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ENGINEERING MONOSODIUM TITANATE FOR ADSORPTION COLUMN PROCESSES

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ABSTRACT

Monosodium titanate (MST) is an inorganic adsorbent powder that effectively removes strontium, plutonium, neptunium, and other trace elements from alkaline high-level waste (HLW) supernate. This work tested one commercial titanate and four general methods to engineer MST into particles large enough to use in adsorption columns. The most successful of the engineered products selected from batch contact and chemical stability testing succeeded in treating 2900 bed volumes (BV) of simulated salt waste containing dissolved plutonium and strontium. There was no detectable strontium breakthrough and only 6% plutonium breakthrough – well within the processing goal – at the end of the demonstration which operated at 5.3 BV/hour. Additional column tests at nominally 15 BV/hr demonstrated similar removal performance.

Batch testing of adsorbents used both actual Savannah River Site (SRS) tank supernate as well as simulated salt solutions spiked with strontium, neptunium, and plutonium. In tank waste tests, internal gelation beads produced by the Oak Ridge National Laboratory (ORNL) demonstrated a batch distribution coefficient of 35,000 +/- 4,000 mL/g for plutonium at a phase ratio of 1970 mL/g. In the same batch the sorbent demonstrated a batch distribution coefficient of 99,000 +/- 7,500 mL/g for strontium. These results indicate that this material should be able to process thousands of bed volumes of SRS salt waste before column breakthrough.

INTRODUCTION

Current plans at the Savannah River Site (SRS) use MST in batch contact processes to remove ⁹⁰Sr and actinides from HLW supernates. Engineering MST into nominally 500 micron particles would allow steady column processing of salt solution. Benefits of column operation include enhanced loading due to equilibration with feed adsorbate levels versus product levels, and a smaller facility footprint relative to that of a batch contact tank. Since MST is considered non-elutable its deployment is simplified over a column needing elution with, perhaps, acid. In addition, a column could be deployed inside a HLW tank reducing the required capital investment.

MST features particles of nominally 0.5-35 micron in size, which are much too fine for use in an adsorption column. An extensive review of the literature and consultation with technical experts identified candidate methods to produce engineered forms of MST. Specific criteria for the candidate materials included freedom from organic content, 30 x 60 mesh particle size,

effective porosity, toleration of high sodium and alkaline solutions, and minimal fines generation. Researchers selected five candidate methods for further study. Laboratory syntheses produced representative samples for characterization and performance testing. Testing identified two suitable methods. The two methods include internal gelation, which is a patented technology of Oak Ridge National Laboratory (ORNL), and internal hydrolysis, a method in which the MST is produced within a porous substrate. A commercial sodium titanate, SrTreat[®], produced by Fortum Engineering (Finland), demonstrated good performance as well although plutonium (Pu) removal kinetics appeared much slower than observed for the other engineered MST materials.

EXPERIMENTAL

Particle Engineering of Monosodium Titanate

Monosodium titanate (empirical formula of $\text{NaTi}_2\text{O}_5\text{H}$) was first prepared by Lynch, et al. [1]. The preparation method consists of mixing a methanol solution of NaOH with a methanol solution of titanium isopropoxide. The MST is then precipitated by addition of water. The white MST solids are amorphous. This compound was found to exhibit high selectivities for strontium and several actinides [2]. Subsequently, much work has been reported on strontium uptake by similar materials prepared by treatment of hydrated TiO_2 with NaOH [3,4,5].

Commercially Available Baseline Materials

One batch of commercially produced MST (Optima batch 00-QAB-417) was used as a baseline for batch testing and portions were also provided to all investigators for the production of engineered MST samples that initiate with MST solids. MST from the same container was thus used at Savannah River National Laboratory (SRNL); UOP, Inc.; ORNL; and Clemson.

A sample of SrTreat[®] was obtained from Fortum Engineering (Fortum, Finland). The sample was stated to have the commercially available particle size range of 300 to 850 microns. The particle shape is non-spherical and granular.

UOP, Inc. Samples

UOP, Inc. pelletized MST by proprietary methods. The binder included zirconium oxide. One sample was of spherical beads within the requested 30 x 60 mesh size range. The other sample was of elongated pellets in what UOP called their “trilobe” shape. UOP, Inc. indicated that each sample contained 80 wt % MST. Titanium analyses at SRNL confirmed the MST content within +/-2 wt %.

