*Effects of Soluble Organic Complexants and
Their Degradation Products on the Removal of
Selected Radionuclides from High-Level Waste*

*Part II: Distributions of Sr, Cs, Tc, and Am
onto 32 Absorbers from Four Variations of
Hanford Tank 101-SY Simulant Solution*

Los Alamos
NATIONAL LABORATORY

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*Edited by Mary Mann, Group CIC-1
Photocomposition by Kathy E. Valdez, Group CIC-1*

*This work was supported by the U.S. Department of Energy,
Office of Environmental Restoration and Waste Management.*

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TRADEMARKS

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EFFECTS OF SOLUBLE ORGANIC COMPLEXANTS AND THEIR DEGRADATION PRODUCTS ON THE REMOVAL OF SELECTED RADIONUCLIDES FROM HIGH-LEVEL WASTE

Part II: Distributions of Sr, Cs, Tc, and Am onto 32 Absorbers from Four Variations of Hanford Tank 101-SY Simulant Solution

by

S. Fredric Marsh, Zita V. Svitra, and Scott M. Bowen

ABSTRACT

Many of the radioactive waste storage tanks at U.S. Department of Energy facilities contain organic compounds that have been degraded by radiolysis and chemical reactions during decades of storage. In this second part of our three-part investigation of the effects of soluble organic complexants and their degradation products, we measured the sorption of strontium, cesium, technetium, and americium onto 32 absorbers that offer high sorption of these elements in the absence of organic complexants. The four solutions tested were (1) a simulant for a 3:1 dilution of Hanford Tank 101-SY contents that initially contained ethylenediaminetetraacetic acid (EDTA), (2) this simulant after gamma-irradiation to 34 Mrads, (3) the unirradiated simulant after treatment with a hydrothermal organic-destruction process, and (4) the irradiated simulant after hydrothermal processing. For each of 512 element/absorber/solution combinations, we measured distribution coefficients (Kds) twice for each period for dynamic contact periods of 30 min, 2 h, and 6 h to obtain information about sorption kinetics. On the basis of our 3,072 measured Kd values, the sorption of strontium and americium is significantly decreased by the organic components of the simulant solutions, whereas the sorption of cesium and technetium appears unaffected by the organic components of the simulant solutions.

EXECUTIVE SUMMARY

Successful remediation of the large quantities of radioactive hazardous waste stored in underground tanks at the Hanford Reservation near Richland, Washington, requires the identification of reliable partitioning agents and suitable technologies. To address this need, we previously completed three screening studies that collectively provide more than 12,000 measured distribution coefficients (Kds) for sorption of 16 elements onto more than 100 absorber materials from 5 simulated Hanford tank waste solutions. The simulants we tested were acid-dissolved sludge (pH 0.6), acidified supernate (pH 3.5), and alkaline supernate (pH 13.9) solutions for Hanford Tank 102-SY,¹ generic Hanford double-shell slurry feed

(DSSF),² and generic Hanford neutralized current-acid waste (NCAW).³

None of these simulants, however, contained any of the degraded organic compounds known to be present in numerous other Hanford waste tanks.⁴ Because many of the organic compounds in Hanford waste interact with multivalent cations, such organic compounds can form aqueous-soluble complexes with metal ions that would otherwise be sorbed.

Our preliminary study⁵ of the effects of ethylenediaminetetraacetic acid (EDTA) and its radiolysis products on the sorption of strontium, cesium, and technetium evaluated 18 absorbers that had shown high sorption of strontium in our previous studies.¹⁻³ In that preliminary study, we used a simulated leachate from an

irradiated simulated slurry for Hanford Tank 101-SY to demonstrate that degraded organic complexants dramatically decrease the sorption of strontium on even the best absorbers for this element.

For our present study, we used a realistic EDTA-containing simulant for Hanford Tank 101-SY and three variations of this simulant. The first variation is the initial simulant solution after gamma-irradiation to 34 Mrads with ^{60}Co . The second variation is the unirradiated simulant after treatment with a hydrothermal organic-destruction process.⁶ The final variation is the irradiated simulant after treatment with the hydrothermal process. Thus, the four simulant solutions represent the range of organic-containing Hanford waste from which key radionuclides must be removed.

The absorbers included in our tests either are already commercially available or could be produced in commercial quantities within a reasonable time. We measured distribution coefficients for each element/absorber/solution combination after dynamic contact periods of 30 min, 2 h, and 6 h to obtain information about the sorption kinetics of each system. Moreover, each element/absorber/solution combination was measured twice for each contact period.

Of the 32 tested absorbers, four were found to offer at least triple-digit K_d values for removing strontium from all four simulant solutions. Although americium was not as strongly sorbed on these absorbers as strontium, we nevertheless identified several absorbers that are capable of removing americium from waste that contains representative soluble organic compounds before the waste is subjected to any organic-destruction treatment.

Thus, existing absorbers appear to be capable of removing strontium from organic-containing waste with no prior organic-destruction treatment. However, for both strontium and americium, the measured K_d values are generally at least an order of magnitude higher and often two orders of magnitude higher for sorption from simulant solutions that have been treated with the hydrothermal organic-destruction process.⁶

In contrast, the sorption of cesium and technetium appears unaffected by the organic components in the four tested simulant solutions.

We plan to test the best absorbers with actual Hanford organic-containing waste, which is to be shipped to us within the next year, to confirm their suitability for removing selected radioactive elements from Hanford waste. Identification of reliable partitioning agents could allow processing of Hanford high-level-waste (HLW) tanks that contain soluble organic complexants to begin and be completed sooner, and at a lower cost, than would otherwise be possible. Thus, the

findings of our experimental studies could benefit radioactive waste processing, environmental remediation, and decommissioning efforts at Hanford and elsewhere within the U.S. Department of Energy (DOE) complex.

I. INTRODUCTION

The Hanford Reservation near Richland, Washington, harbors 177 underground tanks that store more than 65 million gallons of radioactive waste containing some 165 million curies. These HLWs are a byproduct of the production of nuclear materials for national defense needs during the past half-century. Because Hanford operating contractors used numerous chemical processes during this period and procedures for storing the resulting waste were not standardized, many different reagents and waste streams were combined in these underground tanks. The resulting mixtures of sludges, salt-cakes, slurries, and supernates are complex and unstable wastes whose disposal is a serious environmental challenge. Adding significantly to the stored-waste problem is the fact that 68 of these underground waste storage tanks are known, or are presumed, to have leaked.⁴

DOE has directed Los Alamos National Laboratory (LANL) and Sandia National Laboratories/New Mexico (SNL/NM) to support the Hanford Tank Waste Remediation Systems (TWRs) mission, which is to store, treat, and dispose of all tank waste in a safe, cost-effective, and environmentally sound manner. An essential prerequisite for achieving this goal is the identification of suitable partitioning agents and technologies.

In support of the TWRs mission, we previously completed three absorber screening studies that collectively provide more than 12,000 measured distribution coefficients (K_d s) for sorption of 16 elements onto more than 100 commercially available and experimental absorber materials from five simulated Hanford waste solutions. The individual simulants we tested were acid-dissolved sludge (pH 0.6), acidified supernate (pH 3.5), and alkaline supernate (pH 13.9) solutions for Hanford Tank 102-SY;¹ generic Hanford double-shell slurry feed (DSSF);² and generic Hanford neutralized current-acid waste (NCAW).³ None of these five simulants contained any of the degraded organic complexants known to be present in numerous generic Hanford waste tanks.⁴

Although identification of the degraded organic compounds present in the four simulant solutions was far beyond the scope of our study, other investigators have identified many of the organic compounds that form under conditions that approximate the storage of nuclear

waste in underground waste tanks. More than 60% of the initial organic compounds were found to degrade during the storage of an alkaline simulant solution containing EDTA, nitrilotriacetic acid (NTA), N-(2-hydroxyethyl) ethylenediaminetriacetic acid (HEDTA), and citrate at ambient temperature for 171 days.⁷ The effects of gamma-irradiation on simulated mixed waste containing EDTA, NTA, HEDTA, and citrate have been reported,⁸ and the effects of radiolysis and chemical forces on the degradation of citric acid in simulated mixed nuclear waste have been compared.⁹ Measurements of actual Hanford radioactive waste¹⁰ successfully characterized nearly 95% of the total organic carbon (TOC) in neutralized cladding removal waste (NCRW), but only 1.2% of the TOC in double-shell slurry (DSS) waste. Reanalysis of the organics in a Hanford organic complexant waste recently identified 58 individual organic compounds that account for more than 80% of the TOC.¹¹

Because many of the organic complexants in Hanford waste significantly alter the chemistry of multivalent cations by forming aqueous-soluble complexes, these complexants directly compete with sorption processes that would otherwise occur. We recently documented a preliminary study⁵ of the effects of EDTA and its radiolysis products on the sorption of strontium, cesium, and technetium by 18 absorbers that had shown high sorption of strontium in our previous screening studies.¹⁻³ In that preliminary study, we tested a simulated leachate from an irradiated simulated slurry for Hanford Tank 101-SY that had been prepared from a simplified recipe.¹²

The study reported here used a realistic EDTA-containing simulant for the contents of Hanford Tank 101-SY and three variations of this simulant. The first variation is the initial simulant solution after gamma-irradiation to 34 Mrads with ⁶⁰Co. The second variation is the unirradiated simulant after treatment with a hydrothermal organic-destruction process.⁶ The final variation is the irradiated simulant after treatment with the hydrothermal process. Thus, we used four simulant solutions that represent the range of organic-containing Hanford tank waste from which key radionuclides must be removed.

II. EXPERIMENTAL PARAMETERS

A. Simulant Solutions

The four simulant solutions were prepared as follows.

- (1) Unirradiated/untreated: The procedure of Hohl¹³ was used to prepare a realistic 3:1 diluted simulant for Hanford Tank 101-SY. Degradation of the initial

EDTA into other organic compounds in this highly alkaline solution was expected⁷ during the many months between its preparation and experimental testing.

- (2) Irradiated/untreated: A portion of the simulant solution described in (1) subsequently was gamma-irradiated, while exposed to air, at 1.35 Mrads/h to a total of 34 Mrads with ⁶⁰Co in the Gamma Irradiation Facility at Sandia/NM. Irradiation is known to degrade most organic compounds into simpler compounds and carbon dioxide, which forms carbonate in alkaline solutions. Moreover, the solution heating that occurred during the gamma-irradiation may have contributed to the organic degradation. Although we obtained no temperature profile during the irradiation, the solution temperature at the end of the 24-h irradiation was measured to be 49°C.
- (3) Unirradiated/hydrothermal-treated: A portion of the unirradiated simulant solution described in (1) was treated with a hydrothermal organic-destruction process.⁶ The small-scale hydrothermal unit used to process this solution consisted of a 5-ft reactor operating at 450°C and 15,000 psi. The typical residence time of solution passing through this reactor was approximately 25 s.
- (4) Irradiated/hydrothermal-treated: A portion of the irradiated simulant solution described in (2) was treated with the same hydrothermal organic-destruction process described in (3).

The variations of the initial Hanford Tank 101-SY simulant solution differed significantly in appearance. Whereas the unirradiated solution appeared green, the gamma-irradiated portion appeared less green and more yellow. Both hydrothermal-treated portions were bright yellow. These differences are reflected in the visible absorbance spectra of the four solutions in Fig. 1.

Fig. 1. Visible absorbance spectra of the four variations of Hanford Tank 101-SY simulant solution.

The compositions of the initial simulant solution and the three variations of the initial simulant solution are presented in Table 1. Because the measurement of TOC can be uncertain when many unusual organic compounds might be present, the analytical technique used for TOC was tailored to the specific compositions of organics in Hanford tank waste.¹⁴

B. Radiotracers

Because gamma spectrometry would be used to measure the K_d values of the four elements, we added appropriate quantities of radiotracers for strontium, cesium, technetium, and americium to each simulant solution. The four radiotracers and their gamma energies, branching ratios, and concentrations are shown in Table 2. Each simulant solution was initially passed through an Acrodisc™ LC13 PVDF 0.45-μm filter* to remove any

insoluble materials, and the four radiotracers were then added. The simulant and radiotracers were stirred thoroughly and then left undisturbed for at least one week to allow adequate time for soluble complexes or insoluble compounds to form. Every radiotracer-containing simulant solution was refiltered through an Acrodisc™ filter just before its absorber-contact phase began.

C. Absorbers

The 32 selected absorbers (Table 3) offer high sorption of the four elements from previously tested alkaline simulant solutions.¹⁻³ Their suppliers or preparation details are included in the tables for the individual absorbers (section IV.B).

To put all absorbers on a comparable basis, we intentionally air-dried all moist resins and composites at room temperature before testing. Air-drying also prevented unintentional dilution. Had we added as-received wet resins, they could have diluted the simulant solution

*See the list of all trademarked products on p. viii.

Table 1. Compositions of the Four Variations of Hanford Tank 101-SY Simulant Used in This Study

Constituent	Concentration (M)			
	Unirradiated/ untreated	Irradiated/ untreated	Unirradiated/ hydrothermal-treated	Irradiated/ hydrothermal-treated
Cations^a				
Na	3.45	3.82	3.85	3.69
K	0.035	0.035	0.041	0.037
Rb	5×10^{-7}	3×10^{-7}	5×10^{-7}	4×10^{-7}
Cs	9.0×10^{-5}	9.8×10^{-5}	9.8×10^{-5}	9.8×10^{-5}
Al	0.37	0.48	0.48	0.37
Ca	0.001	0.001	0.001	0.001
Cr	0.0062	0.0035	0.0083	0.0042
Fe	2.7×10^{-4}	1.0×10^{-4}	4.5×10^{-5}	3.8×10^{-5}
Sr	3.1×10^{-6}	1.5×10^{-6}	7×10^{-8}	1×10^{-7}
Anions^b				
F				
Cl	0.096	0.091	0.089	0.091
NO ₃	0.67	0.67	0.55	0.54
NO ₂	0.89	1.03	0.48	0.59
PO ₄	0.027	0.028	0.023	0.023
SO ₄	0.016	0.016	0.015	0.015
CO ₃	1.64	1.63	2.03	2.18
Oxalate	BDL ^c	0.005	BDL	BDL
Total organic carbon	0.71	0.59	0.13	0.16
pH	13.7	13.7	13.6	13.5

^aCations, except Na and K, were measured by inductively coupled plasma/mass spectrometry (ICP/MS).

^bAnions, as well as Na and K cations, were measured by ion chromatography (IC).

^cBelow detection limit.

Table 2. Radiotracers Used in This Study

Radiotracer	Gamma Energy (MeV)	Gamma Branching (%)^a	Estimated Concentration
⁸⁵ Sr	0.514	100	3 µg/L
¹³⁷ Cs	0.662	85	6 µg/L
^{95m} Tc	0.204	100	2 pg/L
²⁴¹ Am	0.0595	35.3	3–30 µg/L

^aGamma branching information is provided to indicate the assay sensitivity.

Table 3. Absorbers Evaluated in This Study

Absorber Type/Source	Trademark^a or Other Identification)
Commercially Available Absorbers	Amberlite™ IRC-718 cation exchange resin Bone char absorber Clinoptilolite absorber Diphonix™ cation exchange resin Duolite™ C-467 cation exchange resin Duolite™ CS-100 cation exchange resin Ionac™ SR-6 anion exchange resin Ionsiv™ IE-910 (CST) powder absorber Ionsiv™ IE-96 zeolite absorber Ionsiv™ TIE-96 TiO ₂ -loaded zeolite absorber Nusorb™ Ferroc carbon A absorber Purolite™ A-520-E anion exchange resin Resorcinol/formaldehyde resin (BSC-210) Reillex™ HPQ anion exchange resin Tannin absorber TRU-Spec™ extractant resin
Developmental Absorbers	KCoFC (potassium hexacyanoferrate) crystals NiFC-PAN nickel hexacyanoferrate Sodium nanotitanate (8104-170) ^b Sodium nanotitanate (8225-127) ^b Superlig™ 644 TRW CS-WA treated coal TRW CS-SA treated coal
Experimental Absorbers	Aliquat™ 336 extractant beads Clinoptilolite (purified) absorber Cyanex™ 923 extractant beads PNPK-1 extractant beads PNPK-2 extractant beads N-Butyl-HP anion exchange resin N-Hexyl-HP anion exchange resin N-Octyl-HP anion exchange resin SNL/HTO amorphous hydrous titanium dioxide

^aTrademark owners are identified on p. viii.

^bAlthough at the time of this writing Allied Signal, Inc., had not assigned tradenames to these absorbers, additional information about them can be obtained from Stephen Yates, Tel. 708-391-3446.

by as much as 23%. Although several of our colleagues expressed concern that air-drying might have adverse effects, our earlier studies demonstrated that the air-dried absorbers performed well.¹⁻³

Because air-drying these absorbers under other temperature, humidity, and altitude conditions could yield different moisture contents, knowing the ratio of the weight of each air-dried absorber to its weight after oven-drying at 110°C for 18 h is useful (see Table 4). Drying resins at 110°C will achieve the same moisture content in any laboratory; therefore, these ratios reflect the relative quantities of easily lost water in the air-dried absorbers we used and provide a basis for normalizing our data to those of other studies done with absorbers having different moisture contents.

D. Solution/Absorber Contacts

In most cases, a 250-mg portion of each air-dried absorber was contacted with a measured 6-mL volume of the simulant solution in a specially modified 20-mL disposable hypodermic syringe. We modified these syringes by inserting cylindrical plugs cut from 0.25-in.-thick porous Kynar™ (obtained from Porex Technologies, Fairburn, Georgia) into the tapered tips as filters, which permitted only liquid to pass through.

For each set of experiments, we prepared six syringes, each containing a different absorber. A measured volume of the simulant solution was transferred by pipet into a disposable 15-mL plastic beaker and then drawn into a syringe through the Kynar™ filter. We then sealed the tip of each syringe with a solid Luer cap and placed

the syringes on a 48-rpm tube rotator for dynamic contact periods of 30 min, 2 h, and 6 h. At the end of each contact period, approximately 25% of the total solution volume was expelled through an attached Acrodisc™ LC13 PVDF 0.45-μm filter into a tared counting vial. We then weighed the dispensed solution and used that figure in the calculation of the K_d values.

The described filter-tip syringes were unsuitable for those absorbers whose particle size was smaller than the pores of the Kynar™ filters. For these finely divided materials, we transferred 6 mL of solution by pipet directly into a 30-mL centrifuge tube that contained 250 mg of absorber and mixed each absorber/solution on the rotator for the prescribed times. At the end of each contact period, the tubes were centrifuged at high speed for 5 min. We then transferred approximately 1.5 mL of the centrifuged solution into a 3-mL hypodermic syringe with an Acrodisc™ LC13 PVDF 0.45-μm filter attached to its Luer-lock tip. The centrifuged solution was expelled through this filter into a tared counting vial, from which we determined the solution weight.

E. Calculation of K_d Values

Distribution coefficients (K_d values) normally are calculated in terms of the dry absorber weight;¹⁵ optionally, K_d values can be calculated in terms of the wet-absorber volume. Because most of the absorbers we tested were received in dry form, we tested all absorbers on a comparable dry basis to conform to a consistent K_d convention. We recognize that air-drying moist absorbers to constant weight at room temperature causes some

Table 4. Ratios of Air-Dried Weight to Oven-Dried Weight for Each Dried Absorber

Absorber	Air-dried weight/110°C-dried weight
Amberlite™ C-467	1.136
Amberlite™ IRC-718	1.100
Diphonix™	1.100
Duolite™ CS-100	1.033
Ionac™ SR-6	1.060
NiFC-PAN	1.321
Purolite™ A-520-E	1.137
Resorcinol/formaldehyde (BSC-210)	1.205
Reillex™ HPQ	1.076
N-Butyl-HP	1.132
N-Hexyl-HP	1.113
N-Octyl-HP	1.131
TRW CS-SA	1.091
TRW CS-WA	1.069

polymers to temporarily shrink as they lose physically sorbed water; however, such polymers swell again when they recontact aqueous solution. Therefore, the only adverse effect of air-drying might be a temporary decrease in polymer porosity that could result in slower kinetics until the polymer regains its lost water. To protect the air-dried absorbers from any irreversible damage, we never applied heat while drying the absorbers tested in this study.

Each portion of postcontact solution was assayed by gamma spectrometry using the characteristic gamma energies of the added radionuclides. The fraction of each element sorbed was determined indirectly from the difference in the measured gamma activity of the selected radionuclide before and after contact with each specified absorber.

All Kd values were calculated as follows:

$$Kd = \frac{Pr - Po}{Po} \frac{S}{A},$$

where Pr is the measured precontact activity per milliliter,
Po is the measured postcontact activity per milliliter,
S is the milliliters of solution contacted, and
A is the grams of dry absorber contacted.

F. Corrections

Because the solution weight of the postcontact assay portion usually differed slightly from that of the precontact standard solution, each postcontact solution was normalized to the same weight basis as the standard.

We also made appropriate corrections to account for the fact that the liquid-to-solid ratio and the remaining activity of every radionuclide decreased as successive assay portions were removed.

To determine the quantities of sorbed radionuclides, we compared the activity of each radionuclide in the postcontact solution with the activity of that same radionuclide in the precontact standard. Because most absorbers were initially added in air-dried form, the aqueous solvent was often preferentially sorbed, thereby decreasing the aqueous volume and simultaneously increasing the concentration of the solutes. Consequently, the activity of certain radionuclides was at times slightly higher in the postcontact solution than in the precontact solution. Whenever the postcontact/precontact solution activity ratio of a radionuclide exceeded unity, we assigned a value of 1.000 to the ratio having the highest value and normalized all other radionuclides in that portion of solution accordingly.

G. Measurement Precision

We measured each of the 1,536 element/absorber/solution/contact-time combinations twice for each contact period and calculated the relative standard deviation for each pair of measurements. We then used these individual relative standard deviations to compute pooled standard deviations for three ranges of Kd values. The resulting pooled relative standard deviation for a single measurement was 69% for Kd values between 0.1 and 0.25, 29% for Kd values between 0.25 and 1.0, and 10% for Kd values higher than 1.0. We report the average of each pair of Kd measurements for some of the absorbers tested in section III.A and the average and associated (absolute) standard deviation for every pair of measurements in section III.B.

We did not measure Kd values in duplicate during our earlier studies.^{1-3,5} However, because the experimental technique we used was essentially identical to the one we used in this study, the pooled standard deviations derived in the present study should also provide a reliable estimate of the precision of the Kd values obtained in previous studies. The difficulties in accurately correcting for the complex background under the low-energy (59.5-keV) gamma peak of ²⁴¹Am resulted in Kd values with unusually large uncertainties for this element. Additional uncertainty for ²⁴¹Am measurements in the two hydrothermal-treated simulant solutions resulted from the tenfold-lower initial concentration of americium in these solutions than in the two untreated simulant solutions.

H. Data Transfer and Processing

To save time and minimize the human errors that can be introduced in transcribing large quantities of data, we automated the data transfer and calculation process. We used the *Gamanal*¹⁶ computer program to determine the area of each gamma peak from the raw data in the multichannel analyzer memory. *Gamanal* provides a complete qualitative and quantitative analysis of mixtures of radionuclides by interpreting high-resolution gamma spectra. *Gamanal* determines background, fits and resolves complex peak groupings, determines the energies and absolute intensities of the gamma rays, and calculates the quantities of the source radionuclides. All interferences are resolved by a least-squares solution of the matrix of equations for the gamma intensities.

The *Gamanal* output was electronically imported into an EXCELTM spreadsheet in which the described calculations and corrections were applied. Finally, all calculated Kd values were combined into one master

spreadsheet and converted to a database to provide tabulated comparisons of the measured Kd values according to element and absorber. Because small Kd values always have large associated uncertainties, we rounded all Kd values to not more than one place beyond the decimal point.

I. Calculation of Detection Limits

When peaks in a gamma spectrum were too small to be detected by the *Gamanal* program, we used a simple method¹⁷ to calculate the appropriate detection limits. First, for each radionuclide of interest, we used the same detector to determine the gamma-peak position and full width at half maximum (FWHM) from a spectrum with a strong gamma signal. We then doubled the FWHM to include the entire gamma-peak region (always rounding up to the next integer), which defines the position and width of the region of interest to be applied to the background spectra. After summing the number of background counts in this region, we multiplied the square root of the sum by a confidence-limit scale factor. Finally, we applied a scale-factor value of 2.772 to ensure that 95% of the signal peaks of this magnitude would be detected on the observed background.

The scale factor was determined as follows. Counting statistics dictate that the square root of the number of counts in a region be an estimate of the standard deviation uncertainty of those counts. The detection of a peak requires that the background in the region of interest be subtracted; the single standard deviation of their difference is the square root of the sum of the squares of the single-standard-deviation values for each of the two spectra. Thus, the standard deviation is the square root of the sums of the counts in the two corresponding regions of interest.

Because the hypothetical sample spectrum contains no detectable peak signal, the number of counts in the sample region of interest is equal to the number of counts in the background. Therefore, an estimate of the single-standard-deviation uncertainty (the confidence limits of detection) is the square root of twice the number of background counts, or 1.414 times the square root of the number of background counts. Scaling to exactly two standard deviations (95.44% confidence) would yield a scale factor of 2.828. Standard math tables permit the scale factor to be calculated to any desired confidence limit.

Using these calculated minimum peak area values that could be detected with a 95% confidence level to calculate the corresponding minimum Kd values, we report Kd values in terms of *greater than* (>) the computed minimum Kd values.

III. RESULTS AND DISCUSSION

A high-temperature hydrothermal treatment⁶ was used to process portions of the unirradiated and gamma-irradiated Hanford Tank 101-SY simulant solutions. The hydrothermal system consisted of a reactor 5 ft long and 0.125 in. in diameter, maintained at 450°C and 15K psi. Back-pressure was maintained by a 100-ft length of 0.010-in. stainless steel capillary tubing on the reactor outlet. A needle valve on the outlet of the capillary tubing controlled the flow rate to maintain a solution residence time in the reactor of about 25 s. (This small-scale hydrothermal system, located in the Los Alamos Radiochemistry Facility where the rest of our experimental work was done, had been built the previous year as part of a TWRS-funded development program.)

Although the hydrothermal process destroyed more than 70% of the organic carbon (see Table 1), the organic destruction was not complete. In nearly all cases, the Kd values for strontium and americium from hydrothermal-treated simulants were higher than those from the corresponding untreated simulants. In some cases the Kd values from irradiated solutions are lower than those from their unirradiated counterparts. We speculate that the EDTA initially present was decomposed by gamma irradiation to simpler organic compounds that effectively compete with these absorbers for specific cations. Although we made no attempt to identify the surviving organic species, they are likely to include acetate, oxalate, and alkylamines.

A. Individual Elements

Tables 5 through 8 list some of the best absorbers for sorbing each element from the four simulant solutions; the absorbers are ranked in order of their measured Kd values for sorption from the initial Hanford Tank 101-SY simulant solution. Although absorbers that perform poorly generally are omitted from these tables, the tables in section III.B provide averages and standard deviations for each of the 1,536 sets of duplicate measurements that include all four elements.

1. Strontium. As expected, the sorption of strontium on all absorbers tested is dramatically affected by organic compounds in the two untreated simulant solutions. The fact that the Kd values are generally higher in the irradiated/untreated solution indicates that some of the organic compounds that interfere with the sorption of strontium are destroyed or decomposed into simpler organic compounds by the irradiation.

However, even in the unirradiated/untreated simulant solution where the interference of organic compounds appears greatest, four of the tested absorbers provide triple-digit Kd values for sorbing strontium.

Table 5. Kd Values for Sorbing Strontium from Four Variations of Hanford Tank 101-SY Simulant Solution

Absorber	Unirradiated/ untreated			Irradiated/ untreated			Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h
SNL/HTO	340	530	653	519	880	1312	>16K	>16K	>16K	>19K	>19K	>19K
Na titanate (8225-127)	107	197	248	212	398	554	>16K	>16K	>16K	>18K	>18K	>18K
Na titanate (8104-170)	43	68	100	91	150	217	>16K	>16K	>16K	>18K	>18K	>18K
KCoFC crystals	45	146	280	67	87	135	2352	5K	>25K	2117	5K	6K
Bone char	27	50	80	56	127	225	918	3666	4604	829	2310	6K
TRW CS-SA	3.7	3.5	3.3	5.1	5.7	5.6	674	872	849	822	964	980
Ionsiv™ TIE-96	2.0	3.6	4.8	4.1	8.3	12	146	419	754	128	393	704
Nusorb™ Ferrocabon A	1.9	4.4	7.3	4.7	9.4	16	470	1033	1683	404	840	1420
NiFC-PAN	1.8	5.0	8.4	13	289	1378	964	6K	>22K	1608	3444	6K
Diphonix™	1.8	2.7	3.0	4.1	6.7	8.3	506	925	1243	1090	1986	3222
Ionsiv™ IE-910 (CST)	1.5	2.4	3.7	4.4	7.2	11	944	1383	2159	1012	1298	2134
Amberlite™ IRC-718	1.5	1.7	1.7	6.7	7.1	7.1	493	1149	1609	479	1067	1925
Clinoptilolite	0.3	0.7	0.7	1.4	1.3	1.7	20	24	29	17	20	23
Purified clinoptilolite	0.3	0.4	0.4	0.7	0.7	0.8	6.0	5.5	5.3	5.0	4.8	4.8
Tannin	0.2	0.2	0.1	0.6	1.1	1.5	177	425	681	164	403	631
Duolite™ C-467	0.5	0.6	0.6	1.8	1.9	1.8	271	487	705	411	843	1262
Resorcinol/formaldehyde	0.2	0.3	0.2	0.4	0.4	0.5	54	114	219	41	86	148
TRW CS-WA	0.2	0.6	0.9	1.4	2.1	2.7	624	737	774	747	868	911
Superlig™ 644	<0.1	<0.1	<0.1	0.7	0.8	1.0	11	27	52	9.0	22	39
Ionsiv™ IE-96	<0.1	<0.1	<0.1	<0.1	<0.1	0.1	6.6	12	24	5.5	10	23

2. Cesium. The sorption of cesium on most of the tested absorbers appears unaffected by the varying levels of organic compounds in the four simulant solutions. Many of the tested absorbers provide at least triple-digit Kd values for sorbing cesium from all four simulant solutions.

NiFC-PAN exhibits time-dependent decreasing Kd values in the two untreated simulant solutions. This pattern is attributed to a dissolution of nickel by organic compounds, based on the observed rapid development of the characteristic color of this metal ion during contact of these absorbers with the untreated simulant solutions. Similarly, KCoFC rapidly imparts the characteristic color of cobalt to the solution, although there is no corresponding decrease in Kd values with increased contact time.

Table 6. Kd Values for Sorbing Cesium from Four Variations of Hanford Tank 101-SY Simulant Solution

Absorber	Unirradiated/ untreated			Irradiated/ untreated			Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h
Ionsiv™ IE-910 (CST)	1822	2591	3072	1909	2933	3359	2359	3410	4242	2219	2946	3748
NiFC-PAN	1442	124	53	311	74	25	2646	738	300	1974	393	243
Superlig™ 644	776	2244	3783	626	2151	3821	785	2379	3993	621	2016	3418
Resorcinol/formaldehyde	718	1720	2783	593	1523	2731	883	2000	3489	727	1882	3272
KCoFC crystals	708	1709	2496	188	71	220	20K	>65K	>86K	17K	25K	27K
Duolite™ CS-100	188	220	212	177	214	213	203	248	235	174	205	208
Ionsiv™ IE-96	68	107	121	64	111	130	74	128	158	72	125	152
Clinoptilolite	59	60	59	64	64	64	76	76	76	73	73	72
Purified clinoptilolite	54	53	54	57	56	56	67	66	66	64	63	61
Tannin	51	56	50	49	57	53	53	59	56	47	53	50
Ionsiv™ TIE-96	39	76	96	40	79	102	48	95	129	47	94	124
TRW CS-WA	11	9.3	8.0	12	11	10	12	12	10	12	11	10

3. Technetium. The sorption of technetium on most of the tested absorbers appears relatively unaffected by the varying levels of organic compounds in the four simulant solutions. The absorbers that sorb technetium best are either anion exchange resins or compounds that incorporate functionalities that are closely

related to those of anion exchange resins. Eight of the tested absorbers provide at least triple-digit K_d values for sorbing technetium from all four simulant solutions.

Table 7. K_d Values for Sorbing Technetium from Four Variations of Hanford Tank 101-SY Simulant Solution

Absorber	Unirradiated/ untreated			Irradiated/ untreated			Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h
N-Butyl-HP	683	1208	1406	705	1342	1331	997	1823	2085	952	1772	2258
Aliquat™ 336	649	1262	1422	742	1459	2204	1073	2011	2140	1002	2140	2575
N-Hexyl-HP	602	1165	1406	693	1185	1392	908	1713	2376	890	2000	2506
Purolite™ A-520-E	517	1007	1301	618	1141	1705	721	1408	1887	673	1573	2148
Reillex™ HPQ	465	648	669	511	696	714	569	899	931	545	863	963
Ionac™ SR-6	320	757	1171	351	827	1415	411	1010	1637	434	1171	1714
N-Octyl-HP	251	530	780	327	691	1022	409	903	1397	432	942	1444
Cyanex™ 923	190	269	256	198	315	320	246	279	262	219	292	288
PNPK-1	20	40	55	19	31	40	18	35	41	18	34	40
PNPK-2	18	43	73	26	72	108	23	64	93	16	42	69
TRU-Spec™	17	16	14	17	17	16	18	17	16	19	18	16

4. Americium. Like the sorption of strontium (the other multivalent cation in our study), the sorption of americium is dramatically decreased by organic compounds in the two untreated simulant solutions. Americium often exhibits higher Kd values for sorption from the unirradiated solutions than from their

irradiated counterparts. This pattern, which contrasts with that observed for strontium (Table 5), indicates that the simpler organic compounds such as oxalate and acetate, formed by radiolysis of EDTA, effectively complex americium in highly alkaline solution.

Table 8. Kd Values for Sorbing Americium from Four Variations of Hanford Tank 101-SY Simulant Solution

Absorber	Unirradiated/ untreated			Irradiated/ untreated			Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h	30 min	2 h	6 h
NiFC-PAN	38	176	399	78	770	1321	277	629	870	111	220	409
KCoFC	28	66	101	21	35	49	361	451	551	188	342	389
SNL/HTO	27	53	97	16	27	42	178	335	325	294	527	>753
Bone char	16	44	99	9.2	19	32	333	432	461	835	>838	>849
Na titanate (8225-127)	11	21	38	7.8	14	23	125	229	275	224	377	421
TRW CS-SA	8.4	13	17	6.3	10	14	117	185	200	139	221	348
Na titanate (8104-170)	7.6	12	20	5.7	9.3	14	225	226	326	240	466	>766
Clinoptilolite	5.1	7.0	8.4	6.1	8.1	9.9	107	258	299	113	313	441
Nusorb™ Ferroc carbon A	2.3	5.7	12	1.9	4.3	9.1	88	212	299	142	332	615
Duolite™ CS-100	2.2	3.7	5.3	2.0	3.7	5.2	95	155	212	103	210	274
Purified clinoptilolite	1.9	2.5	3.0	2.6	3.2	4.2	359	>645	>558	72	171	339
Ionsiv™ IE-910 (CST)	1.0	2.1	3.7	0.8	1.7	3.0	22	31	54	19	29	50
Ionsiv™ TIE-96	1.0	3.3	6.8	1.0	3.5	6.9	70	161	209	90	242	476
Diphonix™	0.9	4.1	8.8	<0.1	1.2	2.4	41	66	118	75	141	259
TRW CS-WA	0.7	1.0	1.4	0.4	0.6	1.0	93	126	177	160	216	278
Amberlite™ IRC-718	0.6	1.0	1.2	0.6	1.4	1.8	28	60	120	39	97	196
Superlig™ 644	0.4	0.1	0.1	<0.1	<0.1	<0.1	43	173	238	6.7	26	60
Ionsiv™ IE-96	0.3	0.5	0.9	0.5	0.6	1.0	164	327	433	22	66	234
Duolite™ C-467	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	35	76	108	56	126	179

B. Individual Absorbers

Tables 9 through 40 present Kd values for sorption of the four measured elements onto each of the 32 tested absorbers from each of the four variations of Hanford Tank 101-SY simulant solution. The presentation of sorption data for four elements in each of these tables often provides information about the selectivity for one of the elements in the presence of the other three. Because we present three different contact times, these tables also provide information about sorption kinetics for each element on specific absorbers.

