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Small-Scale Ion Exchange Removal of Cesium and Technetium from Hanford Tank 241-AN-102

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SMALL-SCALE ION EXCHANGE REMOVAL OF CESIUM AND TECHNETIUM FROM HANFORD TANK 241-AN-102

March 29, 2000

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SUMMARY

To demonstrate the flow-sheet parameters of the Hanford River Protection Project facility being designed by BNFL, Inc., a sample of Envelope C salt solution was decontaminated for cesium and technetium by ion exchange. The sample, a supernate from Hanford Tank AN-102, had previously been subjected to entrained solids removal and Sr/TRU precipitation and filtration processes before the ion exchange process was performed. Radioactive cesium and technetium removal was accomplished with two sets of ion exchange columns. Each set was connected as a series of two columns in lead-lag configuration. The cesium and technetium removal columns were previously used for decontamination of Envelope A (Hanford Tank AN-103) salt solution and stored thereafter for two months in de-ionized water. The ion exchange process steps for decontamination of Envelope C were the same as previously described[1] and briefly include: 1) resin preconditioning, 2) loading cycle, 3) caustic wash and water rinsing, 4) elution, and 5) resin regeneration. To determine the number of column volumes of feed sample to process, batch distribution coefficients (K_d) and wet resin density of the ion exchange resins were measured. The K_d values were determined over a wide range of cesium and technetium equilibrium concentrations in order to generate equilibrium isotherms. The isotherm data were fitted to a Freundlich-Langmuir Hybrid equation to determine parameters needed for successful modeling of the breakthrough behavior. The modeling results were used to aid in scale up of the design of the ion exchange columns. The decontaminated sample product was used in demonstration of the Low Activity Waste vitrification process, and concentrated cesium and technetium eluate products were blended with other sample streams for High Activity Waste vitrification demonstration.

The cesium distribution coefficient (mean K_d value) for SuperLig® 644 resin with Envelope C (Tank AN-102) was 759 mL/g. Based on resin density of 0.4 g/mL, the estimated number of column volumes to process at 50% breakthrough ("lambda value") was ~ 304. Prior work[2] indicated that the 50% breakthrough would normally be reached at 37% of this value (i.e. 112 column volumes). To obtain an equilibrium isotherm over a wide range of cesium concentrations, small volumes of Envelope C samples were twice re-contacted with fresh resin and additional K_d values were determined at ambient cell temperature (26±1 °C). Also, a primary and duplicate sample were spiked with non-radioactive cesium to obtain K_d values at higher initial concentrations. It was determined that the batch distribution coefficients generally decrease with increasing cesium concentration. As expected, the K_d values decreased at the higher end of the cesium concentration range studied.

The performance of the cesium ion exchange columns exceeded the pretreatment flow-sheet design criteria. Only 11 % breakthrough of cesium was observed after processing 85 column volumes through the lead column. The lag column removed the remaining cesium to achieve an overall decontamination of 99.93 ± 0.03% (DF = 1790) The elution of cesium from the loaded column with 0.5 M HNO_3 was effective. The elution data indicated 78% removal of adsorbed cesium within the first 5 column

volumes and a 99.8% removal of elutable cesium after 19 column volumes. Only trace cesium remains on the resin after elution.

The technetium distribution coefficient (mean K_d value) for SuperLig® 639 was 15.5 mL/g. However, the resin is not expected to remove technetium that is not present as the pertechnetate ion, and this sample contained 77% as non-pertechnetate. The mean K_d value calculated for the pertechnetate ion species was 142 mL/g. The estimated number of column volumes to process at 50% breakthrough (lambda value) for the extractable pertechnetate fraction was 67 column volumes. Our repeated contact experiments show that only 22.5% of the technetium in Envelope C (AN-102) is pertechnetate ion and could readily be extracted by SuperLig® 639 resin. The equilibrium reaction between pertechnetate and other form(s) of technetium in Hanford wastes is not known. Therefore, it is not known if the repeated batch contacts altered the ratio between pertechnetate and other form(s) of technetium present in the AN-102 sample. Similarly, it is not known if the Sr/TRU removal process, which was done prior to the batch and column tests after different storage duration, altered the ratio of ionic states of technetium.

The technetium column loading on SuperLig® 639 showed an immediate breakthrough. This was expected since Envelope C sample contains a relatively large fraction of technetium that is not present as the pertechnetate (anion) species and therefore could not be extracted by SuperLig® 639 resin. The percent removal of technetium from Envelope C (AN-102) salt solution was 45.74 ± 2.08%, which is higher than the expected maximum 22.5%. The cause of the better-than-expected removal of technetium has not been identified. Elution of technetium from the resin with de-ionized water at 1 CV/h was efficient with over 97% of the removable technetium eluted within the first 10 column volumes. Since the concentration of technetium in the eluate could not be monitored online, elution was continued for additional 30 column volumes to ensure less than 1% of the initial concentration of technetium is eluting from the resin. Although this long elution was not originally expected, the extended elution period should not affect the overall waste processing time cycle, since the loading cycle is also longer than projected. Several proposals are considered to improve the elution process.

Characterization results show that the cesium and technetium content in the final decontaminated product were 2.03E-05 Ci/ mole Na and 8.29E-06 Ci/mole Na, respectively. BNFL Inc. plans to vitrify the AN-102 waste, achieving a sodium oxide incorporation of 11.8 wt% in the immobilized low-activity waste (ILAW) form. For this hypothetical glass, the concentration of Cs-137 and Tc-99 are estimated to be 0.2 Ci/m^3 and 0.082 Ci/m^3, respectively. Per US DOE contract specification (2.2.2.8), the average concentrations of ^{137}Cesium (^{137}Cs), ^{90}Strontium (^{90}Sr), and ^{99}Technetium (^{99}Tc) are limited to less than 3, 20, and 0.1 Ci/m^3, respectively.[6] The average concentrations are calculated by summing the actual inventories of each of the above radionuclides in waste packages and dividing by the total volume of waste in these packages. Thus, a minimum of 25% to 30% removal of the ^{99}Tc present in the feed from AN-102 was required. The radionuclide contents of Cs and Tc in decontaminated Envelope C product sample are respectively 0.106 and 0.043 μCi/mL as shown in Table 11. The chemical compositions

in the final decontaminated product and eluate concentrates were as expected with the exception of uranium and TOC, which were concentrated by the cesium ion exchange resin and chloride ions which were concentrated by the technetium resin.

1.0 INTRODUCTION

The pretreatment process for BNFL, Inc.'s Hanford River Protection Project is to provide decontaminated low activity waste and concentrated eluate streams for vitrification into low and high activity waste glass, respectively. The pretreatment includes sludge washing, filtration, precipitation, and ion exchange processes to remove entrained solids, cesium, transuranics, technetium, and strontium. The cesium (Cs-137) and technetium (Tc-99) ion exchange removal is accomplished by using SuperLig® 644, and 639 resins from IBC Advanced Technologies, American Fork, Utah. The resins were shown to selectively remove cesium and technetium (as anionic pertechnetate) from alkaline salt solutions. The efficiency of ion exchange column loading and elution is a complex function involving feed compositions, equilibrium and kinetic behavior of ion exchange resins, diffusion, and the ionic strength and pH of the aqueous solution. A previous experimental program completed at the Savannah River Technology Center[2] demonstrated the conceptualized flow sheet parameters with an Envelope C sample from Hanford Tank 241-AN-107. Those experiments also included determination of Cs and Tc batch distribution coefficients by SuperLig® 644 and 639 resins and demonstration of small-scale column breakthrough and elution. The experimental findings were used in support of preliminary design bases and pretreatment flow sheet development by BNFL, Inc.

The objectives of this ion exchange study are the following:

(1) Determine the batch distribution coefficients (K_d values) and percent removal for cesium (Cs-137) and technetium (Tc-99) ions on SuperLig® ion exchange materials (SuperLig® 644 and 639) with Envelope C from Hanford Tank AN-102 salt solution.

(2) Provide equilibrium isotherm data to determine the parameters for successful modeling of the ion exchange column breakthrough and elution performance.

(3) Demonstrate Cs-137 and Tc-99 loading and elution profiles for Envelope C using SuperLig® 644 and 639 ion exchange resins.

(4) Provide decontaminated (pretreated) product samples for vitrification into low activity waste glass and concentrated eluate product samples for vitrification into high activity waste glass.

(5) Provide information on the composition of caustic displacement and water wash solutions used between loading, elution, and regeneration cycles.

The batch contact experiments were performed to determine the equilibrium distribution coefficients (K_d values) and percent removal for cesium and technetium ions. The K_d values represent a measure of the equilibrium distribution between the aqueous phase and the ion exchanger at a specific solid to liquid ratio. Also the K_d values provide

valuable information on selectivity, capacity, and affinity of an ion exchange material to remove ions from complex aqueous solutions. Specific tests are performed by the addition of a small quantity of ion exchange material into a small volume of salt solution containing a known exchangeable ion(s).

The factors that effect the K_d values and percent removal of the ion(s) include the temperature, initial concentration of the ions in solution, and the solid/liquid ratio during the contact. Duration of the contact must be sufficient to achieve equilibrium or a constant comparative value must be used. The loading capacity for the ion exchange columns is determined from batch K_d experiments conducted at the same conditions as those of the columns. The K_d values are also determined at different equilibrium concentrations to establish a robust ion exchange decontamination process across the broad range of cesium and technetium concentrations that will be encountered as the solution passes through the columns. These data will be used as input to a computer model to determine the scale-up parameters. The computer modeling work will be reported in a future document.

The small ion exchange column experiments were performed in the Intermediate Level Cells at Savannah River Technology Section, allowing remote handling of materials. The experiments were conducted at ambient temperature (26 ± 1 °C) using two sets of cesium and technetium ion exchange columns. Each set consisted of two columns connected in series, one as the lead column and the other as the lag column. The cesium columns were each packed with 6.2 mL of SuperLig® 644 and the technetium columns each contained 5.6 mL of SuperLig® 639 resin. The feed samples were allowed to pass downflow through the lead column and collected from the lag column in small fractions. The lead and lag columns were periodically sampled during the column loading to detect early breakthrough and ensure that the effluent concentration does not exceed 50% of the influent concentration. Once the analytical results were received and confirmed to meet the LAW acceptance criteria, the fractions were combined and further characterized to verify that the effluent met the vitrification process acceptance criteria.

