Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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Removal of High Specific Activity Fission Products from Uranyl Sulfate Waste Solutions

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August 2020
SRNL-STI-2020-00199, Revision 0

SRNL.DOE.GOV

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Prepared for
U.S. Department of Energy

SRNL-STI-2020-00199
Revision 0

Keywords: *Uranyl Sulfate, Fission Product Removal, Waste Treatment*

Retention: *Varies*

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EXECUTIVE SUMMARY

The Savannah River National Laboratory (SRNL) is currently providing support to SHINE Medical Technologies (SHINE) which plans to deploy a low energy, accelerator-based neutron source to fission low enriched U in a uranyl sulfate target solution for ^{99}Mo production. The ^{99}Mo is initially separated from the fission products and target solution by an extraction column. Subsequent washing of the column will generate waste solutions containing residual U and fission product activity. A small number of high specific activity fission products (e.g., ^{90}Sr, ^{137}Cs, and ^{144}Ce) in these streams will likely control the classification of the low level waste (LLW). If a sufficient amount of the high specific activity isotopes are separated from the SHINE waste streams and concentrated into a waste form, it would be possible to treat a majority of the wash solutions from the column operations as a lower class of LLW (Class A versus Class B or C or Class B versus Class C). The high specific activity fission product elements could then be dispositioned as a much smaller volume of waste rather than requiring the disposal of the entire waste stream at the higher waste classification.

The Savannah River Site (SRS) has experience with using monosodium titanate (MST) and crystalline silicotitanate (CST) to remove Cs and Sr from high salt content waste solutions generated during the reprocessing of nuclear fuels and targets. Both of these materials have worked very well for their intended purposes at the SRS where the fission product elements are removed from highly alkaline waste. On the other hand, SHINE waste streams from the extraction column contain H_2SO_4 which makes the solution acidic. Additionally, the SRS waste streams do not contain other fission product elements such as transition metals or lanthanides because they precipitate upon neutralization of the SRS waste and are not present in the supernate which is dispositioned as LLW following treatment. As such, there are inherent differences between SHINE and SRS waste treatment strategies. Savannah River National Laboratory was tasked with performing scoping studies to see if MST and CST would remove Sr, Cs, and Ce from an acidic mixed metal simulant solution.

Batch contact experiments were performed using MST and two CST type materials. The MST material is a 15 wt % powder in 0.15 M NaOH slurry. The MST showed low adsorption for elements of interest from acidic solution. Furthermore, the powder size makes MST non-ideal for column operations. A CST IE-911 ion exchange material had high Cs adsorption, moderate Sr, and marginal Ce adsorption. Based on adsorption of all species, the ion exchange capacity was found to be 0.032 meq/mL. A bench-top column experiment to measure elemental breakthrough curves was performed using CST IE-911 where chromatographic separations of the mixed simulant were expected to occur. While most elements behaved as expected, the lanthanide series, containing Ce, broke though the column earlier than expected. The second CST material, CST R9120, displayed high adsorption for all elements in the acidic mixed simulant solutions in a batch contact study, and had a calculated loading capacity of 0.091 meq/mL. Future studies to develop a waste treatment flowsheet should focus on CST R9120 to treat SHINE waste solutions.

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CST	crystalline silicotitanate
HEU	highly enriched uranium
LLW	Low Level Waste
MST	monosodium titanate
NNSA	National Nuclear Security Administration
PTFE	polytetrafluoroethylene
SHINE	SHINE Medical Technologies
SRNL	Savannah River National Laboratory
SRS	Savannah River Site

1.0 Introduction

The U.S. Department of Energy's National Nuclear Security Administration (NNSA) has entered into cooperative agreements with commercial businesses to establish a reliable domestic source of ^{99}Mo that will be produced without the use of highly enriched uranium (HEU).[1] Molybdenum-99 is the parent isotope of ^{99m}Tc, the most widely used radioisotope in nuclear medical diagnostic imaging. It is employed in about 14 million procedures per year.[2] The United States currently produces very little ^{99}Mo and therefore imports essentially all of its supply from foreign producers, some of which still use HEU in the production process. The continuity of the ^{99}Mo supply has also been an issue in the recent past. A majority of the research reactors used to irradiate targets that produce much of the world's supply of ^{99}Mo are over 40 years old. As a result of the aging infrastructure, planned and unplanned outages of some of these reactors have resulted in ^{99}Mo supply interruptions.[3]

The NNSA's support to the cooperative agreement participants is based on a (50%/50%) government/commercial cost-share basis,[1] with initial NNSA contributions up to a total of $25 million for each project. In addition to the financial support provided directly to the commercial businesses, NNSA has also funded national laboratory support to assist in the commercialization of the ^{99}Mo production processes. The Savannah River National Laboratory (SRNL) is currently providing support to SHINE Medical Technologies (SHINE) which plans to deploy a low energy, accelerator-based neutron source to fission low enriched U in a uranyl sulfate target solution. The ^{99}Mo is initially separated from the fission products and target solution by an extraction column. Subsequent washing of the column will generate waste solutions containing residual U and fission product activity. A small number of high specific activity fission products (e.g., ^{90}Sr, ^{137}Cs, and ^{144}Ce) in these streams will likely control the classification of the low level waste (LLW). Regulatory limits for the isotopes of interest are summarized in Table 1-1.[4]

Table 1-1. Regulatory Limits for Selected High Specific Activity Fission Products

Radionuclide	Concentration (Ci/m^3)		
	Class A	Class B	Class C
Nuclide with a half-life <5 years (e.g., Ce-144)	700	(a)	(a)
Sr-90	0.04	150	7000
Cs-137	1	44	4600

(a) No established limits – treat as Class B unless restricted by another isotope.

If a sufficient amount of the high specific activity isotopes are removed from the SHINE waste streams, it would be possible to treat a majority of the wash solutions from the column operations as a lower class of LLW (Class A versus Class B or C or Class B versus Class C). The high specific activity fission product elements could then be dispositioned as a much smaller volume of waste rather than requiring the disposal of the entire waste stream at the higher waste classification.