1. Commercially Available Absorbers. The 16 commercially available absorbers listed in Table 3 were included in this study.

a. Amberlite™ IRC-718 Cation Exchange Resin. Amberlite™ IRC-718, manufactured by Rohm & Haas, Philadelphia, Pennsylvania, is a weak-acid cation exchange resin consisting of a styrene/divinylbenzene copolymer with iminodiacetic acid functionality. We air-dried this resin before testing for reasons detailed in section II.C.

The Kd values for all four elements are low for sorption from the two untreated solutions. Strontium and americium are sorbed well from the two hydrothermal-treated solutions.

Table 9. Distribution of Americium, Cesium, Strontium, and Technetium onto Amberlite™ IRC-718 Cation Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.6 ± 0.1	1.0 ± 0.1	1.2 ± 0.1	0.6 ± 0.2	1.4 ± 0.1	1.8 ± 0.1
Cs	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.1 ± 0.1	0.1 ± 0.1	<0.1 ± 0.1
Sr	1.5 ± 0.2	1.7 ± 0.0	1.7 ± 0.1	6.7 ± 0.0	7.1 ± 0.0	7.1 ± 0.0
Tc	3.0 ± 0.4	3.0 ± 0.3	2.9 ± 0.4	4.0 ± 0.7	3.7 ± 0.2	3.4 ± 0.3

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	28 ± 2.5	60 ± 3.1	120 ± 10	39 ± 1.5	97 ± 12	196 ± 26
Cs	0.1 ± 0.2	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	<0.1 ± 0.0	0.1 ± 0.1
Sr	493 ± 46	1149 ± 186	1609 ± 37	479 ± 12	1067 ± 39	1925 ± 320
Tc	3.0 ± 0.0	3.0 ± 0.2	2.9 ± 0.0	3.5 ± 0.0	3.5 ± 0.1	3.0 ± 0.1

b. Bone Char Absorber. Bone char, produced by calcining cattle bones in the absence of air, was obtained from Stauffer Chemical Company, Westport, Connecticut. This absorber, which is predominantly calcium phosphate, was used as received.

Bone char sorbs strontium and americium well from the two untreated simulants and even better from the two hydrothermal-treated solutions. The ">" signs assigned to relatively low values is a consequence of the greater uncertainty introduced by a 10-fold lower initial concentration of americium in the two hydrothermal-treated solutions.

Table 10. Distribution of Americium, Cesium, Strontium, and Technetium onto Bone Char Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	16 ± 0.2	44 ± 0.5	99 ± 0.7	9.2 ± 0.5	19 ± 0.5	32 ± 0.3
Cs	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	0.1 ± 0.2	0.1 ± 0.2
Sr	27 ± 0.1	50 ± 1.2	80 ± 2.3	56 ± 3.5	127 ± 4.7	225 ± 2.2
Tc	1.7 ± 0.0	2.5 ± 0.1	3.9 ± 0.3	1.7 ± 0.2	2.1 ± 0.2	2.2 ± 0.2
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	333 ± 58	432 ± 23	461 ± 23	835 ± 177	>838 ± 7.1	>849 ± 3.5
Cs	0.2 ± 0.2	<0.1 ± 0.1	<0.1 ± 0.1	<0.1 ± 0.1	0.1 ± 0.1	0.2 ± 0.0
Sr	918 ± 101	3666 ± 143	4604 ± 803	829 ± 39	2310 ± 5.9	6148 ± 2043
Tc	2.4 ± 0.0	2.5 ± 0.1	2.4 ± 0.2	2.9 ± 0.1	3.0 ± 0.1	3.6 ± 0.1

c. Clinoptilolite Absorber. Clinoptilolite, a natural zeolite mineral from Castle Creek, Idaho (batch #27034), was obtained from Minerals Research, Clarkson, New Jersey. This absorber was ball-milled to <200 mesh before use.

Clinoptilolite sorbs cesium at useful levels from all four solutions. From the two hydrothermal-treated solutions, strontium sorbs at useful levels and americium sorbs well.

Table 11. Distribution of Americium, Cesium, Strontium, and Technetium onto Clinoptilolite Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	5.1 ± 0.0	7.0 ± 0.3	8.4 ± 0.4	6.1 ± 0.2	8.1 ± 0.2	9.9 ± 0.2
Cs	59 ± 2.3	60 ± 1.1	59 ± 2.0	64 ± 0.9	64 ± 0.2	64 ± 0.7
Sr	0.3 ± 0.3	0.7 ± 0.1	0.7 ± 0.1	1.4 ± 0.0	1.3 ± 0.1	1.7 ± 0.1
Tc	<0.1 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	107 ± 4.3	258 ± 46	299 ± 33	113 ± 11	313 ± 100	441 ± 97
Cs	76 ± 0.4	76 ± 0.1	76 ± 0.6	73 ± 1.4	73 ± 0.1	72 ± 0.8
Sr	20 ± 0.1	24 ± 0.3	29 ± 3.3	17 ± 0.6	20 ± 0.1	23 ± 0.4
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	0.1 ± 0.2

d. Diphonix™ Cation Exchange Resin. Diphonix™, a polyfunctional cation exchange resin that contains diphosphonic acid, sulfonic acid, and carboxylic acid functional groups, was obtained from Eichrom Industries, Inc., Darien, Illinois. We air-dried this resin before testing for reasons detailed in section II.C.

None of the four elements sorbs at useful levels from the two untreated solutions. From the two hydrothermal-treated solutions, strontium and americium sorb well.

Table 12. Distribution of Americium, Cesium, Strontium, and Technetium onto Diphonix™ Cation Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.9 ± 0.0	4.1 ± 0.3	8.8 ± 0.1	<0.1 ± 0.0	1.2 ± 0.0	2.4 ± 0.1
Cs	0.4 ± 0.2	0.4 ± 0.4	0.4 ± 0.1	0.8 ± 0.0	0.7 ± 0.1	0.6 ± 0.1
Sr	1.8 ± 0.1	2.7 ± 0.3	3.0 ± 0.0	4.1 ± 0.2	6.7 ± 0.1	8.3 ± 0.2
Tc	1.3 ± 0.0	1.4 ± 0.1	1.1 ± 0.0	1.8 ± 0.0	1.7 ± 0.1	1.7 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	41 ± 2.7	66 ± 5.7	118 ± 9.3	75 ± 5.2	141 ± 7.4	259 ± 37
Cs	0.8 ± 0.1	0.9 ± 0.0	1.0 ± 0.1	0.9 ± 0.0	1.0 ± 0.0	1.0 ± 0.1
Sr	506 ± 2.2	925 ± 11	1243 ± 2.5	1090 ± 87	1986 ± 105	3222 ± 419
Tc	0.7 ± 0.0	0.7 ± 0.0	0.7 ± 0.1	2.3 ± 0.2	2.6 ± 0.2	2.1 ± 0.2

e. Duolite™ C-467 Cation Exchange Resin. Duolite™ C-467, a chelating cation exchange resin composed of a polystyrene/divinylbenzene copolymer with aminophosphonic acid functionality, is manufactured by Rohm & Haas, Philadelphia, Pennsylvania. We air-dried this resin before testing for reasons detailed in section II.C.

None of the four elements sorbs at useful levels from the two untreated solutions. From the two hydrothermal-treated solutions, strontium and americium sorb well.

Table 13. Distribution of Americium, Cesium, Strontium, and Technetium onto Duolite™ C-467 Cation Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Cs	0.3 ± 0.1	0.3 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	0.5 ± 0.2	0.4 ± 0.2
Sr	0.5 ± 0.0	0.6 ± 0.1	0.6 ± 0.0	2.0 ± 0.1	2.0 ± 0.3	1.9 ± 0.3
Tc	3.7 ± 0.3	5.1 ± 0.0	6.5 ± 1.0	3.7 ± 0.2	4.4 ± 0.3	5.6 ± 0.1
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	35 ± 2.4	76 ± 13	108 ± 8.6	56 ± 4.3	126 ± 1.2	179 ± 21
Cs	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Sr	271 ± 7.5	487 ± 14	705 ± 64	411 ± 15	843 ± 81	1262 ± 95
Tc	3.1 ± 0.4	5.3 ± 0.5	7.7 ± 1.4	3.6 ± 0.0	5.0 ± 0.1	5.4 ± 0.3

f. Duolite™ CS-100 Cation Exchange Resin. Duolite™ CS-100, a phenol-formaldehyde condensate cation exchange resin, was manufactured by Rohm & Haas, Philadelphia, Pennsylvania. We air-dried this resin before testing for reasons detailed in section II.C.

Cesium sorbs consistently well from all four solutions. Strontium and americium also sorb well from the two hydrothermal-treated solutions.

During all contacts with Duolite™ CS-100 resin, the simulant solutions acquired a dark color whose intensity was proportional to the length of the contact time. The change in color indicates some leaching of this absorber in strong alkaline solution.

Table 14. Distribution of Americium, Cesium, Strontium, and Technetium onto Duolite™ C-100 Cation Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	2.2 ± 0.0	3.7 ± 0.1	5.3 ± 0.2	2.0 ± 0.1	3.7 ± 0.1	5.2 ± 0.0
Cs	188 ± 0.6	220 ± 1.1	212 ± 2.8	177 ± 1.4	214 ± 3.7	213 ± 5.3
Sr	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.1 ± 0.2	0.3 ± 0.0	0.3 ± 0.0
Tc	0.3 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	95 ± 3.9	155 ± 42	212 ± 16	103 ± 1.2	210 ± 31	274 ± 48
Cs	203 ± 2.6	248 ± 1.9	235 ± 3.1	174 ± 2.0	205 ± 7.3	208 ± 6.2
Sr	78 ± 1.0	109 ± 0.5	118 ± 2.3	61 ± 1.9	83 ± 1.9	92 ± 2.0
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.3 ± 0.1	0.3 ± 0.3	0.4 ± 0.1

g. Ionac™ SR-6 Anion Exchange Resin. Ionac™ SR-6, a macroporous anion exchange resin with tributyl amine as the functional group, is manufactured by Sybron Chemicals, Inc., Birmingham, New Jersey. We air-dried this resin before testing for reasons detailed in section II.C.

Technetium sorbs strongly and selectively from all four solutions.

Table 15. Distribution of Americium, Cesium, Strontium, and Technetium onto Ionac™ SR-6 Anion Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	<0.1 ± 0.0	<0.1 ± 0.0	<0. ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Cs	0.2 ± 0.1	0.3 ± 0.1	0.3 ± 0.1	0.2 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0
Sr	0.3 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	0.4 ± 0.0	0.4 ± 0.0	0.4 ± 0.0
Tc	320 ± 19	757 ± 41	1171 ± 5.4	351 ± 11	827 ± 7.1	1415 ± 134

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	4.6 ± 0.6	10 ± 1.2	15 ± 0.2	4.2 ± 0.2	8.5 ± 0.6	15 ± 0.2
Cs	<0.1 ± 0.0	<0.1 ± 0.1	<0.1 ± 0.0	0.3 ± 0.0	0.3 ± 0.1	0.2 ± 0.0
Sr	0.2 ± 0.1	0.3 ± 0.1	0.3 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Tc	411 ± 55	1010 ± 87	1637 ± 47	434 ± 42	1171 ± 64	1714 ± 81

h. Ionsiv™ IE-96 Absorber. Ionsiv™ IE-96 is a zeolite absorber manufactured by UOP Molecular Sieves Division, Mobile, Alabama. This absorber was used as received.

