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Title: DISTRIBUTIONS OF 14 ELEMENTS INTO 10 LIQUID EXTRACTANTS FROM SIMULATED ACID-DISSOLVED SLUDGE AND ACIDIFIED SUPERNATE SOLUTIONS OF HANFORD HIGH-LEVEL WASTE

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DISTRIBUTIONS OF 14 ELEMENTS INTO 10 LIQUID EXTRACTANTS FROM SIMULATED ACID-DISSOLVED SLUDGE AND ACIDIFIED SUPERNATE SOLUTIONS OF HANFORD HIGH-LEVEL WASTE

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ABSTRACT

The distributions of 14 elements into ten extractants were measured from simulant solutions that represent acidic dissolved sludge and acidified supernate from Hanford HLW Tank 102-SY. The extractants: LIX™-26, LIX™-54, LIX™-84, LIX™-1010, Cyanex™ 272, Cyanex™ 923, Aliquat™ 336, DHDECMP, DHDECMP-DIPB, and CMPO-DIPB, were sorbed on porous carbon beads to provide dry-appearing beads that would be suitable for column operations. The selected elements, which represent fission products: Ce, Cs, Sr, Tc, and Y; actinides: U, Pu, and Am; and matrix elements: Cr, Co, Fe, Mn, Zn, and Zr; were traced by radionuclides and measured by gamma spectrometry. Distribution coefficients for each of 280 element/absorber/solution combinations were measured for dynamic contact periods of 30 minutes, 2 hours, and 6 hours to provide sorption kinetics information for the selected elements from these complex media. The resulting 840 measured distribution coefficients are presented.

INTRODUCTION

The Hanford Reservation in Washington State incorporates 177 underground storage tanks that contain over 165 million Curies of radioactivity in more than 65 million gallons of waste. Many different processes were used at Hanford during the half-century these wastes accumulated from nuclear materials processing for national defense requirements. Consequently, many different reagents and waste streams were neutralized and combined in underground tanks, often with insufficient concern about compatibility of the various components being stored. The resulting wastes are a complex and sometimes unstable mixture of sludges, saltcakes, slurries, and supernates. Adding significantly to the stored waste problem is the fact that 67 tanks are known, or are suspected, to have leaked.

The US Department of Energy is committed to the environmental remediation of hazardous wastes stored at the Hanford Site. Los Alamos National Laboratory has been directed by DOE to support the Hanford Tank Waste Remediation System (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 the TWRS mission is the identification of suitable partitioning agents and technologies to accomplish this challenging goal.

As part of a larger study for the Tank Waste Remediation System (TWRS) at the Los Alamos National Laboratory,[1] we measured the distribution coefficients of 14 elements representing fission products (Ce, Cs, Sr, Tc, and Y); matrix elements (Cr, Co, Fe, Mn, Zn, and Zr); and actinides (U, Pu, and Am) into ten selected extractants. Each extractant was sorbed on porous carbon support beads to facilitate eventual use as solid column-packing material. Each element/ extractant combination was tested from two simulant solutions representing acid-dissolved sludge and acidified supernate for Hanford HLW Tank 102-SY. Each element/extractant/solution combination was measured for dynamic contact times of 30 min, 2 h, and 6 h to provide information about sorption kinetics.

EXPERIMENTAL PARAMETERS

Radiotracers

Because gamma spectrometry was selected as the technique for measuring the distribution of the selected elements, suitable radiotracers were obtained and added to the simulant solutions. To minimize the possibility of interference among gamma rays with similar energies, we divided these radiotracers into two easily resolved groups. Each group of tracers was added to a separate portion of each simulant solution, after which each of the four resulting solutions was evaluated with each of the six absorbers.

Solutions

Detailed compositions of the two simulant solutions used in this study are published elsewhere.[1] Both solutions were essentially saturated with the salts typical of these types of waste solutions. The acid-dissolved sludge solution was approximately 0.6 M nitric acid and 6 M total nitrate, whereas the acidified supernate solution was approximately 1.9 M total nitrate at a pH of about 4.