2.0 EXPERIMENTAL

2.1. Materials

Envelope C (Hanford Tank 241-AN-102) salt solution was used for determination of batch distribution coefficients and column breakthrough performance tests. The solution received from Hanford was subjected to entrained solids removal and Sr/TRU precipitation filtration processes reported elsewhere.[5] The sample was diluted during Sr/TRU precipitation filtration processes to ~ 6 M [Na^+]. After dilution, the filtrate had a measured density of 1.234 g/mL and initial Cs-137 and Tc-99 concentrations of 190 μCi/mL and 0.079 μCi/mL, respectively. The diluted sample was used as received for batch distribution coefficient (K_d values) and column performance tests. Table 1 shows the chemical and radionuclide compositions of Envelope C samples from Hanford Tank AN-102 and Tank AN-107. The latter sample was previously tested at Savannah River

Technology Center for cesium and technetium removal with SuperLig® resins.[2] The characterization data for the two samples from Hanford Tanks AN-102 and AN-107 appear to be similar, with the exception of the free hydroxide ion concentration in Tank AN-102, which is ~ 4 times higher than that in Tank AN-107. However, the free hydroxide content of the AN-107 sample shown in Table 1 includes buffering by the organic salts, and the true value of hydroxide ion concentration is much lower. Prior to ion exchange testing with the AN-107 sample, the free hydroxide concentration was increased by 0.3 M by addition of sodium hydroxide solution. Similar to previous work, the chemical reagents used in the AN-102 tests for resin pretreatment and column elution were sodium hydroxide and nitric acid solutions, respectively. These reagents were prepared from ACS certified reagents from Fisher Scientific, Inc.

The ion exchange materials used for cesium and technetium removal from Envelope C sample were SuperLig® 644 (batch # 981020MB48-563) and 639 (batch # 981015DHC720011), respectively. SuperLig® 644 is a polymerized proprietary organic material supplied as 20-70 mesh granules. SuperLig® 639 is composed of polystyrene beads with an attached proprietary organic compound. IBC Advanced Technologies, American Fort, Utah, supplied the resin for testing. The physical properties of the SuperLig® 644 and 639 batches used in this study were measured and the results are presented in Table 2. The F-factor values shown in the last column (Table 2) were obtained by drying a sub-sample of the resin over night in a vacuum oven at 105 °C. The sieve analyses for SuperLig® 644 and 639 resin particles are presented in Tables 3 and 4, and have been previously reported.[4]

2.2 Equipment

The equipment used for the ion exchange column tests was assembled remotely in the shielded Intermediate Level Cells in a laboratory module at the Savannah River Technology Center. Each column had been previously used for the Envelope A sample processing, and were reused after being filled with deionized water and storing for ~2 months. For each experiment, two columns (SuperLig® 644 or 639) were connected in series, with a sampling port between. The columns were made of a medium wall Pyrex glass tubing with an inside diameter of 11 mm and a total length of 30 cm. A plastic coating was applied to the outside walls of the columns to contain shattered glass in case of a rupture. A 200 mesh stainless steel screen was fitted into bottom of each column. Column top assemblies included a fill reservoir, a pressure gauge, a pressure relief valve, and feed inlet ports. The fill reservoir on column top assemblies also served as a vent to permit draining liquid, if necessary. The top assemblies were connected to the glass columns by glass ground joints and tightly fitted screw caps. A ruler affixed to the column wall was used to allow observation of resin bed height changes and liquid level. All tubing connections were made of transparent polyethylene lines that had quick-connect fittings attached to each end; the lines had an inside diameter of 0.125-inch.

The equipment used for batch contact tests consists of 30-mL polyethylene bottles, a Mix-Max® orbital shaker, nylon filter units (0.45 μm), plastic filter holders, and an analytical balance accurate to ± 0.001 g. A house-supplied vacuum and a trap assembly were used during sample filtration. All experiments were performed in the shielded Intermediate Level Cell, allowing remote handling of materials.

2.3. Procedure

2.3.1 Batch contact

All batch contact experiments were conducted in duplicate and at cell ambient temperature of 26 ± 1 °C. A standard batch contact test, followed by two sequential re-contacts of the filtrates with fresh resin and an independent cesium spiked batch test were conducted in the shielded cell. The technetium and cesium tests each used the same batch of "as received resin".

In the "standard" cesium and technetium batch tests, a known volume of salt solution (~18 mL) was added into polyethylene bottle with a known quantity of ion exchange resin (~0.18 g). The phase ratio, solution-to-resin, was ~100:1 for most of the experiments. The bottles containing the solution and the resin were placed on the orbital shaker and gently shaken for 24 ± 1 hours at cell ambient temperature (26 ±1 °C). Control samples (~18 mL of salt solution) were treated in the same way as those of duplicate test samples without the addition of the resin. The concentrations of Cs-137 and Tc-99 in control samples were used as the starting initial concentrations to determine the K_d values and percent removed by the resin. The ambient cell temperature was recorded at the beginning and end of each test. After the contact period, the resin was separated from the sample solution by filtration through individual 0.45-micron nylon filter unit. A 1-mL sub-sample of the filtrate was removed from the cell and analyzed by the Analytical Development Section of the Savannah River Technology Center. Sub-samples were often diluted (10:1) with de-ionized water to reduce the radiation dose rate before the samples are transferred from hot cell to analytical laboratories. All dilutions and measurements were performed based on mass and corrected for density of the solution to ensure accuracy. Concentration of the ion(s) in solution is determined by radioactive counting (Cs-137) or inductively coupled plasma-mass spectroscopy (technetium at mass 99) before and after contact with the resin; the quantity of the ion(s) on the exchanger is determined by difference. No correction for any ruthenium isotope was needed, based on the distribution of masses 100-104. Results were corrected for dilution, where appropriate.

Following the "standard" batch test, two sequential re-contact tests were performed on filtrates. In the first re-contact test, a known volume (~ 12 mL) of the filtrate that had been separated from the resin in the standard test was re-contacted with fresh ion exchange resin (~0.12 g). The fresh resin and the filtrate were gently shaken for 24 ±1 hours. After equilibration, the resin was separated from the solution by filtration under vacuum with a 0.45-micron nylon filter unit. A 1-mL sub-sample of the first re-contact filtrate was

removed from cell and submitted for analysis by the radioactive counting technique for Cs-137 or Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) for Tc-99. The second re-contact was conducted by gently shaking a known volume (~ 10 mL) of the first re-contact filtrate with fresh ion exchange resin (~ 0.1 g) for 24 ±1 hours. After equilibration, the resin was separated from the solution by filtration under vacuum with 0.45-micron nylon filter unit. A 1-mL sub-sample of the second re-contact filtrate was submitted for analysis.

Following the two sequential batch contact tests, small quantities of cesium nitrate (in water) were spiked into a known volume of the original Envelope C salt solution. This was done to increase the initial concentration of Cs in test solutions by a factor of ~10. Because of the size of the spike sample and the density difference of the spike and the test solution, it was necessary to add the test solution into a polyethylene bottle containing the spike. The spiked solution was then shaken manually in the cell to get good mixing before contacting with the resin. The spiked test solution (~ 10 mL) was gently shaken with fresh ion exchange resin (~ 0.1 g) for 24 ±1 hours. A control spike sample was also treated in identical steps as the duplicate test samples. After the contact period, the solution was separated from the resin by filtration under vacuum using 0.45-μm nylon filter. A 1-mL sub-sample of the filtrate (spiked solutions) was removed from the shielded cell and analyzed for total cesium by inductively coupled plasma –mass spectroscopy (ICP-MS). Radioactive counting technique was employed for measurement of Cs-137 content in the samples.

2.3.2 Cesium column operation

The cesium ion exchange columns were previously packed and pretreated in a chemical hood and subsequently used for the AN-103 sample test. The pretreatment of the "as received" SuperLig® 644 resin had been conducted according to the protocol developed by Savannah River Technology Center.[4] This protocol, necessary for new or stored resin, included an acid-caustic cycle that resulted in a fully swollen resin bed in the sodium hydroxide form. After pretreatment, the volumes of the resin in the lead and lag columns were each approximately 6.4 mL .

The pretreatment procedure designated for used of columns that had been stored for more than two weeks storage of the SuperLig® 644 resin in de-ionized water include the following:

1. Pump 3 column volumes (CV) of 0.5 M HNO_3 in 1 hour
2. Pump 3 CV of de-ionized water in 1 hour
3. Pump 6 CV of 0.25 M NaOH in 1 hour
4. Initiate the test by pumping feed at 3 CV/h to the ion exchange columns

Two months after the cesium ion exchange column run with Envelope A (Tank AN-103) salt solution, the columns were pretreated according to above procedure. On the day of the run, 6 CV of 0.25 M sodium hydroxide solution was pumped down flow through the

lead into the lag column at 3 CV/h. After this pre-conditioning, the feed was introduced into the lead column at 3 CV/h at ambient cell temperature (26±1 °C). The effluent was collected in 20 column volume fractions from the lag column. The first two CV of effluent were discarded to prevent dilution of the effluent by residual sodium hydroxide solution. Sub-samples were collected from the lead column after 5 and 10 column volumes of feed had passed through the lead column using a three-way Teflon® valve to direct liquid into the sampling line. The line was flushed for 3 minutes before each 10-minute sample collection. Subsequent sub-samples were collected in 10 column volume intervals. The volume of flush and sample liquid taken from the lead column effluent reduced the total volume of liquid pumped into the lag column and caused episodes of zero flow in the lag column. The flow stops for 13 minutes approximately every 3.3 hours.

The effluent from the lag column was routed to 120-mL polyethylene collection bottles located at the lag column exit. Approximately 20 column volumes of effluent were collected in each bottle. The weight of effluent fractions was measured using an analytical balance; the corresponding volume was determined, dividing the effluent weight by the density of the feed. Sub- samples (~3 mL) were collected from lag column after 20 column volumes of effluent were collected. The samples were counted at-line with a gamma counter to monitor the progress of the column loading. The samples from the lead and lag columns were submitted without dilution to Analytical Development Section of Savannah River Technology Center for analysis by radioactive counting. The loading cycle was terminated when the sample feed was consumed.

After termination of the loading cycle, the liquid hold-up was drained from the columns until the liquid level was 2 cm above the resin. The columns were flushed with 2 TAV (total apparatus volume) of 0.1 M sodium hydroxide, followed by 2 TAV of de-ionized water at 3 CV/h. The flush and rinse solutions were collected from the lag column in 1 CV increments. After the wash and rinse cycle, the two columns were disconnected; the tubing to the lead column was filled with 0.5 M nitric acid. The pump dial set was then adjusted to provide a flow of 1 CV/h through the lead column. Eluate fractions were collected in 2 CV increments. Sub-samples (~1 mL) were collected periodically into dilution bottles containing 10 ml of de-ionized water and analyzed by radioactive counting. The column was eluted with 19 column volumes of 0.5 M HNO_3, followed by 3 column volumes of de-ionized water. The resin was then stored in de-ionized water.