Cesium sorbs consistently well from all four solutions. Americium sorbs well and strontium sorbs at moderate levels from the two hydrothermal-treated solutions.

Table 16. Distribution of Americium, Cesium, Strontium, and Technetium onto Ionsiv™ IE-96 Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.3 ± 0.1	0.5 ± 0.1	0.9 ± 0.3	0.5 ± 0.3	0.6 ± 0.1	1.0 ± 0.2
Cs	68 ± 5.9	107 ± 5.5	121 ± 3.5	64 ± 4.5	111 ± 3.4	130 ± 0.2
Sr	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	<0.1 ± 0.0	0.1 ± 0.1
Tc	1.2 ± 0.6	1.8 ± 0.2	2.3 ± 0.1	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.2
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	164 ± 41	327 ± 33	433 ± 121	22 ± 2.3	66 ± 6.5	234 ± 8.2
Cs	74 ± 1.4	128 ± 3.4	158 ± 2.0	72 ± 1.2	125 ± 0.2	152 ± 1.7
Sr	6.6 ± 0.2	12 ± 0.4	24 ± 7.0	5.5 ± 0.5	10 ± 0.1	23 ± 0.1
Tc	<0.1 ± 0.1	0.2 ± 0.3	<0.1 ± 0.1	1.0 ± 0.1	0.8 ± 0.2	0.7 ± 0.3

i. Ionsiv™ IE-910 (CST) Powder Absorber. Ionsiv™ IE-910 is a crystalline silico-titanate powder manufactured by UOP Molecular Sieves Division, Mobile, Alabama, under license from its developers at Sandia/NM and Texas A&M University. This absorber was used as received.

Cesium is strongly and consistently sorbed from all four solutions. From the two hydrothermal-treated solutions, strontium is strongly sorbed and americium is sorbed at useful levels.

Table 17. Distribution of Americium, Cesium, Strontium, and Technetium onto Ionsiv™ IE-910 (CST) Powder Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	1.0 ± 0.1	2.1 ± 0.1	3.7 ± 0.1	0.8 ± 0.2	1.7 ± 0.3	3.0 ± 0.1
Cs	1822 ± 69	2591 ± 2.9	3072 ± 20	1909 ± 3.3	2933 ± 128	3359 ± 135
Sr	1.5 ± 0.1	2.4 ± 0.1	3.7 ± 0.0	4.4 ± 0.2	7.2 ± 0.3	11 ± 0.2
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	22 ± 0.2	31 ± 7.2	54 ± 3.5	19 ± 1.2	29 ± 3.0	50 ± 11
Cs	2359 ± 39	3410 ± 71	4242 ± 88	2219 ± 51	2946 ± 20	3748 ± 9.1
Sr	944 ± 56	1383 ± 2.9	2159 ± 0.2	1012 ± 37	298 ± 114	2134 ± 299
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.2 ± 0.1	0.2 ± 0.0

j. Ionsiv™ TIE-96 Absorber. Ionsiv™ TIE-96 is a titanium-loaded zeolite manufactured by UOP Molecular Sieves Division, Mobile, Alabama. This absorber was used as received.

Cesium is sorbed consistently well from all four solutions. From the two hydrothermal-treated solutions, strontium and americium also sorb well.

Table 18. Distribution of Americium, Cesium, Strontium, and Technetium onto Ionsiv™ TIE-96 Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	1.0 ± 0.0	3.3 ± 0.1	6.8 ± 0.0	1.0 ± 0.2	3.5 ± 0.1	6.9 ± 0.0
Cs	39 ± 0.5	76 ± 1.1	96 ± 0.1	40 ± 0.8	79 ± 1.5	102 ± 0.8
Sr	2.0 ± 0.2	3.6 ± 0.0	4.8 ± 0.0	4.1 ± 0.1	8.3 ± 0.2	12 ± 0.3
Tc	<0.1 ± 0.0	0.1 ± 0.1	0.1 ± 0.1	<0.1 ± 0.1	<0.1 ± 0.0	0.1 ± 0.1

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	70 ± 1.8	161 ± 31	209 ± 12	90 ± 11	242 ± 18	476 ± 16
Cs	48 ± 3.0	95 ± 3.9	129 ± 4.9	47 ± 6.7	94 ± 8.7	124 ± 6.3
Sr	146 ± 11	419 ± 11	754 ± 82	128 ± 17	393 ± 43	704 ± 83
Tc	0.2 ± 0.0	0.2 ± 0.1	0.2 ± 0.1	0.9 ± 0.0	0.8 ± 0.1	0.9 ± 0.1

k. Nusorb™ Ferrocarbon A Absorber. Nusorb™ Ferrocarbon A is a carbon/iron oxide composite manufactured by Nucon International, Inc., Columbus, Ohio. This absorber was used as received.

All four elements sorb somewhat from the two untreated solutions. From the hydrothermal-treated solutions, strontium and americium are strongly sorbed.

Table 19. Distribution of Americium, Cesium, Strontium, and Technetium onto Nusorb™ Ferrocarbon A Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	2.3 ± 0.3	5.7 ± 0.7	12 ± 1.9	1.9 ± 0.5	4.3 ± 1.1	9.1 ± 1.4
Cs	2.4 ± 0.0	4.8 ± 0.0	4.9 ± 0.0	2.1 ± 0.0	4.4 ± 0.2	4.8 ± 0.0
Sr	1.9 ± 0.2	4.4 ± 0.2	7.3 ± 0.4	4.7 ± 0.6	9.4 ± 1.4	16 ± 1.8
Tc	1.7 ± 0.0	3.1 ± 0.1	5.1 ± 0.5	2.1 ± 0.0	3.2 ± 0.1	5.1 ± 0.1

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	88 ± 30	212 ± 33	299 ± 7.9	142 ± 5.6	332 ± 46	615 ± 77
Cs	2.1 ± 0.1	4.7 ± 0.4	5.4 ± 0.4	1.8 ± 0.1	4.1 ± 0.1	4.9 ± 0.1
Sr	470 ± 164	1033 ± 316	1683 ± 704	404 ± 67	840 ± 146	1420 ± 419
Tc	2.3 ± 0.2	3.6 ± 0.3	6.5 ± 0.3	2.8 ± 0.1	5.9 ± 2.2	9.5 ± 4.5

I. Purolite™ A-520-E Anion Exchange Resin. Purolite™ A-520-E, a macroporous strong-base anion exchange resin with triethyl amine as the functional group, was obtained from the Purolite Company, Bala Cynwyd, Pennsylvania. We air-dried this resin before testing for reasons detailed in section II.C.

Technetium is strongly and selectively sorbed from all four solutions with Kd values increasing somewhat with the decomposition of the initial organic component.

Table 20. Distribution of Americium, Cesium, Strontium, and Technetium onto Purolite™ A-520-E Anion Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Cs	0.4 ± 0.0	0.4 ± 0.0	0.4 ± 0.1	0.2 ± 0.1	0.3 ± 0.0	0.2 ± 0.1
Sr	0.4 ± 0.1	0.4 ± 0.0	0.5 ± 0.1	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.1
Tc	517 ± 53	1007 ± 47	1301 ± 31	618 ± 1.1	1141 ± 122	1705 ± 10
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	1.9 ± 1.0	3.9 ± 0.5	4.8 ± 0.6	1.9 ± 0.1	2.8 ± 1.0	4.2 ± 1.0
Cs	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	0.4 ± 0.1	0.4 ± 0.2	0.4 ± 0.1
Sr	$<0.1 \pm 0.1$	0.1 ± 0.0	0.2 ± 0.0	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Tc	721 ± 53	1408 ± 13	1887 ± 153	673 ± 114	1573 ± 301	2148 ± 394

m. Resorcinol/Formaldehyde Resin (BSC-210). Resorcinol/formaldehyde resin (BSC-210), manufactured by Boulder Scientific Company, Mead, Colorado, was provided to us by Jane Bibler of Westinghouse Savannah River Company, whose directions we followed to convert it from the as-received potassium form to a sodium form. We air-dried this resin before testing for reasons detailed in section II.C.

Cesium is sorbed strongly and selectively from all four solutions. From the two hydrothermal-treated solutions, strontium sorbs well and americium sorbs at useful levels.

During all contacts with the resorcinol/formaldehyde resin, the simulant solutions acquired a dark color whose intensity was proportional to the length of the contact time. The change in color indicates some leaching of this absorber in strong alkaline solution.

Table 21. Distribution of Americium, Cesium, Strontium, and Technetium onto Resorcinol/Formaldehyde Resin (BSC-210) from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Cs	718 ± 13	1720 ± 5.5	2783 ± 16	593 ± 28	1523 ± 4.3	2731 ± 103
Sr	0.2 ± 0.0	0.3 ± 0.1	0.2 ± 0.0	0.4 ± 0.1	0.4 ± 0.1	0.5 ± 0.0
Tc	0.8 ± 0.0	1.3 ± 0.1	2.4 ± 0.2	1.5 ± 0.6	1.6 ± 0.1	3.9 ± 0.7

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	22 ± 1.8	40 ± 2.5	46 ± 1.8	26 ± 2.3	45 ± 9.4	51 ± 1.4
Cs	883 ± 50	2000 ± 43	3489 ± 140	727 ± 28	1882 ± 21	3272 ± 90
Sr	54 ± 2.0	114 ± 3.9	219 ± 2.7	41 ± 1.0	86 ± 0.4	148 ± 5.0
Tc	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.2$	0.4 ± 0.1	0.4 ± 0.0	0.5 ± 0.2

n. Reillex™ HPQ Anion Exchange Resin. Reillex™ HPQ anion exchange resin, a copolymer of 1-methyl-4-vinylpyridine and divinylbenzene, was manufactured by Reilly Industries, Inc., Indianapolis, Indiana. We air-dried this resin before testing for reasons detailed in section II.C.

Technetium is strongly and selectively sorbed from all four solutions.

Table 22. Distribution of Americium, Cesium, Strontium, and Technetium onto Reillex™ HPQ Anion Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Cs	0.3 ± 0.1	0.3 ± 0.0	0.4 ± 0.0	0.2 ± 0.0	0.2 ± 0.1	0.3 ± 0.0
Sr	0.4 ± 0.0	0.3 ± 0.1	0.4 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	0.3 ± 0.1
Tc	465 ± 9.8	648 ± 10	669 ± 16	511 ± 36	696 ± 17	714 ± 101

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	7.5 ± 0.7	16 ± 1.3	27 ± 3.1	8.7 ± 0.6	15 ± 0.1	24 ± 0.8
Cs	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Sr	1.4 ± 0.2	1.3 ± 0.2	1.2 ± 0.2	1.6 ± 0.2	1.4 ± 0.1	1.3 ± 0.1
Tc	569 ± 0.6	899 ± 23	931 ± 56	545 ± 18	863 ± 50	963 ± 24

o. Tannin Absorber. Tannin absorber was obtained from Mitsubishi Nuclear Fuels Corporation, Japan. We air-dried this resin before testing for reasons detailed in section II.C.

Cesium sorbs consistently well from all four solutions. From the two hydrothermal-treated solutions, strontium and americium sorb well.

Table 23. Distribution of Americium, Cesium, Strontium, and Technetium onto Tannin Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Cs	51 ± 2.0	56 ± 0.4	50 ± 0.2	49 ± 2.3	57 ± 0.1	53 ± 0.2
Sr	0.2 ± 0.1	0.2 ± 0.1	0.1 ± 0.1	0.6 ± 0.0	1.1 ± 0.0	1.5 ± 0.1
Tc	3.5 ± 0.3	4.4 ± 0.1	6.7 ± 0.3	3.3 ± 0.1	4.6 ± 0.2	6.6 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	25 ± 2.5	53 ± 3.7	78 ± 1.4	67 ± 1.4	129 ± 24	256 ± 6.8
Cs	53 ± 0.2	59 ± 1.3	56 ± 1.2	47 ± 5.7	53 ± 1.0	50 ± 0.2
Sr	177 ± 6.3	425 ± 6.8	681 ± 21	164 ± 40	403 ± 44	631 ± 14
Tc	1.4 ± 0.1	1.9 ± 0.2	1.3 ± 0.2	1.4 ± 0.2	1.4 ± 0.2	1.0 ± 0.2

p. TRU-Spec™ Extractant Resin. TRU-Spec™ extractant resin, a mixture of 13% CMPO (octylphenyl-N, N-diisobutylcarbamoylmethylphosphine oxide) and 27% TBP (tributyl phosphate) on polyacrylic beads, was obtained from Eichrom Industries, Inc., Darien, Illinois. This absorber was used as received.