Solution/Extractant Contacts

In all cases, a 250-mg portion of each sorbed extractant was contacted with a measured 6- or 8-ml volume of each solution in a specially modified 20-ml disposable hypodermic syringe. We modified these syringes by inserting cylindrical plugs cut from 1/4"-thick porous Kynar (obtained from Porex Technologies, Fairburn, Georgia) into the tapered tips as filters, which permitted liquid, but not solid absorber materials to pass through. A 250-mg portion of each dried absorber was weighed beforehand into an appropriately labeled syringe.

To begin a set of experiments, a measured volume of the selected solution was pipetted into a plastic beaker and was then drawn from the beaker into the syringe through the Kynar filter. After closing off the tip of each syringe with a solid Luer cap, a set of six syringes, prepared as described, was mixed on a 48-rpm tube rotator for dynamic contact periods of 30 minutes, 2 hours, and 6 hours. At the end of each contact period, approximately 25% of the total solution volume was expelled through the filtered tip of the syringe

into a tared counting vial. The weight of the dispensed solution was determined and recorded for later use in the calculation of Kd values.

Calculation of Kd Values

Each portion of post-contact solution was assayed by gamma spectrometry for the specific gamma energies of the added radionuclides. The portion of each element sorbed on an absorber was determined indirectly from the difference in the measured gamma activity of the selected radionuclide before and after contact with the specified absorber.

Distribution coefficients (Kd) were calculated for each element as:

$$Kd = \frac{(pre\text{-}contact\ conc.) - (post\text{-}contact\ conc.)}{post\text{-}contact\ concentration}\ \frac{(ml\ solution)}{g\ absorber}.$$

Corrections

Because the weight of the post-contact assay solutions usually differed slightly from that of the pre-contact standard solution, each post-contact solution was first normalized to the same weight basis as the standard. Appropriate corrections also were made in the Kd calculations to account for the fact that the liquid/solid ratio and the total activity of every radionuclide changed after each sequential assay portion was removed.

The activity of each radionuclide in the post-contact solution was then compared with the activity of that same nuclide in the pre-contact standard. Because most absorbers were initially added in air-dried form, however, they sometimes preferentially absorbed the aqueous solvent, which had the effect of increasing the concentration of the solutes in the decreased solution volume. This resulted, at times, in the activity of certain radionuclides being higher in the post-contact solution than in the pre-contact solution. Whenever the post-contact/pre-contact solution activity for any radionuclide exceeded unity, we assigned a value of 1.000 to the solution having the highest value, and all other radionuclides in that same portion of solution were normalized accordingly.

RESULTS

Liquid Extractants

We prepared and evaluated porous carbon beads impregnated with 10 different liquid extractants. The measured distribution coefficients are presented in Tables I through X.

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LIX™-1010

Ten ml of LIX™-1010 (di-2-ethylhexylphosphoric acid) was diluted with 20 ml of cyclohexane, after which 25.76 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 34.7 g of the loaded Ambersorb™ beads.

Zirconium is moderately sorbed and technetium is weakly sorbed from acid-dissolved sludge solution, both with slow sorption kinetics. From acidified supernate solution, americium sorption is significantly greater than the lanthanides, which demonstrates a potential for an efficient trivalent actinide/lanthanide group separation. Uranium, zinc, and manganese are moderately sorbed from acidified supernate solution.