The SRNL has developed flowsheets for the removal of Cs and Sr from high salt content waste solutions generated during the reprocessing of nuclear fuels and targets at the Savannah River Site (SRS). High specific activity fission products such as Sr and Cs are removed to permit immobilization of LLW solutions in saltstone for onsite disposal.[5] The removal of these isotopes has been accomplished using inorganic ion exchange materials which have a high specificity for these elements. Monosodium titanate (MST) is used for Sr removal prior to immobilization of LLW solutions in saltstone[6-8] and crystalline silicotitanate (CST) has been deployed for in situ removal of Cs from SRS waste tank solution prior to final treatment and immobilization.[9-11] Both of these materials have worked very well for their intended purposes at the SRS where the fission product elements are removed from highly alkaline waste. However, the SHINE waste streams generated during ^{99}Mo recovery have significant differences. The SHINE waste streams from the extraction column contain dilute H_2SO_4 which makes the solution acidic. In addition, other fission product elements such as transition metals or lanthanides precipitate upon neutralization of the SRS waste and are not present in the supernate, which is dispositioned as LLW following treatment. The application of MST and CST in H_2SO_4 media has not been previously studied. Both materials are ion exchangers which allow the exchange of a Na cation in the MST or CST with a Sr or Cs cation, respectively, from solution. Depending on the crystal lattice and ionic radii, other cations can be removed by the ion exchange material. This work provides a scoping assessment of the ability of MST and CST to remove Cs, Sr, and Ce from a dilute H_2SO_4 solution. The studies include batch contact experiments with MST, CST IE-911, CST R9120, and a column experiment with CST IE-911 using a non-radioactive simulant solution containing representative fission product elements. Uranium was not present in the simulated SHINE waste solution during these initial scoping experiments.

2.0 Experimental Procedure

2.1 Waste Simulants

The composition of the waste simulants used in the experimental programs was based on estimated concentrations provided by SHINE for a H_2SO_4 wash of the extraction column. The elements selected for waste simulants, listed in Table 2-1, were elements with the highest concentrations in the wash solution. Some elements also serve as surrogates for other elements found in the waste stream. Strontium is a surrogate for Ba, Re is a surrogate for Tc, and Eu is a surrogate for heavier lanthanides. The batch contact simulant was made to be roughly 15 times more concentrated than the concentrations in the estimated SHINE H_2SO_4 wash stream. The sodium nitrate and H_2SO_4 were maintained at the estimated concentrations. Making the metal concentrations more concentrated was necessary to ensure analytical measurements were above the detection limits of the inductively-coupled plasma mass spectrometer (ICP-MS) used for the analyses. The column simulant was made roughly 38 times more concentrated than the estimated SHINE H_2SO_4 wash stream, but sodium nitrate and H_2SO_4 were maintained at similar concentrations. The column simulant was concentrated to address detection limits and to minimize the amount of time it would take to process the simulant through a 1-cm x 10-cm column.

2.2 Batch Experiments

The MST (Harrell Industries) batch experiments were conducted by adding an MST (15 wt % MST in 0.15 M NaOH) slurry to 30 mL of waste simulant. The slurry was stirred to create an even suspension of MST in the slurry. Details of the experimental procedure follow. Approximately 0.10 g of 15 wt % slurry was weighed into a 50-mL Falcon® tube. A 30.00-mL aliquot of the SHINE batch simulant was subsequently added to the tube. A total of six samples were prepared in this manner. The final concentration of the mixture was 0.5 g/L MST. The six mixtures were placed on a rotisserie shaker and rotated for 1, 2, 4, 5, 24, and 48 hours. After the desired time had passed, the mixture was collected in a syringe and filtered through a 0.45-µm PTFE filter. Samples were analyzed by ICP-MS and have a ±20% uncertainty in the concentration measurements.

The CST batch experiments were conducted by first wetting the ion exchange material. The CST material was fully converted to the Na^+ form prior to receiving[12] and was mixed with pH 1.4 H_2SO_4/69 mM $NaNO_3$ solution in a 1:1.5 ratio (material:solution). The pH was determined by measuring total acid in units of [molarity] and converting by equation $pH = -\log[H]$. The mixture was rotated on a mixer for 7 hours and filtered by aspirator to remove excess supernatant for 1 hour. The treated material had a tap density of 1.16 g/mL for IE-911 and 1.18 g/mL for R9120. Approximately 0.5 g of wetted material was weighed into a 50-mL Falcon® tube. A 10.00-mL aliquot of the SHINE batch simulant was subsequently added to the Falcon® tube. A total of six samples were prepared in this manner. The samples were rotated for 1, 2, 4, 7, 24, and 48 hours. After the desired mixing time had passed, the supernatant was filtered through a 0.45-µm PTFE syringe filter. Samples were analyzed by ICP-MS and have a ±20% uncertainty in the concentration measurement. The same experimental procedure was used for UOP-Honeywell products CST IE-911 (Lot 36232-1-4) and CST R9120 (Lot 2099000034).

Table 2-1. Simulant for SHINE Extraction Column Waste Stream.

Element	Compound Used	SHINE's Estimated H_2SO_4 Waste Concentration (µg/L)	Batch Contact Simulant Concentration (µg/L)	Column Simulant Concentration (µg/L)
Ru	$Ru(NO)(NO_3)_3$	2400	40400	118000
Ce	$Ce(NO_3)_3$ $6H_2O$	2400	35500	90000
Cs	$CsNO_3$	2300	34600	85500
Nd	$Nd(NO_3)_3$ $6H_2O$	1700	25300	64200
Sr	$Sr(NO_3)_2$	1400	21200	53200
Pr	$Pr(NO_3)_3$ $6H_2O$	1100	16600	41600
Sm	$Sm(NO_3)_3$ $6H_2O$	450	6800	16900
Rb	$Rb(NO_3)$	400	6200	15200
Pd	$Pd(NO_3)_2$ $2H_2O$	180	2600	6500
Eu	$Eu(NO_3)_3$ $5H_2O$	100	1500	3600
La	$La(NO_3)_3$ $6H_2O$	96	1360	3400
Re	$NaReO_4$	62	2140	5400
H_2SO_4	H_2SO_4	1.19E+06 (pH 1.3)	pH 1.4	pH 1.6
$NaNO_3$	$NaNO_3$	5.90E+06 (69 mM)	66 mM	59 mM
Total NO_3^-			69 mM	69 mM

2.3 Column Experiment

A column experiment was performed using CST IE-911 only due to availability. There was not a sufficient amount of CST R9120 available during the experimental time period available to perform a column test. The CST IE-911 material was wetted as previously described. A 1-cm x 10-cm column was filled with a slurry of the wetted CST IE-911 material (Figure 2-1). The solution was slowly drained and backfilled with more slurry to properly load the column and minimize void spaces that can cause channeling. The total bed volume was 7.85-mL of CST IE-911 material. The column simulant was prepared as previously described. The simulant was loaded into a 25-mL syringe and placed on a syringe pump to control flowrate. The Tygon® tubing between the syringe and column was filled with simulant prior to starting column operation. The raffinate was collected in a graduated cylinder and 1-mL grab samples were collected at desired intervals. The 25-mL syringe was changed when it was running low on simulant. Bubbles in the line were avoided during this procedure. In total, 208 mL of simulant were transferred through the column. The flowrate was arbitrarily chosen as 3 bed volumes per hour (23.56 mL per hour). The residence time of simulant in the column was approximately 20 minutes with a 0.5 cm/min linear velocity. The grab samples were analyzed by ICP-MS and have a ±20% uncertainty in the concentration measurements.