ORNL Samples

ORNL provided two samples of engineered MST produced by the internal gelation method. These spherical materials were reported to contain 32 and 50 wt % MST. Actual MST content could not be measured by titanium content because the binder used to prepare these samples also contained titanium. We thus used the reported values for the MST content to establish quantities to use in the performance testing.

Cements

SRNL researchers prepared a number of cement-bound MST samples. These include aluminosilicate, magnesium phosphate, magnesium aluminum phosphate and calcium phosphate cements. Best engineering judgment and scoping tests were used to select samples from each effort. Researchers checked scoping samples qualitatively for friability and rejected many attempts because they crumbled easily between the fingers. Following is a summary of the findings from the synthesis and characterization of these materials.

Aluminum Silicate Cements

An easy way to make an aluminosilicate cement was found to be by mixing sodium aluminate with DuPont Ludox[®]. The 40 wt % silica form of Ludox[®] was used to reduce the amount of water in the sample. Sodium aluminate was mixed in at a Si:Al molar ratio of 2:1. This approach produced a viscous and almost homogeneous slimy-looking liquid. It was kept warm in an oven overnight at 50 to 60 °C. The product was brittle, creamy white, and homogeneous except for small voids. It contained very small glassy bubbles visible under a microscope. One piece was stored at room temperature over a weekend while another piece was baked at 50 to 60 °C concurrently. X-Ray diffraction found that the piece stored at room temperature was amorphous while some Zeolite A had grown into the piece kept warm in the oven for the additional 3 days.

After initial trials researchers made MST-filled material by mixing 8.3096 g of sodium aluminate with 22.7387 g of air-dried MST cake. After these solids were thoroughly ground and mixed with a mortar and pestle, researchers put the mixture in a beaker with 31.1752 g of Ludox[®] having 40 wt % silica. The material set up well overnight at room temperature and was saved for later grinding and sieving.

Magnesium Phosphate and Magnesium Aluminum Phosphate Cements

Researchers made magnesium aluminum phosphate cements by mixing magnesium oxide (MgO) powder with a syrup of aluminum orthophosphate (AOP). They first prepared the AOP syrup by mixing aluminum trihydroxide powder with 85 wt % phosphoric acid. Such a mixture reacts with some heating and the solids dissolved into the acid to make clear syrup. The Al:P molar ratio for the mixture was 1:3, so the syrup probably contains dihydrogen phosphate ion. MST was introduced into these recipes as moist de-ionized water-washed cake mixed with the MgO. The mixture was extremely viscous but it thinned substantially upon addition of the AOP.

Ceramicrete[™] cement powder as-produced from Argonne National Laboratory was available and was tested. Researchers made samples according to directions. Researchers made samples of the unfilled cement by adding water to the powder and mixing. The cement paste took about 10 minutes to self-heat. Additionally researchers made MST-filled samples by mixing Ceramicrete[™] powder with damp MST cake in a 1:1 weight mixture (dry Ceramicrete[™]: MST

on a dry basis). Damp cake contained 2/3 mass of water so researchers added less than 1.3 mL additional water to 20.35 g Ceramicrete™ and 61.1 g of damp cake at the start of mixing.

Simple soak testing quickly eliminated Ceramicrete™ products from consideration. Researchers discovered that alkaline 5.6 M sodium solution caused swelling and disintegration of pieces of the cement within two days. This occurred despite the fact that the initial cured cement was rock-hard.

Calcium Phosphate Cements

The researchers spent much time attempting to make samples from various calcium phosphate cements. One motivating factor was that both bone char and cured calcium phosphate cements have in common the component hydroxyapatite. Bone char had been the focus of past efforts to find an inexpensive actinide adsorbent solid.

The main difficulty with calcium phosphate cements is the complexity of calcium phosphate crystal forms and morphology. The cements are also sensitive to the environmental conditions during curing. Researchers investigated cement recipes including mixtures of calcined calcium oxide, calcium dihydrogen phosphate, calcium hydroxide, disodium hydrogen phosphate solution, and various calcium:phosphate ratios [6,7]. In addition a researcher contacted medical cement companies and obtained samples from these commercial sources.

Stryker® Corporation (Allendale, NJ) provided about 50 grams of BoneSource® cement powder. Bone surgeons use this medical product to promote bone growth in areas of breakage. The powder makes an easy-to-use white putty when 0.32 mL of de-ionized water is added per gram of powder. The researcher found that mixing was easy and that the powder wet and handled well. Personnel discovered though that the cement formed cracks and made a physically weak product when allowed to set at room temperature overnight. The vendor advised that this material needs conditions exactly as found in the human body for good setting: 100% humidity and a temperature of 37 °C. These conditions were indeed found to improve the apparent strength of the product.

After many trials with the Stryker® and other calcium phosphate cements, we selected the Stryker Bonesource® material loaded with 25 wt % MST for further testing. The selection was made because the Bonesource® material was least friable.

Clemson Sample

The Clemson Environmental Technologies Laboratory (CETL) used a hot-oil column to produce particles for this work. This sol-gel process used colloidal silica in most cases but we used colloidal zirconium hydroxide to engineer MST because chemical stability of a zirconium oxide binder was expected to be much better than that of silica. CETL created a well-mixed slurry of air-dried MST cake with 20 wt % zirconium (VI) oxide colloidal solution. They adjusted the ratio of MST solids to solution so that the slurry was thick but still pumpable in the hot oil rig.

The rig used a peristaltic pump to deliver slurry through a needle into the top of the 27-foot tall silicone oil column. Oil at 70 °C set and partly dehydrated the slurry. The set droplets (beads) were collected on a screen at the bottom of the column. They were washed with hexanes to remove the oil. CETL reported that the slurry was easy to handle and set up well as desired in the process. The pear shape of the beads showed that the slurry was quick-setting out of the needle. We found that the beads contained 7.8 wt % Ti and 14.2 wt % Zr. The titanium level translates to an MST content of 16.1 wt %.

Internal Hydrolysis Samples

The internal hydrolysis method of engineering MST requires a porous chemically stable solid substrate. Five inorganic substrates were screened for use using a simple alkali salt soak test. Porous glass and Florisil[®] (a porous magnesium silicate) were quickly eliminated from consideration because they showed visible degradation and caking in one day of soaking in the salt solution.

Porous alumina samples were provided by Saint-Gobain Norpro[®] (Akron, OH) and also by Porocel[®] Corporation (Little Rock, AR). The Saint-Gobain material had sample number 2002650354, type SA52124. The sample consisted of 1300 micron diameter spheres. Smaller spheres were not available because the catalyst industry served by this company does not have interest in a smaller product. The Porocel[®] material was their Dynocel 600[®] 30 x 60 mesh granules. Both of these materials showed no degradation when soaked in alkali salt solutions.

K28 high temperature insulating firebrick was obtained from Morgan-Thermal Ceramics. The firebrick was pulverized and sieved to obtain 30 x 60 mesh granules. Slightly larger pieces showed no visible degradation in caustic salt soaking.

The hydrolysis of tetraisopropyl titanium (TPT) in highly alkaline alcohol solution forms MST solids. This requires an anhydrous solution "A" of 30 mL TPT, 11.6 mL of 4.4 M sodium methoxide in methanol, and 120 mL isopropyl alcohol. Solution "B", of 15.4 wt % water and isopropyl alcohol, forms MST solids by a hydrolysis reaction with solution "A". MST is engineered by first soaking a dried porous substrate in solution "A" using several cycles of vacuum to make sure that there is no air in the pores. The researcher then drains the excess solution "A" and adds excess solution "B" to form MST within the pores of the solid. The solids are then heated to near boiling for an hour followed by an overnight soak. Excess MST is washed away the next day and the product is air dried at room temperature.

The first effort used the St. Gobain spheres and the Porocel[®] Dynocel granules. The researcher initially measured MST content of the products by a dry-basis mass balance on substrate before and after hydrolysis. The St. Gobain material was ruled out at this step because it only picked up 2 wt % MST. The Porocel[®] material gained 10 wt % MST. That material was analyzed for titanium content and the result was used as the quantitative measurement for batch contact work.

An effort was made to increase the loading of MST in Porocel[®] substrate, and the K28 (JM-28) firebrick granules were also processed. Solution "A" contained only 10 mL of isopropyl

alcohol rather than 120 mL so that all reacting chemicals were more concentrated. The water content of solution “B” was set to 50 wt % in this case. Titanium content indicated that the Porocel substrate was 12.6 wt % MST and the K28 firebrick was 11.8 wt % MST. The more highly loaded Porocel[®] material was used in the second set of spiked simulant batch contacts and is named the “new” internal hydrolysis sample.

