to treat an entire tank of HLW solution for ^{90}Sr and alpha activity removal.

Introduction

Approximately 140 million liters of high-level nuclear wastes (HLW) are presently stored in 49 underground carbon steel tanks at the Savannah River Site (SRS). About 9% (11 million liters) of the waste consists of precipitated solids, referred to as sludge, that contains about 60% of the radioactivity and settles to the bottom of the HLW storage tanks. The remaining volume of HLW is stored as concentrated salt solution and saltcake produced from evaporation of the waste solutions. The liquid/saltcake fraction of the HLW contains about 40% of the radioactivity and is comprised of principally cesium isotopes (^{134}Cs and ^{137}Cs), smaller amounts of strontium-90 (^{90}Sr), and alpha-emitting isotopes of uranium (U), plutonium (Pu), neptunium (Np), and other actinide elements.[1] Based on SRS HLW tank concentrations and current disposal limits,[2] plutonium is the key actinide.

The basic approach for disposition of this waste is to concentrate the radioactive components from the bulk wastes into a small volume fraction that is then vitrified in the Defense Waste Processing Facility (DWPF). For the saltcake (which is to be dissolved) and salt solution, two treatment steps are planned: removal of Sr and alpha-emitting isotopes of Pu and Np, and removal of Cs. After these treatments, the separated radioactive components would be transferred into the DWPF for vitrification with the sludge fraction of the HLW. The decontaminated liquid waste would be transferred into the Saltstone Facility for incorporation into a cement wasteform.

Disposition of the large volume of HLW will be a multi-year effort. To accelerate HLW disposition, in-tank or in-situ equipment, such as ion exchange columns or “tea bags” could be used. In-tank deployment would allow treatment of multiple tanks simultaneously, and would take advantage of the long treatment time available. In addition, in situ treatment would enable sending more salt to Saltstone without first requiring the actinide processing unit operation step in the Actinide Removal Process (ARP) or Salt Waste Processing Facility (SWPF) projects. Because of these potential advantages, the Savannah River National Laboratory (SRNL) Independent Research and Development program sponsored research for the in situ treatment of actinides. The column method tested in this study for removal of Pu and Sr is a continuous type process which differs from the batch treatment processes that are the current baseline for the ARP and the SWPF.

Previous Work

In the 1960’s and 1970’s, the Savannah River Plant (now SRS) used zeolite columns, called Cesium Removal Columns (CRC), to remove radioactive Cs from evaporator overheads and tank waste.[3] Elsewhere, in-tank zeolite ion exchange columns were also used at the West Valley Demonstration Project during 1988-1990 to process 2.34 million liters of supernate containing radioactive Cs.[4,5] In the early 1990’s, the SRNL, known then as the Savannah River Technology Center (SRTC), developed and installed in-tank ion exchange columns in two of its own waste tanks (F and K). Columns were filled with

Duolite GT-73 resin and provided sufficient mercury decontamination to render the waste non-hazardous and allow shipment to an evaporator.[6] The columns were successful for years.

In related work, a skid-mounted ion exchange system was successfully demonstrated at Oak Ridge National Laboratory (ORNL) in 1997 for removing Cs from ~30,000 gallons of Melton Valley Storage Tank supernate.[7] That demonstration led to the use of a modified system to treat more than 215,000 gallons of supernate from 1997 to 2000 in 14 operational campaigns.[8] The success of the ORNL work led to development efforts aimed at Cs treatment of SRS HLW.[9] As for tea bags, a journal article[10] published near the end of the current study showed that others have considered the bag concept for treatment of fission products.

The current SRNL study examines the treatment of actinides and Sr by adsorption - processes that could be installed inside tanks. The work was made more feasible by the recent development of an engineered sorbent material described later. To minimize analytical costs, Pu was the only actinide used in this study since it is the most important actinide in the SRS HLW system.

Experimental

Simulated Waste Solution

A 20 L batch of caustic simulated waste solution was made with the target concentrations shown in Table 1, which total 5.6M sodium (Na) ions. This recipe was developed by Hobbs.[11] Analysis of the solution by ICP-Emission Spectroscopy (ICPES) yielded results shown in Table 2. Note in Table 2 that the measured values for Na, aluminum (Al), and sulfur (S) are very close to the targeted values.

Table 1. Target Composition of Simulated Waste Solution

Component	Concentration (M)
Free NaOH	1.33
Total $NaNO_3$	2.60
$NaAl(OH)_4$	0.429
$NaNO_2$	0.134
Na_2SO_4	0.521
Na_2CO_3	0.026
Total Na	5.59

Table 2. Measured Non-radioactive Components of Simulated Waste Solution

Analyte	Feed Concentration	
	(mg/L)	(M)
Al	11,100	0.411
Cr	2.19	---
Fe	1.00	---
Na	127,000	5.52
S	16,400	0.511
Sb	30.3	---
Sn	15.7	---
Sr	0.72	---

During formulation of this simulant, Pu was added at a target concentration of 200 μg/L. Periodic samples were taken over weeks to determine stability. Initially, Pu content was determined by a gross alpha count of a dried plate of solution. This screening method had high uncertainties due to the high salt content of the solution. Nevertheless, it confirmed that plutonium was present. Later, Pu was measured by a spiked extraction method termed Pu-TTA or by liquid scintillation counting. These two methods have much lower uncertainties, typically 6% at the 1-sigma or 1-standard deviation level. Note that starting with the 14-week sample, all Pu/alpha samples (3.00 mL each) were acidified with 3.00 mL of 5 M nitric acid. Acidic samples are more conducive to the analytical methods used for Pu or alpha activity determinations. A final feed sample was taken before the simulant solution (20 L) was split into 10 L batches for the two tests.

Before testing, ^{85}Sr radiotracer was added to the simulant at an initial value of 122 counts/min/mL. Measurement of ^{85}Sr throughout this study was by a low-temperature germanium detector using count times of 10 minutes and samples of 3.00 mL. Over the course of this study, the average value of ^{85}Sr in 16 blank samples was 0.8 counts/min/mL (cpm/mL) and the highest blank reading was 1.33. The half-life of ^{85}Sr and the time between sampling and counting were used to adjust counting results to reflect activity at the time of sampling. Note that the total Sr in the solution, 0.72 mg/L, is appreciable, and the amount of ^{85}Sr removed corresponds directly to the amount of total Sr removed. Also, note that the ICPES detection limit for total Sr in this aqueous solution was 0.16 mg/L.

Tea Bag with Sorbent

The current baseline material for treatment of actinides and Sr in SRS HLW is monosodium titanate (MST). Several attempts were made in this study to construct a porous, flow-through device, an engineered “tea bag”, filled with MST powder. However, because the particle size range of MST is nominally 0.5-35 μm, it was difficult to construct a device that both promoted flow and contained the MST powder.

To resolve this problem, a newly developed form of MST, referred to as engineered MST, was used. Specifically, the engineered MST material consists of beads containing 50 wt% MST in a titanium oxide matrix. The beads were made at ORNL[12,13] through an internal gelation process. This form of engineered MST compared favorably to other forms of aggregated MST, which were prepared and tested for performance in small-scale batch testing with simulated and real waste.[14]

For the current study, the beads were sieved, passing through a #30 sieve and retained on a #60 sieve. Hence, the dry particle size range was 250 – 595 microns. The beads were loaded into a tea bag made of stainless steel wire cloth (70 x 70 mesh) with 165-μm openings. The loaded tea bag was soaked in a non-radioactive, Sr-free caustic waste simulant for 16 days prior to use. The soaking solution was made from the same base recipe as the spiked simulant used for testing. A minimum of three to five days of soaking is recommended. Longer times were used in this study due to equipment delays.

Figure 1 shows the tea bag in operation. Once testing began, samples were taken after 2, 4, 24, 168, 336, and 672 hours. The 168-hr (or 7-day) sample was analyzed for ^{85}Sr but not Pu. During tea bag testing, mild agitation of the bulk (10 L) solution was provided by a magnetic stirrer.