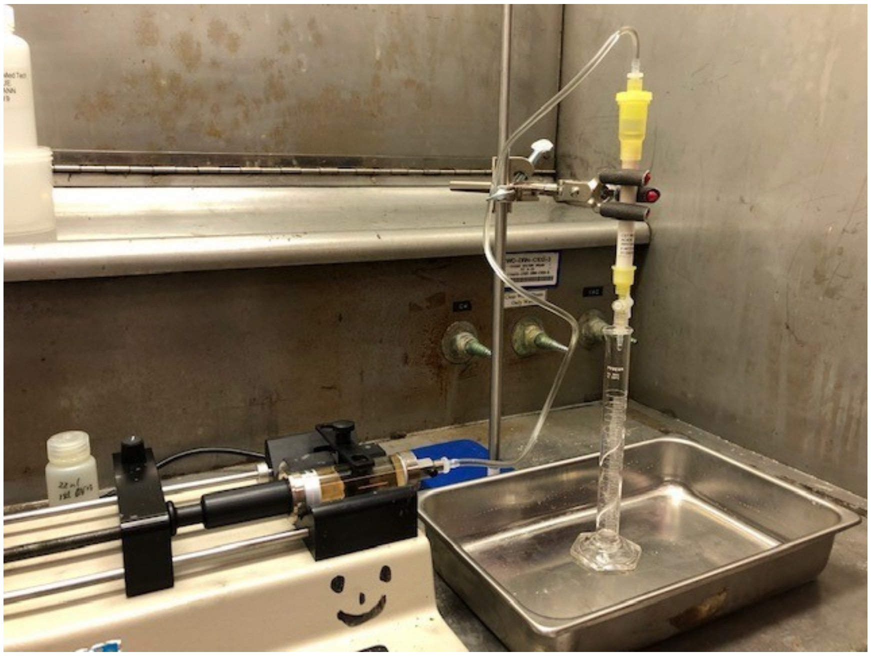

Figure 2-1: Photograph of column experiment showing syringe pump, column, and graduated cylinder to measure raffinate volume.

3.0 Results and Discussion

3.1 Batch Contact Study with MST Slurry

Monosodium titanate is supplied as a slurry of 15 wt % MST in 0.15 M NaOH solution. Initially, an effort was made to convert the basic MST slurry to an acidic slurry. Ideally, the MST would be filtered and then dried in an oven and diluted to the desired concentration. However, MST cannot be completely dried in a vacuum oven as it is difficult to rewet the material after completely drying. Instead, the MST slurry was filtered, rinsed with pH 1.4 H_2SO_4/69 mM $NaNO_3$ solution (similar to SHINE waste conditions), and diluted in the same rinse solution to approximately 15 wt %. After repeated attempts, the supernatant pH did not drop below 5, and a proper mass concentration could not be determined since the MST was a wet, clay-like material with an unknown density. Since attempts to convert MST to an acidic form were unsuccessful the MST slurry was used as is (i.e., 15 wt % MST in 0.15 M NaOH) for batch contacts with SHINE waste simulant.

Batch contact of SHINE test simulant with MST resulted in low removals for all elements (Figure 3-1). The highest adsorption of nominally 25% was for Ru followed by Pd and the lanthanide series. Lastly the alkali and alkaline earth metals showed little to no removal from solution by MST. If more 15 wt % MST slurry was added to the 30 mL simulant, the percent removals would increase simply because more MST binding sites are available due to increased MST mass. However, the overall trend shows that the primary elements of concern (Cs, Sr, and Ce) displayed the lowest adsorptions, with Cs and Sr having effectively zero removal. The average K_d's (Appendix A, Figure A-1) of Sr and Cs across contact times are 1.1 mL/g and 4.5 mL/g, respectively. Cerium has a higher calculated average K_d of 117.1 mL/g. The low values for Sr and Cs indicate MST does not recover Sr and Cs ions under the tested simulant conditions.

The pH of the waste simulant was 1.38, and the addition of 0.1 grams of 15 wt % slurry did not change the pH significantly (pH 1.38 to pH 1.42 after addition). Typically, MST is used to adsorb alkali and alkaline earth metals from basic solutions. The high acidity, pH 1.4, affects complexation of elemental species with the MST material. In order to use MST, the waste stream would need to undergo a pH adjustment from acidic to basic solution. This will increase overall liquid waste volumes and precipitate some of the waste stream components. Furthermore, the MST material is a slurry consisting of fine MST particulates. As such, it is not suitable for column separations but could be deployed using a mixer/decanter or mixer/filter flowsheet.

