

REMOVAL OF RADIONUCLIDES FROM THE WATER-SOLUBLE FRACTION OF HANFORD NUCLEAR DEFENSE WASTES

Denis M. Strachan
Battelle Pacific Northwest Laboratory

Wallace W. Schulz
Rockwell Hanford Operations

INTRODUCTION

The current Hanford Waste Management Program has operated since 1968 to remove the bulk of the long-lived heat emitters ^{90}Sr and ^{137}Cs from stored high-level wastes ^{1,2}. The liquid waste remaining after removal of ^{90}Sr and ^{137}Cs is returned to underground tanks for eventual evaporation to damp solid salt cake. Hanford salt cake is an admixture of large amounts of NaNO_3 with minor amounts of $\text{NaAl}(\text{OH})_4$, Na_2CO_3 , Na_2CrO_4 , Na_3PO_4 , Na_2SO_4 , and NaOH ⁽³⁾. Small amounts of ^{90}Sr , ^{137}Cs , ^{99}Tc and other radionuclides are also associated with the salt cake.

Approximately 95,000 m³ of salt cake and 49,000 m³ of "sludge" will eventually (1980's) accumulate in approximately 50 underground single-shell tanks ³. In addition, about 45,000 m³ of residual liquid are anticipated for storage in double-shell tanks. Hanford evaporator-crystallizer units are operated to severely restrict the amount of hygroscopic NaOH in salt cake. The highly alkaline (4 to 6M NaOH) residual liquid phase resulting from such limited evaporation contains large amounts of soluble sodium salts (e.g. NaNO_3 , NaNO_2 , $\text{NaAl}(\text{OH})_4$, etc.), ^{137}Cs , and ^{90}Sr .

Sludge is defined to mean all water-insoluble solids stored in underground tanks irrespective of their origin and composition. Sludge includes the hydrated oxides (manganese, iron, aluminum, etc.) which precipitated when BiPO_4 , