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Small-Scale Ion Exchange Removal of Cesium and Technetium from Hanford Tank 241-AN-103

by

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April 12, 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 Hanford Tank-241-AN-103 (Envelope A) salt solution was decontaminated. Radioactive cesium and technetium removal was accomplished with two sets of ion exchange columns connected in lead-lag configuration. The cesium and technetium removal columns were packed with SuperLig® 644 and 639 ion exchange resins, respectively. Process steps included: 1) resin preconditioning, 2) loading cycle, 3) caustic washing and water rinsing, 4) elution, and 5) resin regeneration. To determine the size of the columns needed to treat the sample, the capacity of the resin had been estimated from the batch distribution coefficients (K_d) and the resin density. The K_d values were determined over a range of radionuclide concentrations and were used to generate an equilibrium isotherm. The overall resin capacity and equilibrium constant, which can be determined from Langmuir or Freundlich equation fit of the equilibrium data, are important parameters for successful modeling of the column breakthrough and elution behavior. The modeling can then be used to aid in scale up of the design of the ion exchange columns. The decontaminated Tank 241-AN-103 sample will be used in a demonstration of the Low Activity Waste vitrification process, and the concentrated radionuclides will be used for High Activity Waste vitrification.

The Tank 241-AN-103 sample was diluted to 5.25 M sodium ion concentration and filtered. The average cesium equilibrium distribution coefficient (K_d value) for SuperLig® 644 was 1564 ± 49 mL/g. The estimated 50% breakthrough point was 626 ± 20 column volumes, (lambda (λ), i.e. K_d value multiplied by the resin wet density). Prior work[2] indicated that 50% breakthrough would be reached at 37% of this value (231.1 ± 7.3 column volumes). To obtain the adsorption isotherm over a range of cesium concentrations, the samples were twice recontacted with fresh resin and additional K_d values were determined. Also, duplicate initial samples were spiked with non-radioactive cesium to obtain K_d values at higher initial concentrations. The equilibrium isotherm data for SuperLig® 644 were shown to fit the Freundlich equation over the cesium concentration range studied. The batch distribution coefficients increased with equilibrium cesium concentration then precipitously dropped (i.e. reduced by 55%) to less than half of that in the low concentration range when the equilibrium concentration was increased by more than 5 orders of magnitude. (The 241-AN-103 sample was mixed with another sample from this tank to achieve a final concentration of 4.99 M sodium ion prior to both column experiments).

The cesium-removal column performance greatly exceeded the design criteria and the estimated performance. Only 11 % breakthrough of cesium was observed after passing 164 column volumes of sample through the lead column. The lag column removed the remaining cesium to achieve an overall decontamination of 99.38%. However the effluent was slightly contaminated with cesium during handling and the level exceeds the target cesium concentration. Cesium elution from the lead column at 1

CV/h using 0.5 M HNO_3 was effective, despite the observed long tailing, which probably resulted from incomplete loading. After passing 17 column volumes of 0.5 M nitric acid through the column at 1CV/h, the residual cesium content in the eluate was ~ 0.04 uCi/mL

For the diluted (to 5.25M Na^+) and filtered sample, the average technetium K_d value was 471 ± 35 mL/g for SuperLig® 639. The estimated 50% breakthrough point (λ value) for technetium was 221± 16 column volumes. Similar to cesium, the distribution coefficient was determined over a range of technetium concentrations. The equilibrium data for technetium show a Freundlich isotherm. Based on the ability of the resin to remove most of the Tc from the sample, there does not appear to be an appreciable amount of a non-pertechnetate form of technetium. SuperLig® 639 is not expected to remove technetium that is not present as the pertechnetate ion. The repeated contact experiments show that at least 97.6% of the technetium could be extracted by SuperLig 639 resin. Other Hanford tank wastes are known to contain technetium as pertechnetate (extractable by SuperLig 639 resin) and a sizeable fraction of another form(s) of technetium that is not an anion. 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 equilibrium between pertechnetate and other form(s) of technetium present in the AN-103 sample.

Technetium removal from Hanford Tank 241-AN-103 by the SuperLig® 639 resin column was very effective and exceeded the design criteria. Only 47% breakthrough occurred after processig 270 column volumes of sample. The lag column maintained less than 8% breakthrough during the course of the run to achieve an overall technetium decontamination of 91.5%. Despite this 8% breakthrough, the effluent contains less technetium than the target concentration. The 8% breakthrough was attributed to problems with resin floating during the early part of the experiment. Elution of the technetium resin was slow and required a larger volume of water (40 column volumes) than anticipated. However, given the longer than anticipated loading cycle, 270 column volumes (90-hours) versus BNFL Inc. flowsheet value of 100 column volumes (33.3-hours), the extended elution period should not affect the waste processing time cycle. Several proposals are included to investigate improvements to the elution process.

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 activity and high level waste glass, respectively. The pretreatment includes sludge washing, filtration, precipitation, and ion exchange processes to remove entrained solids, cesium, transuranics, technetium, and strontium. The ion exchange removal of cesium (Cs) and technetium (Tc) ions 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 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[1] demonstrated the conceptualized flow sheet parameters with a similar Hanford tank sample (241-AW-101). Those experiments 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 the current small-scale ion exchange study were the following:

(1) Determine the batch distribution coefficients (K_d values) and percent removal for cesium (Cs) and technetium (Tc) ions on SuperLig® ion exchange materials (SuperLig® 644 and 639) using Tank 241-AN-103 (Envelope A) salt solution.

(2) Demonstrate Cs and Tc loading, breakthrough, and elution profiles for Envelope A sample using SuperLig® 644 and 639 ion exchange resins.

(3) Provide decontaminated (pretreated) feed and concentrated eluate streams for vitrification into low level and high activity glass.

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

The small ion exchange column and batch contact experiments were performed in a shielded cell, allowing remote handling of materials. The experiments were conducted at ambient temperature using two cesium or technetium ion exchange columns connected in series. The columns were designated as lead and lag and were packed with ~ 6.2 mL of SuperLig® 644 for cesium removal and ~ 5.3 mL SuperLig® 639 for technetium removal. The sample was first diluted to ~5 M sodium ion concentration and filtered to remove insoluble solids. The solution was passed through the technetium removal columns first, and the effluent was collected in fractions. (The planned pretreatment process will remove the cesium first, but schedule conflicts dictated that technetium removal precede cesium removal for this experiment) Once Tc analysis results were received and confirmed to meet the acceptance criteria, the effluent fractions were combined and passed through the cesium columns. Similarly, the cesium effluent was collected in fractions and composited after analysis confirmed that the effluent met the vitrification process acceptance criteria. The loading capacity for the ion exchange columns was determined from batch K_d experiments conducted at the same conditions as those of the columns. The K_d values were also determined at different equilibrium concentrations to establish a robust ion exchange decontamination process across a 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 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). Concentration of the ion(s) in solution is determined by radioactive counting (cesium) 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. 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.

2.0 EXPERIMENTAL

2.1. Materials:

Hanford Tank 241- AN-103 salt solution was used for determination of batch distribution coefficients and column breakthrough efficiency tests. Half of the solution received from Hanford was diluted with 0.01 M sodium hydroxide solution to 5.25 M [Na^+] and filtered with 0.45-μm nylon filters.[4] After dilution and filtration, the filtrate had a density of 1.26 g/mL and initial Cs-137 and Tc-99 concentrations of 194 μCi/mL and 0.033 μCi/mL, respectively. This solution was used for the distribution coefficient (Kd) tests. Prior to the column tests, the second half of the sample was diluted, filtered, and combined with the first half. The composited solution was 4.99 M [Na^+]. Table 1 compares characterization of the first half of the sample, the composited filtrate, and that for Tank-241-AW-101.[3] The characterization data for the two tanks were similar, except that the technetium concentration in Tank-241-AN-103 (0.033 μCi/mL) is less than half of that found in Tank-241-AW-101. Thus, the technetium K_d values are expected to be less than those found in Tank-241-AW-101, despite the fact that the nitrate concentration in Tank -241-AN-103 is 11% higher than that in Tank-241-AW-101. The chemical reagents used for resin pretreatment and column elution were, respectively, sodium hydroxide and nitric acid solutions prepared from ACS certified reagents from Fisher Scientific, Inc.

The ion exchange materials used for cesium and technetium removal from Hanford Tank 241-AN-103 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 both resins for testing. The physical characteristics of SuperLig® 644 and 639 different batches of the resins were measured at Savannah River Technology Center (SRTC) and the results are presented in Table 2. The F-factor values shown in the table were obtained by drying the resin over night in a vacuum oven at 105 °C. Results of a sieve analysis of the SuperLig® 639 resin batches used in the tests are given in Table 3.

2.2 Equipment:

The equipment used for the ion exchange column tests was assembled remotely in a shielded-cell in a Savannah River Technology Center laboratory module. Two identical ion exchange columns were used per column test. The columns were made of a medium wall Pyrex glass tube 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 stainless steel screen with a mesh size of 200 mesh was fitted into the bottom of the column. The column top assemblies had a fill reservoir, a pressure gauge, a pressure relief valve, and feed inlet port. The fill reservoir on column top assemblies also served as a vent. The top assembly was connected to the lower section by a glass ground joint and was tightly fitted by a screw cap. 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 polypropylene lines that had Teflon® quick-connect fittings attached to each end; the lines had inside diameter of 0.125-inch.

The equipment used for batch contact tests consists of 15-mL polyethylene bottles, a Mix-Max® orbital shaker, nylon filter units, 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 cell, allowing remote handling of materials.

2.3. Procedure

2.3.1 Batch Contact Procedure: The ion exchange materials used in this study for removal of cesium and technetium ions were SuperLig® 644 (batch # 981020MB48-563) and 639 (bath # 981015DHC720011). The resins were supplied by IBC Advanced Technologies, American Fork, Utah. All batch contact experiments were conducted in duplicate. A standard batch contact test, followed by two re-contacts of each batch filtrate with fresh resin and an independent cesium or technetium spiked test were conducted in the shielded cell. All tests 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) in which no ion exchange resin was added, was treated in identical process steps as the actual test samples. The concentrations of Cs and Tc in control samples were used as the starting initial concentrations for determining 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 an 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 (1:10) with de-ionized water to reduce the radiation dose rate during transfer from the shielded cell. All dilutions and measurements were performed based on mass and corrected for density of the solution to ensure accuracy. The concentration of cesium (Cs-137) was determined before and after contact with SuperLig® 644 resin by gamma (Ba-137m) radioactive counting technique. The technetium concentration was determined before and after contact with SuperLig® 639 resin by inductively coupled plasma-mass spectroscopy (ICP-MS) at mass 99. 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 conducted with fresh ion exchange resin using the filtrate that had been separated from the resin in the preceding test. In the first re-contact test, a known volume (~ 12 mL) of the filtrate that had been separated from the resin used 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 spent resin was separated from the solution 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 (Cs-137) or Inductively Coupled Plasma-Mass Spectroscopy (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 sequential tests, small quantities of cesium nitrate (in water) or sodium pertechnetate (in ~1 M HNO_3) were spiked into another sample of known volume of Hanford Tank 241-AN-103 filtrate. This was done to increase the initial concentration of Cs or Tc in test solutions by a factor of ~10. Because of the size of the spike sample and the density difference the spike and the test solution, it was necessary to add the test solution into polyethylene bottles 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 spike filtrate was removed from the shielded cell and submitted for analysis by ICP-MS for total cesium and by radioactive counting technique for Cs-137 content in spike and control samples. Similarly, ICP-MS was used for determination of Tc-99 mass in spiked filtrate and control samples.