TABLE I. DISTRIBUTION OF 14 ELEMENTS INTO LIX™-1010
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.8	0.9	0.6
Sludge	Cs	<0.1	0.3	0.1
	Sr	<0.1	0.2	0.1
	Tc	1.8	2.7	4.0
	Y	1.1	0.5	0.4
	Cr	0.8	0.8	0.7
	Co	0.4	0.1	0.2
	Fe	0.4	0.3	0.5
	Mn	<0.1	0.2	0.1
	Zn	<0.1	0.4	0.2
	Zr	3.6	10	25
	U	0.1	<0.1	0.5
	Pu	0.8	0.7	0.9
	Am	0.1	0.1	0.1
Acidified	Ce	0.5	1.5	1.7
Supernate	Cs	<0.1	<0.1	0.2
	Sr	0.7	0.7	0.4
	Tc	1.0	1.7	3.0
	Y	1.0	1.2	1.3
	Cr	0.3	0.6	0.6
	Co	1.0	0.9	0.6
	Fe	<0.1	0.1	<0.1
	Mn	4.9	5.7	5.3
	Zn	3.6	4.0	4.0
	Zr	0.1	0.8	0.5
	U	4.3	7.1	15
	Pu	0.1	<0.1	0.1
	Am	10	27	64

LIX™-54

Ten ml of LIX™-54 (a beta diketone) was diluted with 20 ml of cyclohexane, after which 26.80 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 34.6 g of the loaded Ambersorb™ beads.

Zirconium is weakly sorbed from acid-dissolved sludge solution. From acid-ified supernate solution, the sorption of americium is significantly greater than that of lanthanides, which demonstrates a potential for an efficient trivalent actinide/lanthanide group separation. Uranium, iron, manganese, plutonium, and technetium are weakly-to-moderately sorbed from acidified supernate solution.

TABLE II. DISTRIBUTION OF 14 ELEMENTS INTO LIX™-54
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved Sludge	Ce	0.7	0.8	0.8
	Cs	0.5	0.5	0.3
	Sr	0.5	0.3	0.2
	Tc	0.5	0.5	0.5
	Y	0.7	0.6	0.6
	Cr	0.8	1.0	0.8
	Co	0.9	0.4	0.5
	Fe	0.7	0.3	0.4
	Mn	0.3	0.2	0.2
	Zn	0.5	0.3	0.2
	Zr	1.4	1.7	3.4
	U	<0.1	0.1	0.1
	Pu	0.5	0.1	0.1
	Am	0.3	0.2	0.1
Acidified Supernate	Ce	<0.1	0.4	0.4
	Cs	<0.1	0.3	0.1
	Sr	0.8	0.3	0.6
	Tc	1.4	2.8	4.1
	Y	0.4	0.1	0.3
	Cr	0.5	0.3	0.7
	Co	0.5	1.0	0.9
	Fe	2.9	3.4	4.7
	Mn	2.4	2.1	2.6
	Zn	0.4	0.3	0.6
	Zr	0.1	0.3	0.4
	U	11	12	12
	Pu	2.2	2.8	3.0
	Am	34	32	30

<u>LIX™-84</u>

Ten ml of LIX™-84 (a ketoxime) was diluted with 20 ml of cyclohexane, after which 26.13 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 34.8 g of the loaded Ambersorb™ beads.

None of the 14 elements was significantly sorbed from either solution.

TABLE III. DISTRIBUTION OF 14 ELEMENTS INTO LIX™-84
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.7	0.7	0.9
Sludge	Cs	0.4	0.1	0.4
	Sr	<0.1	0.3	0.1
	Tc	0.7	1.2	1.7
	Y	0.7	0.6	0.8
	Cr	0.4	0.7	0.8
	Co	0.6	1.0	0.7
	Fe	1.0	0.5	0.8
	Mn	0.9	0.3	0.5
	Zn	0.6	0.2	0.3
	Zr	0.8	1.4	1.8
	U	<0.1	0.3	0.5
	Pu	0.6	0.4	0.3
	Am	0.7	0.2	0.2
Acidified	Ce	0.9	0.4	0.5
Supernate	Cs	<0.1	0.4	0.2
	Sr	0.5	0.4	0.6
	Tc	0.2	0.7	0.7
	Y	0.2	0.4	0.4
	Cr	1.0	1.3	1.3
	Co	0.8	0.8	1.1
	Fe	0.3	0.6	0.5
	Mn	0.9	0.9	0.6
	Zn	0.6	0.4	0.2
	Zr	<0.1	<0.1	0.4
	U	0.8	0.8	0.7
	Pu	0.7	0.5	0.4
	Am	<0.1	<0.1	<0.1

LIX™-26

Ten ml of LIX™-26 (8-hydroxyquinoline) was diluted with 20 ml of cyclohexane, after which 26.90 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 37.1 g of the loaded Ambersorb™ beads.