Basic properties of the engineered materials are shown in Tables I and II. Standard deviation of Table 1 is a measure of the dispersity of particle size about the mean. The ORNL products had the tightest particle size distribution relative to the other materials. The ORNL 50 wt% MST beads are shown in Figure 1. Weight percent MST of Table 2 was measured by dissolving samples and measuring titanium, then converting its mass to an MST equivalent (mass MST/mass titanium = 2.08). The values for ORNL materials were as-provided by ORNL because the titanium oxide binder was not counted as MST. The adsorption properties of pure ORNL titanium oxide binder are unknown. SrTreat[®] contains titanium but its chemical composition is proprietary.



Figure 1. Photograph of ORNL 50 wt % MST Beads

Table I. Mean Volume-Weighted Particle Size and Dispersity

Engineered Sample	Mean Value (microns)	Standard Deviation (microns)
UOP Beads	523	112
ORNL 50 wt % MST Beads	573	105
SrTreat [®]	5*	4*
ORNL 32 wt % MST Beads	520	81
Mg-AOP Cement	407	165
Al-Si Batch 1 Cement	450	166
Al-Si Batch 2 Cement	457	156
Hydroxyapatite Cement	*	*
Internal Hydrolysis	530	145

* Turbidity prevented accurate measurement

Table II. Weight Percent MST and Dry Bed Density of Engineered Materials

Sample	Weight Percent MST	Dry Bed Density Average g/mL	+/- Error, percent
SrTreat [®]	n/a	0.863	1.74
UOP Beads	78.6	1.13	1.18
UOP Trilobe	80.5	Not measured	
ORNL 50 wt % Beads	50.	0.991	0.97
ORNL 32 wt % Beads	32.	1.05	0.00
Mg-AOP	39.6	0.747	0.01
Al-Si Batch 1	35.7	0.736	0.00
Al-Si Batch 2	38.4	0.813	0.83
Hydroxyapatite	25	0.840	0.84
Internal Hydrolysis	9.9	0.642	1.25
New Internal Hydrolysis	12.6	0.669	0.33
K-28 Firebrick	11.8	0.746	0.02

Batch Contact Work

The work used the salt simulant solution of Table III for all spiked simulant testing. This simulant recipe has been used uniformly for much work with MST at SRNL in the past.

Table III. Composition of the Salt Simulant

Component	Molarity
Free hydroxide	1.33
Nitrate	2.60
Aluminum (as aluminate)	0.429
Nitrite	0.134
Sulfate	0.521
Carbonate	0.026
Total Sodium	5.60

Table IV presents the $^{239}\text{Pu}/^{240}\text{Pu}$ and ^{237}Np data for the simulant for indicated number of days after makeup. All soluble species entries are in microgram/L.

Table IV. Actinide Levels in the Salt Simulant, microgram/L*

Time (d)	$^{239}\text{Pu}/^{240}\text{Pu}$	$^{239}\text{Pu}/^{240}\text{Pu}$ Target	[Np]	[Np] Target	[U]	[U] Target
0	179 (14.)	200	530 (23.)	500		
11	197 (15.)	200	549 (25.)	500		
27	194 (15.)	200	568 (21.)	500		
48	211 (10.)	200	508 (46.)	500	12,500 (2,500)	10,000

* Values in parentheses are the single standard deviation estimate of error.

The tank composite solution consisted of supernate samples from SRS Tanks 11H, 30H, 32H and 39H. We diluted the composite material to 5.8 M in sodium using 1.66 M NaOH solution. The composite also contained 9,350 mg/L aluminum. The diluted composite was allowed to stand while changes in radiochemical analyses were noted. Table V displays the key analytical data over 48 days.

Table V. Key Radioactive Element Levels in the Tank Composite Salt Solution

Time (d) after compositing	[Total Pu], microg/L	$^{239}\text{Pu}/^{240}\text{Pu}$], microg/L	^{90}Sr], dpm/mL	[U] , mg/L
2	105 (6.6%)	94 (6.9%)	3.83 E+06 (7.8%)	3.0 (9.6%)
9	102 (8.0%)	91 (8.4%)	3.96 E+06 (7.8%)	2.6 (12.7%)
21	89 (8.3%)	78 (8.8%)	3.97 E+06 (7.8%)	2.6 (18.1%)
21 filtered	52(12.0%)	47 (12.8%)	1.01 E+06 (8.2%)	2.1 (22.8%)
32	90(13.8%)	81 (14.8%)	1.51 E+06 (7.5%)	
48	98 (6.6%)	88 (6.9%)	3.88 E+06 (8.2%)	
48 filtered	64. (9.1%)	58. (9.6%)	1.23 E+06 (8.2%)	

Values in parentheses are the single standard deviation estimate of error.