2.3.3 Technetium column operation

The technetium removal from Envelope C (Hanford Tank AN-102) salt solution was accomplished with a set of two ion exchange columns in series. The columns were previously employed for technetium removal from Envelope A (Hanford Tank AN-103) salt solution. The wet volume of the resin in the columns was ~ 5.6 mL and the resin bed height was 5.9 cm (i.e. an L/D ratio of 6.2). The purpose of the lag column was to further decontaminate the feed sample in order achieve the low activity waste vitrification acceptance criteria. The resin was held in place with a small amount of quartz wool and

glass beads. Previous testing with Envelope A had shown that a portion of the resin floats unless restrained.

On the day of the run, the resin was pre-conditioned with 3 CV of 1 M NaOH in one hour. The feed to technetium ion exchange columns, which had been decontaminated for Cs, was pumped down-flow through the lead column at 3 column volumes per hour. The flow rate was adjusted during the early stages of the run to accommodate for the density change between the conditioning solution and the feed. The flow rate was periodically checked by weighing samples collected from the lag column for 10 minutes. Also, the weight of effluent fractions at 20 column volume intervals was measured and using the density of the effluent solutions, the flow rate was verified.

Sub-samples (3 mL) were collected from the lead column after 5 and 10 column volumes of feed were processed through the lead column. Subsequent sub-samples were collected after each 10 column volumes. Sub-samples were collected through a three-way Teflon® valve at the lead column exit sampling line. The effluent from the lead column was routed to sample vials located under the sampling line. The line was flushed for 3 minutes, followed by 10-minute sample collection. Effluent sub-samples (~ 3 mL) were submitted to Analytical Development Section of Savannah River Technology Center for analysis of Tc-99 mass by inductively coupled plasma-mass spectroscopy. When not in sampling mode, the effluent from the lead column was allowed to pass down flow into the lag column that served to further decontaminate the feed sample. Sampling the lead column resulted in a discontinuous flow rate for the lag column, where flow stops for 13 minutes approximately every 3.3 hours. The lag column effluent was collected in 20 column volume increments, then sampled for 10 minutes. Sub-samples (~1 mL) from the lag column were submitted without dilution for analysis to Analytical Development Section of Savannah River Technology Center. The column loading was terminated when the available feed was consumed.

Upon completion of the technetium loading cycle, the feed hold up was drained from the columns until the liquid level was 2 cm above the surface of the glass beads. The columns were then washed in series with 2 TAV (TAV = total apparatus volume; the volume of tubing and liquid headspace in the lead column) of 0.1 M sodium hydroxide, followed by two TAV of de-ionized water. The wash and rinse effluent solutions were collected from the lag column in small increments of 2 column volumes each. Sub-samples (~ 1 mL) were collected each time after one column volume of solution had passed through the lag column.

Upon completion of the column wash and rinse, the lead and lag columns were disconnected. The flow rate through the lead column was adjusted to 1 CV/hr. The eluent (de-ionized water) was pumped at ambient temperature down flow into the lead column and collected in two column volume fractions. The elution was conducted at cell ambient temperature of 26 ± 1°C. Eluate fractions were collected into 30-mL polyethylene bottles. The elution was continued until 40 column volumes of water had passed through the

column. Sub-samples of the eluate were collected between fractions and were analyzed for Tc-99 by ICP-MS.

3.0 RESULTS AND DISCUSSION

3.1. Batch Distribution Coefficients: Batch contact experiments were performed to determine equilibrium distribution coefficients (K_d values) and percent removal for Cs-137 and Tc-99. The batch experiments include the addition of a small quantity of ion exchange materials into a known volume of the salt solution containing known quantities of Cs and Tc ions. The concentration of Cs-137 in solution was determined by radioactive counting before and after contact with the SuperLig® 644 resin. The concentration of Tc-99 before and after contact was determined by inductively coupled plasma-mass spectroscopy (ICP-MS). The quantity of the ions on the resins was determined by difference.

Batch distribution coefficients (K_d values) and percent removal were calculated from the following equations:

$$K_d = \frac{(C_i / C_f - 1)\, V}{(M \cdot F)} \qquad (1)$$

and

$$\%R = \frac{100\,(C_i - C_f)}{C_i} \qquad (2)$$

where C_i is the initial concentration of salt solution, C_f is the final (equilibrium) concentration, V is the volume of liquid sample, M is the "as received" mass of the resin, and F is the mass of dried resin divided by the mass of "as received" resin (i.e., the dry weight correction or F-factor).

The K_d values and percent removals (%R) for cesium are presented in Table 5. The values shown in last column (Table 5) represent the mean of duplicate samples. The percent relative standard deviations (% RSD) for the standard batch contact, first and second re-contacts, and spiked batch test K_d values were 4.07, 0.7, 1.19, and 3.86 %, respectively. This indicates that duplication results were generally good. It is noted that the mean standard K_d value for adsorption of Cs from Envelope C on SuperLig® 644 resin was 759 mL/g. The mean K_d values for first and second re-contacts of prior filtrates with fresh ion exchange resin were 1380 and 1366 mL/g, respectively. Thus, decreasing the concentration of cesium had a significant effect on the K_d values for SuperLig® 644 resin, as expected. For example, the K_ds reduced from 1380 to 759 mg/L and from 759 to 302 mg/L as the cesium concentration in the solution phase increased from 7.74E-06 to 6.33E-5 M and from 6.33E-5 to 1.0 E-3 M, respectively. This may be clearly illustrated by plotting on a log-log scale the K_d values as a function of Na/Cs ratios as shown in Figure 1. It can

be seen that the K_d values increase logarithmically with increasing Na/Cs ratio and the relationship may be represented by the following expression:

$$\log(K_d) = \log(66.638) + 0.17 \log[Na/Cs] \quad (3)$$

It is worth noting that the data fit this logarithmic relation reasonably well with R-squared value of 0.911. Equation (3) gives a quick and simple means for calculating approximate K_d values for this solution. This expression is invalid for solutions containing different concentrations of potassium and free hydroxide, which have the greatest effect on the cesium K_d values for SuperLig® 644 resin. The measured K^+ and OH^- concentrations of the AN-102 feed sample (that had been processed through Sr/TRU removal and filtration were 0.029 and 1.69 M, respectively.

The uptake of cesium by the SuperLig® 644 resin was calculated from concentration of cesium on the resin, the volume of the solution, and the mass of the resin. Figure 2 shows a log-log plot of cesium uptake vs. equilibrium concentration for Envelope C (AN-102) salt solution. Thus, a linearized Freundlich equation may be written as

$$\log q = \log K + \frac{1}{n} \log C_e \quad (4)$$

where q is the cesium uptake per unit mass of resin, C_e is the cesium concentration in the solution phase at equilibrium, The empirical constants, K and n can be determined by a regression package. It can be seen that the equilibrium data for cesium on SuperLig® 644 are very well represented ($R^2 = 0.996$) by the Freundlich expression

$$q = 84.252 C_e^{0.83} \quad (5)$$

A detailed analysis of the data correlation and column modeling will be reported in a separate document.

The loading of cesium on SuperLig® 644 up to 50% breakthrough was estimated from the mean standard K_d value and the wet density[1] of the resin. The Lambda (λ) value for Envelope C (AN-102) loading on the resin was calculated as

$$\lambda = K_d \times \rho = 759 \text{ mL/g} \times 0.4 \text{ g/mL} = 303 \quad (6)$$

Thus, the estimated column volumes of feed sample that can be processed before a 50% breakthrough occurs was 303. Previous work[2] with an Envelope C sample, however, indicated that the breakthrough would occur at 37% of this value. Therefore, the adjusted lambda value for Envelope C was 112 column volumes.

Table 6 shows the mean K_d values and percent removals for SuperLig® 639 resin. This resin will extract only the pertechnetate form of technetium. It can be seen that the mean standard K_d value for total Tc-99 was 15.5 mL/g. It is evident from these results that as the exchangeable pertechnetate species in solution depletes, the amount adsorbed onto the resin decreases. While 87% of the extractable technetium (pertechnetate) was removed during the standard batch contact, the first re-contact appears to have removed the remaining amount. Thus, after the first recontact, ~3550 µg/L of the technetium remained in solution. Since this amount is indistinguishable from that after the second recontact, a second "pertechnetate only" K_d cannot be accurately calculated. The results indicate that a maximum of 1032 µg/L or 22.5% of the total Tc present in Envelope C salt solution was pertechnetate (anion) species. Using this value as the total pertechnetate concentration, the corresponding standard K_d value for the "pertechnetate-only" ion species is 142 mL/g. This assumes that sequential batch contacts do not affect the ratio of pertechnetate to non-pertechnetate. Other factors, such as the presence of Ru-99 masking the Tc-99 analysis are unlikely interferences due to the absence of masses 100-104 amu. Another possibility is that the technetium is all present as pertechnetate, but an unknown chelant prevents adsorption onto the resin. This explanation is also extremely unlikely, as the resin is a very strong absorbent for pertechnetate. If another chelant interfered, it would have to form such a strong bond that <10% of it partitions to the resin. It is possible that the Sr/TRU precipitation process impacted the ratio of pertechnetate to non-pertechnetate, since a powerful oxidant (sodium permanganate) is added to the solution. The batch contacts were performed 4 weeks after the Sr/TRU removal and the column tests were performed 6 weeks after the removal.

The pertechnetate loading capacity at 50% breakthrough was estimated from the batch distribution coefficient and the wet density of the SuperLig® 639 resin. The estimated column volume to process at 50% breakthrough or the Lambda (λ) was

$$\lambda = K_d \times \rho = 142 \text{ mL/g} \times 0.47 \text{ g/mL} = 67 \qquad (7)$$

Thus, up to 67 column volumes of Envelope C (Tank AN-102) salt solution could be processed with SuperLig® 639 resin before 50% breakthrough occurs.