2.3.2 Technetium column run

The technetium removal from Hanford Tank 241-AN-103 filtrate was accomplished by operating two ion exchange columns in series. Two glass columns with an inside diameter of 11 mm each and a total height of 30 cm were employed. The first column was designated as the lead column and the second as the lag column to further decontaminate the sample to achieve the vitrification acceptance criteria (0.029 µCi/mL). In a 30-mL polyethylene bottle, a known amount of dry SuperLig® 639 resin was soaked in de-ionized water for two days. A portion of the mixture was slurried into the 1.1-cm i.d. glass columns. The walls of the columns were tapped while slurring the resin to ensure uniformly packed beds. The wet volume of the resin in each column was approximately 5.6 mL and the resin bed was 5.9 cm high (i.e. an L/D ratio of 6.2). A sieve analysis of the resin shows that over 95% of the resin beads had a particle size range less than 20 mesh. The measured SuperLig® 639 dry bulk density was 0.468 g/mL and the resin particle density was 1.147 g/mL. The bottom of the resin bed was retained in place by stainless steel screen rated at 200 mesh. The top of the resin bed was initially free, but introduction of the salt solution caused floating. After passing ~10 column volumes of sample through the lead column, the experiment was stopped and the resin was re-settled with dilute caustic. The test was restarted, and the resin floated after another 10 column volumes of sample had passed. At this point, the resin bed was covered with 1-2 cm of quartz wool. Glass beads (1-2 cm) were put on top of the quartz wool to provide additional mass to hold the resin in place.

After packing the columns with SuperLig® 639, the columns were flushed with 3 column volumes of de-ionized water, followed by 3 column volumes of 1.0 M sodium hydroxide solution. The resin bed was then flushed with 3 column volumes of de-ionized water and stored in water for several days. Just prior to initiation of the run, the resin was conditioned by pumping 3 column volumes of 1.0 M NaOH through the lead and into the lag column. The pump (FMI metering pump, model RP-G5, from Fluid Metering, Inc. Oyster Bay, New York) dial was used to adjust the flow rate to the desired value. The solution was allowed to pass from the lead to the lag column through a three-way Teflon® valve connected to a glass sampling tube. The valve was primarily used to route the solution into the sampling tube during sample collection. After 3 column volumes of the 1.0 M NaOH effluent solution were collected from the bottom of the lag column, the liquid hold up in the column was reduced to 1-2 cm above the glass beads.

On the day of the run, the feed (Hanford Tank 241-AN-103 filtrate) was checked to ensure that it contains no visible solids. The feed was then pumped through the lead column at a flow rate of 3 CV/h (15.3 mL/h). Due to resin floating and bed re-packing during the early part of the column loading, the resin bed volumes in the lead and lag columns were reduced from 5.6 to 5.1 mL and from 5.6 to 5.4 mL, respectively. Therefore, 1 CV was defined as 5.1 mL for the lead column and 5.4 mL for the lag column. 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 through the lead into lag column was calculated from the weight of the samples collected during a 10-minute sampling period and the density of the feed. Similarly, the flow rate through the lag column was calculated from the weight of the effluent fractions collected from the lag column at specified column volume intervals and the density of filtrate effluent from the column.

The feed was introduced down flow into the lead column at 3 CV/h. Sub-samples, 3 mL each, were collected from the lead column after 5 and 10 column volumes of feed had passed through the lag column. Sub-samples were collected after each subsequent 10 column volume interval. 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. A sub-sample (~ 1 mL) of effluent collected was diluted 1:10 with de-ionized water in order to reduce the dose rate to allow the sample removal from the shielded cell. Effluent sub-samples 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 sampling the lead column, the effluent from the lead column was allowed to pass down flow into the lag column that served to further decontaminate the feed solution. 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 stopped when the analysis of periodic samples of effluent indicated a nearly 45% breakthrough for Tc-99. At this point 80% of 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) or 22.95 mL of 0.1 M sodium hydroxide, followed by 2 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 termination of the column wash and rinse, the lead and lag columns were disconnected. The pump dial set was then adjusted to reduce the flow rate to 1 CV/hr through the lead column. The eluent (de-ionized water) was pumped down flow into the lead column and collected in two column volume fractions. Eluate fractions were collected into 30-mL polyethylene bottles containing approximately 5 mL of de-ionized water. The lead column elution was stopped after 18 column volumes of water had pumped through the column. The column was left in de-ionized water over the next 3 weeks during which time, sub-samples of the eluate that had been collected between fractions were analyzed for Tc-99 by ICP-MS.

Analysis results of eluate samples revealed an incomplete elution of Tc from the SuperLig® 639 resin. A second elution cycle was performed three weeks after the first elution cycle using an additional 22 column volumes of de-ionized water at 2 CV/hr. At the end of the second elution cycle, the column was flushed with 3 CV of 1.0 M NaOH in preparation for storage.

2.3.3 Cesium column run

The cesium ion exchange columns were packed and pretreated in a non-radioactive chemical hood. The pretreatment of the resin (SuprLig® 644) was conducted according to protocol developed by Savannah River Technology Center.(3) This protocol (see below) 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 and were stored in de-ionized water. The columns were transferred into the shielded cell two days before the column run was initiated.

New SuperLig® 644 Resin Pretreatment Procedure:

1. Soak in 1.0 M NaOH (~10:1 vol/:vol) for 2 hrs (gentle shaking)
2. Decant NaOH and add de-ionized water

3. Slurry resin into column while tapping side of column
4. Pump 3 column volumes (CV) of 0.5 M HNO_3 in 1 hour
5. Pump 3 CV of de-ionized water in 1 hour
6. Pump 6 CV of 0.25 M NaOH in 2 hours
7. Pump 3 CV of de-ionized water in 1 hour (only if stored >48 hours)

Several weeks after the technetium column run, the effluent fractions from the technetium removal column were combined and thoroughly mixed. 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. After this pre-conditioning, the feed was introduced into the lead column at 3 CV/h (18.3 mL/h) and the effluent was collected from the lag column. Due to resin bed shrinking in the lead column from 6.4 to 6.1 mL during the early part of the column loading, 1 CV was defined as 5.1 mL. 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 was collected. 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 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 after 20 column volume increments and counted at-line with a gamma counter to monitor the progress of the column loading. The samples from the load and lag columns were submitted for analysis by radioactive counting without dilution to Analytical Development Section of Savannah River Technology Center. The loading cycle was terminated when the feed solution 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 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 10 column volumes of 0.5 M HNO_3, followed by 3 column volumes of de-ionized water. The resin was stored in de-ionized water.

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 lambda (λ; i.e. K_d value multiplied by the resin wet density) values represent an estimate of the volume of solution that can be processed by a quantity of ion exchange resin. A plot of K_d values as a function of equilibrium concentration of cesium and technetium provides important information on selectivity and overall capacity of the ion exchange resin. The batch experiments include the addition of a small quantity of ion exchange materials into a small volume of the salt solution containing known quantities of Cs or Tc ion. 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 was analyzed by 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:

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

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 results for cesium K_d values and percent removal (%R) are presented in Table 4. The results show that SuperLig® 644 resin is very effective for removal of cesium from Hanford Tank 241-AN-103 salt solution. The standard K_d value was 1564±49 mL/g (average of duplicate samples and the ± refers to the deviation from the average) as compared to the previous value of 944 mL/g observed for another sample of Envelope A from Tank-241-AW-101.[2] The source of this disparity in K_d values between the two salt solutions is probably related to the difference in potassium concentration.

The loading capacity for cesium on SuperLig® 644 was estimated from average K_d values and the wet density[1] of the resin. The estimated volume at 50% breakthrough, Lambda (λ), was as follows:

$$\lambda = K_d \times \rho = 1564 \text{ mL/g} \times 0.4 \text{ g/mL} = 626 \qquad (3\text{-a})$$

Thus, the estimated volume of Tank-241-AN-103 salt solution that can be processed per mL of SuperLig® 644 before 50% breakthrough of the feed occurs was 626 ± 20 column volumes. Previous work[2] with an Envelope A sample indicated that breakthrough would occur at 37% of this value. The adjusted lambda value was used to calculate the quantity of resin needed to decontaminate the sample.

Figure 1 shows the batch distribution coefficient (K_d values) and equilibrium capacity for cesium as a function of equilibrium concentration. It can be seen that the K_d values increase from 1150 mL/g to 1560 mL/g and after a short plateau, the K_d value decreases to about 720 mL/g. The figure also shows a log-log plot for cesium uptake by SuperLig® 644 as a function of the equilibrium concentration for cesium. The data shown in Figure 1 are fitted with the Freundlich equation in the form

$$\log q = \log K + \frac{1}{n} \log C_e \qquad (3-b)$$

Where q is the uptake cesium per unit mass of resin, Ce is the equilibrium concentration corresponding to q, K is a constant for the cesium-resin system, and n is another constant that is restricted to values greater than 1. The data correlation with equation (3) yielded a straight line with a slope of 1/n and an intercept equal to log K. A detailed analysis of the data correlation and column modeling will be presented in a separate document.

Technetium K_d results are presented in Table 5. This resin will extract only the pertechnetate form of technetium. It is evident from data in Table 5 that as the exchangeable pertechnetate species in solution depletes, the amount absorbed onto the resin decreases, as expected. The results also indicate that a maximum of 57 µg/L of Tc is present as non-pertechnetate ion, which is only 2.3% of the total Tc present. This assumes that sequential batch contacts do not effect the ratio of pertechnetate to non-pertechnetate. The loading capacity for SuperLig® 639 was estimated from the batch distribution coefficient and the density of the resin. The estimated volume to process at 50% breakthrough, Lambda (λ), was calculated as follows:

$$\lambda = K_d \times \rho = 471 \text{ mL/g} \times 0.47 \text{ g/mL} = 221 \qquad (3\text{-c})$$

Thus, the lambda value suggests that up to 221 ± 16 column volumes of Envelope A (Tank-241-AN-103) salt solution could be processed with SuperLig® 639 resin before 50% breakthrough occurs.

Figure 2 shows a semi-logarithmic plot of equilibrium distribution coefficients (K_d values) and a log-log plot for technetium uptake as a function of the equilibrium concentration. The technetium K_d values, shown by the square symbols, rise by a factor of 5 from 70 to 355 mL/g as the initial technetium concentration increases from 57 to 96 μg/L. The K_d values then approach a constant value as the available sites on the ion exchange resin are fully utilized. Thus, a further increase in the initial technetium concentration results in a moderate decrease of the K_d values as indicated by the technetium-spike test.

3.2. Technetium Column Loading

The planned treatment process removes cesium before technetium. However, due to scheduling constraints, the technetium column loading using Envelope A (Tank 241-AN-103) salt solution was conducted prior to cesium decontamination. About 1400 mL of the salt solution was passed down-flow through the two sequential technetium columns at 3 CV/h. The results for Tc-99 breakthrough using SuperLig® 639 resin are shown in Tables A-7 and and A-8 for the lead and lag columns, respectively.

The cumulative volume of effluent (column 2, Table A-7) was calculated from the weight of the samples collected divided by the density of the feed, and the time interval between successive samples. The cumulative volume of effluent from the lag (column 2, Table A-8) was determined from the weight of effluent fractions divided by the density of the solution. The ICP-MS results in μg/L at mass 99 are presented in Tables A-7 and A-8 (column 3). The results were converted into μCi/mL (column 4) using a conversion factor of 5.88E+04 (μg/L)/(μCi/mL).

Figure 3 shows cumulative amounts of technetium in the feed, effluent, and on the resin as a function of column volumes of feed or effluent that passed through the lead column. The amount of technetium pumped into the column over time (square symbols in Figure 3) was calculated from the concentration of feed (2.55E-3 mg/mL) multiplied by the volume of solution pumped into the column as the run progressed.

The amount of technetium in the effluent (diamond symbols in Figure 3) was calculated from the effluent volume multiplied by the average concentration of successive measurements in a short time interval. The total amount of technetium in the effluent was calculated from the equation

$$\sum_{i=0}^{i=N} [ave.(C_i + C_{i+1})\cdot \Delta V] \qquad (4)$$

Where C_i and C_{i+1} are consecutive effluent concentrations (mg/L), ΔV is the effluent volume collected over pre-determined intervals and N is the number of technetium effluent samples. The loading curve (circles) represents the amount technetium loaded onto the resin as run progressed and it was calculated by difference (i.e. total amount of technetium fed into the column minus total quantity in the effluent).