Plutonium is moderately sorbed, whereas uranium and technetium are weakly sorbed from acid-dissolved sludge solution. Technetium, cerium, americium, and uranium sorb at useful levels from acidified supernate solution.

TABLE IV. DISTRIBUTION OF 14 ELEMENTS INTO LIX™-26
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.4	0.5	0.4
Sludge	Cs	0.4	0.5	0.2
	Sr	0.7	0.4	0.5
	Tc	1.1	1.4	1.4
	Y	0.5	0.6	0.4
	Cr	0.5	0.6	0.5
	Co	0.4	0.4	0.5
	Fe	0.3	0.6	0.5
	Mn	<0.1	<0.1	<0.1
	Zn	0.5	0.4	0.3
	Zr	0.6	0.6	0.7
	U	2.0	2.7	3.4
	Pu	3.9	7.9	15
	Am	0.4	0.5	0.4
Acidified	Ce	32	40	42
Supernate	Cs	<0.1	<0.1	<0.1
	Sr	0.6	0.7	0.6
	Tc	54	112	190
	Y	4.1	4.7	4.9
	Cr	<0.1	0.2	0.3
	Co	1.3	1.1	1.1
	Fe	0.3	0.3	0.3
	Mn	<0.1	<0.1	0.1
	Zn	0.3	0.2	0.1
	Zr	0.8	0.9	0.7
	U	12	21	24
	Pu	0.2	0.1	0.2
	Am	24	33	30

<u>CYANEX™ 923</u>

Ten ml of Cyanex™ 923 (trialkylphosphine oxide) was diluted with 20 ml of cyclohexane, after which 26.83 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 36.8 g of loaded Ambersorb™ beads.

From acid-dissolved sludge solution, plutonium, zirconium, and technetium sorb moderately with slow sorption kinetics. Technetium, lanthanides, americium, and uranium sorb strongly from acidified supernate solution. This extractant demonstrates an outstanding ability to recover many targeted elements from acidified supernate solution.

TABLE V. DISTRIBUTION OF 14 ELEMENTS INTO LIX™-1010
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.5	0.2	0.5
Sludge	Cs	0.5	0.3	0.4
	Sr	0.8	0.5	0.5
	Tc	1.8	2.6	4.3
	Y	0.7	0.4	0.7
	Cr	0.8	0.6	0.9
	Co	0.5	0.2	0.3
	Fe	<0.1	0.4	0.6
	Mn	<0.1	<0.1	<0.1
	Zn	0.2	0.1	0.2
	Zr	2.0	3.1	7.6
	U	<0.1	1.0	1.9
	U*	2.7	6.5	14
	Pu	3.0	7.6	21
	Pu*	21	100	199
	Am	0.3	0.3	0.3
	Am*	0.4	0.4	0.4
Acidified	Ce	744	987	1596
Supernate	Cs	0.8	0.5	0.4
	Sr	0.8	0.5	0.6
	Tc	300	551	582
	Y	1078	2014	2981
	Cr	<0.1	<0.1	0.3
	Co	0.6	0.7	0.7
	Fe	<0.1	<0.1	<0.1
	Mn	0.4	0.9	0.7
	Zn	2.9	4.4	4.3
	Zr	0.9	0.4	0.8
	U	122	233	356
	Pu	1.4	1.9	2.0
	Am	538	751	802

* These solutions contained 60 mg Pu/L; all others contained 3 g Pu/L.