3.2. Cesium Column Loading

The cesium decontamination experiment for Envelope C (AN-102) was conducted ~2 months after completion of the Envelope A (AN-103) cesium column run. Envelope C feed was subjected to Sr/TRU precipitation and filtration prior to the ion exchange column run. The Sr/TRU precipitation was performed ~6 weeks prior to the ion exchange column test. In this regard, the test did not mimic plant operations as it is unlikely that any oxidant from the Sr/TRU precipitation would have still been present after the long storage period. A separate test has been performed to confirm that the resin is not significantly impacted by the Sr/TRU removal process, and will be reported in a subsequent document. The filtrate from Sr/TRU precipitation and filtration was a dark,

brown solution with a measured density of 1.234 g/mL. Analysis of the filtrate solution indicated that the nitrate (NO_3^-), potassium (K^+), and hydroxide (OH^-) concentrations were 1.89E+00, 2.92E-02, and 1.69E+00 M, respectively. The total organic carbon (TOC), uranium and chloride ion concentrations in the filtrate were 2.25E+04, 1.23E+01, and 4.35E-02 mg/L, respectively. The Na^+ concentration measured by inductively coupled plasma – atomic emission spectroscopy (ICP-AES) was 5.98 M, thus giving Na/Cs and K/Cs ratios of 9.42E+04 and 4.62E+02, respectively. The breakthrough data for cesium on SuperLig® 644 resin are shown in Table 7. Figure 3 shows the Cs-137 concentration profile as a function of the effluent column volumes from the lead ion exchange column. It can be seen that an 11% breakthrough occurred for Envelope C (AN-102) solution at 85 column volumes and the guard column exhibited a less than 0.1% breakthrough at 53 column volumes; the run was terminated due to insufficient feed. The breakthrough curves in Figure 3 indicate that the cesium removal from Envelope C (AN-107) was slightly better than from feed sample (AN-102). This may be attributed to slightly higher potassium concentration in this sample (AN-102), compared to AN-107, or the lower Na/Cs mole ratio for the AN-107 sample (6.89E+04) versus that of the AN-102 sample (9.42E+04, assuming Cs^{137} is 25% of the total cesium in each sample).

Figure 4 shows the results of the sodium and free hydroxide analyses for the feed displacement and rinse effluent samples. The feed displacement or wash solution (2 CV of 0.1 M NaOH) reduced the Na^+ and hydroxide concentrations from ~ 6 to 1.3 M and ~1.69 to 0.27 M, respectively. The de-ionized water rinse that followed the feed displacement had residual Na^+ and free hydroxide concentrations of 0.088 M and 0.11 M, respectively, indicating additional water rinsing may be necessary to achieve good displacement of the 0.1M NaOH solution from the column. The percent volume change of the resin between the acid and caustic cycles was in the range 30 to 40%; a 10% reduction of the resin volume observed during the loading (vs. pre-conditioning) showed no detrimental effect on the resin performance. This swelling and shrinking behavior is consistent with previous data.[1] There was no visible indication of resin decomposition as the characteristic brown color of the resin was absent from column effluent, rinse, and regenerate solutions.

3.3. Cesium Column Elution

The results for cesium elution from SuperLig® 644 resin with dilute nitric acid (0.5 M HNO_3) at 1 CV/h are shown in Table 8 and Figure 5. Figure 5 reveals that the dilute nitric acid (0.5 M) was effective for nearly complete removal of cesium from the resin. The elution curve exhibits four regions, namely: neutralization of residual 0.1M NaOH solution, peak, exponential decay, and tailing. The neutralization region at the left bottom end of the curve represents several column volumes of acid eluting very little cesium. During this period, the acid is probably reacting with the residual hydroxide ions in the liquid and the hydrogen ions are starting to replace sodium ions on the resin. The peak region of the curve shows a sharp peak at 5 column volumes, during which almost 78% of the removable cesium in the column was eluted. The exponential decay region that follows the peak region shows a significant drop in cesium concentration within a

few column volumes of the acid. The final region of elution curve shows a small tailing effect. Less than 2% of the initial cesium concentration remained in the eluent after 13 column volumes had passed through the column. The small tailing effect may be attributed to incomplete and uneven cesium loading on the resin. Due to high affinity of the resin, the cesium may have been loaded in a narrow layer on the top of the resin bed. It has been observed that when the resin is fully loaded with cesium, less difficulty is observed in completely eluting the cesium. Also, during elution a rapid shrinkage of the resin could cause a diminished diffusion velocity of cesium ions in the resin and, consequently, lower the rate of elution. The volume of the resin decreased from 6.2 to 4.3 mL at the end of elution.

3.4. Technetium Column Loading

The planned treatment process removes cesium before technetium. Thus, at the conclusion of the cesium column run and after reviewing the cesium concentration profile data and confirming that the decontaminated effluent fractions from guard column met pretreatment radionuclide acceptance limits, all effluent fractions were combined. About 500 mL of the combined solution was pumped down-flow at 3 CV/h into the technetium ion exchange columns. The sodium (Na^+) and nitrate (NO_3^-) concentrations in the feed sample (AN-102) to the ion exchange columns were measured by inductively coupled plasma – atomic emission spectroscopy (ICP-AES) and ion chromatography (IC), respectively. The values measured for Na^+ and $NO3^-$ were 5.98 and 1.89 M, giving a NO_3/Tc ratio of 2.59E+04 in the feed. The nitrate anion ($NO3^-$) primarily competes with the pertechnetate anion to bind on the SuperLig® 639 resin. The results for Tc-99 breakthrough the ion exchange columns are presented in Table 9. The reported results assume all observed m/z 99 is due to technetium and any observed ruthenium spectra are not typical of natural ruthenium, but may be exclusively fission product ruthenium.

Figure 6 shows a plot of the effluent Tc-99 concentration in µg/L as a function of the number of column volumes processed. It can be seen, as expected, that the large portion of non-pertechnetate form of technetium in the feed (up to 70%) had passed through both the lead and lag columns. This result indicates that slightly more was removable than was shown by the batch recontact distribution coefficient results, which revealed that less than 22.5% of the technetium is extracted by the resin. The data in Figure 6 show a significant scatter, the cause of which is not known but may be related to wall effects or dilution effects. The average resin-particle diameter for SuperLig® 639 was reported by the vendor to be approximately 500 µm. With a column inside diameter of 11 mm, the bed diameter is about 20 resin-particle diameters. Although this situation is less than ideal (>30 particle diameters) it was necessary due to the limited sample size and the desire to obtain a breakthrough profile. The results thus yield a conservative estimate of the full-scale column performance. Smaller resin particles could enhance the kinetics of the exchange process and prevent wall effects, but it could not be directly scaled up. Using smaller particle sizes for the full-scale unit would cause a higher pressure drop. The lead column loading data indicated an anomalous low Tc concentration at 20 column volumes. There is no apparent cause for this, but may be the result of analytical or dilution error.

Figure 7 shows the results of the sodium and free hydroxide analyses for the feed displacement and rinse effluent samples from technetium ion exchange columns. The feed displacement or wash solution (3 CV of 0.1 M NaOH) reduced the Na^+ and free hydroxide concentrations from ~ 5.57 and 0.82 M to 0.795 and 0.31 M, respectively. The de-ionized water rinse that followed the feed displacement had reduced the Na^+ and free hydroxide concentrations to 0.11 and 0.1 M, respectively.

3.5. Technetium Column Elution

The results for technetium elution from SuperLig® 639 lead column are presented in Table 10. The elution was performed with de-ionized water at approximately 1 CV/h. The elution profile of the SuperLig 639® is shown in Figure 8. It should be pointed out that the first three column volumes in the abscissa of Figure 8 represent a de-ionized water "rinse" at 3 CV/h, which in effect is the beginning of the elution cycle. Figure 8 shows that the technetium concentration of the eluate reached a peak corresponding to 2.7 times the feed concentration at 6 column volumes, then exponentially decreased to less than 4% of the feed concentration up to 12 column volumes. Elution is considered complete when less than 1% of the total feed technetium concentration remains in the eluate, and this occurred at 22 CV. The elution efficiency for Tc-99 was ~ 97% in the first 10 column volumes of the eluate. This is in agreement with previously observed elution efficiency scheme for SuperLig 639® with Envelope C, in which 98% of Tc-99 loaded on the resin was stripped within 10 column volumes.[5] Due to the unavailability of at-line Tc monitor/analyzer, it was not possible to determine when elution was complete, so the elution was continued for up to 41.9 column volumes and samples were submitted for analysis. The last 18 column volumes of elution volume stripped the technetium remaining on the resin from 0.4 to 0.16% (i.e. 4.0E-03 C/Co to 1.6E-03 C/Co). Although the technetium elution rate for SuperLig® 639 resin seems to be better than expected, the resin was not fully loaded with technetium, and therefore, the data need to be interpreted with caution. Also, basing the elution on "pertechnetate only", rather than total technetium, shows that the pertechnetate elution was not complete until 41.9 column volumes.

3.6. Product Characterization

The characterization results for decontaminated product and concentrated cesium and technetium eluate products are shown in Tables 11, 12, and 13, respectively. In the tables, the radionuclides, elemental compositions, and anion compounds of the final products were also presented. The percent removal of Cs-137 and Tc-99 from Envelope C (AN-102) salt solution were 99.93± 0.03% and 45.7 ± 2.08%, respectively. The percent removals of cesium and technetium were calculated from the mean concentrations of two measurements each for Cs-137 and Tc-99 both in the feed and product solutions. The standard deviation (σ) for the percent removal was calculated from equation (8). The error in the percent removal was calculated assuming a variance of two standard deviations (2σ) in the average.

$$\sigma^2 = \left(\frac{100}{C_i}\right)^2 x\left[\left((S_{Cf})^2 + \left(\frac{C_f}{C_i}\right)^2 x(S_{Ci})^2\right)\right] \quad (8)$$

where S_{cf} = standard deviation of final concentration,
S_{ci} = standard deviation of initial concentration,
C_f = final concentration, and C_i = initial concentration.

The concentrations of Cs-137 and Tc-99 in the decontaminated product were 1.10E-01 and 4.3E-02 μCi/mL, respectively. The concentration of both radionuclides are lower than the maximum content allowed by immobilized Law activity waste (ILAW) acceptance criteria. The chemical species in the final decontaminated product were generally as expected. Uranium concentration in the final decontaminated product by inductively coupled plasma - mass spectroscopy (ICP-MS) was only 6.2E-01 mg/L, which indicates a significant removal of uranium as compared to initial concentration of 1.23E+01 mg/L. The concentration of uranium in the composite eluate solution was 1.68E+01 mg/L.

The results for characterization of cesium and technetium eluate products are shown in Tables 12 and 13. The chemical species in the eluate products were generally as expected, with the exception of uranium, total organic carbon and chloride ion. The chloride appears to be concentrated both by the cesium and technetium ion exchange resin. Observations of high chloride ion concentration by the SuperLig® resins was previously reported by us. [4] Similarly, concentration of uranium by SuperLig® 644 from Envelope C has also been observed previously. [4] However, the amount of uranium in this eluate is much lower than from tank AN-107. Presumably, the initial concentration of uranium is much lower in this AN-102 sample, although it was not directly measured in the previous work. In addition, the Sr/TRU removal process further reduced the uranium concentration in this AN-102 sample. Chromium has often been observed to be concentrated in cesium eluate, but was not observed in this case, based on comparison to other residuals from the feed such as aluminium.