BATCH CONTACT PROCEDURES

The batch contact procedure for this work was closely aligned with past procedures for MST except that batches of engineered material required a change in the sampling procedure.⁸ To provide the typical phase ratio of 2,500 mL/g or 0.4 g of sorbent per liter, 250 mL of salt solutions were contacted with sorbent having the equivalent of 0.1 g MST. Researchers used

500 mL bottles to provide sufficient headspace for good agitation of each batch at the shaker cycle rate of 175 rpm. Oven shaker temperature was controlled to 27 ± 2 °C.

Technicians sampled the batches at least a half hour after agitation stopped to allow settling of the engineered materials. They withdrew approximately 5 mL of salt solution, filtered it through 0.45 micron nylon syringe tip filters, and pipetted 4 mL of the filtrate into 4 mL of 5 M nitric acid. They agitated the mixtures and allowed at least 2 hours of time before processing the samples further to allow complete acid dissolution. Three mL of acidified filtrate were measured into gamma vials for counting. Five mL were sampled separately for Pu analysis by thenoyltrifluoroacetone (TTA) spiked extraction and strontium-85 gamma counting.

For tests in the Shielded Cells with actual tank wastes, we agitated the contents of test bottles using a five-station magnetic stirrer and small Teflon[®] magnet bars. Batch bottles in cells work were 250 mL because headspace was not needed for good agitation. A water bath at 27 ± 2 °C provided temperature stability. Technicians measured 1.5 grams of filtrate into 60 mL plastic sample bottles containing pre-measured 25 mL portions of 3 M nitric acid. Acidified filtrate samples sat for more than two hours before further sample pouring and cell exit for chemical and radiochemical analyses.

ADSORPTION COLUMN PROCEDURES

Three adsorption column tests were conducted to remove Pu and Sr from simulated waste solutions containing 5.6 M sodium. The simulant recipe was the same as for batch testing and ⁸⁵Sr was used as a radiotracer. Multiple feed analyses were taken to determine Pu content. Engineered MST was soaked in non-radioactive, Sr-free simulant for at least five days before loading into the column. The ORNL internal gelation beads swelled to twice their original size in the first two days (or less) of soaking in caustic simulant solution. The SRNL internal hydrolysis material did not swell more than 10%. Neither material exhibited a change in size after the initial soaking period or during testing.

All three column tests treated the waste in a once-through fashion and operated continuously (i.e., overnight) using the same equipment. The column and tubing were rinsed with water and with non-radioactive simulant solution between tests. The first column test lasted 23 days and treated 10 L of simulated waste solution. The latter two tests continued for 5 days at a higher flow rate. Most of the solution (8 L) from the first test was recovered and spiked again with Pu and Sr. This latter solution was split for the remaining two tests. Details for the different column tests are provided in Table VI. Though the latter two tests did not contain significant total Sr, all tests contained sufficient ⁸⁵Sr radiotracer for measurement purposes. The column was 0.833 cm in diameter and was jacketed. The circulating water temperature through the jacket was maintained between 23.0 and 26.0°C for the three tests.

For the three column tests, ⁸⁵Sr was measured by a low temperature germanium detector. However, one detector was used for Test 1 and a separate detector was used for Tests 2 and 3. Both instruments yielded values with no more than 10% uncertainty. Because of the relatively short half-life of ⁸⁵Sr, measured values were time-adjusted based on the natural decay of ⁸⁵Sr and the time between sampling and analysis. In the Results and Discussion section, all ⁸⁵Sr

values represent activity at time of sampling. Also, ^{85}Sr data were normalized to a constant feed level of 100 to facilitate comparison.