<u>CYANEX^{TM} 272</u>

Ten ml of Cyanex^{TM} 272 (trimethylpentylphosphoric acid) was diluted with 20 ml
of cyclohexane, after which 26.49 g of Ambersorb^{TM} 563 porous carbon beads,
obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the
liquid. The cyclohexane evaporated to yield 37.1 g of the loaded Ambersorb^{TM}
beads.

Zirconium is strongly and selectively sorbed from acid-dissolved sludge
solution, although the sorption kinetics are slow. Uranium is selectively
sorbed at a useful level from acidified supernate solution.

TABLE VI. DISTRIBUTION OF 14 ELEMENTS INTO CYANEX^{TM}-272
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.4	0.2	0.5
Sludge	Cs	0.3	0.2	0.3
	Sr	0.5	0.2	0.4
	Tc	0.4	0.6	0.7
	Y	0.8	0.7	0.7
	Cr	0.5	0.6	0.8
	Co	0.5	0.4	0.4
	Fe	<0.1	0.3	0.3
	Mn	<0.1	<0.1	<0.1
	Zn	0.1	0.3	0.2
	Zr	8.0	32	199
	U	0.3	1.4	1.4
	Pu	0.9	1.3	2.8
	Am	<0.1	0.2	0.1
Acidified	Ce	0.1	0.5	0.2
Supernate	Cs	0.4	0.1	0.1
	Sr	0.9	0.6	0.9
	Tc	0.5	0.3	0.5
	Y	0.8	1.0	1.1
	Cr	<0.1	0.7	0.7
	Co	0.8	0.7	0.7
	Fe	0.5	0.6	0.6
	Mn	<0.1	<0.1	<0.1
	Zn	1.1	1.3	1.6
	Zr	0.5	0.7	0.6
	U	23	41	58
	Pu	0.2	0.1	0.3
	Am	1.2	1.1	1.3

DHDECMP

Ten ml of DHDECMP (dihexyl-N,N-diethylcarbamoylmethylphosphonate), obtained from Occidental Chemical Corporation, Specialty Products Division, Niagara Falls, NY, was diluted with 20 ml of cyclohexane, after which 27.99 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 39.5 g of the loaded Ambersorb™ beads.

Plutonium is moderately sorbed, whereas uranium and technetium are weakly sorbed from acid-dissolved sludge solution. From acidified supernate solution, technetium, cerium, americium, and uranium are well sorbed, whereas yttrium is weakly sorbed.

TABLE VII. DISTRIBUTION OF 14 ELEMENTS INTO DHDECMP
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.2	0.3	0.4
Sludge	Cs	0.4	0.3	0.4
	Sr	0.3	0.4	0.4
	Tc	0.7	1.0	1.6
	Y	0.5	1.0	0.6
	Cr	0.8	0.8	0.7
	Co	0.5	0.4	0.2
	Fe	0.2	0.3	0.3
	Mn	<0.1	<0.1	<0.1
	Zn	0.2	0.3	0.2
	Zr	0.9	0.6	0.9
	U	1.3	2.2	3.2
	Pu	4.4	8.2	15
	Am	0.4	0.6	0.4
Acidified	Ce	26	45	51
Supernate	Cs	<0.1	<0.1	<0.1
	Sr	0.5	0.6	0.7
	Tc	43	112	222
	Y	3.4	5.1	5.8
	Cr	0.3	0.2	0.6
	Co	0.8	0.9	1.1
	Fe	<0.1	<0.1	<0.1
	Mn	0.6	0.5	0.4
	Zn	0.7	0.4	0.5
	Zr	<0.1	0.5	0.8
	U	22	31	33
	Pu	0.4	0.3	0.4
	Am	25	34	35

DHDECMP-DIPB

Ten ml of DHDECMP was diluted with 10 ml of DIPB (diisopropylbenzene) and 30 ml of cyclohexane, after which 48.45 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 65.3 g of the loaded Ambersorb™ beads.