The total organic carbon (TOC) in the cesium and technetium eluate products were reported as 470 and 221 mg/L, respectively. However, duplicate samples varied by a factor of 2.8 for both cesium and technetium eluates. The poor precision between the samples and duplicates for TOC analyses indicates the difficulty in measuring this parameter in this complex matrix.

3.7 Spent Resin Digestion and Analyses

The spent SuperLig® 644 and 639 resins that were used for decontaminating Envelope A (AN-103) and C (AN-102) salt solutions were digested and the residual analytes were measured by ICP-ES, ICP-MS, AA, and gamma scan. Prior to resin digestion, the resins were regenerated with 6 CV of 1.0 M NaOH (SuperLig® 639) and

0.25M NaOH (SuperLig® 644), then rinsed with 9 CV of de-ionized water. The resins were then slurried out from the columns into filter units and subjected very briefly to vacuum just long enough to remove standing liquids. The resin digestion was performed by heating 0.1 g of the resin with 3 ml of conc. HNO_3 in the Parr bomb at 150 °C for 1.5 hours to destroy the organic. The digested solution was then cooled to ambient temperature and diluted with water to 10 mL final volume.

The analytical results for digested resins are presented in Table 14. The residual radionuclides (Cs-137 and Tc-99) on SuperLig® 639 resin were as expected. However, the Cs-137 content in the spent SuperLig 644 resin was 7.4 µCi/g despite the fact that the elution data showed less than 1% of initial Cs in Envelope C salt solution (i.e. 1.80 µCi/mL) was in the eluate at the end of elution. The residual concentrations of RCRA characteristically hazardous elements were below the theoretical TCLP limit. The theoretical TCLP limit was calculated assuming all of the metal ion would dissolve in the TCLP test and a solid: liquid ratio of 50 g to 1 L.

4.0 CONCLUSION

Cesium and technetium removal from Envelope C salt solution from Hanford Tank AN-102 was accomplished using IBC manufactured SuperLig® ion exchange resins. The cesium distribution coefficient for SuperLig® 644 resin was 759 mL/g. The projected column volume of feed to process at 50% breakthrough was estimated as 303 column volumes, although prior work indicated that only 37% of this value (112 CV) would be reached. The resin achieved 11% breakthrough by the time the available feed sample was consumed at 85 column volumes. Nitric acid (0.5 M) was effective to elute cesium from the resin with less than 0.2% of the cesium remaining in the effluent from the column at 18 column volumes. The elution profile exhibited a small tailing effect. The percent removal of cesium from Envelope C by two-column (lead/lag) configuration containing SuperLig® 644 resin was 99.93± 0.03%.

The distribution coefficient for technetium on SuperLig® 639 resin was 16 mL/g. The calculated K_d value for the pertechnetate (anion) form was 142 mL/g. The projected column volume to process at 50% breakthrough for extractable pertechnetate was 67. As expected, an immediate breakthrough of the non-extractable, non-pertechnetate form of technetium was observed through both lead and lag columns. Elution of technetium from the resin with de-ionized water at 1 CV/h was efficient with 97% recovery up to 10 column volumes and less than 1% of Tc remaining on the resin after 22 column volumes. The percent removal of technetium from Envelope C by SuperLig® 639 was 45.74 ± 2.08 % . This is approximately twice the percent pertechnetate in the salt solution (22.5%) determined through sequential batch re-contacts.

Characterization results showed that the Cs/Na and Tc/Na mole ratios in the final decontaminated product were 2.03E-05 Ci/mole Na and 8.29E-06 Ci/mole Na, respectively. These results are within the range of Cs and Tc limits for the immobilized

law activity waste glass[6]. The chemical compositions in the final decontaminated product and eluate concentrates were as expected with the exception of uranium, TOC, and chloride ions which were concentrated by the cesium ion exchange resin and later eluted by nitric acid. The residual analytes in the digested spent resins were generally below their detection limits, with the exception of Cs in the spent SuperLig® 644 resin.

5.0 APPROVALS

Design Check

M. C. Thompson 5/9/00

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Actinide Technology Section

Date

6.0 REFERENCES

6.0 REFERENCES

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Table 1. Small Envelope C Filtrate characterization Data

Constituent ions	Tank-241-AN-102 concentration [M]	Tank-241-AN-107 Concentration [M]
Cations, M		
[Na^+]	5.98	5.71
[Al]	2.72E-01	2.1E-3
[Ca]	5.14E-03	8.6E-3
[Cr]	1.69E-03	1.1E-3
[Pb]	4.03E-04	7.6E-3
[Zn]	< 5.05E-05	1.0E-4
[K^+]	2.92E-02	2.6E-2
U-238 mass (mg/L)	1.23E+01	n. m
Anions, M		
[NO_3^-]	1.89E+00	2.48E+00
[NO_2^-]	8.32E-01	8.29E-01
[Cl^-], by IC	4.35E-02	3.3E-02
[F^-], by IC	4.11E-02	6.3E-02
[SO_4^{-2}]	6.20E-02	6.3E-02
[PO_4^{-2}]	< 1.26E-02	6.3E-02
formate	nm	1.33E-01
OH^- (Free)	1.69E+00	3.94E-01
Radionuclides, uCi/mL		
Cs-137	1.90E+02	246*
Am-241	1.50E-02	< 0.29*
Sr-90	1.65E+00	70.3*
alpha	1.78E-02	0.55*
Tc (calc'd, mass 99)	7.80E-02	0.051*
Total carbon		
TIC	0.65 M	0.95 M*
TOC	1.88 M	2.68 M*
Density (g/mL)	1.23E+00	1.276

* Characterization before dilution, corrected for dilution by calculation

Table 2. Physical properties of "as received" SuperLig® resin

Resin	Batch #	Particle density (g/mL)	Bulk density (g/mL)	% Water content	F-Factor
SuperLig 644	981020MB48-563	1.549	0.758	9.6	0.904
SuperLig 639	981015DHC-720011	1.147	0.468	1.3	0.987

Table 3. Sieve analysis of SuperLig 639 resin batch batch #981015DHC-720011

		wt. of fractions (g)		wt. %		ave. wt. %
Screen Mesh	Opening	Sample 1	Sample 2	Sample 1	Sample 2	(%)
10	> 200 mm					
30	> 600	4.37	4.169	87.35	67.80	77.57
40	> 425	0.574	0.722	11.47	11.74	11.61
50	> 300	0.035	0.054	0.70	0.88	0.79
70	> 212	0.01		0.20		0.10
80	> 180	0.007				
100	> 150					
230	> 62					
	total	5.00	4.945	99.7	98.7	99.2

wt. original sample # 1 = 5.003 g; wt. sample #2 = 5.008 g

Table 4. Sieve analysis of SuperLig 644 resin batch batch # 981020mb48-563

		wt. of fractions (g)		wt. %		ave. wt. %
Screen Mesh	Opening	Sample 1	Sample 2	Sample 1	Sample 2	(%)
10	> 200 mm	0	0	0		0
30	> 600	2.9342	2.8406	59.77	54.89	57.33
40	> 425	1.1143	1.2821	22.70	24.77	23.73
50	> 300	0.6389	0.7456	13.01	14.41	13.71
70	> 212	0.2115	0.3072	4.31	5.94	5.12
80	> 180	0.0105	0	0.21	0.00	0.11
100	> 150					
230	> 62					
	total	4.9094	5.1755	99.8	100	99.9

wt. original sample # 1 = 5.003 g; wt. sample #2 = 5.1758 g

Table 5. Batch distribution coefficient data for Cs on SuperLig® 644 resin

Sample ID	solution mass (g)	resin mass (g)	solution vol.(mL)	phase ratio	[Cs –137] (uCi/mL)	[Cs –137] Removal (uCi/mL)	Kd (mL/g)	ave. Kd (mL/g)	ave. (% R)
Standard Kd									
BNF-M1800-AN102-1	14.37	na	11.68	na	199.3	na	na	na	na
BNF-M1800-AN102-2	14.726	na	11.97	na	181.4	na	na	na	na
BNF-M1800-S644-1	14.799	0.1207	12.01	99.5	23.52	166.8	780.8		
BNF-M1800-S644-1D	14.552	0.1204	11.81	98.1	24.43	165.9	737.1	758.9	88
1st. Recontact Kd									
BNF-M1800-S644-1R-1	10.33	0.1003	8.398	83.7	1.486	22.034	1373		
BNF-M1800-S644-1R-ID	10.991	0.1004	8.936	89.0	1.619	22.81	1387	1380	94
2nd. Recontact Kd									
BNF-M1800-S644-2R-1	6.18	0.0513	5.024	97.9	0.106	1.380	1410		
BNF-M1800-S644-2R-2	6.203	0.0509	5.043	99.1	0.124	1.495	1321	1366	93
Cs spiked Kd test‡					Cs (ug/L)	Cs (ug/L)			
BNF-C320-S644-spk-1	12.73	0.1003	10.35	103	57402	147458	293.3		
BNF-C320-S644-spk-1D	12.78	0.1009	10.39	103	55093	149767	309.7	301.5	73
BNF-C320-Cs-spk-F2	11.47	na	9.324	na	204860	na			

‡~ 20 µL of 1 M Cs(NO3) was added into 30 mL of the diluted AN-102 sample.
F-factor = 0.904; Temperature = 26 ± 1°C; contact time = 24 ±1 h; batch #: 981020MB48-563
na = not applicable

Cs Isotopic ratio	avg. values (ppb)
Cs-133	4.03E+03
Cs-135	1.35E+03
Cs-137	1.92E+03
Total Cs	7.28E+03

Table 6. Batch distribution coefficients for technetium on SuperLig® 639 resin

Sample ID	Solution wt. (g)	Resin mass (g)	Volume (mL)	phase ratio	[Tc –99] (μg/L)	Tc-99 Removed (ug/L)	Kd (mL/g)	ave. Kd (mL/g)	% (R)
Standard Kd									
BNF-M1800-F-1	14.37	NA	11.68	na	4713	na	na	na	na
BNF-M1800-F-1D	14.726	NA	11.9	na	4451	na	na	na	na
BNF-M1800-S39-1	15.173	0.1204	12.34	102.5	3951	632	16.60		
BNF-M1800-S39-1D	15.474	0.1203	12.58	104.6	4032	550	14.45	15.53	87
1st. Recontact Kd									
BNF-M1800-S39-1R-1	10.913	0.1007	8.872	88.11	3641	309	na	na	na
BNF-M1800-S39-1R-1D	11.076	0.1004	9.005	89.69	3498	534	na	na	na
2nd. Recontact Kd									
BNF-M1800-S39-2R-1	6.160	0.0510	5.008	98.20	3548	na	na	na	na
BNF-M1800-S39-2R-1D	6.162	0.0510	5.01	98.23	3553	na	na	na	na