The amounts of technetium in the feed, effluent and on the resin in the lag column are shown in Figure 4. The curve on the top (squares) represents the calculated cumulative amounts of technetium fed into the lag column as the run progressed. The bottom curve (diamonds) shows the measured technetium that passed through the column and the middle curve (circles) represents the calculated amount of technetium progressively loaded onto the resin as the run continued

The concentration breakthrough profile (C/Co) for technetium was plotted as a function of the volume of effluent processed through the column; and the results are shown in Figure 5. The normalized curve for the lead column shows that ~ 270 column volumes of feed were processed before a 47% breakthrough occurred. The lag column, on the other hand, maintained less than 8% breakthrough during the course of the column run. Also, up to 2.3% of the technetium may be as a non-pertechnetate form which is expected to pass the column unchanged. The data in Figure 5 show a significant scatter, the cause of which is not known but may be related to wall 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. 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.

It is unlikely that significant channeling occurred during the course of the run as the breakthrough of technetium (8%) was continuous and lower than previously experienced with channeling of SuperLig® 644 columns (>20%). However, some operational difficulties were encountered during the initial stages of the run. In addition to the difficulty of adjusting the liquid level before initiating the feed sample, a portion of the resin in the lead column started to float after the salt solution had filled the bed. The SuperLig® 639 batch used in this study was very light resin, having a particle density of 1.147 g/mL; the density of the salt solution was 1.267 g/mL. The excess solution in the resin bed was drained and the resin was resettled with the introduction of 0.1 M NaOH. After this failed to prevent floating upon re-introduction of the feed, the resin bed was re-packed with 1-2 cm of quartz wool added on top of the bed and another 1-2 cm of glass beads was added on top of the wool to provide a restraining weight. Although this diluted the feed with sodium hydroxide solution, it appears to have had a negligible effect on the results. The piston of a second pump that was used intermittently for pumping the salt solution had seized in the cylinder, and the pump had to be removed from service and the pump head replaced. There was no evidence of solids or residues present in the lines and other visible parts of the flow path. Despite these disruptions, the results presented in Fig. 5 show minimal adverse effects or column performance loss from the resin bed floating and re-packing during the initial stages of the run.

3.3. Technetium Column Elution

The results for Tc-99 elution from SuperLig® 639 lead column are presented in Table A-9. The elution was performed with de-ionized water at approximately 1 CV/h. Figure 6 shows the elution produced a sharp peak (28 C/Co) at 13 column volumes, followed by an exponential decrease to C/Co = 5 at 18 column volumes. Due to the unavailability of an at-line Tc analyzer it was not possible to determine when elution was complete, so the elution was terminated after 18 column volumes and samples were submitted for analysis. The resin column was left in deionized water for storage. After results were received, the elution was continued several weeks later at higher flow rate (2 CV/h). Changes in the bed density caused by shrinkage of the resin (~10%) during the column elution, pump behavior, and extremely low pumping rates made the flow rate control difficult. The "second" elution curve exhibited a long tailing, which required 22 column volumes of water to reduce the remaining technetium from 5 C/Co to 0.07C/Co. The sudden drop in Tc concentration between the elution periods is a function of the way samples were collected. The first eluate sample ("28 CV") after the resumption of the elution was collected as a grab sample after 5 CV had passed through the bed. The Tc had probably desorbed from the resin during the several week storage period, and was initially at a very high concentration in the storage solution. By the time the 5 CV of eluate had passed, the Tc-containing storage solution was washed out. This indicates that reduction in the volume of eluate may be accomplished by soaking the resin, rather than continuously pumping eluate through it. However, a controlled test is needed to determine if the duration of elution is extended. Overall, the technetium elution rate for SuperLig® 639 resin was very slow, requiring an additional 22 column volume of water for a total of 40 column volumes. Characterization of composited technetium eluate is given in Table 6. The ICP-MS results are used for reference values. Methods are desired to increase the rate of elution of technetium from SuperLig® 639.

3.4. Cesium Column Loading

The cesium decontamination experiment was conducted several weeks after the technetium test was completed. The feed to the cesium columns was combined effluent fractions (Tank 241-AN-103) that had been decontaminated through the technetium columns. About 1000 mL of the effluent was passed in down-flow mode through the cesium ion exchange columns at 3.0 CV/h. Prior to initiating the ion exchange cesium column run, SuperLig® 644 resin was pretreated according to the protocol developed at Savannah River Technology Center.[3] The pretreatment was found critical to successful decontamination of various salt solutions from Hanford Tank Waste by SuperLig® 644. Previous testing of Hanford tank samples indicated that pretreatment for Envelope A was different from that for Envelopes B and C. However, we recently demonstrated that a single pretreatment scheme could be employed to decontaminate various simulated salt solutions of Hanford Tank Waste. The scheme includes a two-hour soak of SuperLig® 644 resin in 1.0 M NaOH (1:10 ~v/v). Following decanting and rinsing, the resin is slurried into the column and packed by gently tapping the sides. An acid treatment cycle is conducted with three column volumes of dilute acid (0.5 M HNO_3). The acid is rinsed off from the column with three column volumes of de-ionized water. A caustic cycle utilizing six column volumes of 0.25 M NaOH is used to regenerate the resin into the sodium ion form and adjust the pH to prevent aluminum hydroxide precipitation. Following regeneration, a column run is initiated or the resin is rinsed with de-ionized water until neutral then stored in de-ionized water. It is apparently critical to repeat the acid washing/caustic regeneration cycle shortly before initiating the column run. Otherwise, storage for more than a few days appears to cause channeling after start up of the test.

Figures 7 and 8 show the calculated amounts of cesium in the feed, effluent, and on the resin for SuperLig® 644 resin in the lead and lag columns. The curies of cesium accumulated on resin as loading progressed was plotted as a function of the liquid that was pumped through the columns.The breakthrough data for cesium lead and lag columns as a function of effluent column are shown in Figure 9. The data plotted in the figure show that 11% breakthrough occurred for the lead column at 160 column volumes and the run was terminated due to an insufficient volume of feed available to reach 50% breakthrough. Thus, the cesium breakthrough curve indicated that the cesium removal from Envelope A was better than expected.

The percent volume change of the resin between the acid and caustic cycles was in the range of 30 to 40%; a 10% reduction of the resin volume observed during the breakthrough showed no detrimental effect on the resin performance. This swelling and shrinking behavior is consistent with previous data.[1] There was no indication of resin decomposition as the characteristic brown color of the resin was absent in column effluent, rinse, and regenerate solutions.

3.5. Cesium Elution

The results for cesium elution from SuperLig® 644 resin are shown in Figure 10. The data in the figure show that elution with dilute nitric acid (0.5 M HNO_3) at 1 CV/h was effective for nearly complete removal of cesium from the resin. The elution curve exhibits four regions, namely: neutralization, 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 sodium ions on the resin. The peak region of the curve shows a sharp peak at 4 column volumes, during which almost 70% of the cesium in the column was eluted. The exponential decay region that follows the peak region shows a significant drop in cesium concentration with few column volumes of the acid. The final region of elution curve shows a tailing effect. Less than 2% of the cesium remained in the column after 13 column volume of the eluent had passed through the column. The unexpected tailing 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. During elution, a rapid shrinkage of the resin could have caused a diminished diffusion velocity of cesium ions in the resin and, consequently, lowered the rate of elution. The volume of the resin decreased from 6.2 to 4.3 mL at the end of elution. Some difficulty was experienced in maintaining the eluent flow rate at the desired level (i.e. 1 CV/h) due to the low flow rate.

The results for characterization data of the final product and cesium eluate are shown in Tables 7 and 8. The final products are the decontaminated AN-103 salt solution from which the cesium and technetium radionuclides are removed, and the composited cesium eluate product solution. The percent removals of the cesium and technetium, were calculated from the average concentrations of Cs-137 (Table 4) and Tc-99 (Table 5) and the average concentrations of these isotopes in decontaminated AN-103 product (Table 7). The percent removal of Cs-137 from Envelope A salt solution was calculated from the area under curve for lag column in Figure 9 (118 µCi) and the initial activity of Cs-137 fed into the two-column configuration (i.e. 164 CVx 6.1 mL/CVx 207.1 µCi/mL= 3.41E+05 µCi). Similarly, the percent removal of Tc-99 was calculated from the area calculated under the curve for the lag column in Figure 5 (245 µg) and the initial activity of Tc-99 fed into the two-column configuration (i.e. 270 CV x 5.4 mLx 2554 µg/L). The percent removal calculated for Cs-137 and Tc-99 were 99.94% and 93.5%, respectively.

The cesium content in the composited effluent is higher than expected, based on the grab samples discussed above. The maximum Cs-137 content in those samples was 0.45 µCi/mL which is lower than 1.2 µCi/mL for the composited effluent. Apparently, the effluent was contaminated with excess liquid from one or more lead column grab samples. Comparison of the total organic carbon (TOC) in

Table 1 (AN-103 salt solution) to that in Table 7 (decontaminated AN-103 product solution) shows a 4-fold increase in the total organic carbon concentration in the decontaminated product. The increase in the TOC values may be attributed to organic species leaching from the resin, although this is thought to be unlikely. The total organic carbon in cesium eluate product (Table 8) is 75 mg/L or less than 21% of the amount in the AN-103 feed sample. One explanation is that the organic components in the resin are solubilizing into the feed solution, but not the eluate. However, this large increase in organic carbon corresponds to nearly 1 gram of carbon leaching from the resin during the run. Loss of this much resin should be reflected in a reduction in the volume of the resin beds by more than 2 mL, which was not observed to occur. Also, no color change in the effluent was observed. The analytical analysis results are therefore suspect. Although there are other possible explanations, further work is needed to confirm this result and examine resin degradation.

The chemical species present in cesium eluate product (Table 8) are higher than expected, particularly chromium and uranium. Both of these species have previously been observed to concentrate in the eluate. The chemical form of the chromium is not known for certain. If it is chromate, the resin may be degraded during the acidic elution cycle. Further work is needed to speciate the chromium compound and evaluate its effect on the process.

4.0 CONCLUSION

Cesium and technetium removal from Hanford Tank-241-AN-103 salt solution was accomplished using IBC manufactured SuperLig® ion exchange resins. The cesium batch distribution coefficient for SuperLig® 644 resin was 1564 mL/g. The projected column volume of feed to process at 50% breakthrough was estimated as 626 column volumes, although prior work indicated that only 37% of this value would be reached (231 CV). The resin performance exceeded expectations, and only 11% breakthrough was attained by the time the feed was consumed at 164 column volumes. Elution of cesium to 2% of the feed concentration was accomplished with 17 column volumes of 0.5 M nitric acid. The elution profile exhibited unexpectedly long tailing. A comparison of Cs-137 in diluted Envelope A feed sample from Tank-241-AN-103 and that in composite treated sample using two columns in a lead/lag configuration containing SuperLig® 644 resin show 99.38% cesium removal. However, cesium removal was more effective than this value indicates because the effluent composite sample was apparently contaminated with lead column samples. The calculated area beneath the lag breakthrough curve at 143 column volumes was 118 μCi and the Cs content in the processed volume was 2.07E+05 μCi (i.e. 164 CV x 6.1 mL/CV x 207 μCi/mL). Thus, the calculated percent removal for Cs was 99.94%.

The technetium distribution coefficient by SuperLig® 639 resin was 471 mL/g. The projected column volume of feed solution to process at 50% breakthrough was 221. A slightly better than expected 270 column volume of Hanford Tank 241-

AN-103 solution was processed before 47% breakthrough occurred, but this discrepancy is probably within the error of the lambda estimate. Elution of technetium from the resin was accomplished with de-ionized water at 1 CV/h. The elution was slow, requiring up to 40 column volumes of water to remove up to 96.5 % of technetium that was loaded onto the resin. Technetium decontamination of up to 93.5% was accomplished with two columns of SuperLig® 639 resin connected in a lead/lag configuration. A continuous breakthrough of ~8% of the technetium was attributed to resin floating problems near the beginning of the experiment. This led to contamination of the lag column and caused the breakthrough.

The elution cycle for the SuperLig® 639 resin was longer than expected. This may cause operational delays and will generate considerable eluate volume. The resin vendor (IBC Advanced Technologies, Inc.) has recommended increasing the temperature of the water used for elution, and we concur with examining the feasibility of this approach. The thermal shock stability should be examined concurrently to verify that the resin does not degrade due to rapid changes in temperature. Another approach to reducing the elution cycle duration may be to terminate the elution earlier, leaving more technetium on the column. The technetium would then be slowly released from the lead column when the feed cycle resumes, and caught on the lag column. Alternatively, a reduction in the volume of eluate may be realized by stopping the elution after 10-15 column volumes, thereby allowing the resin to sit in the eluent and allow technetium desorption without generating more liquid volume. A periodic displacement of eluate every few hours may be beneficial. This approach would not decrease the duration of the elution, but may decrease the volume of eluate generated. The use of smaller resin particles may be another method to enhance the elution, although this would also increase the pressure drop during operation. All of these proposed method need to be tested prior to implementation.

5.0 APPROVALS

Design Check

M. C. Thompson 5/10/00
M. C. Thompson Date
Actinide Technology Section

6.0 REFERENCES

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2. Hassan, N. M. and McCabe D. J., Hanford Envelope A Tank Waste Ion Exchange Column Study (U), SRTC-BNFL-019, Revision 0, October 24, 1997.

3. Hassan, N. M, King, W. D., and McCabe, D. J, SuperLig Ion Exchange Resin Swelling and Buoyancy Study (U), BNF-003-98-0051, Revision 0, March 25, 1999

4. Hay, M.S., Chemical Characterization of an Envelope A Sample from Hanford Tank 241-AN-103, BNF-003-98-0248, May 2, 2000.

Table 1. Filtrate characterization for Envelope A salt solutions

Constituent ions [M]	Tank-241-AN-103 First filtrate [M]	Tank-241-AN-103 combined filtrate [M]	Tank-241-AW-101 concentration [M]
[Na^+]	5.25	4.99	5.07
[Al]	0.70	0.84	0.468
[Ca]	<0.000	0.0002	0.0002
[Cr]	0.0014	0.0014	8.9E-4
[P]	0.0097	0.0099	0.0083
[Si]	0.0045	0.0034	0.00143
[K^+]	0.121	0.117	0.448
Anions, M			
[NO_2^-]	1.037	0.866	0.875
[NO_3^-]	1.571	0.998	1.39
[Cl^-]	0.061	0.085	0.0769
[SO_4^{-2}]	0.008	0.009	< 0.010
[PO_4^{-2}]	0.0069	0.0059	--
[C_2O_4=]	0.0059	0.0069	--
Free OH-	2.166	1.869	2.33
Radionuclides			
[Cs-137]	194 µCi/ml	200 µCi/ml	204 µCi/ml
[Tc-99] (calc'd) mass 99	(0.033 µCi/ml)	(0.052 µCi/mL)	0.06 µCi/mL (LSC) 0.088 µCi/mL (ICPMS)
TIC	2515 ppm	3681	0.150 (M)*
TOC	376 ppm	568	0.279 (M)*

* characterization before dilution, corrected for dilution by calculation

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

ResinType	Batch #	Particle density (g/mL	Bulk density (g/mL)	Water content (%)	F-Factor
SuperLig 644	644BZ	1.611	0.793	10	0.9
SuperLig 644	981020MB48	1.549	0.758	9.6	0.904
SuperLig 639	980624001DC	1.219	0.489	1.4	0.986
SuperLig 639	981015DHC-720011	1.147	0.468	1.3	0.987

Table 3. Sieve analysis of SuperLig 639 resin batch batch #981015DHC720011

		wt. of fractions (g)		wt. %		
Screen Mesh	Opening	Sample 1	Sample 2	Sample 1	Sample 2	Avg. (%)
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				
	total	5.00	4.945	99.7	98.7	99.2

Original wt. of sample 1 = 5.003 g; Original wt. of sample 2 = 5.008 g

Table 4. Batch Distribution Coefficient Data for Cesium on SuperLig® 644 Resin

Sample ID	solution mass (g)	resin mass (g)	Solution vol.(mL)	phase ratio	[Cs -137] (μCi/mL)	Cs-137 Removal (μCi/mL)	Kd (mL/g)	ave. Kd (mL/g)	avg. % removal
Standard Kd									
BNF-A325-F44-1	18.233	na	14.47	na	192.2	na	na	na	na
BNF-A325-S644-1	18.153	0.180	14.41	80.04	10.01	182.2	1.61E+3		
BNF-A325-S644-1d	17.344	0.180	13.77	76.47	10.17	182.1	1.52E+3	1564	95
First Recontact Kd									
BNF-A325-S644-1R-1	13.983	0.120	11.10	92.48	0.62	9.40	1552		
BNF-A325-S644-1R-1d	14.152	0.120	11.23	93.60	0.62	9.50	1582	1567	94
Second Recontact Kd									
BNF-A325-S644-2R-1	10.024	0.100	7.96	79.56	0.04	0.575	1155		
BNF-A325-S644-2R-1d	10.598	0.100	8.41	84.11	0.045	0.580	1210	1157	93
Cs spiked Kd[‡] (gamma scan)									
BNF-A325-Cs-sp-F2	11.542	na	9.16	na	222.1	na	na	na	na
BNF-A325-S644-sp-1	12.663	0.100	10.05	100.5	30.69	191.4	693		
BNF-A325-S644-sp-1d	12.655	0.100	10.04	100.4	33.10	189.0	634	664	86
(total Cesium by ICP-MS)					units (mg/L)	units (mg/L)	units (mL/g)	units (mL/g)	
BNF-A325-Cs-sp-F2	11.542	na	9.16	na	132.5	na	na	na	na
BNF-A325-S644-sp-1	12.663	0.100	10.05	100.5	16.95	115.5	757.6		
BNF-A325-S644-sp-1d	12.655	0.100	10.04	100.4	18.66	113.8	677.6	718	87

F-factor = 0.904; Temperature = 26 ± 1°C; contact time = 24 ±1 h; batch #: 981020MB48-563
[‡]~ 20 μL of 1 M Cs(NO3) was added into 30 mL of the diluted AN-103 sample. "-1" and "-1d" refer to the first sample and a duplicate. "F1" and "F2" refer to the feed (initial) and spiked feed samples. na = not applicable

Table 5. Batch distribution coefficients for technetium on SuperLig® 639

Sample ID	sample wt. (g)	Resin mass (g)	Volume (mL)	phase ratio	[Tc -99] (μg/L)	Tc-99 (μg/L) removed	Kd (mL/g)	avg. (mL/g)	avg. % removal
Standard Kd									
BNF-A325-F-1	22.76	na	18.06	na	2407	na	na	na	na
BNF-A325-S39-1	24.43	0.1804	19.39	107.5	481	1925	435.6		
BNF-A325-S39-1d	22.07	0.1803	17.52	97.2	392	2015	506.2	470.9	82
First Recontact Kd									
BNF-A325-S39-1R-1	14.56	0.1201	11.55	96.2	95.21	386.1	395.2		
BNF-A325-S39-1R-1d	15.63	0.1202	12.40	103.2	97.23	294.5	316.7	356	78
Second Recontact Kd									
BNF-A325-S39-2R-1	10.026	0.0810	7.96	98.2	56.10	39.11	69.38		
BNF-A325-S39-2R-1d	10.572	0.0802	8.39	104.6	57.86	39.37	72.12	70.7	41
Tc-99 Spike Kd[†]									
BNF-A325-Tc-spr-F-2	15.90	na	12.62	na	25306	na	na	na	na
BNF-A325-S39-spr-1	11.63	0.1018	9.23	90.7	4683	20623	404.7		
BNF-A325-S39-spr-1d	11.59	0.1011	9.20	91.0	4725	20581	401.4	403.1	81

F-factor =0.987; Temperature = 26 ± °C; contact time = 24 ±1 h; batch #: 981015DHC720011
[†] 0.5 mL of 20 μCi/mLTc was added into 30 mL of the AN-103 sample. na = not applicable

Table 6. Characterization Data for Tc-99 Eluate Composite Solution

Sample ID		AN-103-TcELU-1		AN-103-TcELU-2		avg. values
Dilution factors		1.44E+01		1.35E+01		1.39E+01
Cs-137 (µCi/mL)		7.45E-01		6.99E-01		7.22E-01
Co-60 (µCi/mL)	<	3.69E-03	<	8.71E-04	<	2.28E-03
Eu-154 (µCi/mL)	<	7.48E-03	<	7.63E-03	<	7.56E-03
Eu-155 (µCi/mL)	<	1.12E-02	<	1.10E-02	<	1.11E-02
Am-241 (µCi/mL)	<	1.97E-02	<	1.84E-02	<	1.91E-02
Tc-99 (mg/L) by ICP-MS		9.85E+00		9.59E+00		9.72E+00
Mass 232 (mg/L)	<	1.72E-04	<	1.62E-04		1.67E-04
233	<	1.72E-04	<	1.62E-04	<	1.67E-04
234	<	1.72E-04	<	1.62E-04	<	1.67E-04
235		1.69E-02	<	1.62E-04	<	8.55E-03
236	<	1.72E-04	<	1.62E-04	<	1.67E-04
237	<	1.72E-04	<	1.62E-04	<	1.67E-04
238		1.25E-01		1.40E-01		1.32E-01
239	<	1.72E-04	<	1.62E-04	<	1.67E-04
240	<	1.72E-04	<	1.62E-04	<	1.67E-04
241	<	1.72E-04	<	1.62E-04	<	1.67E-04
242	<	1.72E-04	<	1.62E-04	<	1.67E-04
243	<	1.72E-04	<	1.62E-04	<	1.67E-04
244	<	1.72E-04	<	1.62E-04	<	1.67E-04
245	<	1.72E-04	<	1.62E-04	<	1.67E-04
total alpha (µCi/ml)		2.18E-02		1.72E-02	<	1.95E-02
Sr-90 (µCi/mL)		1.29E-02		6.18E-03		9.56E-03
K (mg/L) by AA		1.61E+07		6.16E+01		8.05E+06
Carbon						
inorganic (TIC), mg/L		6.32E+01		6.46E+01		6.39E+01
Organic (TOC), mg/L		5.46E+01		6.19E+01		5.82E+01
IC (mg/L)						
NO_3		1.38E+03		1.33E+03		1.36E+03
NO_2		1.15E+02		1.21E+02		1.18E+02
PO_4	<	1.44E+02	<	1.35E+02	<	1.39E+02
SO_4	<	7.18E+01	<	6.73E+01	<	6.96E+01
Oxalate	<	1.44E+02	<	1.35E+02	<	1.39E+02
Cl	<	2.17E+01	<	1.35E+01	<	1.76E+01
F	<	1.44E+01	<	1.35E+01	<	1.39E+01
Chem-Check						
U (mg/L)	<	1.44E-01	<	1.35E-01	<	1.39E-01
ICP-ES (mg/L)						
Al		6.48E+00		1.30E+02		6.81E+01
B	<	5.74E+00	<	5.38E+00	<	5.56E+00
Ba	<	2.87E-01	<	2.69E-01	<	2.78E-01
Cd	<	1.13E+01	<	9.07E+00	<	1.02E+01
Co	<	2.87E-01	<	2.69E-01	<	2.78E-01
Cr	<	7.18E-01	<	6.73E-01	<	6.96E-01
Cu	<	7.18E-01	<	6.73E-01	<	6.96E-01
Ca		8.62E-01		8.08E-01		8.35E-01
Fe	<	5.74E-01	<	1.87E+00	<	1.22E+00
La	<	1.87E+00	<	1.75E+00	<	1.81E+00
Li	<	4.31E-01	<	4.04E-01	<	4.17E-01
Mg	<	1.44E-01	<	1.35E-01	<	1.39E-01
Mn	<	1.44E-01	<	1.35E-01	<	1.39E-01
Mo		4.31E-01		4.04E-01		4.17E-01
Na		7.69E+02		7.44E+02		7.56E+02
Ni	<	1.87E+00	<	1.75E+00	<	1.81E+00
P		6.42E+00		4.27E+00		5.34E+00
Pb	<	4.16E+00	<	3.90E+00	<	4.03E+00
Re	<	7.32E+00	<	6.86E+00	<	7.09E+00
Si		4.06E+00		6.49E+00		5.28E+00
Sn	<	1.87E+00	<	1.75E+00	<	1.81E+00
Sr	<	1.44E-01	<	1.35E-01	<	1.39E-01
Ti	<	4.31E-01	<	4.04E-01	<	4.17E-01
V	<	5.74E-01	<	5.38E-01	<	5.56E-01
Zn	<	4.31E-01	<	4.04E-01	<	4.17E-01
Zr	<	7.18E-01	<	6.73E-01	<	6.96E-01