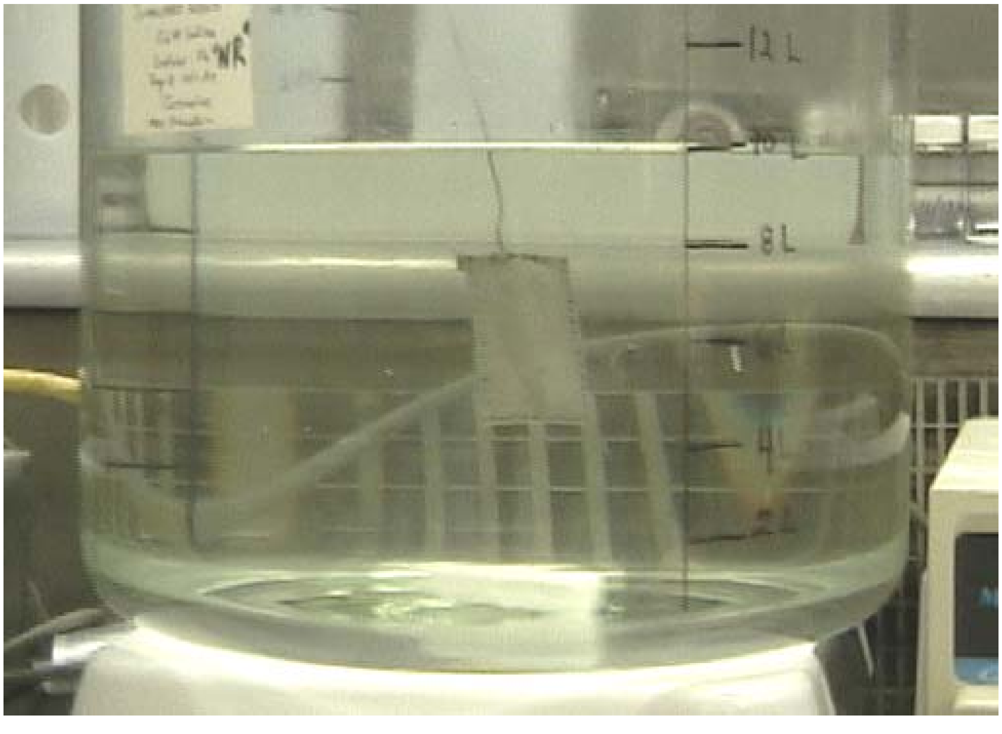

Figure 1. Tea bag with MST beads in 10 L of simulated waste solution.

Adsorption Column

An adsorption (or ion exchange) column was used for once-through treatment of the other 10 L of spiked simulated waste solution. The column test was operated continuously (i.e., overnight). The column, 0.833 cm in diameter, was loaded with 1.86 g of ORNL 50 wt% MST beads that had soaked for 22 days in the non-radioactive Sr-free simulant. A liquid head of about 5 cm (i.e., 8.2 mL) was maintained above the beads. After loading, it was observed that the beads occupied almost twice the volume that was anticipated given their dry bed density (lightly tapped) of 0.991 g/mL. Based on column graduations and diameter, the 1.86 g of beads occupied 3.38 cm^3, yielding a wet density of 0.55 g beads/mL. A related study[15] reports that the "swelling" of these beads occurs within the first 2 days (or less) of soaking in caustic salt solution, and no change in size is observed thereafter. In the current study, the size of the beads did not change once loaded into the column. The cause for the increased size of the beads in caustic solution is not known, but further investigation is recommended. The length-to-diameter (L/d) ratio of the loaded column was 7.2. During testing, the spiked simulant was pumped through the column at 0.3 mL/min, which converts to 5.3 bed volumes (BV)/hour. The column was jacketed and the circulating water through the jacket was kept at 25±1°C.

A photograph of the column during operation is shown in Figure 2. Most of the MST beads are within a thin glass insert, which was installed so that the L/d ratio would be greater than one. The unexpected swelling of the beads before loading resulted in a taller than expected bed. In fact, at the top of the bed, the MST beads exceed the height of the insert and occupy the full diameter (1.45 cm) of the column. Near the top of the insert, the beads appear slightly colored. This is likely due to both the lighting effects of the photo and an initial color change in the beads, since the photo was taken after processing began. Color change of the beads is discussed in the Results section of this report.

Figure 2. Column loaded with ORNL 50 wt% MST beads.

Results

Plutonium Content in Feed

Table 3 shows the plutonium content in the simulant prior to and during testing. The final feed sample (simulant age: 16 wks) was obtained within 24 hours of starting the tea bag test. A feed concentration of 108 μg Pu/L was determined by averaging the three feed values and the first two samples taken after testing began. Test results also showed that the plutonium was predominantly $^{239/240}$Pu. ^{238}Pu contributed only 2% of the Pu activity (and 0.01% of the Pu mass).

Table 3. Plutonium Content Before and During Tea Bag Test

Sample	Plutonium μg/L	% Uncertainty (1-sigma)
Feed @ 12 wks	95	5
Feed @ 14 wks	97	5
Feed @ 16 wks	120	7
Time after Tea Bag Addition	----	--
2 hr	108	6
4 hr	119	6
24 hr	101	6
336 hr (14 days)	72	3
672 hr (28 days)	22	5

Tea Bag Test

The tea bag containing engineered MST beads removed both Sr and Pu from the spiked simulated waste solution. For Pu removal, results are shown in Figure 3, where the initial Pu concentration represents the average value described above. After 28 days, 80% of the Pu had been removed.

At the beginning of the tea bag test, a small amount of fines was released into the bulk solution and settled to the bottom of the reservoir. The slow stir rate of the bulk solution did not generate additional fines during the 4-week test. After the test was complete, agitation of the bulk solution was increased and more fines were observed coming from the tea bag into the reservoir. Since the sorbent beads were constrained within the tea bag, fines generation is attributed to the shearing action of fluid motion, not to fluidization/collision of the beads.

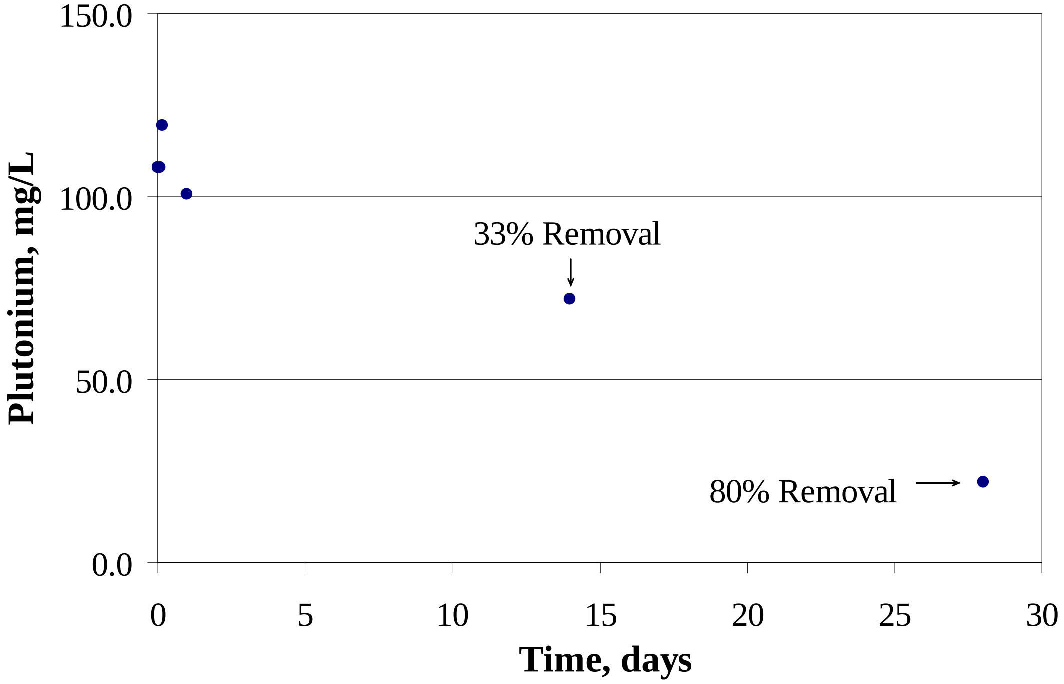

Figure 3. Removal of plutonium by tea bag with ORNL 50 wt% MST beads.

For ^{85}Sr removal, results are shown in Figure 4. The control sample represents analysis of the simulated waste solution that was not contacted with the MST beads. The gradual decline in the control values represents the natural decay of ^{85}Sr. Recall that a significant amount of non-radioactive Sr (0.71 mg/L) was present in pre-test solutions. The ^{85}Sr activity in the tank is indicative of total Sr present. As mentioned before, all ^{85}Sr values represent activity at time of sampling. Measured values are time-adjusted based on the natural decay of ^{85}Sr and the time between sampling and analysis.

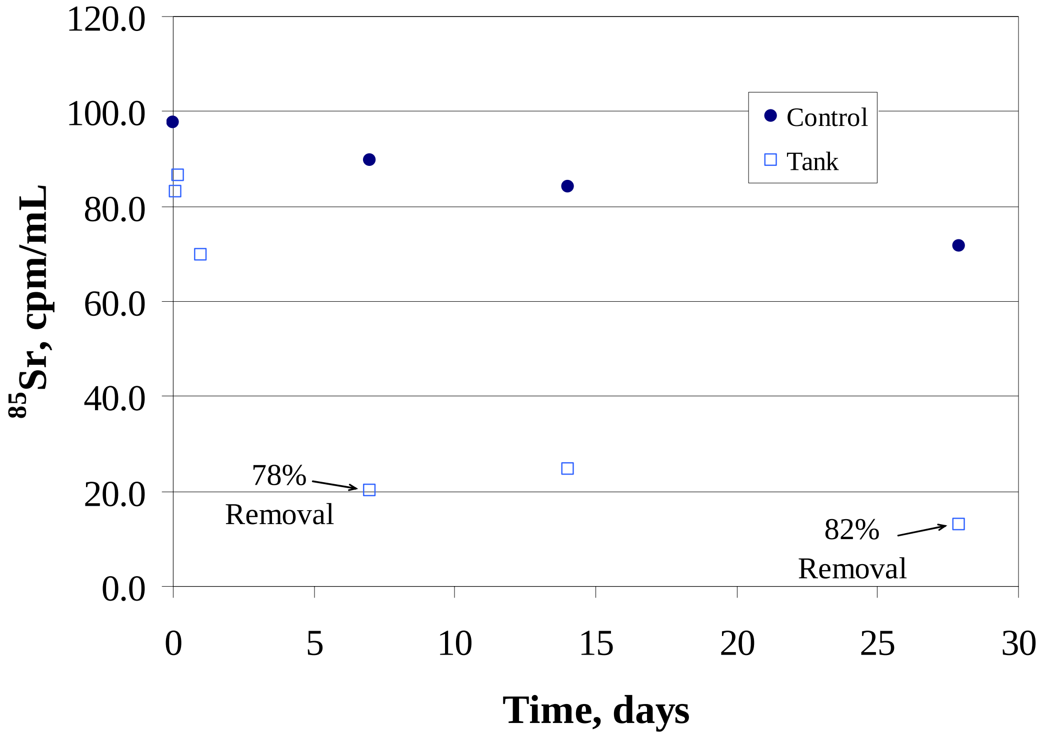

Figure 4. Removal of strontium by tea bag with ORNL 50 wt% MST beads.

Sorption Column Test

The sorption column loaded with ORNL 50 wt% MST beads removed Pu and ^{85}Sr as shown in Figures 5 and 6. The flow rate through the column was 5.3 BV/hr and the column was maintained at 25±1°C. The column operated continuously for 23 days without any observed problems.

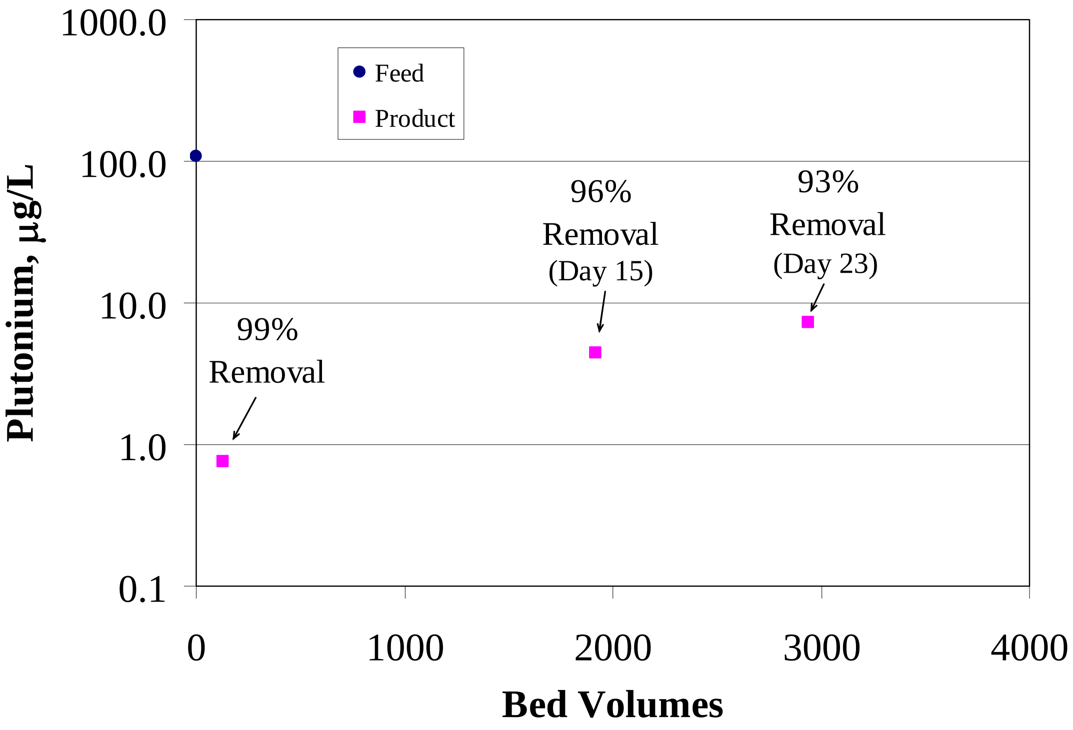

Figure 5. Removal of plutonium by sorption column of ORNL 50 wt% MST beads.

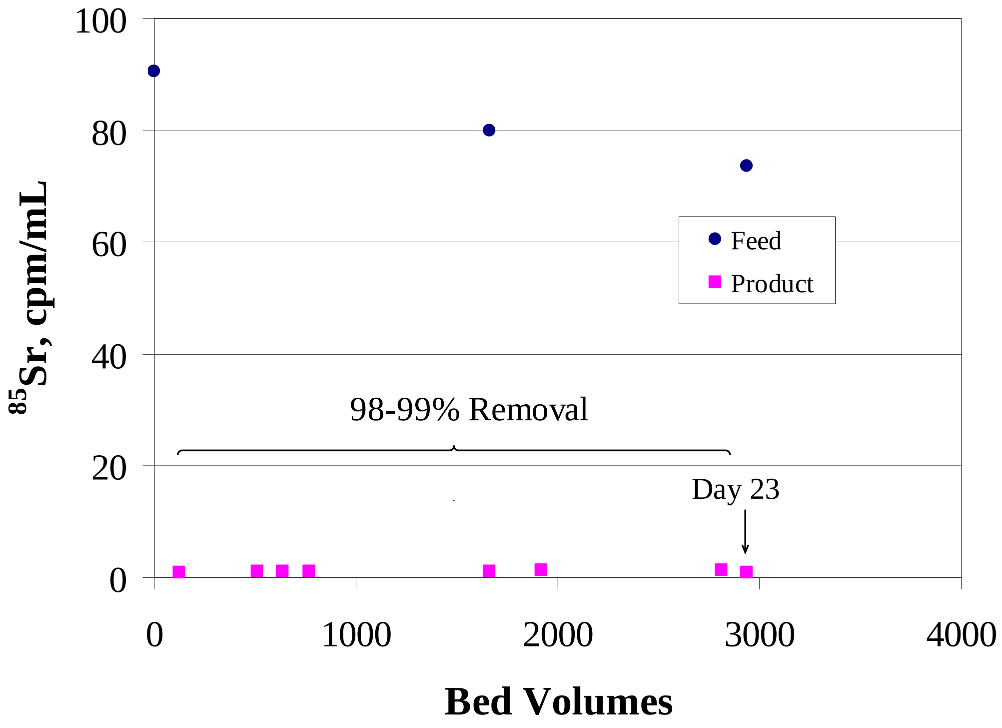

Figure 6. Removal of strontium by sorption column of ORNL 50 wt% MST beads.