Plutonium is moderately sorbed from acid-dissolved sludge solution. From acidified supernate solution, technetium, cerium, americium, and uranium are sorbed at useful levels, whereas yttrium is weakly sorbed.

TABLE VIII. DISTRIBUTION OF 14 ELEMENTS INTO DHDECMP-DIPB FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	<0.1	0.4	0.6
Sludge	Cs	0.1	<0.1	0.2
	Sr	0.2	0.1	0.4
	Tc	0.6	1.1	1.5
	Y	0.3	0.4	0.6
	Cr	0.5	0.7	0.9
	Co	0.1	0.4	0.5
	Fe	0.3	0.5	0.3
	Mn	<0.1	<0.1	<0.1
	Zn	0.3	0.5	0.2
	Zr	0.4	0.8	1.0
	U	1.2	2.3	2.7
	Pu	2.9	5.5	10
	Am	0.4	0.6	0.3
Acidified	Ce	18	28	28
Supernate	Cs	0.1	<0.1	0.2
	Sr	0.5	0.7	0.4
	Tc	27	49	62
	Y	2.9	3.3	3.1
	Cr	0.6	0.5	0.5
	Co	0.6	0.9	0.6
	Fe	0.1	0.3	0.4
	Mn	<0.1	0.3	0.2
	Zn	0.2	0.3	0.1
	Zr	0.4	0.5	0.5
	U	9.3	16	18
	Pu	0.7	0.6	0.4
	Am	14	21	19

CMPO-DIPB

Ten ml of CMPO (octylphenyl-N,N-diisobutylcarbamoylmethylphosphine oxide), obtained from M&T Chemicals, Inc., Rahway, NJ, was diluted with 10 ml of DIPB (diisopropylbenzene) and 30 ml of cyclohexane, after which 46.83 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 64.1 g of the loaded Ambersorb™ beads.

Technetium, zirconium, and uranium are weakly sorbed from acid-dissolved sludge solution. Technetium, cerium, americium, uranium, and yttrium are well sorbed from acidified supernate solution.

TABLE IX. DISTRIBUTION OF 14 ELEMENTS INTO CMPO-DIPB
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.3	0.3	0.6
Sludge	Cs	0.5	0.2	0.3
	Sr	0.5	0.4	0.3
	Tc	0.6	1.1	2.2
	Y	0.5	0.6	0.5
	Cr	0.7	0.8	0.9
	Co	0.2	0.1	0.1
	Fe	0.6	0.6	0.5
	Mn	<0.1	<0.1	<0.1
	Zn	0.4	0.3	0.3
	Zr	0.8	0.8	1.4
	U	0.4	0.8	1.4
	Pu	0.4	0.4	0.7
	Am	0.4	0.5	0.5
Acidified	Ce	78	370	499
Supernate	Cs	0.8	0.3	0.3
	Sr	<0.1	0.2	0.3
	Tc	41	111	196
	Y	15	45	89
	Cr	0.8	0.7	0.5
	Co	0.1	1.0	0.8
	Fe	<0.1	<0.1	0.2
	Mn	1.6	1.6	2.1
	Zn	1.0	0.4	0.5
	Zr	1.1	0.8	0.8
	U	49	183	291
	Pu	0.8	0.5	1.0
	Am	49	355	472

<u>Aliquat 336</u>

Five grams of Aliquat™ 336, (tricaprylmethylammonium chloride) obtained from Aldrich Chemical Co., was diluted with 10 ml of hexane, after which 17.32 g of Ambersorb™ 563 porous carbon beads, obtained from Rohm & Haas Co., Philadelphia, PA, was added to absorb the liquid. The cyclohexane evaporated to yield 26.2 g of the loaded Ambersorb™ beads.

Plutonium is moderately sorbed from acid-dissolved sludge solution. From acidified supernate solution, technetium is strongly sorbed with high selectivity and only uranium is cosorbed to a significant extent. This extractant demonstrates an outstanding ability to selectively recover technetium from acidified supernate solution.