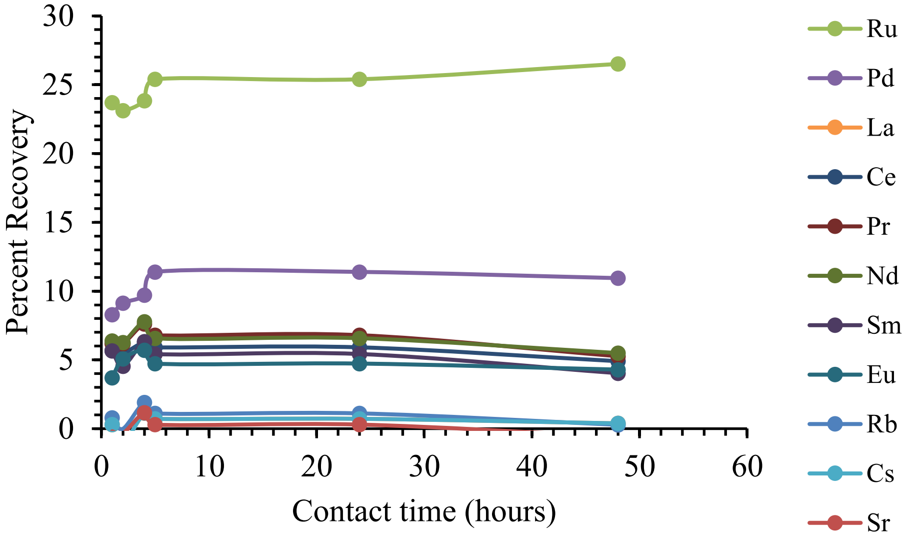

Figure 3-1: Batch contact of SHINE waste simulant with MST over time

3.2 Batch and Column Studies of CST IE-911

Batch contact experiments using CST IE-911 material to remove fission product elements from 10-mL of SHINE waste simulant showed 100% adsorption for Rb and Cs (Figure 3-2). Strontium and Ru had removals of 55% and 42%, respectively. The lanthanide elements each had 9-10% adsorptions on the CST IE-911 followed by 8% of the Pd, and only 2% of the Re. By adding up the masses of all the elements adsorbed on the material, and converting to mmol, the overall capacity was found to be 0.017 mmol/mL. Accounting for the charge of various elements in the simulant, the 0.017 mmol/mL translates to 0.032 meq/mL loading capacity for the ion exchange material. In converting mmol to meq, Ru was assumed to be Ru^{3+} and ReO_4^- was not included due to being an anion and low adsorption. The batch contact data are presented as distribution values and distribution coefficients in Appendix A, Figure A-2. While the CST operates as an ion exchange material, the Cs^+ and Rb^+ cations exchange with Na^+ cations in the CST crystal lattice. Similarly, the Sr^{2+} exchanges with two Na^+ cations. The adsorption mechanism for trivalent elements (i.e., lanthanides and Ru^{3+}) is not known. Logically, trivalent cations would exchange with three Na^+ cations, but the coordination of the trivalent element to CST has not been studied in detail. Although the nonradioactive Ru^{3+} exhibited reasonable adsorption in this test, during treatment of actual waste, the valence chemistry of fission product Ru may be more difficult to control and could convert to volatile RuO_4. Sodium perrhenate was used for rhenium element in the simulant. Perrhenate is an anion (ReO_4^-) and was not expected to adsorb to the material. Results show rhenium adsorption remained low across batch contact times.

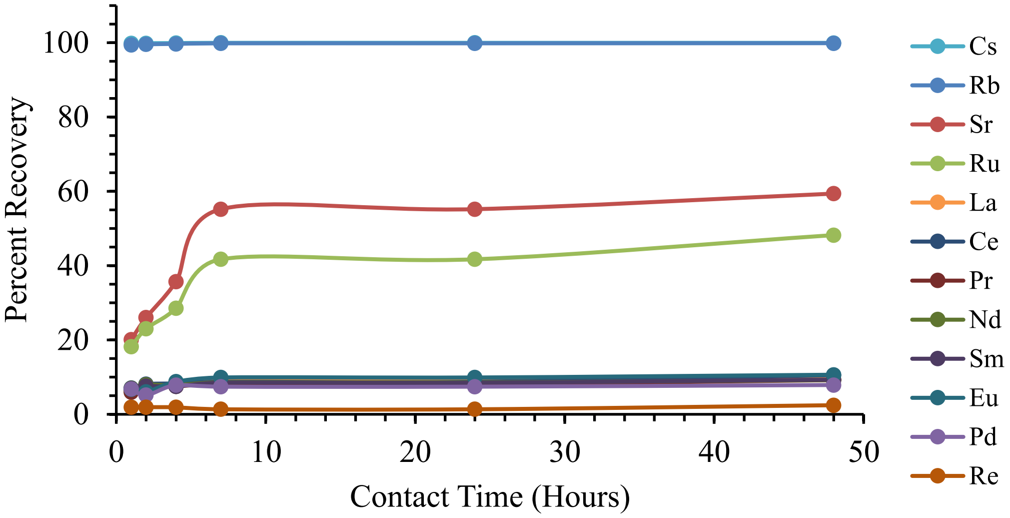

Figure 3-2: Batch contact of CST IE-911 with SHINE waste simulant over time

From the batch contact study, after 48 hours of contact time with the CST material, the loading capacity was calculated to be 0.032 meq/mL. For a 7.85-mL column, the total loading capacity would be 0.25-meq. The batch contacts showed that ReO_4^- was the least adsorbed followed by Pd and the lanthanide series (Figure 3-2). The least adsorbed element, Re, was expected to break through the column first when 0.25-meq of material from the feed solution had passed through the column. Knowing the simulant concentration, breakthrough was predicted to occur after 24-mL of simulant went through the column. Indeed, Re did start to break through the column around 24-mL (Figure 3-3). Eventually, the Re concentration in the raffinate exceeded 100% of the Re concentration in the feed solution. This is most likely due to a concentrated Re band forming in the column as a result of chromatographic separation. The concentrated band eventually fully breaks through and the Re concentration in the raffinate levels out to 100%, indicating that the concentration of Re in the feed simulant into the column was the same as the Re concentration leaving the column in the raffinate. In other words, Re was no longer being removed by the column. Interestingly, the lanthanides did not follow this pattern. The lanthanides were expected to show a similar chromatographic separation and succeed Re breakthrough due to their higher adsorptions observed in the batch contact. However, the raffinate had a lanthanide concentration of 80% of the feed simulant after three bed volumes (24-mL). The 80% concentration in the raffinate remained consistent until 120-mL of the raffinate were collected. The consistent 80% raffinate concentration does indicate there was some retention to the CST or else the raffinate concentration would've reached 100%. Concurrently, as the Re concentration decreased to 100%, the lanthanide raffinate concentration increased to above 100%, indicating a concentrated band did form. The lanthanide band started to break through after the Re band. As stated previously, the coordination of trivalent lanthanide to CST is not well understood. The batch contacts were first sampled at 1 h; whereas, in the column experiment the solution had a 20-minute residence time. The kinetics of lanthanide adsorption to CST IE-911 could be too slow causing the bulk of the lanthanides to pass through. The samples were also analyzed by ICP-OES to support the lanthanide trend determined by ICP-MS and are shown in Appendix A, Figure A-4.