On the eighth day of testing (after 1000 BV), the MST beads in the top portion of the sorption column had become a light tan color. The coloration increased through the remainder of the test. A photograph of the column at the end of testing is shown in Figure 7. The color may be caused by Pu or by another trace metal, such as iron, which was present in the feed at 1 mg/L. For future work, we recommend post-test digestion of the sorbent followed by chemical analyses to identify the source of the color.

Figure 7. Column of MST beads after treating 2900 bed volumes of simulant.

Discussion

Both the column and the tea bag methods were effective at removing Pu and Sr from 10 L of simulated waste solution. The column method removed essentially all of the Sr and about 95% of the Pu in 23 days. The tea bag removed 82% of the Sr and 80% of the Pu in 28 days. Thus, for treating the same amount of simulant in a similar time-frame, the column provided more complete removal. With the tea bag, a minor loss of fines into the bulk solution was observed. Fines released into the bulk solution still adsorb Pu and Sr but are no longer removable via the tea bag. For large-scale application, adsorbent beads would need to be more resistant to shedding or be used with minimal agitation. During the column test, fines were not observed in the treated solution.

For a treatment method to be successful, the treated stream must meet certain activity limits, e.g., <18 nCi/g of total alpha activity. To convert this limit to concentration, the ratio of different isotopes must be known. Since Pu concentration limits are very sensitive to the prevalence of ^{238}Pu, we chose to discuss results in terms of decontamination factor (DF), which is the ratio of the feed concentration divided by the product concentration.

For treated waste to meet Z-Area (Saltstone) limits for low-level disposal, the required Sr DF ranges from 4.8 for the average waste concentration to 28 for the bounding waste concentration. These factors are based on waste solution at 6.44M Na. In the current study, the column with ORNL 50 wt% MST demonstrated Sr DFs ranging from 54 to 80 in eight samples taken over 23 days for the 5.6M Na simulant. So, a column of engineered MST would treat at least 2900 BV sufficiently to meet the ^{90}Sr Saltstone criterion, even at the bounding condition. Similarly, the Pu DF required to meet the Z-area limit ranges from 12 for the average waste concentration to 55 for the bounding waste concentration. The lowest Pu DF for the column was 15, indicating that the column is also able to treat 2900 BV of average waste to meet the Saltstone Pu limit. Further work is needed to determine if a column of engineered MST could treat waste at the bounding Pu concentration.

In comparison, the tea bag method achieved a Sr DF of 5.5 and a Pu DF of 4.9 after 28 days. Thus, for average waste concentrations, the tea bag tested would provide sufficient Sr removal but insufficient Pu removal. It is possible that tea bags with a higher mass of MST per volume of waste, or tea bags used for longer periods of time may be able to provide sufficient removal to meet Saltstone Pu limits. Nevertheless, the column method provides better removal for similar time-frames and sorbent amounts.

To scale up the tea bag to a large, million-gallon tank, some equipment development would be needed to ensure good contact between the sorbent beads and the waste supernate. However, the column method is relatively easy to scale up. An advantage of column scale-up is that if the BV/hr flow rate and length-to-diameter ratio are kept constant, the superficial velocity increases with the cube root of the volume increase. With a higher velocity at the same BV/hr flow rate, the kinetics of adsorption are as good or better in the larger column.