Table VI. Characteristics of Column Tests with Engineered MST

Test No.	Engineered MST	Mass of Sorbent	Volume of Sorbent	Column L/d ratio	Feed rate	Pu	Total Sr
		g	mL		BV/hr	$\mu\text{g/L}$	$\mu\text{g/L}$
1	ORNL 50 wt.% MST (internal gelation)	1.86	3.38	7.2	5.3	108 ± 12	0.72
2	ORNL 50 wt.% MST (internal gelation)	0.92	2.0	4.3	15	120 ± 23	--
3	SRNL 12 wt.% MST (internal hydrolysis)	1.34	2.2	4.9	13.6	120 ± 23	--

RESULTS AND DISCUSSION

Adsorbent performances in the caustic simulant and the high level waste tank solution are compared in Table VII. All values are for 250 mL batches measured after 10 days of agitation. Adsorbent mass was varied to keep the equivalent MST mass a constant at 0.1 g in each batch. Decontamination factors (DF) are simply the ratio of strontium or plutonium activity in untreated solution to that after contact with adsorbent. Batch distribution coefficients (K_d) are calculated using total sample mass, thus, materials with low MST weight percents are penalized. Duplicated experiments are indicated when material names show #1 and #2.

Strontium was relatively easy to remove. All adsorbents provided distribution coefficient (K_d) values exceeding $1.E+04$. Best performing engineered materials were consistent for both strontium and plutonium so these materials (i.e., SrTreat[®], ORNL 50 wt % MST beads, and internal hydrolysis MST in alumina particles) were also tested with the tank waste. Strontium performance was consistent between simulant and tank solution for the chosen materials.

ORNL 50 wt % MST beads material was the only selected adsorbent that did not show solution matrix effects for plutonium. SrTreat[®] and internal hydrolysis particles performed less well in tank waste than in simulant solution.

As shown in Figure 2, the ORNL 50 wt % MST removed Pu slightly better than the Internal Hydrolysis (SRNL) 12 wt % MST. For the ORNL 50 wt % material at 5.3 BV/hr, Pu removal was 99% initially and 94% after 2900 BV, indicating that the sorbent had not yet reached its capacity. Also, for the ORNL 50 wt % material, increasing flow rate from 5.3 to 15 BV/hr did not significantly reduce Pu adsorption performance.

As seen in Figure 3, the Internal Hydrolysis material adsorbed Sr exceptionally well. The points on the chart for Internal Hydrolysis represent detection limits. No Sr was found in the Internal Hydrolysis product solution. Similarly, the ORNL 50 wt % MST material operating at 5.3 BV/hr removed Sr almost completely throughout 2900 BV. Increasing the flow rate to 15 BV/hr did reduce the level of Sr removal.