To predict Ru and Sr breakthrough, the elements with lower adsorption (Re and the lanthanide series) were not included when summing up the mmols of elements fed through the column. Using the same 0.25-meq loading value for the 7.85-mL column, total mmol of Ru, Sr, Cs, and Rb was expected to hit 0.25-meq around a collected raffinate volume of 45-mL. As Figure 3-3: SHINE waste simulant breakthrough curves from CST IE-911 column shows, the Sr and Ru concentration in the raffinate started to increase in the 45 to 70 mL range. Palladium also starts to break through the column slightly above a raffinate volume of 100-mL. As stated previously, Pd was expected to breakthrough between Re and the lanthanide based on Pd's low adsorption in the batch contacts. Interestingly, between a raffinate volume of 130 and 160 mL, the Ru and Pd have a sharp uptick in concentration that does not follow the overall trend. This suggests an interaction may be occurring between palladium nitrate and ruthenium nitrosyl nitrate. It is not apparent why the similar trend is occurring for these elements; however, elucidating the phenomena is beyond the scope of these experiments. The total Cs and Rb calculated to be sent through the column was 0.17-meq (0.17-mmol). Since the column has a 0.25-meq capacity, Cs and Rb breakthroughs were not expected to occur. The data shows Cs and Rb did not breakthrough, maintaining a net zero concentration in the raffinate throughout the column experiment.

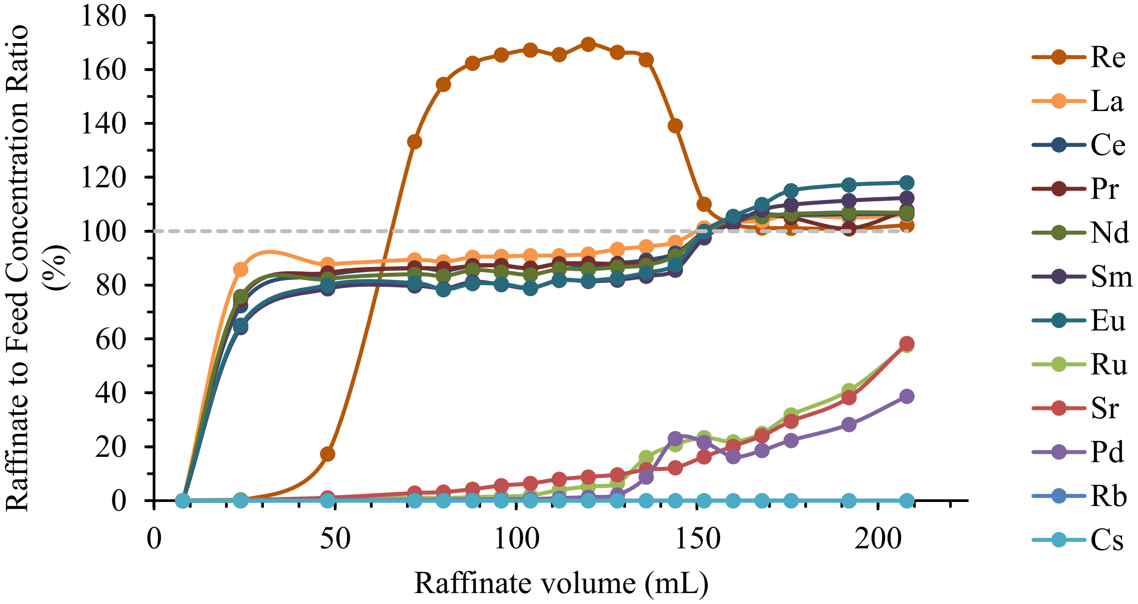

Figure 3-3: SHINE waste simulant breakthrough curves from CST IE-911 column

3.3 Batch Studies of CST R9120

UOP, LLC (a Honeywell company) currently manufactures CST R9120 which has shown high Cs adsorptions over a wide pH range. The CST R9120 series and CST IE-911 are effectively the same material. The CST IE-911 name was changed to CST R9120 when proprietary engineering changes were made. Although both materials are effectively the same, slight variations in the performance of CST R9120 compared to CST IE-911 have been observed at the SRS.[11] A limited amount of CST R9120 material was available for use in the scoping studies; therefore, only batch contact experiments were performed using the CST R9120. The results, shown in Figure 3-4, were very promising. Cesium and Rb were 100% adsorbed after the first hour. Cerium and the other lanthanides also achieved 100% adsorption followed by 100% Sr adsorption and subsequently 98% Ru and 96% Pd adsorption (note that the y-axis starts at 80% on Figure 3-4). The total capacity for all elements absorbed was calculated as 0.091 meq/mL, higher than the 0.032 meq/mL for the IE-911 material. The capacity of the R9120 could be even higher since a majority of the elements reached 100% adsorption from the simulant solution. The results are also presented as distribution values and distribution coefficients in Appendix A, Figure A-5. The titanium concentration in the solution was also measured at the different contact times (Figure 3-5). The trend shows a linear increase of Ti in the solution. For CST IE-911 the amount of Ti in solution increased also (Appendix A, Figure A-3), but was not linear over the same time scale. This could be an indication that the CST material is degrading in the H_2SO_4 solution or due to "loose" Ti in the initial matrix including during the pretreatment and should be studied more thoroughly. Previous studies have demonstrated Cs^+ adsorption by CST in up to 3 M HNO_3.[13] The differences between CST IE-911 and CST R9120 batch contact results, although effectively the same material, are not obvious at this time. There could have been differences in the initial state of the material (Na vs H form) that the pretreatment did not address, or the materials may be more different than the manufacturer has indicated. Regardless, the R9120 material is currently being manufactured by UOP and provides the higher lanthanide recoveries of the two materials in batch contact experiments.