In the column test for the current study, the 50 wt% MST beads were loaded at 0.067 wt% Pu on average but were clearly not fully loaded since no significant breakthrough had occurred. The observed loading would double to 0.13 wt% Pu on an MST-only basis, which is still lower than the previously reported maximum loading of Pu on MST[16] (0.28 wt%). For comparison, the current study employed a phase ratio of about 5,300 mL solution/g sorbent while the 0.28 wt% Pu value was from a batch study at 20,000 mL/g MST. Recently, a separate study showed that for a batch contact, increasing the phase ratio from 2500 to 25,000 mL/g MST increased the Pu loading almost ten-fold, to a value of 0.25 wt% Pu. That study recommended tests at even higher phase ratios to establish the full capacity of MST. High phase ratios are needed because actinides and strontium are sparingly soluble in caustic high sodium solutions. The authors estimate that a phase ratio of 1×10^6 mL waste/g sorbent may be needed to determine the maximum capacity of engineered MST.

In-Tank Deployment of Engineered MST

To apply the bench-scale column results for an in-tank treatment process, we believe the deployment would be similar to that used for treating evaporator overheads (i.e., Cesium Removal Columns (CRC)).[17,18] A CRC-like column would fit through a HLW tank riser.

A column prefilter would be needed to remove suspended solids, which could foul the MST column. The following scenario is for once-through treatment of waste supernate and uses a conservative estimate of 70 gal for the CRC column volume.

Given: supernate volume = 1 x 10^6 gal = 3.785 x 10^6 L
Given: plutonium concentration = 100 µg/L.
Thus, the tank contains 380 g of plutonium

Given: 1 CRC column: ~70 gal (22 in diam., 42 in. tall plus bottom cone)
Measured wet density of 50 wt% MST beads: 0.55 kg/L.
Thus, one column would contain 70 * 3.785 * 0.55 = 150 kg of beads.

Scaling up the results of lab tests without extrapolating to more bed volumes,
2,900 Bed volumes = 2,900*(70 gal) = 200,000 gal.
Avg. removal = 96%.
Thus, 200,000 gal* 3.785 * 100 x 10^{-6} g Pu/L * 0.96 = 73 g Pu removed.

Treating 2,900 bed volumes of the assumed column converts to about 20% of the tank volume. If the column were operated at 5.3 BV/hr, as in the lab test, the flow rate would be 6.2 gal/min and the first 200,000 gal would be treated in 23 days.

In the lab test, the sorbent was not fully loaded after 2,900 bed volumes. Thus, it is expected that a 70-gal column would effectively treat more than 200,000 gal. Capacity measurements are needed to establish the effective volume of waste that can be treated by an MST column. Kinetic studies are also needed, especially at higher flow rates. In a related study yet to be issued, two engineered MST samples showed DFs similar to the current study in column tests operated at nominally 15 BV/hr, which is almost three times faster than used in this study. Accurate capacity and kinetic data would allow for the determination of the total sorbent needed as well as the best flow rate for particular applications.

Conclusions

The results of this study lead to the following conclusions:

1. Both tea bag and column methods using ORNL 50 wt% MST beads were effective at removing plutonium and strontium from a simulated waste solution.
2. The column method proved more effective than the tea bag method in removing Sr and Pu.
3. The column method removes enough Sr and Pu to meet Saltstone limits.
4. The tea bag method may not be effective in removing sufficient amount of Pu to meet the Saltstone limit for alpha activity.
5. The column test demonstrated that for ORNL 50 wt% MST beads, no significant breakthrough of Pu or Sr had occurred at the conclusion of the test, indicating that the sorbent was not fully loaded.

6. Some shedding of fines from the MST beads was observed in the tea bag test.
7. It is plausible that a column of engineered MST deployed inside a waste tank would provide actinide and strontium treatment of at least 200,000 gallons of waste supernate in 23 days.

Future Work

The observations in this study lead to the following recommendations for future research:

1. Conduct experiments to determine the maximum adsorption capacity of ORNL 50 wt% MST for actinides and strontium.
2. Conduct sorption tests at higher flow rates, in terms or bed volumes/hour, to understand sorption kinetics and provide for optimization of full-scale operations.
3. Investigate the cause of swelling of ORNL 50 wt% MST beads in caustic solution and verify that it would not be problematic for large-scale operations.
4. Conduct post-test chemical analyses of the sorbent to determine cause of coloration and confirm adsorption data.

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