Technetium sorbs at a moderate level from all four solutions. Americium also sorbs at low levels from the two hydrothermal-treated solutions.

Table 24. Distribution of Americium, Cesium, Strontium, and Technetium onto TRU-Spec™ Extractant Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Cs	0.2 ± 0.1	0.1 ± 0.1	0.1 ± 0.0	0.3 ± 0.1	0.3 ± 0.0	0.2 ± 0.0
Sr	<0.1 ± 0.0	0.2 ± 0.1	0.2 ± 0.0	0.3 ± 0.1	0.3 ± 0.1	0.2 ± 0.1
Tc	17 ± 0.1	16 ± 0.0	14 ± 0.2	17 ± 0.6	17 ± 0.1	16 ± 0.4

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	7.1 ± 1.2	8.7 ± 0.6	12 ± 0.2	7.3 ± 0.5	10 ± 0.4	13 ± 1.2
Cs	0.3 ± 0.2	0.2 ± 0.1	0.3 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1
Sr	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.3 ± 0.0	0.5 ± 0.1	0.4 ± 0.1
Tc	18 ± 0.7	17 ± 0.4	16 ± 0.4	19 ± 0.1	18 ± 0.4	16 ± 0.1

2. Developmental Absorbers. Seven developmental absorbers were included in our study.

a. KCoFC (Potassium Hexacyanoferrate) Crystals.

These KCoFC (potassium hexacyanoferrate) crystals (150 to 600 μm in diameter) were prepared at Oak Ridge National Laboratory using the procedure described by Prout¹⁸ with a minor modification. This absorber was used as received.

Although cesium, strontium, and americium are sorbed with useful to very high K_d values from all four

solutions, the organic compounds in the two untreated solutions clearly decrease the sorption efficiency for these elements. The rapid appearance of the characteristic red color of cobalt when KCoFC was added to the untreated solutions indicates poor stability of this absorber at high pH and in the presence of certain organic compounds. The lower K_d value for cesium after the 2-h contact in irradiated/untreated solution appears to be valid, based on the agreement between the two measurements for that contact period.

Table 25. Distribution of Americium, Cesium, Strontium, and Technetium onto KCoFC crystals (150 to 600 μm in diameter) from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	28 $\pm$ 0.5	66 $\pm$ 0.2	101 $\pm$ 0.5	21 $\pm$ 0.4	35 $\pm$ 0.2	49 $\pm$ 0.6
Cs	708 $\pm$ 23	1709 $\pm$ 125	2496 $\pm$ 157	188 $\pm$ 11	71 $\pm$ 8.0	220 $\pm$ 50
Sr	45 $\pm$ 1.0	146 $\pm$ 2.4	280 $\pm$ 9.8	67 $\pm$ 0.7	87 $\pm$ 3.2	135 $\pm$ 4.8
Tc	<0.1 $\pm$ 0.0	<0.1 $\pm$ 0.0	<0.1 $\pm$ 0.0	<0.1 $\pm$ 0.0	<0.1 $\pm$ 0.1	<0.1 $\pm$ 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	361 $\pm$ 46	451 $\pm$ 53	551 $\pm$ 94	188 $\pm$ 17	342 $\pm$ 18	389 $\pm$ 64
Cs	21K $\pm$ 1K	>65K $\pm$ 21K	>86K $\pm$ 355	17K $\pm$ 3829	25K $\pm$ 718	27K $\pm$ 2881
Sr	2352 $\pm$ 84	5579 $\pm$ 312	>25K $\pm$ 708	2117 $\pm$ 18	5K $\pm$ 489	6K $\pm$ 683
Tc	0.2 $\pm$ 0.0	<0.1 $\pm$ 0.0	0.2 $\pm$ 0.1	<0.1 $\pm$ 0.0	0.2 $\pm$ 0.2	0.1 $\pm$ 0.1

b. NiFC-PAN (Nickel Hexacyanoferrate) Composite.

This nickel hexacyanoferrate-polyacrylonitrile composite was prepared by Ferdinand Sebesta of the Czech Technical University, Prague, Czech Republic. We air-dried this composite before testing for reasons detailed in section II.C.

Cesium and americium are sorbed well from unirradiated/untreated solution, whereas these two elements and strontium are sorbed well from the two hydrothermal-treated solutions. The higher Kd values for strontium sorption from the irradiated/untreated solution indicates that some of the organic compounds that interfere with the sorption of strontium are destroyed or

decomposed into simpler organic compounds by gamma-irradiation. The generally lower Kd values for americium and strontium sorption from the untreated solutions are attributed to competition from organic complexants.

The decreasing cesium Kd values with increasing contact time in the two untreated solutions coincide with the rapid appearance of the characteristic green color of nickel when NiFC-PAN was added to the these two solutions. The apparent dissolution of NiFC-PAN in these solutions indicates poor stability of this absorber at high pH in the presence of certain organic compounds.

Table 26. Distribution of Americium, Cesium, Strontium, and Technetium onto NiFC-PAN (Nickel Hexacyanoferrate) Composite from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	38 ± 0.7	176 ± 1.5	399 ± 11	78 ± 5.2	770 ± 59	1321 ± 141
Cs	1442 ± 11	124 ± 0.9	53 ± 0.2	311 ± 3.9	74 ± 2.9	25 ± 0.2
Sr	1.8 ± 0.2	5.0 ± 0.1	8.4 ± 0.2	13 ± 1.4	289 ± 8.8	1378 ± 36
Tc	0.2 ± 0.2	0.4 ± 0.1	0.7 ± 0.0	0.3 ± 0.3	0.7 ± 0.1	0.3 ± 0.1

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	277 ± 88	629 ± 130	870 ± 11	111 ± 2.1	220 ± 46	409 ± 32
Cs	2646 ± 348	738 ± 19	300 ± 9.6	1974 ± 335	393 ± 76	243 ± 29
Sr	964 ± 48	6005 ± 21	>22K ± 721	1608 ± 242	3444 ± 1249	6K ± 1145
Tc	0.4 ± 0.0	0.8 ± 0.0	1.0 ± 0.1	1.1 ± 0.3	1.4 ± 0.1	1.3 ± 0.0

c. Sodium Nanotitanate (8104-170) Absorber. A sodium nanotitanate (8104-170), <100 mesh, absorber of proprietary composition was provided by Allied Signal, Inc., Research and Technology, Des Plaines, Illinois. This absorber was used as received.

Strontium is sorbed well and americium is sorbed somewhat from the two untreated solutions. Strontium is sorbed very strongly and americium is sorbed well from the two hydrothermal-treated solutions.

Table 27. Distribution of Americium, Cesium, Strontium, and Technetium onto Sodium Nanotitanate (8104-170) Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	7.6 ± 0.1	12 ± 0.7	20 ± 0.7	5.7 ± 0.3	9.3 ± 0.1	14 ± 0.3
Cs	0.6 ± 0.0	0.6 ± 0.1	0.7 ± 0.1	0.4 ± 0.0	0.3 ± 0.1	0.5 ± 0.0
Sr	43 ± 0.2	68 ± 1.4	100 ± 1.0	91 ± 4.2	150 ± 3.1	217 ± 0.4
Tc	0.2 ± 0.2	<0.1 ± 0.1	<0.1 ± 0.1	0.1 ± 0.1	<0.1 ± 0.0	0.1 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	225 ± 23	226 ± 5.2	326 ± 22	240 ± 69	466 ± 39	>766 ± 12
Cs	0.5 ± 0.1	0.7 ± 0.4	0.6 ± 0.1	0.4 ± 0.0	0.5 ± 0.0	0.4 ± 0.1
Sr	16K ± 141	16K ± 146	16K ± 139	>18K ± 109	>18K ± 129	>18K ± 185
Tc	<0.1 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	0.5 ± 0.2	0.3 ± 0.1

d. Sodium Nanotitanate (8225-127) Absorber. A sodium nanotitanate (8225-127), <40 mesh, absorber of proprietary composition was provided by Allied Signal, Inc., Research and Technology, Des Plaines, Illinois. This absorber was used as received.

Strontium is sorbed well and americium is sorbed somewhat from the two untreated solutions. Strontium is sorbed very strongly and americium is sorbed well from both hydrothermal-treated solutions. Of the 32 absorbers tested, this absorber provides the second-highest sorption of strontium from untreated organic-containing simulants.

Table 28. Distribution of Americium, Cesium, Strontium, and Technetium onto Sodium Nanotitanate (8225-127) Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	11 ± 0.2	21 ± 0.1	38 ± 0.2	7.8 ± 0.1	14 ± 0.1	23 ± 0.4
Cs	1.1 ± 0.1	1.1 ± 0.0	1.1 ± 0.1	0.7 ± 0.1	0.9 ± 0.1	0.8 ± 0.0
Sr	107 ± 2.1	197 ± 2.7	248 ± 9.1	212 ± 0.2	398 ± 5.1	554 ± 14
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	<0.1 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	125 ± 1.8	229 ± 30	275 ± 4.5	224 ± 50	377 ± 20	421 ± 129
Cs	1.2 ± 0.1	1.0 ± 0.0	1.1 ± 0.1	1.0 ± 0.0	0.9 ± 0.1	0.9 ± 0.0
Sr	16K ± 368	16K ± 35	16K ± 211	>18K ± 121	>18K ± 130	>18K ± 74
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.1 ± 0.1	0.4 ± 0.1	0.3 ± 0.1

e. SuperLig™ 644 Particles. SuperLig™ 644, a proprietary macrocycle absorber, was prepared by IBC Advanced Technologies, Inc., American Fork, Utah, and was provided to us by 3M Industrial and Consumer Sector, New Products Department, St. Paul, Minnesota.

Cesium is consistently sorbed very well from all four solutions. Americium and strontium are also sorbed at useful levels from the two hydrothermal-treated solutions. The pattern of highest sorption of americium from the unirradiated/hydrothermal-treated solution corresponds to the lowest level of total organic carbon in this solution (Table 1).

Table 29. Distribution of Americium, Cesium, Strontium, and Technetium onto Superlig™ 644 Particles from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.4 ± 0.6	0.1 ± 0.1	0.1 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Cs	776 ± 41	2244 ± 124	3783 ± 361	624 ± 46	2151 ± 156	3821 ± 13
Sr	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.7 ± 0.2	0.8 ± 0.2	1.0 ± 0.1
Tc	0.8 ± 0.4	0.9 ± 0.0	1.4 ± 0.1	1.1 ± 0.2	1.1 ± 0.1	1.8 ± 0.1
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	43 ± 16	173 ± 3.1	238 ± 25	6.7 ± 1.4	26 ± 1.4	60 ± 4.0
Cs	785 ± 70	2379 ± 263	3993 ± 271	621 ± 86	2016 ± 120	3418 ± 87
Sr	11 ± 0.5	27 ± 1.1	52 ± 0.0	9.0 ± 0.7	22 ± 0.1	39 ± 1.2
Tc	<0.1 ± 0.1	<0.1 ± 0.1	<0.1 ± 0.0	0.5 ± 0.2	0.6 ± 0.0	0.5 ± 0.1

f. TRW CS-WA Treated Coal. TRW CS-WA treated coal is manufactured by TRW Inc., Redondo Beach, California, from various starting coals using the company's patented Molten Caustic Leaching process, which converts the coal to a high-surface-area carbonaceous material with weak-acid ion exchange groups on the surface. The resulting sorbent combines the characteristics of activated carbon and ion exchange resins.

Cesium is sorbed somewhat from all four solutions. Strontium and americium are sorbed well from the two hydrothermal-treated solutions.