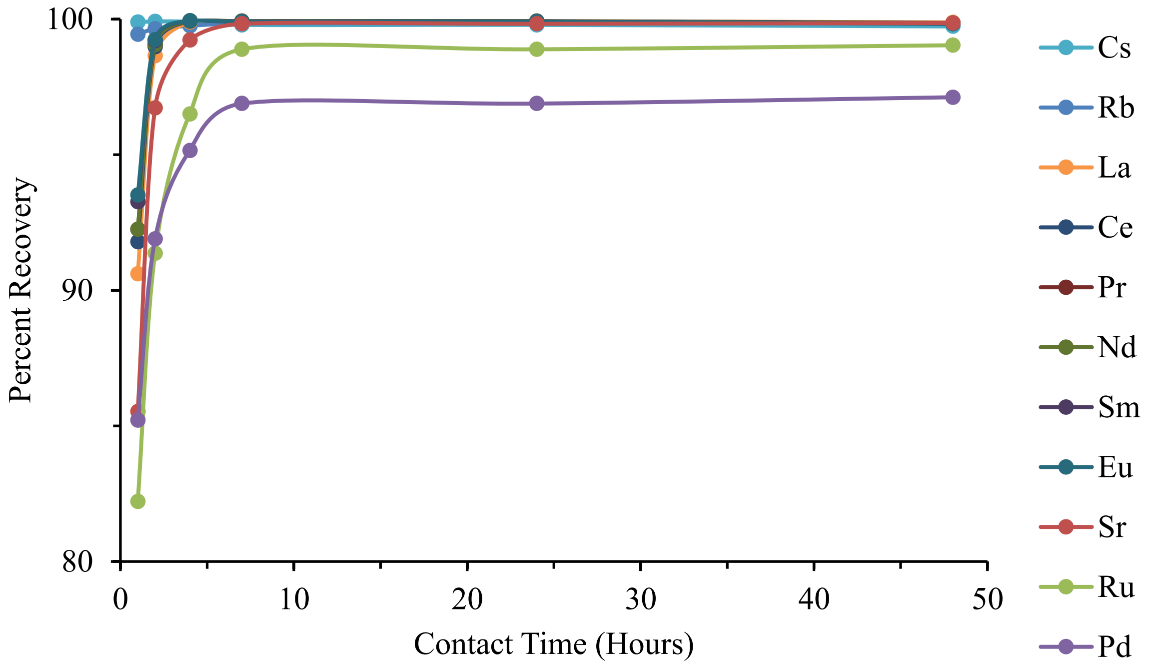

Figure 3-4: Batch contact of CST R9120 with SHINE waste simulant over time

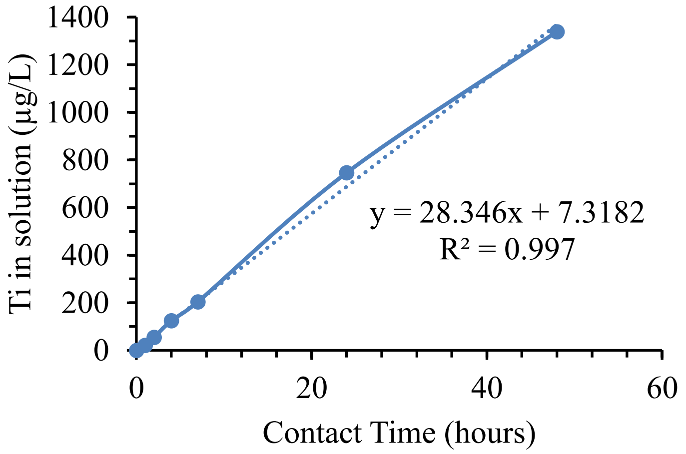

Figure 3-5: Titanium concentration in SHINE waste simulant batch contact with CST R9120 over time

4.0 Conclusions

Three inorganic ion exchange materials were evaluated to remove Sr, Cs, and Ce from a simulated H_2SO_4-containing waste stream generated from SHINE's Mo-99 recovery process. The MST slurry tested is not a viable candidate for waste treatment. The MST, under acidic conditions, did not adsorb either Sr or Cs and showed marginal adsorption of Ce. Furthermore, the MST material is not ideal for waste treatment utilizing column separations. Two CST materials were evaluated. The CST IE-911 material was able to successfully remove Cs and Sr as indicated by batch and column experiments. However, it had marginal Ce and lanthanide adsorptions in batch experiments. From the batch contact, the overall capacity was calculated as 0.032 meq/mL. In the column experiment the lanthanides, including Ce, were not well retained on the column. After 1 bed volume, the raffinate lanthanide concentration was roughly 80% of the feed simulant concentration. After Re broke through the column, the lanthanide raffinate concentration increased to above 100% of the feed concentration, indicating that the lanthanides were somewhat retained on the column. However, the lanthanide behavior in the CST IE-911 column is not ideal for a waste treatment process. The CST R9120 material had high adsorption of Cs, Sr, and Ce in batch contact experiments. In fact, the material had high adsorptions for all elements in the batch simulant and had a calculated loading capacity of 0.091 meq/mL. As such, further studies should focus on CST R9120 material for application in a SHINE waste treatment process with additional attention given to the pretreatment step.

5.0 Recommendations, Path Forward or Future Work

Future experiments should focus on the CST R9120 material due to the high recovery rates observed in the batch contact experiment. Experiments should include column tests to measure breakthrough capacities. Also, these scoping studies maintained similar solution conditions. In application, the SHINE waste stream conditions can fluctuate. As such, solution conditions, like pH, should be varied to measure changes in material adsorption. Furthermore, future studies need to include U in the simulants to measure if any U absorption occurs. Lastly, degradation of the CST material under SHINE waste stream conditions needs to be evaluated. The IE-911 and R9120 can also be dissolved and analyzed to determine if there's different concentrations of elements that make up CST.

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Appendix A Additional MST and CST Data

Data from MST, CST IE-911 and R9120 batch contact experiments are presented as distribution and K_d values in Figure A-1 and A-4. The distribution values were calculated as:

$$D = \frac{C_o - C_f}{C_f}$$

Where D is the distribution value, C_o is the initial concentration, and C_f is the final concentration. The K_d values were calculated as:

$$K_d = \left(\frac{C_o - C_f}{C_f}\right) \times \frac{V}{m}$$

Where V is volume of simulant (mL) and m is mass of ion exchange material (in grams). The K_d values are not true distribution constant values since they were calculated from a mixed element solution. True K_d values would be calculated for an individual element species.