F-factor =0.987; Temperature = 26 ± °C; contact time = 24 ±1 h; batch #: 981015DHC-720011
† 0.5 mL of 20 μCi/mLTc was added into 30 mL of Hanford AN-102 sample.

na = not applicable

Table 7. Cesium Ion Exchange column loading profiles

Lead column sample ID	# CV processed (lead column)	Cs-137 Effluent (μCi/mL)	Concentration profile (C/Co)
BNF-C320-CsIX-Cr4-F-1	na	182.24	na
BNF-C320-CsIX-Cr4-F-2	na	181.95	na
BNF-C320-CsIXEFF-Cr4-LCS1	5	4.097E-02	2.25E-04
BNF-C320-CsIXEFF-Cr4-LCS2	10	1.084E-01	5.96E-04
BNF-C320-CsIXEFF-Cr4-LCS3	20	4.495E-01	2.47E-03
BNF-C320-CsIXEFF-Cr4-LCS4	30	1.692E+00	9.29E-03
BNF-C320-CsIXEFF-Cr4-LCS5	40	7.362E+00	4.04E-02
BNF-C320-CsIXEFF-Cr4-LCS6	50	9.085E+00	4.99E-02
BNF-C320-CsIXEFF-Cr4-LCS7	60	8.236E+00	4.52E-02
BNF-C320-CsIXEFF-Cr4-LCS8	70	1.555E+01	8.54E-02
BNF-C320-CsIXEFF-Cr4-LCS9	80	1.912E+01	1.05E-01
BNF-C320-CsIXEFF-Cr4-LCS10	85	2.107E+01	1.16E-01
Lag column sample ID			
BNF-C320-CsIXEFF-Cr4-GCV-s1	13	1.149E-02	6.31E-05
BNF-C320-CsIXEFF-Cr4-GCV-s2	27	5.287E-02	2.90E-04
BNF-C320-CsIXEFF-Cr4-GCV-s3	39	3.741E-01	2.05E-03
BNF-C320-CsIXEFF-Cr4-GCV-s4	53	2.273E-01	1.25E-03

Table 8. Cesium elution profile for lead column

Lead column sample ID	# CV eluted (lead column)	Cs in eluate (μCi/mL)	Concentration profile (C/Co)
BNF-C320-CsIX-Eluate-Cr4-LCS1	2.02	7.54E-02	4.14E-04
BNF-C320-CsIX-Eluate-Cr4-LCS2	3.19	1.05E+01	5.75E-02
BNF-C320-CsIX-Eluate-Cr4-LCS3	5.29	1.54E+04	8.48E+01
BNF-C320-CsIX-Eluate-Cr4-LCS4	8.70	2.96E+03	1.62E+01
BNF-C320-CsIX-Eluate-Cr4-LCS5	11.84	5.81E+00	3.19E-02
BNF-C320-CsIX-Eluate-Cr4-LCS6	14.09	2.78E+00	1.53E-02
BNF-C320-CsIX-Eluate-Cr4-LCS7	16.36	1.26E+00	6.91E-03
BNF-C320-CsIX-Eluate-Cr4-LCS8	18.78	3.08E-01	1.69E-03

Table 9. Technetium ion exchange column loading profiles for small Envelope C (AN-102) salt solution on SuperLig 639 (Analysis by ICP-MS)

Lead column sample ID	# CV processed (lead column)	Tc-99 Effluent (μg/L)	Concentration profile (C/Co)
BNF-C320-TcIX-Feed-1	na	4.73E+03	na
BNF-C320-TcIX-Feed-2	na	4.63E+03	na
BNF-C320-TcIXEFF-Cr4-LCS1	5	2.154E+03	4.61E-01
BNF-C320-TcIXEFF-Cr4-LCS2	10	2.884E+03	6.17E-01
BNF-C320-TcIXEFF-Cr4-LCS3*	20	2.005E+03	4.29E-01
BNF-C320-TcIXEFF-Cr4-LCS4	30	2.863E+03	6.12E-01
BNF-C320-TcIXEFF-Cr4-LCS5	40	2.901E+03	6.21E-01
BNF-C320-TcIXEFF-Cr4-LCS6	50	3.399E+03	7.27E-01
BNF-C320-TcIXEFF-Cr4-LCS7	60	3.123E+03	6.68E-01
Guard column sample ID			
BNF-C320-TcIXEFF-Cr4-1CV-s1	13	3.036E+03	6.49E-01
BNF-C320-TcIXEFF-Cr4-2CV-s2	36	3.113E+03	6.66E-01
BNF-C320-TcIXEFF-Cr4-3CV-s3	47	3.247E+03	6.94E-01
BNF-C320-TcIXNaOH-Cr4-1CVS	48	9.879E+02	2.11E-01

Table 10. Technetium elution profile (lead column)

Sample ID Lead column	# CV eluate (lead column)	Tc-99 eluted (µg/L)	Concentration profile (C/Co)
BNF-C320-TcIX-Eluate-Cr4-LTc1	1.96	4.95E+03	1.06E+00
BNF-C320-TcIX-Eluate-Cr4-LTc2	4.04	1.09E+04	2.34E+00
BNF-C320-TcIX-Eluate-Cr4-LTc3	5.98	1.26E+04	2.70E+00
BNF-C320-TcIX-Eluate-Cr4-LTc4	8.63	2.21E+03	4.73E-01
BNF-C320-TcIX-Eluate-Cr4-LTc5	9.66	6.85E+02	1.47E-01
BNF-C320-TcIX-Eluate-Cr4-LTc6	11.90	1.92E+02	4.10E-02
BNF-C320-TcIX-Eluate-Cr4-LTc7	14.64	9.98E+01	2.13E-02
BNF-C320-TcIX-Eluate-Cr4-LTc8	18.05	1.54E+02	3.28E-02
BNF-C320-TcIX-Eluate-Cr4-LTc9	21.49	4.45E+01	9.53E-03
BNF-C320-TcIX-Eluate-Cr4-LTc10	24.78	1.88E+01	4.02E-03
BNF-C320-TcIX-Eluate-Cr4-LTc11	28.03	1.37E+01	2.93E-03
BNF-C320-TcIX-Eluate-Cr4-LTc12	31.03	1.23E+01	2.63E-03
BNF-C320-TcIX-Eluate-Cr4-LTc13	34.18	1.60E+01	3.42E-03
BNF-C320-TcIX-Eluate-Cr4-LTc14	36.25	7.56E+00	1.62E-03
BNF-C320-TcIX-Eluate-Cr4-LTc15	37.98	7.98E+00	1.71E-03
BNF-C320-TcIX-Eluate-Cr4-LTc16	39.73	8.61E+00	1.84E-03
BNF-C320-TcIX-Eluate-Cr4-LTc17	41.18	1.19E+01	2.54E-03
BNF-C320-TcIX-Eluate-Cr4-LTc18	41.92	7.42E+00	1.59E-03

Table 11. Characterization for Envelope C decontaminated product

Sample ID		AN-102-Prod-1		AN-102-Prod-2		Avg. value
Cs-137 (μCi/mL)		1.16E-01		9.55E-02		1.06E-01
Co-60 (μCi/mL)		4.27E-02		4.01E-02		4.14E-02
Eu-154 (μCi/mL)		2.92E-02		2.94E-02		2.93E-02
Eu-155 (μCi/mL)		1.73E-02		1.81E-02		1.77E-02
Tc-99 (mg/L)		2.56E+00		2.52E+00		2.54E+00
massTh-232, mg/L		8.76E-01		1.05E+00		9.65E-01
mass U-233	<	7.19E-04	<	7.15E-04	<	7.17E-04
mass U-235	<	7.19E-04	<	7.15E-04	<	7.17E-04
mass U-236	<	7.19E-04	<	7.15E-04	<	7.17E-04
mass Np-237		8.25E-02		7.79E-02		8.02E-02
mass Pu/U-238		6.25E-01		6.17E-01		6.21E-01
mass Pu-239		4.31E-02		1.81E-02		3.06E-02
mass Pu-240	<	7.19E-04	<	7.15E-04	<	7.17E-04
mass Am/Pu-241	<	7.19E-04	<	7.15E-04	<	7.17E-04
mass Am-243	<	7.19E-04	<	7.15E-04	<	7.17E-04
mass Cm-245	<	7.19E-04	<	7.15E-04	<	7.17E-04
K (AA), mg/L		9.09E+02		9.16E+02		9.12E+02
Carbon (mg/L)						
inorganic (TIC)		7.06E+03		6.18E+03		6.62E+03
Organic (TOC)		2.57E+04		3.17E+04		2.87E+04
OH^- (Free)		8.49E-01		8.10E-01		8.3E-01
ICP-ES (mg/L)						
Al		6.64E+03		6.49E+03		6.57E+03
B		2.30E+01		2.11E+01		2.21E+01
Ba	<	1.11E+00	<	1.10E+00	<	1.11E+00
Ca		2.05E+02		2.00E+02		2.02E+02
Cd		2.59E+01		2.46E+01		2.52E+01
Co	<	3.32E+00	<	3.31E+00	<	3.32E+00
Cr		6.92E+01		6.70E+01		6.81E+01
Cu		6.46E+00		5.87E+00		6.17E+00
Fe		3.43E+00		3.61E+00		3.52E+00
La	<	4.43E+00	<	4.41E+00	<	4.42E+00
Li	<	1.66E+00	<	1.65E+00	<	1.66E+00
Mg		2.84E+00		3.17E+00		3.00E+00
Mn	<	5.54E-01	<	5.52E-01	<	5.53E-01
Mo		3.01E+01		2.87E+01		2.94E+01
Na		1.22E+05		1.17E+05		1.20E+05
Ni		1.55E+02		1.47E+02		1.51E+02
P		8.11E+02		8.17E+02		8.14E+02
Pb		5.58E+01		5.77E+01		5.68E+01
Re	<	2.83E+01	<	2.81E+01	<	2.82E+01
Si		6.36E+01		5.62E+01		5.99E+01
Sn		2.45E+01		2.57E+01		2.51E+01
Sr		8.42E+01		7.92E+01		8.17E+01
Ti	<	1.11E+00	<	1.10E+00	<	1.11E+00
V	<	2.22E+00	<	2.21E+00	<	2.21E+00
Zn		9.37E+00		9.33E+00		9.35E+00
Zr		5.72E+00		3.58E+00		4.65E+00