TABLE X.   DISTRIBUTION OF 14 ELEMENTS INTO ALIQUAT™ 336
FROM TWO SOLUTIONS SIMULATING HANFORD HLW TANK 102-SY.

Medium	Element	30-min Kd	2-h Kd	6-h Kd
Acid-Dissolved	Ce	0.4	0.6	0.6
Sludge	Cs	0.3	0.3	0.2
	Sr	0.4	0.3	0.3
	Tc	0.7	0.8	1.0
	Y	0.3	0.3	0.2
	Cr	<0.1	<0.1	0.1
	Co	0.6	0.3	<0.1
	Fe	0.2	0.3	0.5
	Mn	<0.1	<0.1	<0.1
	Zn	0.3	0.1	0.1
	Zr	0.8	0.4	0.6
	U	0.5	1.1	1.7
	Pu	2.3	5.7	14
	Am	0.2	0.2	0.2
Acidified	Ce	1.0	1.6	2.0
Supernate	Cs	1.8	2.2	2.1
	Sr	0.3	0.2	0.3
	Tc	295	444	572
	Y	0.3	0.4	0.2
	Cr	0.1	0.1	0.1
	Co	0.2	0.1	<0.1
	Fe	0.2	0.3	0.3
	Mn	0.3	0.3	0.3
	Zn	<0.1	<0.1	<0.1
	Zr	<0.1	0.1	0.1
	U	3.2	5.5	7.9
	Pu	0.1	0.2	0.4
	Am	0.4	0.6	0.6

CONCLUSIONS

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1. Technetium is sorbed well by all five anion exchange resins. From acid-dissolved sludge solution the order is Reillex™ HPQ > Purolite™ A-520-E > Ionac™ SR-3 > Sybron (Pr)₃N > Ionac™ SR-6. From acidified supernate solution the order is Sybron™ SR-3 > Ionac™ SR-6 > Purolite™ A-520-E > Reillex™ HPQ > Ionac™ SR-3.

2. Plutonium is sorbed well by some of these resins from acid-dissolved sludge solution only. The order is Reillex™ HPQ > Purolite™ A-520-E > Ionac™ SR-3 > Sybron (Pr)₃N > Ionac™ SR-6.

3. Uranium is sorbed somewhat by most of these resins. From acid-dissolved sludge solution the order is Purolite™ A-520-E > Ionac™ SR-3 > Reillex™ HPQ > Sybron (Pr)₃N > Ionac™ SR-6. From acidified supernate solution the order is Reillex™ HPQ > Sybron (Pr)₃N > Purolite™ A-520-E > Ionac™ SR-3 > Ionac™ SR-6.

4. Cerium is sorbed significantly better than yttrium or americium by most of these resins. The order for cerium sorption from acid-dissolved sludge is Purolite™ A-520-E > Ionac™ SR-3 > Reillex™ HPQ > Sybron™ (Pr)₃ > Ionac™ SR-6. From acidified supernate solution the order is the same.

5. Americium is weakly sorbed, and only from acid-dissolved sludge solutions, by Purolite™ A-520-E, Ionac™ SR-3, and Reillex™ HPQ.

6. The increased hydrophobicity of Sybron™ (Pr)₃N and Ionac™ SR-6 resins was reflected by the fact that these two resins floated on the surface of the aqueous solutions during the contacts. The measured Kd values obtained during our dynamic batch contacts may be significantly lower than what might be obtained from column experiments, in which improved liquid/solid contact would be expected as slightly pressurized solution is passed through the resin column.

REFERENCE

1. S. F. MARSH, Z. V. SVITRA, and S. M. BOWEN, "The Distribution of 14 Elements on 60 Selected Absorbers from Simulated Acidic Dissolved Sludge and Alkaline Supernate Solutions for Hanford High-Level Waste Tank 102-SY," Los Alamos National Laboratory report LA-12654 (October 1993).