Table 7. Characterization data for decontaminated AN-103 product

Sample ID		A-103-Prod-1		A-103-Prod-2		avg. values
Cs-137 (µCi/mL)		1.28E+00		1.23E+00		1.26E+00
Co-60 (µCi/mL)	<	2.77E-03	<	6.53E-04	<	1.71E-03
Eu-154 (µCi/mL)	<	6.72E-03	<	6.99E-03	<	6.85E-03
Eu-155 (µCi/mL)	<	1.36E-02	<	1.23E-02	<	1.29E-02
Am-241(µCi/mL)	<	3.80E-02	<	2.27E-02	<	3.04E-02
Tc-99 (mg/L) by ICP-MS		2.31E-01		2.22E-01		2.26E-01
mass 232 (mg/L)		4.92E-02		1.73E-02		3.33E-02
233	<	1.28E-04	<	1.35E-04	<	1.32E-04
234	<	1.28E-04	<	1.35E-04	<	1.32E-04
235	<	1.28E-04	<	1.35E-04	<	1.32E-04
236	<	1.28E-04	<	1.35E-04	<	1.32E-04
237	<	1.28E-04	<	1.35E-04	<	1.32E-04
238		6.11E-01		6.23E-01		6.17E-01
239	<	1.28E-04	<	1.35E-04	<	1.32E-04
240	<	1.28E-04	<	1.35E-04	<	1.32E-04
241	<	1.28E-04	<	1.35E-04	<	1.32E-04
242	<	1.28E-04	<	1.35E-04	<	1.32E-04
243	<	1.28E-04	<	1.35E-04	<	1.32E-04
244	<	1.28E-04	<	1.35E-04	<	1.32E-04
245	<	1.28E-04	<	1.35E-04	<	1.32E-04
K (AA), mg/L		4.23E+03		4.29E+03		4.26E+03
Carbon (mg/L)						
inorganic (TIC)		2.31E+03		2.31E+03		2.31E+03
Organic (TOC)		1.57E+03		1.33E+03		1.45E+03
ICP-ES (mg/L)						
Ag	<	4.28E-01	<	4.50E-01	<	4.39E-01
Al		2.24E+04		2.24E+04		2.24E+04
B	<	1.29E+01	<	1.24E+01	<	1.27E+01
Ba	<	2.14E-01	<	2.25E-01	<	2.19E-01
Cd	<	7.49E-01	<	6.53E-01	<	7.01E-01
Co	<	5.35E-01	<	5.63E-01	<	5.49E-01
Cr	<	7.16E+01	<	7.20E+01	<	7.18E+01
Cu	<	6.42E-01	<	6.75E-01	<	6.58E-01
Ca		1.97E+00		2.08E+00		2.02E+00
Fe	<	4.28E-01	<	4.50E-01	<	4.39E-01
La	<	1.39E+00	<	1.46E+00	<	1.43E+00
Li	<	1.22E+00	<	1.17E+00	<	1.19E+00
Mg	<	1.07E-01	<	1.13E-01	<	1.10E-01
Mn	<	1.93E-01	<	1.58E-01	<	1.75E-01
Mo		4.20E+01		4.08E+01		4.14E+01
Na		1.16E+05		1.15E+05		1.15E+05
Ni	<	1.39E+00	<	1.46E+00	<	1.43E+00
P		3.32E+02		3.39E+02		3.35E+02
Pb	<	3.44E+01	<	3.38E+01	<	3.41E+01
Re	<	5.46E+00	<	5.74E+00	<	5.60E+00
Si		1.51E+02		1.53E+02		1.52E+02
Sn	<	4.22E+01	<	3.89E+01	<	4.06E+01
Sr	<	1.07E-01	<	1.13E-01	<	1.10E-01
Ti	<	3.21E-01	<	3.38E-01	<	3.29E-01
V	<	4.28E-01	<	4.50E-01	<	4.39E-01
Zn	<	3.21E-01	<	3.38E-01	<	3.29E-01
Zr	<	3.25E+00	<	7.31E-01	<	1.99E+00

Table 8. Characterization data for composite cesium eluate solution

Sample ID		AN-103-CsELU-1		AN-103-CsELU-2		avg. duplicate
Cs-137 (µCi/mL)		2.47E+03		2.50E+03		2.49E+03
Co-60 (µCi/mL)	<	7.84E+01	<	3.20E+01	<	5.52E+01
Eu-154 (µCi/mL)	<	1.19E+02	<	9.20E+01	<	1.06E+02
Eu-155 (µCi/mL)	<	1.73E+02	<	1.75E+03	<	9.61E+02
Am-241 (µCi/mL)	<	5.33E+02	<	4.47E+02	<	4.90E+02
Tc-99 (mg/L)	<	1.22E+00	<	1.20E+00	<	1.21E+00
Mass 232 (mg/L)		1.04E+00		6.95E-01		8.69E-01
233	<	4.26E-03	<	4.20E-03	<	4.23E-03
234	<	4.26E-03	<	4.20E-03	<	4.23E-03
235	<	4.26E-03		4.27E-01		2.16E-01
236	<	4.26E-03	<	4.20E-03	<	4.23E-03
237	<	4.26E-03	<	4.20E-03	<	4.23E-03
238		3.31E+01		3.14E+01		3.22E+01
239	<	4.26E-03	<	4.20E-03	<	4.23E-03
240	<	4.26E-03	<	4.20E-03	<	4.23E-03
241	<	4.26E-03	<	4.20E-03	<	4.23E-03
242	<	4.26E-03	<	4.20E-03	<	4.23E-03
243	<	4.26E-03	<	4.20E-03	<	4.23E-03
244	<	4.26E-03	<	4.20E-03	<	4.23E-03
245	<	4.26E-03	<	4.20E-03	<	4.23E-03
Sr-90 (dpm/mL)	<	1.27E+03	<	9.35E+02	<	1.10E+03
Pu-239/240 (µCi/ml)		4.36E-04		1.63E-04		3.00E-04
Pu-238 (µCi/ml)		5.32E-04		2.49E-04		3.90E-04
K (AA), mg/L		4.42E+01		1.00E+02		7.23E+01
Carbon (mg/L)						
inorganic (TIC)	<	1.90E+02		1.87E+02		1.88E+02
Organic (TOC)		9.71E+02		9.07E+02		9.39E+02
U (mg/L)		3.23E+01		2.24E+01		2.73E+01
IC (mg/L)						
NO_3	<	1.90E+04	<	1.87E+04		1.88E+04
NO_2	<	1.90E+04	<	1.87E+04		1.88E+04
PO4	<	1.90E+04	<	1.87E+04		1.88E+04
SO4	<	9.49E+03	<	9.35E+03		9.42E+03
Oxalate	<	1.90E+04	<	1.87E+04		1.88E+04
Cl (ISE)		8.69E+03	<	7.87E+03		8.28E+03
F (ISE)	<	8.16E+01	<	8.04E+01		8.10E+01
ICP-ES (mg/L)						
Ag	<	1.57E+01	<	1.12E+01	<	1.35E+01
Al		6.17E+01		5.65E+01		5.91E+01
B	<	2.28E+01	<	2.24E+01	<	2.26E+01
Ba	<	6.26E+00		3.74E+00	<	5.00E+00
Cd	<	3.79E+00	<	3.74E+00	<	3.77E+00
Co	<	1.14E+01	<	1.12E+01	<	1.13E+01
Cr		1.76E+01		1.23E+01		1.50E+01
Cu		7.59E+00		8.42E+00		8.00E+00
Ca		3.89E+02		1.95E+02		2.92E+02
Fe		1.54E+01		8.23E+00		1.18E+01
La	<	1.52E+01	<	1.50E+01	<	1.51E+01
Li	<	6.45E+00	<	5.61E+00	<	6.03E+00
Mg		1.84E+01		7.48E+00		1.29E+01
Mn	<	1.90E+00	<	1.87E+00	<	1.88E+00
Mo	<	4.55E+00	<	3.74E+00	<	4.15E+00
Na		1.03E+03		1.09E+03		1.06E+03
Ni	<	1.90E+01	<	1.87E+01	<	1.88E+01
P	<	8.35E+01	<	8.23E+01	<	8.29E+01
Pb	<	8.16E+01	<	8.04E+01	<	8.10E+01
Si	<	5.12E+01	<	5.05E+01	<	5.09E+01
Sn	<	3.79E+01	<	3.74E+01	<	3.77E+01
Sr		6.45E+00		4.86E+00		5.66E+00
Ti	<	7.40E+00	<	3.74E+00	<	5.57E+00
V	<	1.21E+01	<	7.48E+00	<	9.81E+00
Zn		2.75E+01		1.61E+01		2.18E+01
Zr	<	1.27E+01	<	9.35E+00	<	1.10E+01

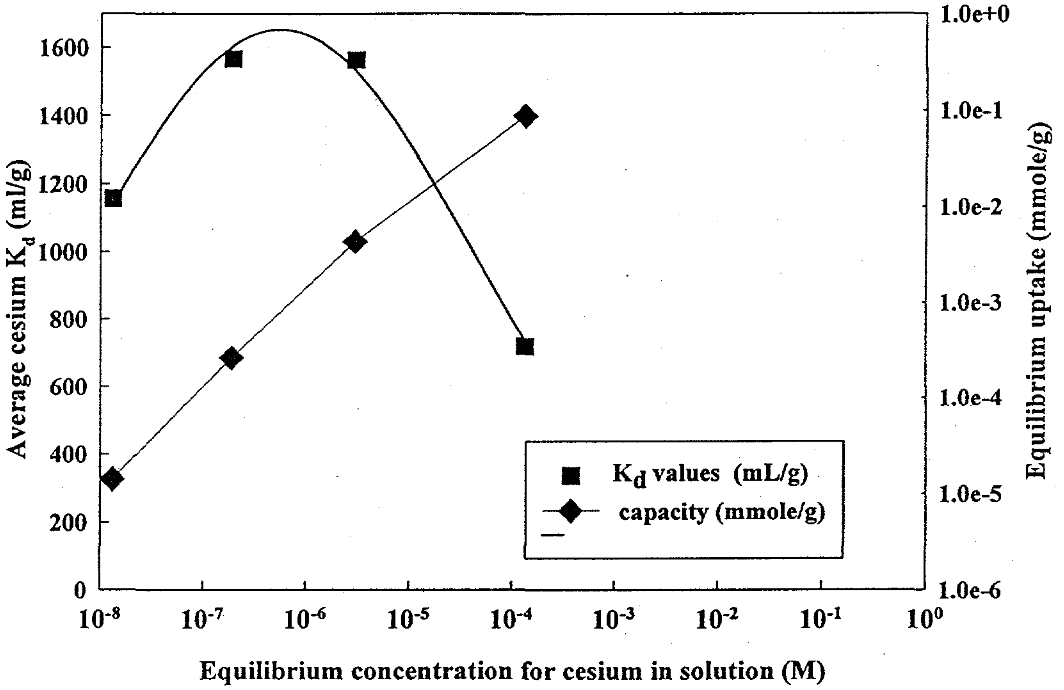


Figure 1. Batch distribution coefficients for cesium on SuperLig 644
(Feed: Tank-241-AN-103, batch # 981020mb48-563
Temperature ~ 26 deg. C; contact time: 24 hrs).