Table 12. Characterization data for cesium eluate product

Sample ID		AN-102-CsELU-1		AN-102-CsELU-2		Avg. values
Cs-137 (μCi/mL)		5.12E+02		5.10E+02		5.11E+02
Tc-99 (mg/L)		6.21E-02		5.08E-02		5.64E-02
total alpha (dpm/ml)		5.04E+07		5.10E+07		5.07E+07
K (AA), mg/L		8.91E+1		7.10E+1		8.01E+1
Carbon (mg/L)						
inorganic (TIC)	<	2.16E+1	<	2.13E+1	<	2.15E+1
Organic (TOC)		6.94E+2		2.45E+2		4.70E+2
IC (mg/L)						
NO_3^-		2.30E+04		2.18E+04		2.24E+04
NO_2^-	<	2.16E+03	<	2.13E+03	<	2.15E+03
PO4	<	2.16E+03	<	2.13E+03	<	2.15E+03
SO4	<	1.08E+03	<	1.07E+03	<	1.07E+03
Oxalate	<	2.16E+03	<	2.13E+03	<	2.15E+03
Cl- by IC	<	4.32E+02	<	4.27E+02	<	4.30E+02
F- by IC	<	4.32E+02	<	4.27E+02	<	4.30E+02
Cl^- (ISE)		5.87E+03		na		5.87E+03
F^- (ISE)	<	2.16E+01		na	<	2.16E+01
Chem-check						
U (mg/L)		1.77E+01		1.58E+01		1.68E+01
ICP-ES (mg/L)						
Al		2.50E+02		2.85E+02		2.68E+02
B		2.03E+02		2.43E+02		2.23E+02
Ba		5.62E-01		5.42E-01		5.52E-01
Ca		6.21E+01		6.94E+01		6.57E+01
Cd		1.23E+00		8.23E-01		1.03E+00
Co	<	1.30E+00	<	1.30E+00	<	1.30E+00
Cr		9.80E+00		8.82E+00		9.31E+00
Cu		2.98E+01		2.94E+01		2.96E+01
Fe		7.27E+00		5.96E+00		6.61E+00
La	<	1.73E+00	<	1.73E+00	<	1.73E+00
Li	<	6.49E-01	<	6.50E-01	<	6.49E-01
Mg		8.48E+00		9.42E+00		8.95E+00
Mn		6.70E-01		5.85E-01		6.28E-01
Mo	<	4.32E-01		4.77E-01	<	4.55E-01
Na		1.50E+03		1.47E+03		1.48E+03
Ni		4.50E+01		4.58E+01		4.54E+01
P	<	9.51E+00	<	9.53E+00	<	9.52E+00
Pb	<	9.30E+00	<	9.32E+00	<	9.31E+00
Re	<	1.10E+01	<	1.10E+01	<	1.10E+01
Si		9.46E+01		1.02E+02		9.85E+01
Sn	<	4.32E+00	<	4.33E+00	<	4.33E+00
Sr		1.04E+00		1.10E+00		1.07E+00
Ti		4.97E-01	<	4.33E-01	<	4.65E-01
V	<	8.65E-01	<	8.67E-01	<	8.66E-01
Zn		3.61E+00		3.79E+00		3.70E+00
Zr	<	1.08E+00	<	1.08E+00	<	1.08E+00

Table 13. Characterization Data for Technetium Eluate Product

Sample ID		A-103-TcELU-1		A-103-TcELU-2		Avg. values
Cs-137 (μCi/mL)		1.08E-02		1.88E-02		1.48E-02
Tc-99 (mg/L)		1.95E+00		1.94E+00		1.95E+00
total alpha (dpm/ml)		4.55E+03		4.30E+03		4.43E+03
K (AA), mg/L		1.01E+1		8.20E+0		9.14E+00
inorganic (TIC)	<	2.23E+1	<	6.98E+1	<	4.60E+1
Organic (TOC)		3.30E+2		1.12E+2		2.21E+02
IC (mg/L)						
NO_3^-		2.23E+03	<	2.24E+03		2.24E+03
NO_2^-		2.23E+03		2.24E+03		2.24E+03
PO_4	<	2.23E+03	<	2.24E+03	<	2.24E+03
SO_4	<	1.12E+03	<	1.12E+03	<	1.12E+03
Oxalate	<	2.23E+03	<	2.24E+03	<	2.24E+03
Cl^-		6.34E+03		5.97E+03		6.15E+03
F^-	<	2.23E+01	<	2.24E+01	<	2.24E+01
Cl (ISE)		6.34E+03		5.97E+03		6.15E+03
F (ISE)	<	2.23E+01	<	2.24E+01	<	2.24E+01
U (mg/L)	<	2.23E-01	<	2.24E-01	<	2.24E-01
ICP-ES (mg/L)						
Al		2.11E+01		2.22E+01		2.16E+01
B	<	2.68E+00	<	2.69E+00	<	2.68E+00
Ba		1.05E+00		9.86E-01		1.02E+00
Ca		9.28E+00		7.93E+00		8.60E+00
Cd		1.07E+00		8.74E-01		9.72E-01
Co		2.54E+00		2.22E+00		2.38E+00
Cr		2.34E+00		1.70E+00		2.02E+00
Cu		1.76E+00		1.59E+00		1.68E+00
Fe		2.21E+00		2.06E+00		2.13E+00
La		9.79E+00		8.71E+00		9.25E+00
Li		1.45E+00		1.39E+00		1.42E+00
Mg		1.43E+00		1.30E+00		1.36E+00
Mn		3.57E-01		2.91E-01		3.24E-01
Mo		9.37E-01		5.60E-01		7.48E-01
Na		3.16E+02		4.18E+02		3.67E+02
Ni		2.68E+00		3.00E+00		2.84E+00
P	<	9.81E+00	<	9.86E+00	<	9.83E+00
Pb		1.07E+01		1.04E+01		1.06E+01
Re	<	1.14E+01	<	1.14E+01	<	1.14E+01
Si		1.47E+01		1.42E+01		1.44E+01
Sn	<	4.46E+00	<	4.48E+00	<	4.47E+00
Sr		5.13E-01		4.48E-01		4.80E-01
Ti		1.72E+00		1.55E+00		1.63E+00
V		2.88E+00		2.67E+00		2.77E+00
Zn		1.38E+00		1.12E+00		1.25E+00
Zr		2.81E+00		2.76E+00		2.78E+00

Table 14. Resin digestion and analyses results

Sample ID		SuperLig 639		SuperLig 639		SuperLig 644		SuperLig 644	TCLP limit
Cs-137 (μCi/g)		2.92E-02		4.41E-02		6.88E+00		7.85E+00	(wt%)
Tc-99 (μg/g)		3.58		3.26		3.97		5.02	
Mass 230 (ug/g)	<	0.44	<	0.43	<	0.47	<	0.44	
231	<	0.44	<	0.43	<	0.47	<	0.44	
232		9.41		7.68	<	0.47	<	0.44	
233	<	0.44	<	0.43	<	0.47	<	0.44	
234	<	0.44	<	0.43	<	0.47	<	0.44	
235	<	0.44	<	0.43	<	0.47	<	0.44	
236	<	0.44	<	0.43	<	0.47	<	0.44	
237	<	0.44	<	0.43	<	0.47	<	0.44	
238	<	0.44	<	0.43		7.73		10.15	
239	<	0.44	<	0.43	<	0.47	<	0.44	
240	<	0.44	<	0.43	<	0.47	<	0.44	
241	<	0.44	<	0.43	<	0.47	<	0.44	
242	<	0.44	<	0.43	<	0.47	<	0.44	
AA analysis, wt. %									
K		2.40E-03		2.50E-03		2.70E-03		3.31E-03	
As		9.07E-05		1.03E-04		1.13E-04		1.13E-04	1.00E-02
Se		3.46E-05		4.18E-05		4.52E-05		4.95E-05	2.00E-03
Hg	<	1.00E-04	<	1.00E-04	<	1.00E-04	<	1.00E-04	4.00E-04
Chem Check									
U (μg/g)	<	9.10E+03	<	8.99E+03	<	9.72E+03	<	9.25E+03	
ICP-ES, wt.%									
Ag	<	5.46E-06	<	5.46E-06	<	5.83E-06	<	5.55E-06	1.00E-02
Al	<	2.46E-05	<	2.46E-05	<	2.62E-05	<	2.50E-05	
B	<	1.09E-05	<	1.09E-05	<	1.17E-05	<	1.11E-05	
Ba	<	1.82E-06	<	1.82E-06	<	1.94E-06	<	1.85E-06	2.00E-01
Ca		2.95E-04		2.95E-04		3.22E-04		4.17E-04	
Cd	<	1.82E-06	<	1.82E-06	<	1.94E-06	<	1.85E-06	2.00E-03
Co	<	5.46E-06	<	5.46E-06	<	5.83E-06	<	5.55E-06	
Cr	<	4.55E-06	<	4.55E-06		1.47E-04		1.98E-04	1.00E-02
Cu	<	2.73E-06	<	2.73E-06		5.22E-05		7.44E-05	
Fe		8.88E-06		8.88E-06		4.34E-06		8.93E-06	
La	<	7.28E-06	<	7.28E-06	<	7.78E-06	<	7.40E-06	
Li	<	2.73E-06	<	2.73E-06	<	2.92E-06	<	2.77E-06	
Mg		5.51E-05		5.51E-05		5.49E-05		7.28E-05	
Mn	<	9.10E-07	<	9.10E-07	<	9.72E-07	<	9.25E-07	
Mo	<	1.82E-06	<	1.82E-06	<	1.94E-06	<	1.85E-06	
Na		1.29E-03		1.29E-03		8.86E-03		1.11E-02	
Ni	<	9.10E-06	<	9.10E-06	<	9.72E-06	<	9.25E-06	
P	<	4.00E-05	<	4.00E-05	<	4.28E-05	<	4.07E-05	
Pb	<	3.91E-05	<	3.91E-05	<	4.18E-05	<	3.98E-05	
Si	<	2.46E-05	<	2.46E-05	<	2.62E-05	<	2.50E-05	
Sn	<	1.82E-05	<	1.82E-05	<	1.94E-05	<	1.85E-05	
Sr	<	1.82E-06	<	1.82E-06	<	1.94E-06	<	1.85E-06	
Ti	<	1.82E-06	<	1.82E-06	<	1.94E-06	<	1.85E-06	
V	<	3.64E-06	<	3.64E-06	<	3.89E-06	<	3.70E-06	
Zn		6.59E-06		6.59E-06		8.40E-06		1.10E-05	
Zr	<	4.55E-06	<	4.55E-06	<	4.86E-06	<	4.62E-06	