During all contacts with TRW CS-WA absorber, the simulant solutions acquired a dark color whose intensity was proportional to the length of the contact time. The change in color indicates some leaching of this absorber in strong alkaline solution.

Table 30. Distribution of Americium, Cesium, Strontium, and Technetium onto TRW CS-WA Treated Coal from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.7 ± 0.2	1.0 ± 0.2	1.4 ± 0.1	0.4 ± 0.2	0.6 ± 0.1	1.0 ± 0.1
Cs	11 ± 0.0	9.3 ± 0.3	8.0 ± 0.1	12 ± 0.4	11 ± 0.1	10 ± 0.0
Sr	0.2 ± 0.2	0.6 ± 0.1	0.9 ± 0.1	1.4 ± 0.0	2.1 ± 0.1	2.7 ± 0.2
Tc	0.4 ± 0.0	0.4 ± 0.2	0.6 ± 0.1	0.9 ± 0.1	0.6 ± 0.1	0.6 ± 0.1

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	93 ± 4.2	126 ± 5.6	177 ± 29	160 ± 4.7	216 ± 6.3	278 ± 39
Cs	12 ± 0.1	12 ± 0.1	10 ± 0.2	12 ± 0.0	11 ± 0.9	10 ± 0.3
Sr	624 ± 20	737 ± 59	774 ± 19	747 ± 16	868 ± 16	911 ± 39
Tc	0.5 ± 0.2	0.8 ± 0.1	1.0 ± 0.0	0.9 ± 0.1	0.6 ± 0.5	1.1 ± 0.3

g. TRW CS-SA Treated Coal. TRW CS-SA treated coal is manufactured by TRW Inc., Redondo Beach, California, from various starting coals using the company's patented Molten Caustic Leaching process, which converts the coal to a high-surface-area carbonaceous material with weak-acid ion exchange groups on the surface. Subsequent treatment adds strong-acid functionality to the surface of the coal to produce a sorbent that combines the characteristics of activated carbon and ion exchange resins.

When compared with the Kd values obtained with TRW CS-WA, the Kd values for cesium sorption from the two hydrothermal-treated solutions are similar, but they are lower for sorption from the two untreated solutions. The Kd values for sorption of strontium and americium on the two TRW absorbers are similar from the hydrothermal-treated solutions but are higher on TRW CS-SA from the untreated solutions.

During all contacts with TRW CS-WA absorber, the simulant solutions acquired a dark color whose intensity was proportional to the length of the contact time. The change in color indicates some leaching of this absorber in strong alkaline solution.

Table 31. Distribution of Americium, Cesium, Strontium, and Technetium onto TRW CS-SA Treated Coal from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	8.4 ± 0.5	13 ± 0.6	17 ± 0.1	6.3 ± 0.2	10 ± 0.3	14 ± 0.3
Cs	5.5 ± 0.3	4.8 ± 0.3	4.5 ± 0.2	5.2 ± 0.1	5.1 ± 0.1	4.8 ± 0.1
Sr	3.7 ± 0.2	3.5 ± 0.4	3.3 ± 0.1	5.1 ± 0.1	5.7 ± 0.0	5.6 ± 0.1
Tc	0.6 ± 0.3	0.6 ± 0.3	0.7 ± 0.3	0.6 ± 0.1	0.6 ± 0.1	0.7 ± 0.3

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	117 ± 3.0	185 ± 0.1	200 ± 24	139 ± 0.2	221 ± 1.0	348 ± 21
Cs	11 ± 0.0	11 ± 0.2	9.2 ± 0.1	11 ± 0.5	11 ± 0.8	9.2 ± 0.5
Sr	674 ± 3.8	872 ± 54	849 ± 72	822 ± 83	964 ± 35	980 ± 134
Tc	0.6 ± 0.2	1.0 ± 0.0	1.0 ± 0.1	1.2 ± 0.1	1.7 ± 0.1	1.5 ± 0.0

3. Experimental Absorbers. Nine experimental absorbers were included in our study.

a. Aliquat™ 336 Extractant Beads. We combined one volume of Aliquat™ 336 (methyltricaprylammonium chloride), obtained from Aldrich Chemical Co., Milwaukee, Wisconsin, with two volumes of hexane and added Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas, Philadelphia, Pennsylvania, to absorb the liquid. After evaporation of the volatile hexane, the dry-appearing loaded beads contained 0.191 g Aliquat™ 336 per g.

These extractant beads sorb technetium strongly and selectively from the two untreated simulants. In addition to sorbing technetium strongly, Aliquat™ 336 also sorbs americium somewhat from the two hydrothermal-treated simulants.

Table 32. Distribution of Americium, Cesium, Strontium, and Technetium onto Aliquat™ 336 Extractant Beads from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	0.5 ± 0.0	0.4 ± 0.1	0.3 ± 0.0
Cs	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	0.3 ± 0.0	0.3 ± 0.1	0.2 ± 0.1
Sr	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.1	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Tc	649 ± 94	1262 ± 133	1422 ± 109	742 ± 92	1459 ± 242	2204 ± 212

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	8.8 ± 1.0	15 ± 0.6	24 ± 0.3	11 ± 1.0	17 ± 1.1	26 ± 3.7
Cs	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	0.1 ± 0.0
Sr	$<0.1 \pm 0.0$	0.3 ± 0.1	0.4 ± 0.1	0.5 ± 0.2	0.7 ± 0.1	0.9 ± 0.0
Tc	1073 ± 21	2011 ± 117	2140 ± 4.0	1002 ± 201	2140 ± 192	2575 ± 215

b. Clinoptilolite (Purified) Absorber. A portion of the <200-mesh clinoptilolite (see section B.1.c, Table 11) was tested as received. A separate portion of clinoptilolite was processed by prolonged ultrasonic washing in distilled water, after which the rapid-settling fraction that contained much of the quartz, feldspar, and clay was discarded. The suspended clinoptilolite fine powder was decanted, allowed to settle, and dried at 110°C to constant weight before use.

Like the unpurified clinoptilolite, purified clinoptilolite sorbs cesium at useful levels from all four solutions. From the two hydrothermal-treated solutions, strontium sorbs at useful levels and americium sorbs well. The highest K_d values for sorbing americium from the unirradiated/treated simulant coincide with the lowest TOC level (see Table 1). The fact that most K_d values for sorption on purified clinoptilolite are lower than those obtained with as-received clinoptilolite (Table 11) indicates that some aspect of the purification process adversely affects the performance of the purified clinoptilolite.

Table 33. Distribution of Americium, Cesium, Strontium, and Technetium onto Purified Clinoptilolite Absorber from Four Variations of Hanford Tank 101-SY Simulant Solution

K _d Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	1.9 ± 0.2	2.5 ± 0.1	3.0 ± 0.3	2.6 ± 0.4	3.2 ± 0.3	4.2 ± 0.2
Cs	54 ± 1.6	53 ± 0.8	54 ± 2.0	57 ± 1.2	56 ± 1.1	56 ± 0.3
Sr	0.3 ± 0.0	0.4 ± 0.2	0.4 ± 0.0	0.7 ± 0.2	0.7 ± 0.2	0.8 ± 0.1
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.7 ± 1.0	0.2 ± 0.3

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	359 ± 36	>645 ± 13	>558 ± 113	72 ± 3.9	171 ± 25	339 ± 23
Cs	67 ± 0.6	66 ± 0.5	66 ± 0.6	64 ± 0.6	63 ± 0.6	61 ± 0.4
Sr	6.0 ± 0.2	5.5 ± 0.1	5.3 ± 0.2	5.0 ± 0.4	4.8 ± 0.2	4.8 ± 0.2
Tc	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	<0.1 ± 0.0

c. Cyanex™ 923 Extractant Beads. We combined one volume of Cyanex™ 923 (trialkylphosphine oxide), obtained from American Cyanamid Company, Wayne, New Jersey, with two volumes of cyclohexane and added Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas, Philadelphia, Pennsylvania, to absorb the liquid. After evaporation of the volatile cyclohexane, the dry-appearing loaded beads contained 0.27 mL Cyanex™ 923 per g.

Technetium sorbs well and selectively from the two untreated solutions and well from the two hydrothermal-treated solutions. Americium also sorbs slightly from the hydrothermal-treated solutions.

Table 34. Distribution of Americium, Cesium, Strontium, and Technetium onto Cyanex™ 923 Extractant Beads from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	0.3 ± 0.1	0.2 ± 0.0	0.2 ± 0.0
Cs	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	0.2 ± 0.1	0.2 ± 0.0	0.2 ± 0.0
Sr	0.4 ± 0.2	0.4 ± 0.1	0.4 ± 0.1	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Tc	190 ± 16	269 ± 12	256 ± 8.3	198 ± 1.8	315 ± 3.4	320 ± 1.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	2.1 ± 0.3	3.8 ± 0.5	6.7 ± 1.2	3.8 ± 0.0	6.0 ± 0.2	8.7 ± 0.8
Cs	0.2 ± 0.1	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	0.2 ± 0.1
Sr	$<0.1 \pm 0.0$	0.1 ± 0.0	0.4 ± 0.1	0.3 ± 0.1	0.6 ± 0.1	0.9 ± 0.1
Tc	246 ± 9.3	279 ± 6.9	262 ± 5.9	219 ± 20	292 ± 4.3	288 ± 11

d. PNPk-1 Extractant Beads. PNPk-1, a proprietary phosphinimine extractant,¹⁹ was prepared and provided by Kattesh V. Katti and coworkers at the University of Missouri, Columbia, Missouri. We dissolved PNPk-1 in chloroform and sorbed the mixture on Amborsorb™ 563 porous carbon beads, obtained from Rohm & Haas, Philadelphia, Pennsylvania. After evaporation of the volatile chloroform, the dry-appearing loaded beads contained 0.208 g PNPk-1 per gram.

Technetium is moderately sorbed from all four solutions. Americium also is moderately sorbed from the two hydrothermal-treated solutions.

Table 35. Distribution of Americium, Cesium, Strontium, and Technetium onto PNPk-1 Extractant Beads from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.2 ± 0.1	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	0.3 ± 0.3	0.1 ± 0.2	0.2 ± 0.2
Cs	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	0.2 ± 0.1	$<0.1 \pm 0.1$	0.1 ± 0.2	0.2 ± 0.2
Sr	$<0.1 \pm 0.0$	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.1	0.2 ± 0.2	0.2 ± 0.2
Tc	20 ± 0.9	40 ± 1.6	55 ± 3.1	19 ± 2.1	31 ± 3.9	40 ± 6.8
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	10 ± 0.6	22 ± 2.1	38 ± 4.2	11 ± 0.7	20 ± 1.3	40 ± 3.6
Cs	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	0.2 ± 0.1	0.2 ± 0.1
Sr	0.7 ± 0.0	1.0 ± 0.2	0.9 ± 0.2	0.4 ± 0.2	0.8 ± 0.0	0.8 ± 0.1
Tc	18 ± 0.7	35 ± 0.1	41 ± 0.8	18 ± 0.1	34 ± 0.7	40 ± 4.3

e. PNPk-2 Extractant Beads. PNPk-2, a proprietary phosphinimine extractant,¹⁹ was prepared and provided by Kattesh V. Katti and coworkers at the University of Missouri, Columbia, Missouri. We dissolved PNPk-2 in chloroform and sorbed the mixture on AmbersorbTM 563 porous carbon beads, obtained from Rohm & Haas, Philadelphia, Pennsylvania. After evaporation of the volatile chloroform, the dry-appearing loaded beads contained 0.205 g PNPk-2 per gram.

Technetium is moderately sorbed from all four solutions. Americium also is moderately sorbed from the two hydrothermal-treated solutions.