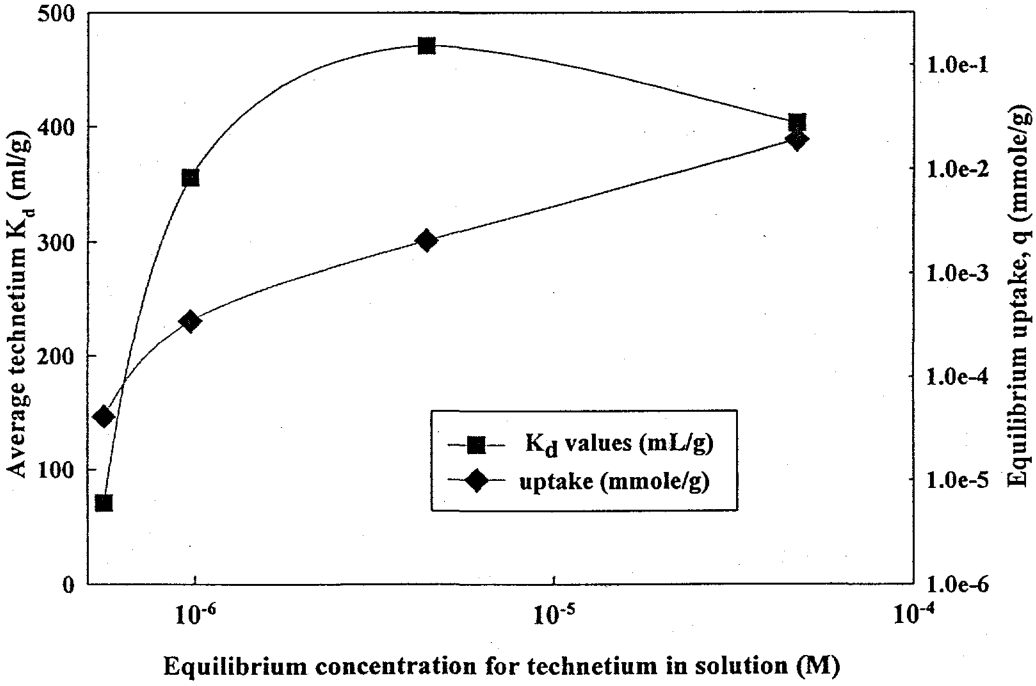


Figure 2. Batch distribution coefficients for cesium on SuperLig 644
(Feed: Tank-241-AN-103, batch: 98101DHC720011,
Temperature: ~ 26 deg. C; Contact time: 24 hours)

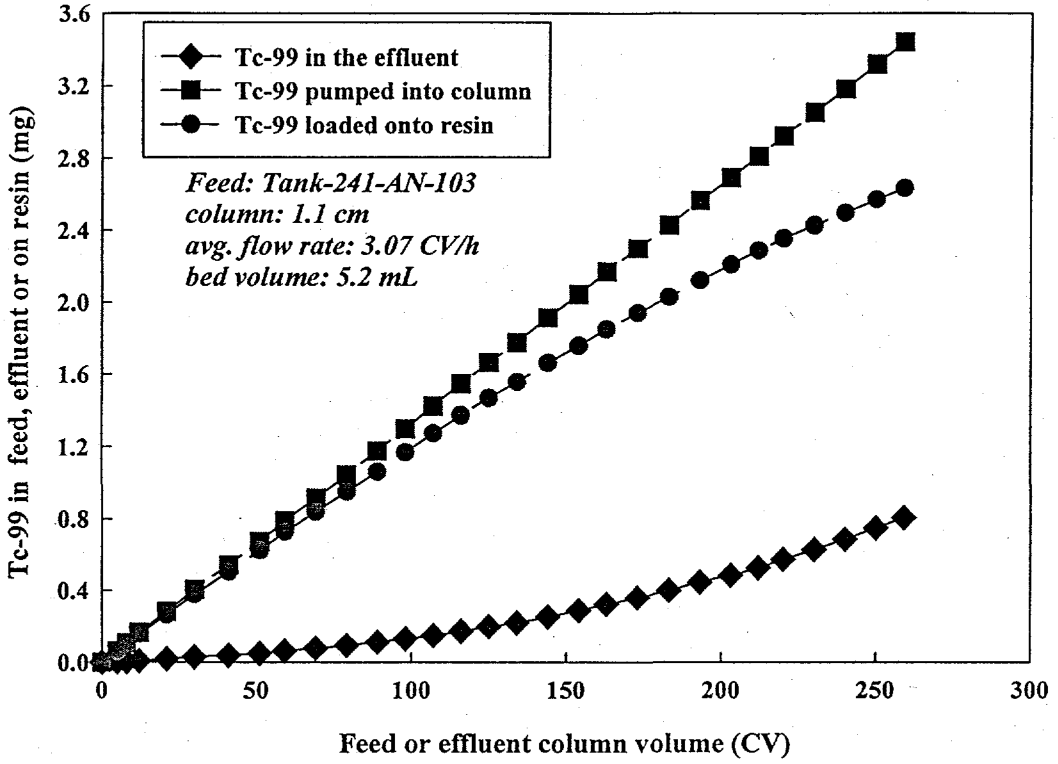


Figure 3. Technetium loading curve for Superlig 639 lead column

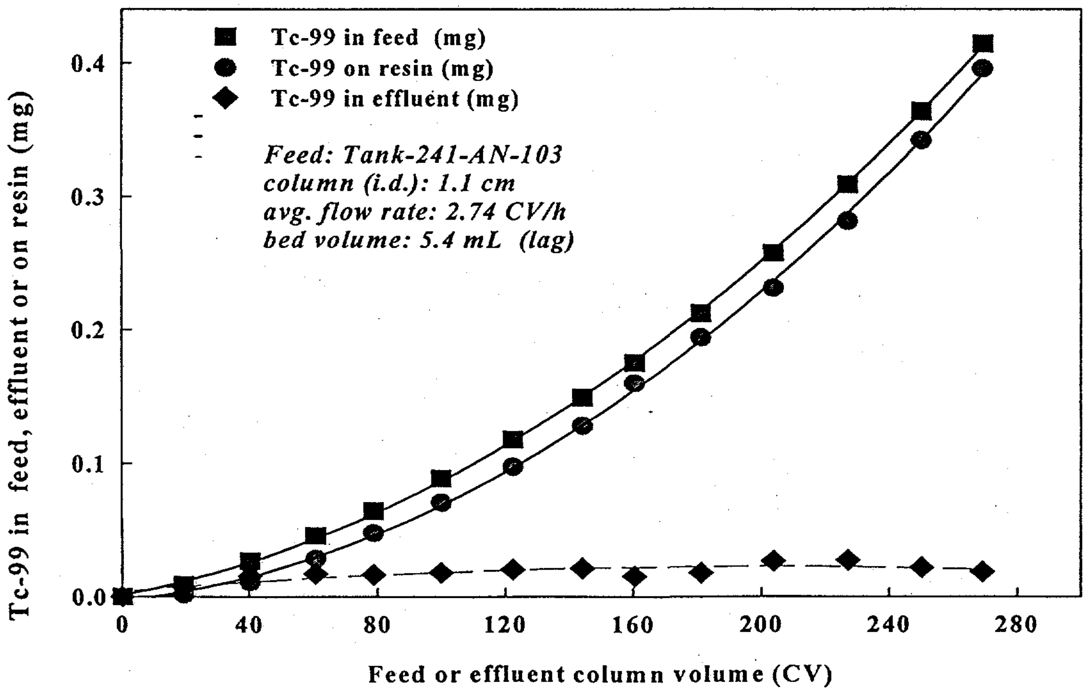
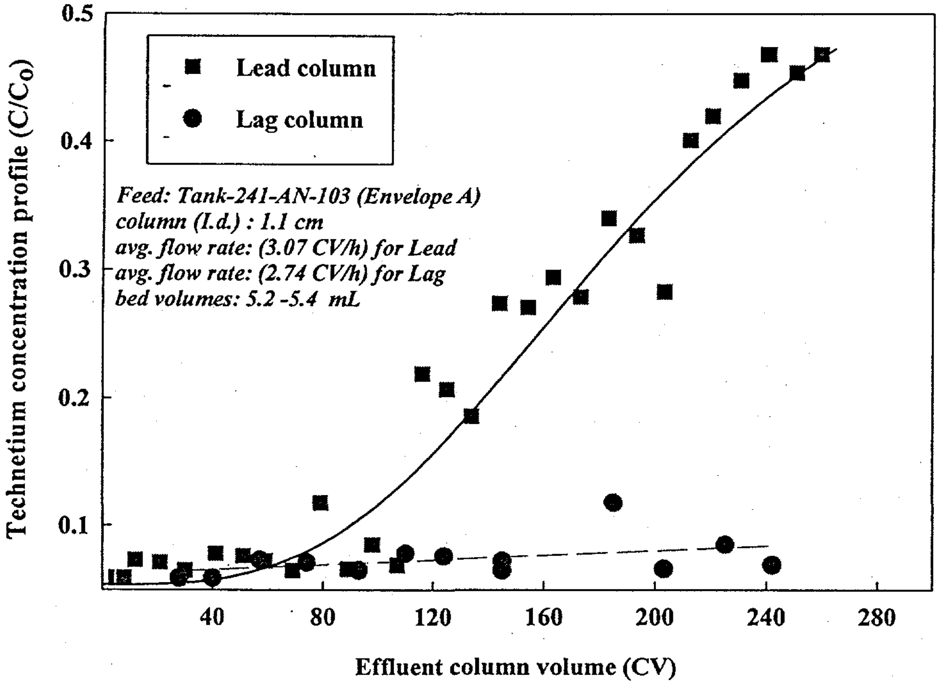


Figure 4. Technetium loading curve for Superlig 639 lag column



Feed: Tank-241-AN-103 (Envelope A)
column (I.d.) : 1.1 cm
avg. flow rate: (3.07 CV/h) for Lead
avg. flow rate: (2.74 CV/h) for Lag
bed volumes: 5.2 -5.4 mL