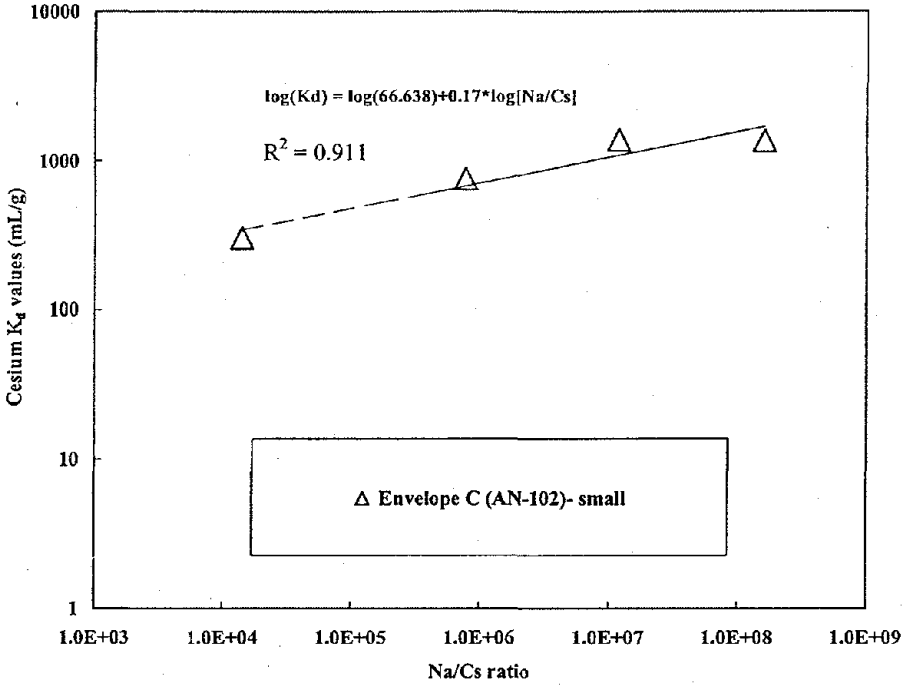


Figure 1. Log-Log plot of cesium equilibrium distribution coefficient

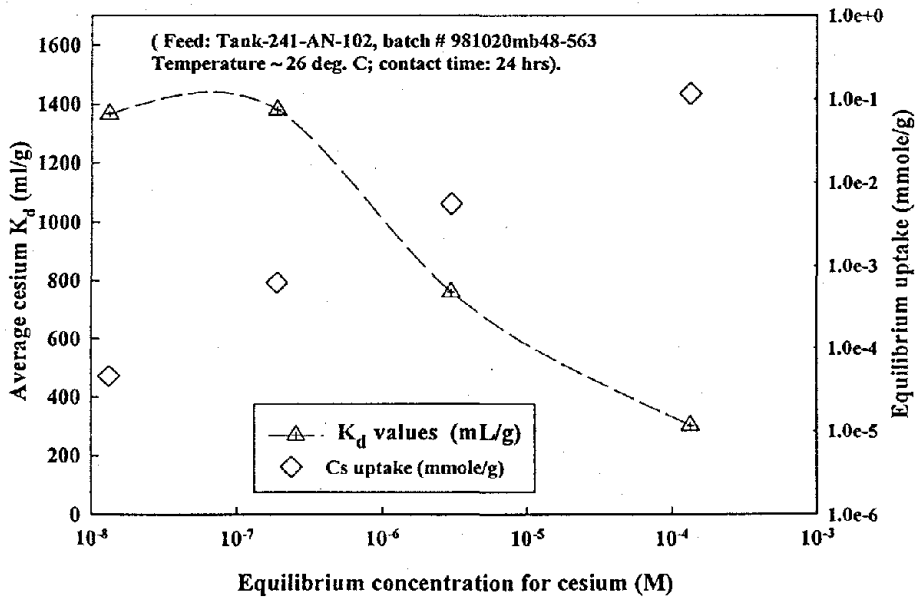


Figure 2 . Batch distribution coefficients for cesium on SuperLig 644

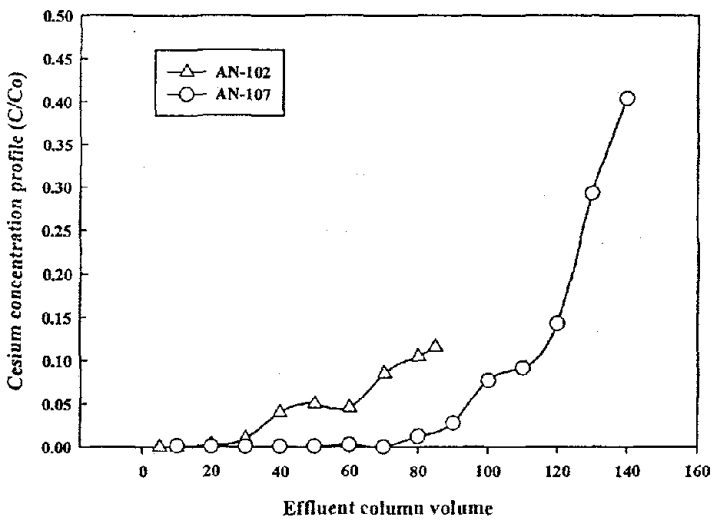


Figure 3. The loading breakthrough profile for Cs-137 on SuperLig 644 resin

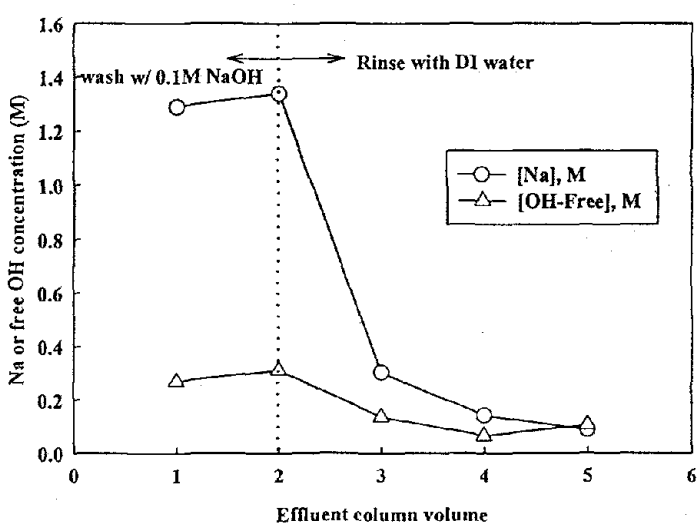


Figure 4. The Na and free OH concentrations in feed displacement and rinse solutions post Cs ion exchange column loading

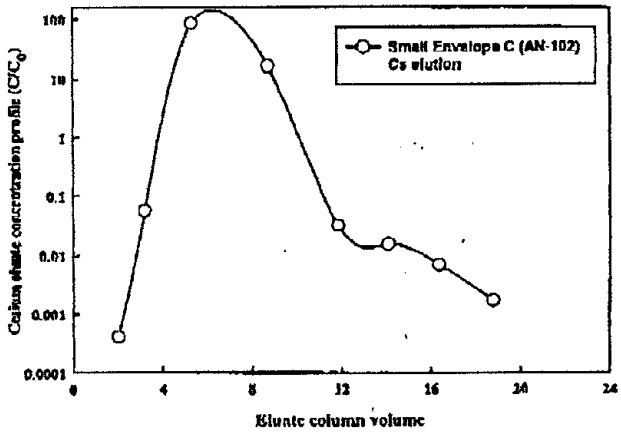


Figure 5. The eluate concentration profile for Cs-137 on SuperLig 644 resin

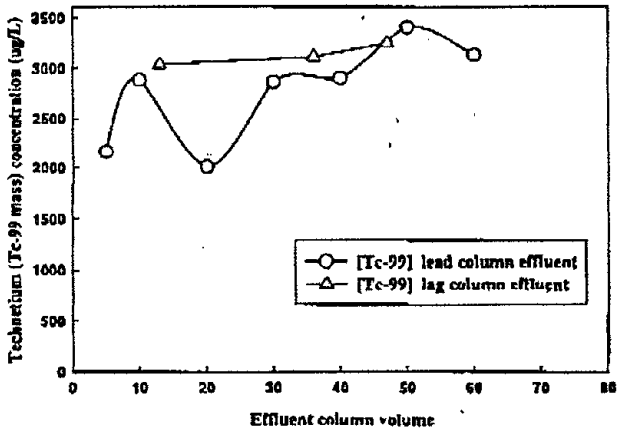


Figure 6. The loading breakthrough profile for Tc-99 on SuperLig 639 resin

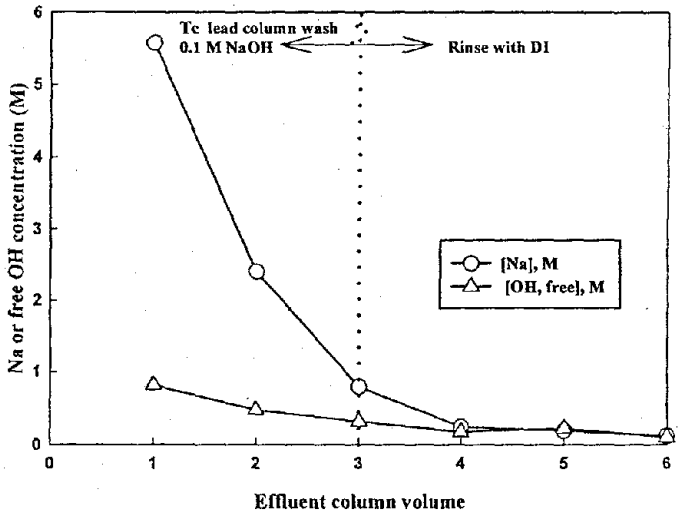


Figure 7. The feed displacement and rinse concentrations of Na and free OH

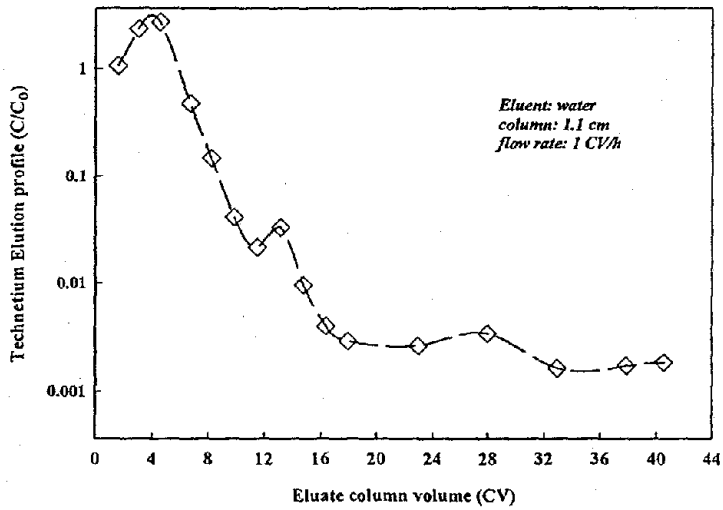


Figure 8. Technetium elution curve for AN-102 from Superlig 639 resin bed