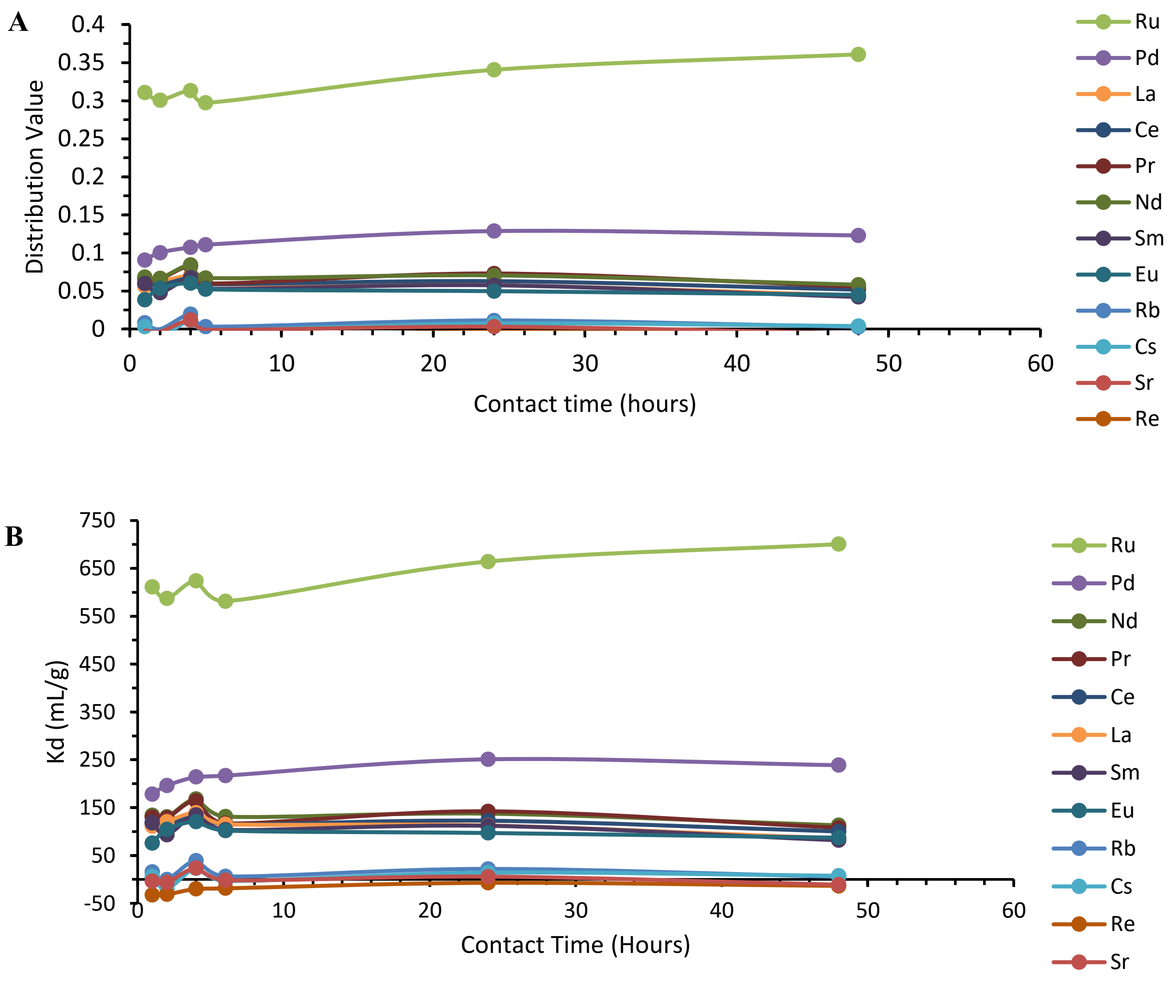

Figure A-1: Data from batch MST experiments presented in alternative way. (A) Distribution value of elements in simulant. (B) Effective K_d of elements absorbed to resin from mixed metal simulant

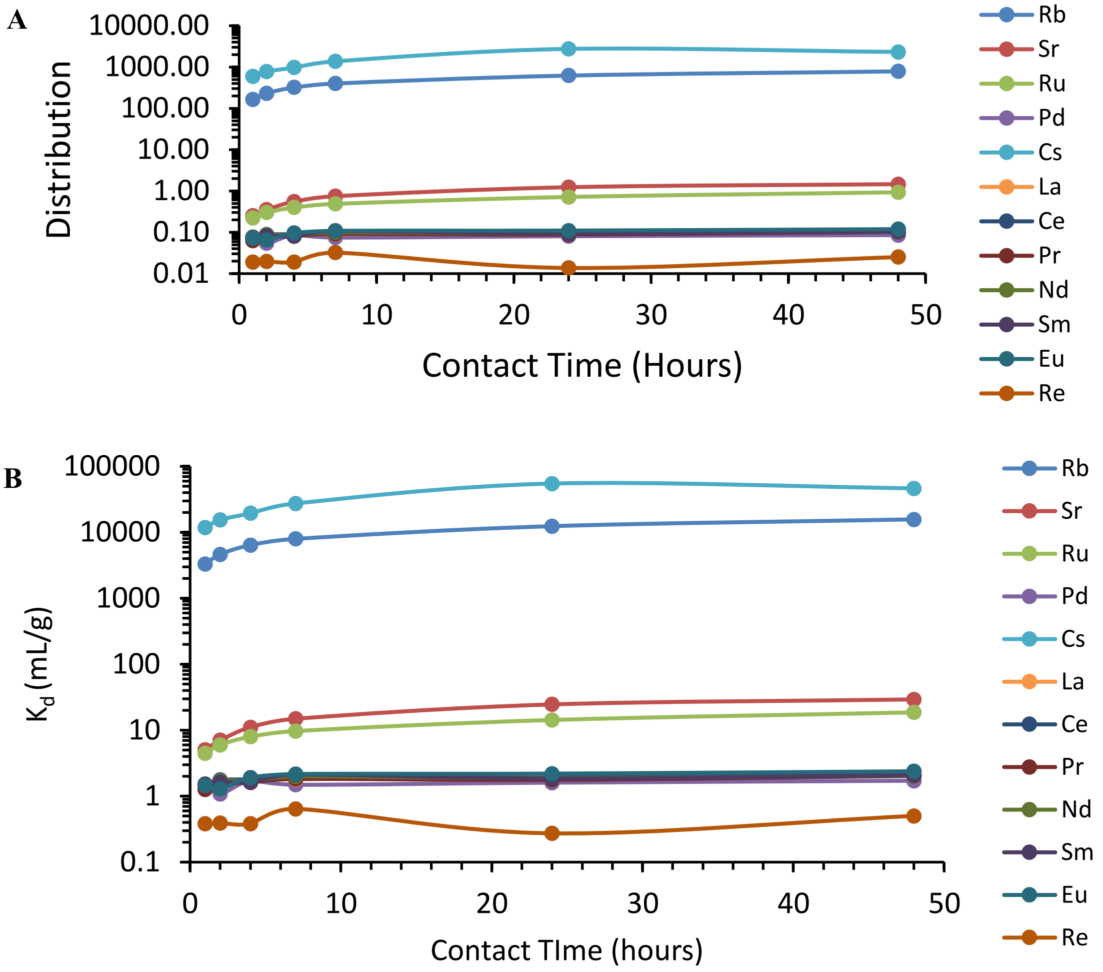

Figure A-2: Data from batch CST IE-911 experiments presented in alternative way. (A) Distribution value of elements in simulant. (B) Effective K_d of elements absorbed to resin from mixed metal simulant