Table 36. Distribution of Americium, Cesium, Strontium, and Technetium onto PNPk-2 Extractant Beads from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	0.3 ± 0.0	0.2 ± 0.0	0.2 ± 0.0
Cs	0.1 ± 0.1	$<0.1 \pm 0.1$	0.1 ± 0.0	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$
Sr	0.2 ± 0.0	0.4 ± 0.0	0.3 ± 0.0	$<0.1 \pm 0.1$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$
Tc	18 ± 1.4	43 ± 6.2	73 ± 9.9	26 ± 6.1	72 ± 15	108 ± 13

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	6.6 ± 1.5	13 ± 3.5	26 ± 4.8	6.9 ± 0.4	12 ± 1.3	21 ± 2.7
Cs	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	0.2 ± 0.0	0.3 ± 0.0
Sr	0.7 ± 0.1	1.1 ± 0.1	1.2 ± 0.1	0.6 ± 0.0	0.9 ± 0.0	1.1 ± 0.0
Tc	23 ± 3.9	64 ± 11	93 ± 11	16 ± 0.1	42 ± 1.0	69 ± 2.5

f. N-Butyl-HP Anion Exchange Resin. N-Butyl-HP is the 1-butyl derivative of Reillex™ HP (poly-4-vinylpyridine/divinylbenzene) prepared by Donald McQuigg of Reilly Industries, Indianapolis, Indiana. On the basis of measured chloride exchange capacities, the product was quaternized to 43% of the theoretical maximum.

Technetium is strongly sorbed from all four solutions, with americium moderately cosorbed from the two hydrothermal-treated solutions.

Table 37. Distribution of Americium, Cesium, Strontium, and Technetium onto N-Butyl-HP Anion Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Cs	0.3 ± 0.0	0.3 ± 0.1	0.3 ± 0.1	0.4 ± 0.1	0.3 ± 0.0	0.3 ± 0.1
Sr	0.3 ± 0.0	0.4 ± 0.0	0.4 ± 0.0	0.3 ± 0.1	0.3 ± 0.0	0.3 ± 0.1
Tc	683 ± 28	1208 ± 20	1406 ± 34	705 ± 45	1342 ± 277	1331 ± 228
Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	8.1 ± 0.5	14 ± 0.7	19 ± 0.2	7.6 ± 0.3	13 ± 0.3	19 ± 0.9
Cs	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	$<0.1 \pm 0.1$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$	$<0.1 \pm 0.0$
Sr	0.9 ± 0.0	0.7 ± 0.0	0.7 ± 0.1	1.1 ± 0.3	1.0 ± 0.1	0.9 ± 0.1
Tc	997 ± 15	1823 ± 16	2085 ± 37	952 ± 33	1772 ± 6.2	2258 ± 88

g. N-Hexyl-HP Anion Exchange Resin. N-Hexyl-HP is the 1-hexyl derivative of Reillex™ HP (poly-4-vinylpyridine/divinylbenzene) prepared by Donald McQuigg of Reilly Industries, Indianapolis, Indiana. On the basis of measured chloride exchange capacities, the product was quaternized to 45% of the theoretical maximum.

Technetium is strongly sorbed from all four solutions, with americium slightly cosorbed from the two hydrothermal-treated solutions. The selectivity of this resin for technetium is better than that offered by the N-Butyl-HP resin.

Table 38. Distribution of Americium, Cesium, Strontium, and Technetium onto N-Hexyl-HP Anion Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Cs	0.2 ± 0.2	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.2	0.1 ± 0.0	0.2 ± 0.0
Sr	0.3 ± 0.1	0.4 ± 0.0	0.5 ± 0.1	0.3 ± 0.1	0.2 ± 0.1	0.4 ± 0.1
Tc	602 ± 3.7	1165 ± 8.4	1406 ± 39	693 ± 145	1185 ± 55	1392 ± 173

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	3.4 ± 0.4	3.9 ± 1.0	5.2 ± 0.7	3.1 ± 0.1	4.1 ± 0.6	5.1 ± 0.4
Cs	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Sr	0.3 ± 0.1	0.2 ± 0.0	0.2 ± 0.0	0.6 ± 0.1	0.3 ± 0.1	0.5 ± 0.0
Tc	908 ± 92	1713 ± 3.7	2376 ± 38	890 ± 0.3	2000 ± 480	2506 ± 165

h. N-Octyl-HP Anion Exchange Resin. N-Octyl-HP is the 1-octyl derivative of Reillex™ HP (poly-4-vinylpyridine/divinylbenzene) prepared by Donald McQuigg of Reilly Industries, Indianapolis, Indiana. On the basis of measured chloride exchange capacities, the product was quaternized to 59% of the theoretical maximum.

Technetium is strongly and selectively sorbed from all four solutions. Americium sorption from the hydrothermal-treated solutions is essentially absent, which makes the selectivity of this resin for technetium better than that offered by either the N-Butyl-HP or N-Hexyl-HP resins.

Table 39. Distribution of Americium, Cesium, Strontium, and Technetium onto N-Octyl-HP Anion Exchange Resin from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	<0.1 ± 0.1	<0.1 ± 0.0
Cs	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Sr	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.0	0.2 ± 0.2	0.2 ± 0.0	0.3 ± 0.0
Tc	251 ± 31	530 ± 68	780 ± 123	327 ± 20	691 ± 12	1022 ± 163

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	0.8 ± 0.5	1.5 ± 0.0	2.4 ± 0.2	<0.1 ± 0.0	<0.1 ± 0.0	0.2 ± 0.0
Cs	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	0.6 ± 0.1	0.5 ± 0.1	0.4 ± 0.1
Sr	0.1 ± 0.2	0.3 ± 0.0	0.4 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0
Tc	409 ± 23	903 ± 51	1397 ± 17	432 ± 16	942 ± 20	1444 ± 67

i. *SNL/HTO*. SNL/HTO is an amorphous hydrous titanium dioxide powder prepared by Sandia/NM. This absorber was used as received.

Strontium is sorbed with at least triple-digit Kd values and americium is sorbed moderately to well from all four solutions. Of the 32 absorbers tested, this absorber provides the highest sorption of strontium from untreated organic-containing simulants.

Table 40. Distribution of Americium, Cesium, Strontium, and Technetium onto SNL/HTO from Four Variations of Hanford Tank 101-SY Simulant Solution

Kd Value for Specified Time						
Element	Unirradiated/ untreated			Irradiated/ untreated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	27 ± 0.3	53 ± 0.0	97 ± 1.1	16 ± 0.4	27 ± 0.6	42 ± 0.6
Cs	1.1 ± 0.1	0.9 ± 0.2	0.7 ± 0.3	1.2 ± 0.4	1.0 ± 0.2	0.9 ± 0.1
Sr	340 ± 1.0	530 ± 17	653 ± 11	519 ± 19	880 ± 60	1312 ± 43
Tc	<0.1 ± 0.0	<0.1 ± 0.0	0.1 ± 0.2	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.0

Element	Unirradiated/ hydrothermal-treated			Irradiated/ hydrothermal-treated		
	30 min	2 h	6 h	30 min	2 h	6 h
Am	178 ± 26	335 ± 5.8	325 ± 18	294 ± 14	527 ± 162	>753 ± 0.0
Cs	1.1 ± 0.1	1.1 ± 0.2	0.8 ± 0.1	0.9 ± 0.0	0.8 ± 0.1	0.7 ± 0.1
Sr	>16K ± 270	>16K ± 0.0	>16K ± 91	>19K ± 207	>19K ± 0.0	>19K ± 70
Tc	<0.1 ± 0.1	<0.1 ± 0.0	<0.1 ± 0.0	<0.1 ± 0.1	0.2 ± 0.3	0.2 ± 0.1

IV. CONCLUSIONS

Our study of the effectiveness of 32 absorbers for removing selected elements from a series of organic-containing simulants for Hanford Tank 101-SY has met the initial objective of measuring the effects of degraded organic compounds on the sorption of strontium, cesium, technetium, and americium.

Cesium and technetium sorptions appear unaffected by the organic components in the four tested simulant solutions. However, the presence of EDTA and its degradation products in the simulant solutions seriously interferes with the sorption of strontium and americium on absorbers that were previously shown to sorb these elements from organic-free simulant solutions. Such organic compounds would also be expected to interfere with the sorption of other targeted multivalent cations, such as plutonium.

Nevertheless, four of the tested absorbers offer at least triple-digit K_d values for removing strontium from all four simulant solutions. Thus, specific existing absorbers are capable of removing strontium from organic-containing waste with no prior organic-destruction treatment.

Although americium is not as strongly sorbed on the 32 tested absorbers as strontium, we identified several absorbers that seem capable of removing americium from organic-containing waste before organic-destruction treatment. Some of the absorbers provide higher K_d values for sorbing americium from the unirradiated/treated solution than from the irradiated/treated simulant solution. This pattern, which is the reverse of that observed for strontium, suggests that some of the simpler organic compounds formed by radiolysis of EDTA form stable americium complexes in highly alkaline solution.

The Los Alamos hydrothermal treatment process⁶ destroys much of the organic carbon and converts the remainder to simpler organic compounds that interfere much less with the sorption of multivalent cations. The measured K_d values for strontium and americium are generally at least an order of magnitude higher and are often two orders of magnitude higher for sorption from simulant solutions that have been treated with the hydrothermal organic-destruction process.

On the basis of solution coloration, KCoFC and NiFC-PAN absorbers appear to rapidly degrade at high pH in the presence of some organic compounds. Duolite™ CS-100 and the resorcinol/formaldehyde (BSC-210) resin both darken the simulant solutions with intensities that appear to parallel the contact times. These observations indicate that the effect of strong alkaline solutions and soluble organic compounds on absorber stability should be evaluated in future absorber-stability studies.

Measurements of important radionuclides from more-realistic simulants, such as a generic Hanford complexant concentrate simulant that contains a mixture of organic compounds, are needed. Similar tests with actual organic-containing Hanford waste solutions should be performed as soon as such waste samples can be sent to us.

V. FUTURE STUDIES

This investigation demonstrates that the type and concentration of degraded organic compounds in a simulant affect the sorption of strontium and americium onto absorbers that perform well in the absence of such organics. These organic compounds would be expected to interfere in the sorption of other targeted multivalent cations, such as plutonium. Therefore, in the next phase of our absorber study, we plan to measure the distribution of strontium, cesium, technetium, americium, and plutonium (if plutonium is sufficiently soluble) from comparable variations of a generic organic-containing Hanford complexant concentrate simulant onto the best identified absorbers. After that, we plan to test the best-performing absorbers with actual waste from Hanford Complexant Concentrate Tanks 101-SY and 103-SY.

ACKNOWLEDGMENTS

This study was supported by the U. S. Department of Energy through the Tank Waste Remediation Systems (TWRS) of DOE/Richland Operations Office and the TWRS Technology Development Program Office (TDPO) of Westinghouse Hanford Company.

Pamela Rogers of the Environmental Systems and Waste Characterization Group at Los Alamos performed the hydrothermal processing of the unirradiated and gamma-irradiated simulant solutions.

Larry Bruckner of the Statistics Group at Los Alamos provided guidance for pooling individual standard deviations from many duplicate measurements.

Donald McQuigg and Eric Scriven of Reilly Industries prepared and provided experimental polyvinylpyridine resins for this study.

Susan Radzinski of the Nuclear and Radiochemistry Group at Los Alamos prepared the PNPk-1 and PNPk-2 extractant beads.

Charles Cotter of the Environmental Systems and Waste Characterization Group at Los Alamos prepared the purified clinoptilolite and provided both clinoptilolite absorbers.

Clifford Mills of the Nuclear Materials Process Technology Group at Los Alamos designed the tip-filter system that allowed disposable hypodermic syringes to be successfully used as container-dispensers during thousands of dynamic contact experiments.

Dennis Fennelly of UOP Molecular Sieves Division provided samples of Ionsiv™ TIE-96 for evaluation.

Ferdinand Sebesta, Department of Nuclear Chemistry, Czech Technical University, Prague, Czech Republic, prepared and provided the NiFC-PAN composite absorber for our evaluation.

Norman Brown of Sandia National Laboratories/New Mexico supplied samples of the Ionsiv™ IE-910 crystalline silico-titanate and SNL/HTO amorphous hydrous titanium dioxide absorbers used in this study, provided valuable advice and suggestions during numerous technical discussions, and also peer-reviewed this manuscript.

We especially appreciate the assistance and cooperation of many members of the Nuclear and Radiochemistry Group at Los Alamos.

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