Figure 5. Technetium breakthrough curve for Superlig 639 resin

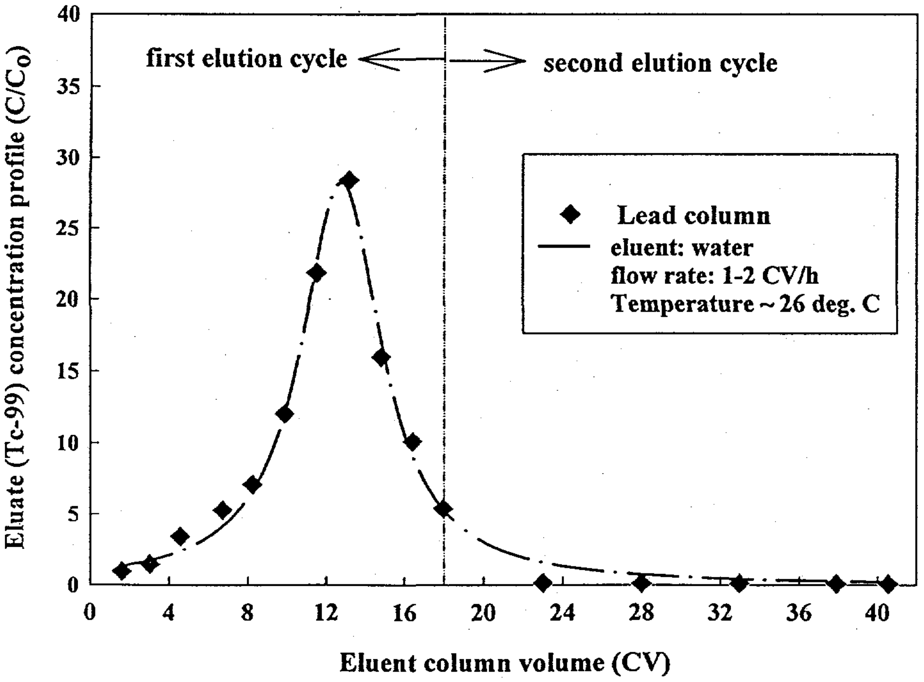


Figure 6. Technetium elution curve for Superlig 639 resin

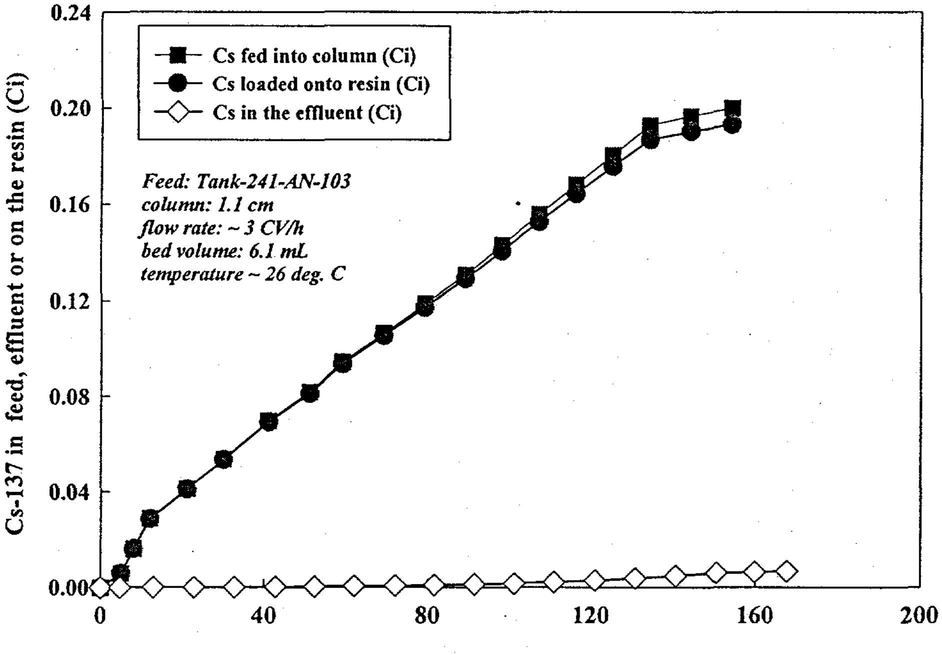


Figure 7. Cesium loading curve for Superlig 644 lead column

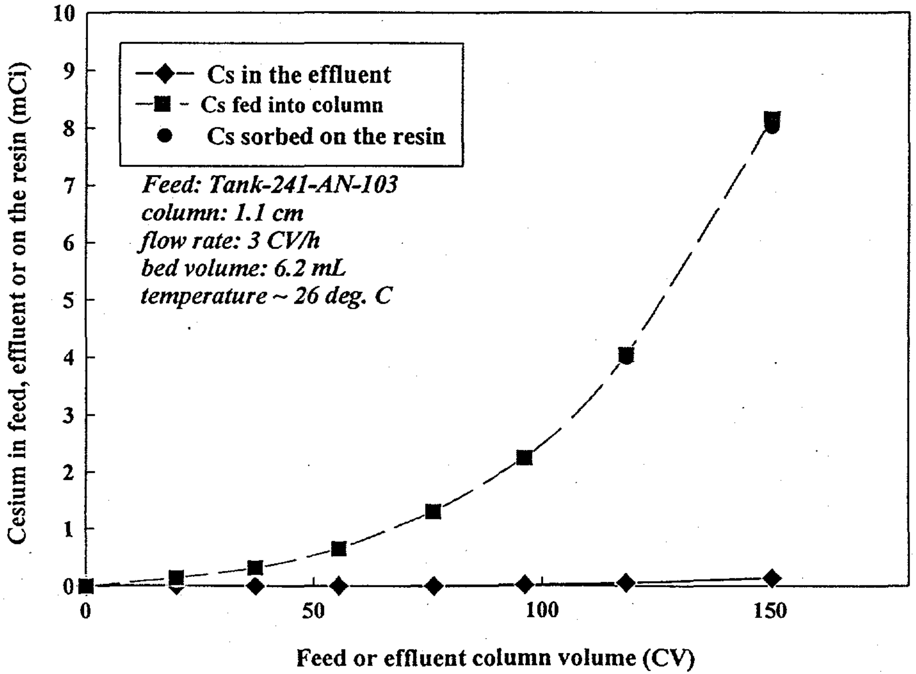


Figure 8. Cesium loading curve for Superlig 644 lag column

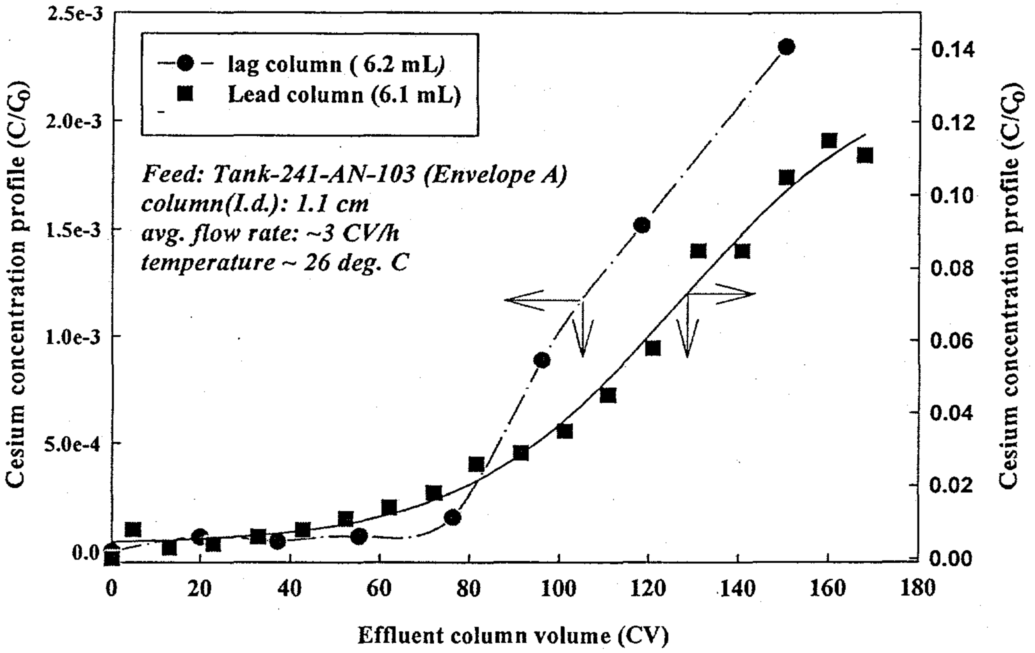


Figure 9. Cesium breakthrough curves for Superlig 644 resin

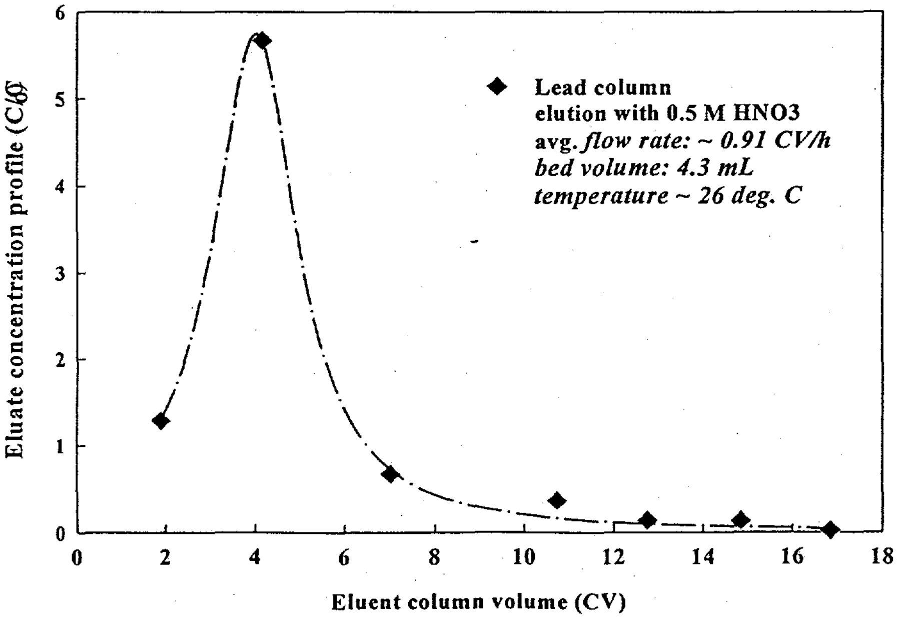


Figure 10. Cesium elution curve for Superlig 644 resin

7.0 Appendix A:

Table A-1. Standard Batch Distribution Coefficient for cesium from Tank-241-AN-103 on SuperLig® 644

Sample ID	**BNF-A325-S644-1**	**BNF-A325-S644-1D**	**BNF-A325-F44-1**
solution Mass (g)	18.153	17.344	18.233
resin mass (g)	0.18	0.18	na
solution Vol. (mL)	14.41	13.77	14.47
phase ratio	80.04	76.47	na
Cs conc. (μCi/mL)	10.005	10.168	16.058
water dil. (mL)	na	na	10.004
sample mass (g)	na	na	1.149
dilution factor	na	na	11.971
Dilution corrected Cs-137			
Cs conc. (μCi/mL)	10.005	10.17	192.2
Cs removed (μCi/mL)	182.2	182.1	na
Kd (mL/g)	1577.7	1482	
ave. Kd (mL/g)	1529.8		
% removal	94.795	94.7	

na = not applicable

Table A-2. Sequential Batch Distribution Coefficients for cesium from Tank-241-AN-103 on SuperLig® 644

Sample ID	**BNF-A325-S644-1R-1**	**BNF-A325-S644-1R-1D**	**BNF-A325-S644-2R-1**	**BNF-A325-S644-2R-1D**
solution mass (g)	13.983	14.152	10.024	10.598
resin mass (g)	0.12	0.12	0.1	0.1
solution volume (mL)	11.10	11.23	7.96	8.41
phase ratio	92.48	93.60	79.56	84.11
[Cs-137] conc. (μCi/mL)	0.6186	0.6245	0.04381	0.04458
No dilution used				
[Cs-137] removed (μCi/mL)	9.4	9.5	0.575	0.580
F-factor	0.924	0.924	0.924	0.924
Kd (mL/g)	1519	1548	1130	1184
Average Kd (mL/g)	1533		1157	
[Cs-137] % removal	93.82	93.86	92.92	92.86
Contact time (hrs)	24±1	24±1	24±1	24±1
Temperature (°C)	27±1	27±1	27±1	27±1

Table A-3. Spiked Batch Distribution Coefficient for cesium on SuperLig® 644

Sample ID	BNF-A325-S644-sp-1	BNF-A325-S644-sp-1D	BNF-A325-Cs-sp-F2
solution mass (g)	12.663	12.655	11.542
resin mass (g)	0.1000	0.1000	na
solution volume (mL)	10.05	10.04	9.16
phase ratio	100.50	100.44	na
[Cs-137] conc. (µCi/mL)	30.686	33.099	18.121
Total cesium (ppb) *	16950	18660	10809
water of dilution (mL)	na	na	10.0042
sample mass (g)	na	na	1.12
dilution factor	na	na	12.255
Dilution corrected			
[Cs-137] (µCi/mL)	30.686	33.099	222.
Total cesium (ppb)	16950	18660	132461
[Cs-137] removed (µCi/mL)	191.4	189	
Total Cs removed (ppb)	115511	113801	
F-factor	0.924	0.924	
Kd (mL/g) – total cesium	741.2	662.9	
Kd (mL/g) –Cs-137	678.4	620.6	
[Cs-137] % removal	86.56		
Total cesium % removal	85.64		

Table A-4. Standard Batch Distribution Coefficient for Tc-99 from Tank-241-AN-103 on SuperLig® 639