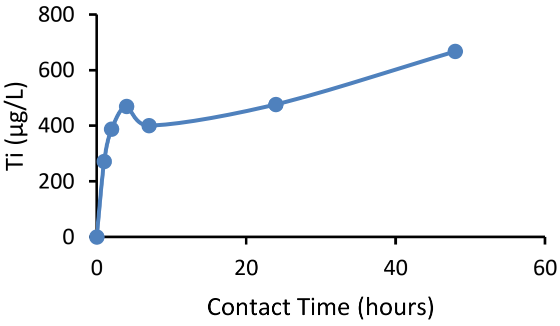

Figure A-3: Titanium concentration found in solution after CST IE-911 batch contact experiment after various time lengths.

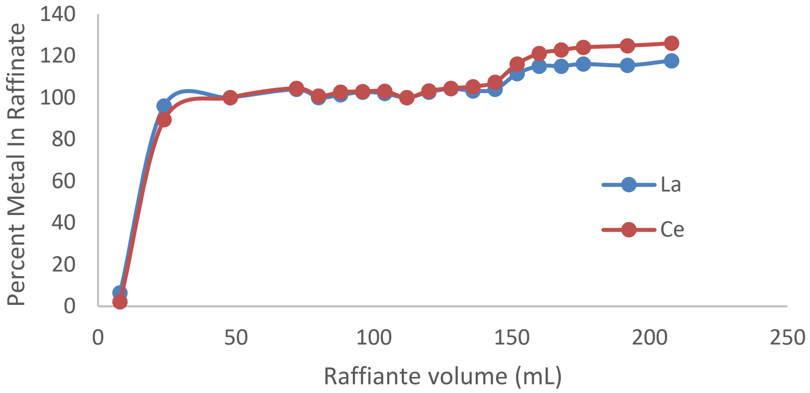

Figure A-4: ICP-OES of La and Ce from CST IE-911 column experiment, confirming trend observed by ICP-MS analysis.

Figure A-4 shows ICP-OES data for the CST IE-911 column experiment. The trend exhibited by the lanthanide series was unexpected and as such the samples were analyzed by ICP-OES to confirm the results by ICP-MS. Of the mixed element solution, only La and Ce were detectable by ICP-OES. The results from ICP-OES confirm results from ICP-MS analysis.

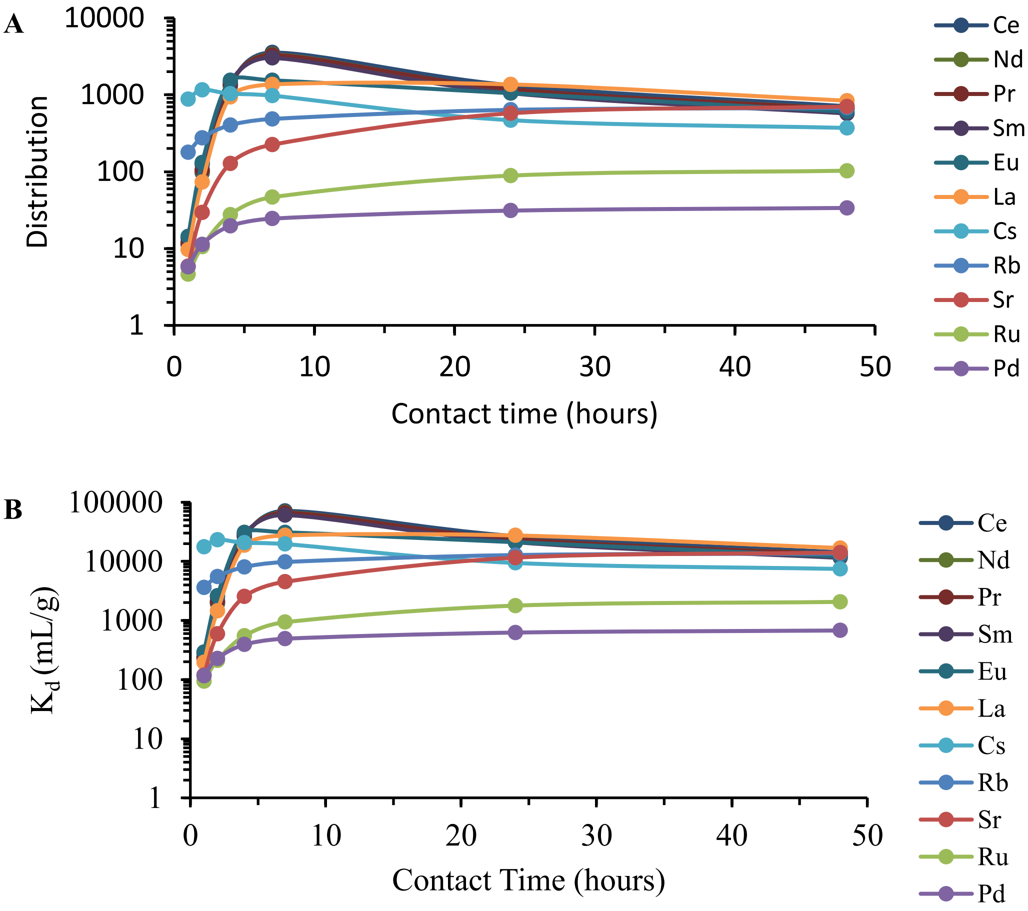

Figure A-5: Data from batch CST R9120 experiments presented in alternative way. (A) Distribution value of elements in simulant. (B) Effective K_d of elements absorbed to ion exchange material from mixed metal simulant