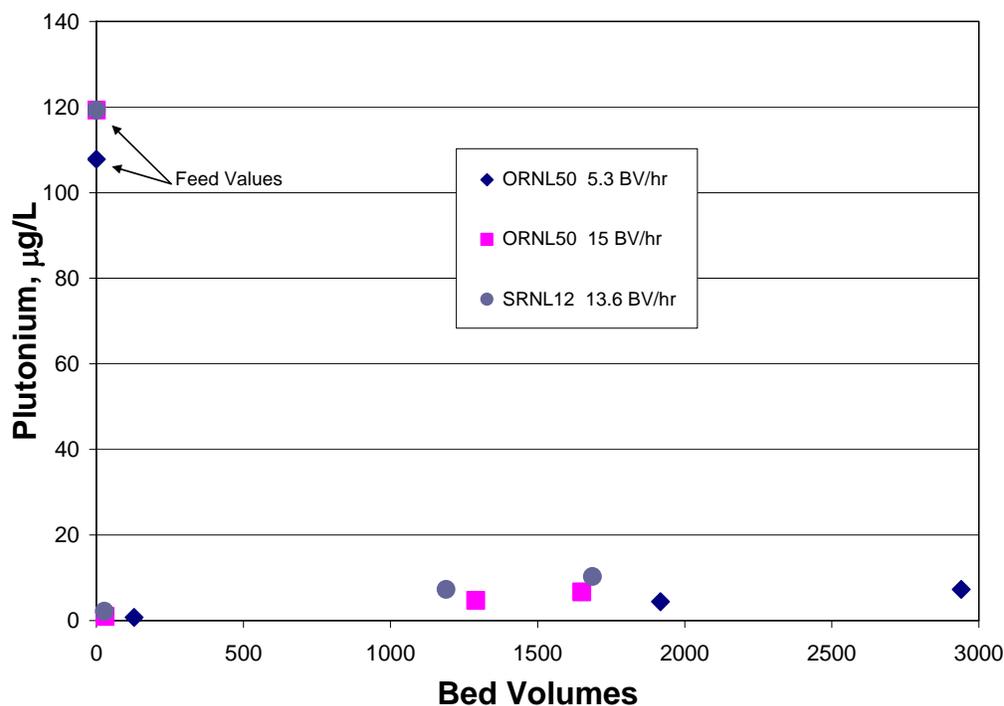


Figure 2. Plutonium removal in column test with Engineered MST

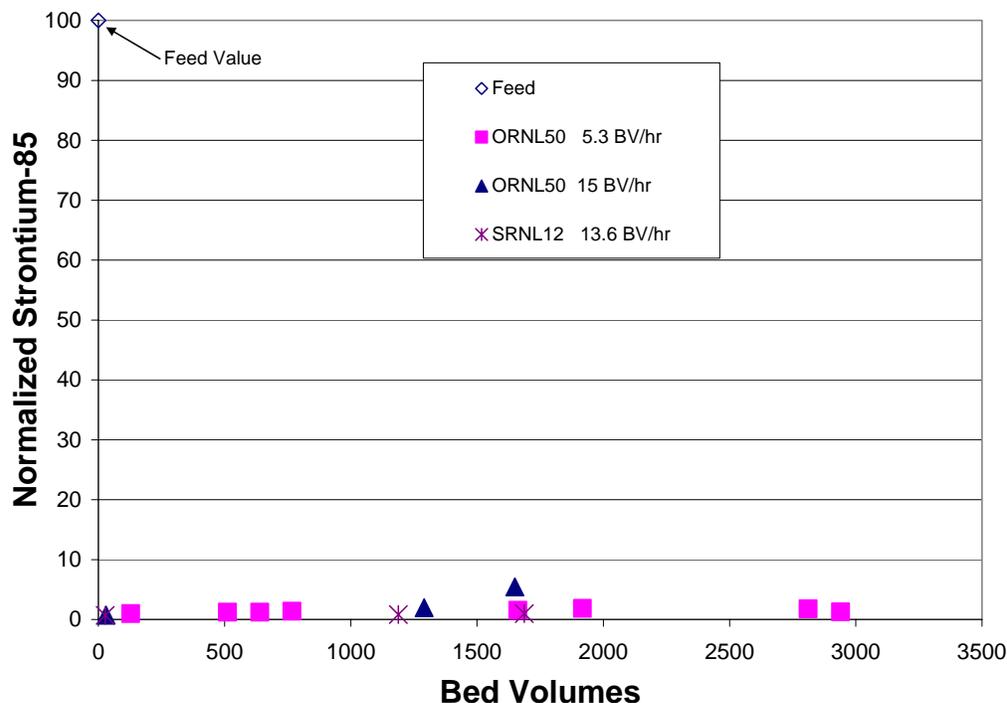


Figure 3. Strontium removal in column tests with Engineered MST.

In the first test, ORNL50 at 5.3 BV/hr, the white sorbent became light tan during testing. A photograph of the column after the full 2900 BV is shown in Figure 4. 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. Qualitatively, Figure 4 shows that the column was not fully loaded, as the sorbent between the 0 and 1.5 cm graduations remained white.



Figure 4. Column of MST beads after treating 2900 bed volumes of simulant.

CONCLUSIONS

The better performing engineered MST materials included that produced by ORNL internal gelation, the internal hydrolysis techniques, and the commercially available SrTreat[®]. Spiked simulant plutonium and strontium data for ORNL 50 wt % beads compared well with those from actual tank composite supernate. The internal hydrolysis technique suffers from inability to load porous material more than about 13 wt % MST, but the adsorbent created demonstrates performance that is at least as good as that of MST powder. SrTreat[®] has good adsorption capacities for strontium and plutonium, but poorer removal kinetics than other materials. There was also a negative matrix effect for its plutonium removal from the tank composite sample, though the experiment was not verified by duplication.

Cements containing MST had chemical stability problems including bleed of phosphate. We strongly recommended that other forms of hydroxyapatite or other phosphate-containing adsorbents be tested for this problem before they are considered further.

Both ORNL 50 wt % beads and internal hydrolysis granules were effective at removing Pu and Sr from simulated waste solutions in once-through column treatments.

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