Sample ID	BNF-A325-S39-1	BNF-A325-S39-1D	BNF-A325-F-1
solution mass (g)	24.431	22.07	22.756
resin mass (g)	0.1804	0.1803	NA
solution volume (mL)	19.39	17.52	18.06
phase ratio	107.48	97.15	nm
[Tc-99] conc. (ug/L)	40.76	34.46	240.18
water of dilution (mL)	10.0025	10.0086	10.006
sample mass (g)	1.122	1.214	1.33
dilution factor	12.233	11.388	10.479
Dilution corrected			
[Tc-99] conc. (µg/L)	481	392	2407
[Tc-99] removed (µg/L)	1925	2015	nm
F-factor	0.824	0.824	
Kd (mL/g)	511.8	594.8	
Average Kd (mL/g)	553.3		
[Tc-99] % removal	80.00	83.72	
Contact time (hrs)	24±1	24±1	
Temperature (°C)	26±1	26±1	

nm = not measured

Table A-5. Sequential Batch Distribution Coefficients for Tc-99 on SuperLig® 639

Sample ID	BNF-A325-S39-1R-1	BNF-A325-S39-1R-1D	BNF-A325-S39-2R-1	BNF-A325-S39-2R-1D
solution mass (g)	14.556	15.627	10.026	10.57
resin mass (g)	0.1201	0.1202	0.081	0.0802
solution volume (mL)	11.55	12.40	7.96	8.39
phase ratio	96.19	103.18	98.24	104.62
[Tc-99] conc. (μg/mL)	8.40	7.45	5.00	4.90
water of dilution (mL)	10.000	10.004	10.006	10.01
sample mass (g)	1.244	1.234	1.233	1.168
dilution factor	11.129	11.215	11.225	11.797
Dilution corrected				
[Tc-99] conc. (μg/L)	95	97	56	58
[Tc-99] removed (μg/L)	386	295	39.1	39.4
F-factor	0.824	0.824	0.824	0.824
Kd (mL/g)	464.4	372.1	81.5	84.7
Average Kd (mL/g)	418.2		83.1	
[Tc-99] % removal	80.22	75.18	41.1	40.5
Contact time (hrs)	24±1	24±1	24±1	24±1
Temperature (°C)	26±1	26±1	26±1	26±1

Table A-6. Spike Batch Distribution Coefficients for technetium on SuperLig® 639

Sample ID	BNF-A325-S39-spr-1	BNF-A325-S39-spr-1D	BNF-A325-Tc-spr-F-2
solution mass (g)	11.634	11.586	15.897
resin mass (g)	0.1018	0.1011	NA
solution volume (mL)	9.23	9.20	12.62
phase ratio	90.70	90.95	NA
[Tc-99] conc. (μg/L)	4682.83	4724.92	2126.51
water of dilution (mL)	nm	nm	10.2946
sample mass (g)	nm	nm	1.19
dilution factor	nm	nm	11.900
Dilution corrected			
[Tc-99] conc. (μg/L)	4683	4725	25306
[Tc-99] removed (μg/L)	20623	20581	
F-factor	0.824	0.824	
Kd (mL/g)	475.5	471.6	
Average Kd (mL/g)	473.6		
[Tc-99] % removal	81.5	81.3	
Contact time (hrs)	24±1	24±1	
Temperature (°C)	26±1	26±1	

Table A-7. Breakthrough data for Tc-99 on Lead column containing SuperLig 639

Sample ID	Lead column effluent (CV)	Tc-99 s (ppb) (ICP-MS)	Tc-99 activity (µCi/mL)	breakthrough profile (C/Co)
BNF-A-TcIXEFF-Cr1-Feed	na	2554	0.043	na
BNF-A-TcIXEFF-Cr1-LCS1	7	56	0.001	0.022
BNF-A-TcIXEFF-Cr1-LCS2	15	203	0.003	0.079
BNF-A-TcIXEFF-Cr1-LCS3	23	231	0.004	0.090
BNF-A-TcIXEFF-Cr1-LCS4	30	240	0.004	0.094
BNF-A-TcIXEFF-Cr1-LCS5	39	263	0.004	0.103
BNF-A-TcIXEFF-Cr1-LCS6	48	252	0.004	0.099
BNF-A-TcIXEFF-Cr1-LCS7	61	119	0.002	0.047
BNF-A-TcIXEFF-Cr1-LCS8	71	252	0.004	0.099
BNF-A-TcIXEFF-Cr1-LCS9	79	311	0.005	0.122
BNF-A-TcIXEFF-Cr1-LCS10	89	324	0.006	0.127
BNF-A-TcIXEFF-Cr1-LCS11	99	315	0.005	0.123
BNF-A-TcIXEFF-Cr1-LCS12	109	440	0.007	0.172
BNF-A-TcIXEFF-Cr1-LCS13	118	332	0.006	0.130
BNF-A-TcIXEFF-Cr1-LCS14	127	403	0.007	0.158
BNF-A-TcIXEFF-Cr1-LCS15	136	559	0.010	0.219
BNF-A-TcIXEFF-Cr1-LCS16	145	529	0.009	0.207
BNF-A-TcIXEFF-Cr1-LCS17	154	474	0.008	0.186
BNF-A-TcIXEFF-Cr1-LCS18	164	700	0.012	0.274
BNF-A-TcIXEFF-Cr1-LCS19	174	692	0.012	0.271
BNF-A-TcIXEFF-Cr1-LCS20	184	750	0.013	0.294
BNF-A-TcIXEFF-Cr1-LCS21	193	712	0.012	0.279
BNF-A-TcIXEFF-Cr1-LCS22	203	868	0.015	0.340
BNF-A-TcIXEFF-Cr1-LCS23	213	836	0.014	0.327
BNF-A-TcIXEFF-Cr1-LCS24	223	724	0.012	0.283
BNF-A-TcIXEFF-Cr1-LCS25	232	1023	0.017	0.401
BNF-A-TcIXEFF-Cr1-LCS26	240	1073	0.018	0.420
BNF-A-TcIXEFF-Cr1-LCS27	250	1145	0.019	0.448
BNF-A-TcIXEFF-Cr1-LCS28	260	1196	0.020	0.468
BNF-A-TcIXEFF-Cr1-LCS29	270	1160	0.020	0.454

Table A-8. Breakthrough data for Tc-99 on Lag column containing SuperLig 639 resin

Sample ID	Column effluent (CV)	Tc-99 (ppb) (ICP-MS)	Tc-99 activity (μCi/mL)	Breakthrough profile (C/Co)
BNF-A-TcIXEFF-Cr1-Feed	na	2554	4.34E-02	na
BNF-A-TcIXEFF-Cr1-1CV/S1	28	151	2.57E-03	0.059
BNF-A-TcIXEFF-Cr1-2CV/S2	40	151	2.57E-03	0.059
BNF-A-TcIXEFF-Cr1-3CV/S3	57	186	3.16E-03	0.073
BNF-A-TcIXEFF-Cr1-4CV/S4	74	181	3.08E-03	0.071
BNF-A-TcIXEFF-Cr1-5CV/S5	93	166	2.82E-03	0.065
BNF-A-TcIXEFF-Cr1-6CV/S6	110	199	3.38E-03	0.078
BNF-A-TcIXEFF-Cr1-7CV/S7	124	194	3.30E-03	0.076
BNF-A-TcIXEFF-Cr1-8CV/S8	145	183	3.11E-03	0.072
BNF-A-TcIXEFF-Cr1-9CV/S9	145	165	2.81E-03	0.065
BNF-A-TcIXEFF-Cr1-10CV/S10	185	301	5.12E-03	0.118
BNF-A-TcIXEFF-Cr1-11CV/S11	203	168	2.86E-03	0.066
BNF-A-TcIXEFF-Cr1-12CV/S12	225	217	3.69E-03	0.085
BNF-A-TcIXEFF-Cr1-13CV/S13	242	177	3.01E-03	0.069

Table A-9. Elution data for Tc-99 from Lead column containing SuperLig 639 resin

Sample ID	Column effluent (CV)	Tc-99 (ppb) (ICP-MS)	Tc-99 activity (μCi/mL)	breakthrough profile (C/Co)
BNF-A-TcLead-Eluate-Cr1-1CV	1.616	2472	4.20E-02	0.968
BNF-A-TcLead-Eluate-Cr1-2CV	3.038	3737	6.40E-02	1.463
BNF-A-TcLead-Eluate-Cr1-3CV	4.556	8761	1.49E-01	3.430
BNF-A-TcLead-Eluate-Cr1-4CV	6.722	13527	2.30E-01	5.296
BNF-A-TcLead-Eluate-Cr1-5CV	8.232	18124	3.08E-01	7.097
BNF-A-TcLead-Eluate-Cr1-6CV	9.866	30732	5.22E-01	12.033
BNF-A-TcLead-Eluate-Cr1-7CV	11.519	55882	9.50E-01	21.880
BNF-A-TcLead-Eluate-Cr1-8CV	13.159	72538	1.23E+00	28.402
BNF-A-TcLead-Eluate-Cr1-9CV	14.790	40784	6.93E-01	15.968
BNF-A-TcLead-Eluate-Cr1-10CV	16.391	25723	4.37E-01	10.072
BNF-A-TcLead-Eluate-Cr1-11CV	17.952	13763	2.34E-01	5.389
BNF-A-TcLead-Eluate-Cr1-1CV-R	22.985	380	6.00E-03	0.149
BNF-A-TcLead-Eluate-Cr1-2CV-R	27.979	309	5.00E-03	0.121
BNF-A-TcLead-Eluate-Cr1-3CV-R	32.952	291	5.00E-03	0.114
BNF-A-TcLead-Eluate-Cr1-4CV-R	37.899	258	4.00E-03	0.101
BNF-A-TcLead-Eluate-Cr1-5CV-R	40.552	168	3.00E-03	0.066

Table A-10. Breakthrough data for Cs-137 on Lead column containing SuperLig 644 resin

Sample ID	Column Effluent (CV)	Cs-137 concentration (µCi/mL)	breakthrough profile (C/Co
BNF-A-CsIXEFF-cr2-Feed	na	2.07E+02	na
BNF-A-CsIXEFF-cr2-LCS1	4.76	1.60E+00	7.70E-03
BNF-A-CsIXEFF-cr2-LCS2	12.93	7.01E-01	3.40E-03
BNF-A-CsIXEFF-cr2-LCS3	22.88	8.74E-01	4.20E-03
BNF-A-CsIXEFF-cr2-LCS4	32.83	1.21E+00	5.90E-03
BNF-A-CsIXEFF-cr2-LCS5	42.78	1.60E+00	7.70E-03
BNF-A-CsIXEFF-cr2-LCS6	52.37	2.33E+00	1.12E-02
BNF-A-CsIXEFF-cr2-LCS7	62.05	2.93E+00	1.41E-02
BNF-A-CsIXEFF-cr2-LCS8	72.00	3.76E+00	1.82E-02
BNF-A-CsIXEFF-cr2-LCS9	81.61	5.43E+00	2.62E-02
BNF-A-CsIXEFF-cr2-LCS10	91.41	5.99E+00	2.89E-02
BNF-A-CsIXEFF-cr2-LCS11	101.2	7.28E+00	3.52E-02
BNF-A-CsIXEFF-cr2-LCS12	110.9	9.39E+00	4.53E-02
BNF-A-CsIXEFF-cr2-LCS13	120.9	1.20E+01	5.78E-02
BNF-A-CsIXEFF-cr2-LCS14	130.8	1.77E+01	8.52E-02
BNF-A-CsIXEFF-cr2-LCS15	140.7	1.76E+01	8.51E-02
BNF-A-CsIXEFF-cr2-LCS16	150.6	2.17E+01	1.05E-01
BNF-A-CsIXEFF-cr2-LCS17	159.9	2.38E+01	1.15E-01
BNF-A-CsIXEFF-cr2-LCS18	164.0	2.30E+01	1.11E-01

Table A-11. Breakthrough data for Cs-137 on Lag column containing SuperLig 644 resin

Sample ID	Column effluent (CV)	Cs-137 concentration (µCi/mL)	breakthrough profile (C/Co
BNF-A-CsIXEFF-cr2-Feed	na	2.07E+02	na
BNF-A-CsIXEFF-cr2-1Cv/S1	20	1.39E-02	6.695E-05
BNF-A-CsIXEFF-cr2-2CV/S2	37	9.47E-03	4.574E-05
BNF-A-CsIXEFF-cr2-3CV/S3	55	1.46E-02	7.032E-05
BNF-A-CsIXEFF-cr2-4CV/S4	75	3.28E-02	1.585E-04
BNF-A-CsIXEFF-cr2-5CV/S5	95	1.84E-01	8.881E-04
BNF-A-CsIXEFF-cr2-6CV/S6	117	3.14E-01	1.518E-03
BNF-A-CsIXEFF-cr2-7CV/S7	143	4.85E-01	2.344E-03

na = not measured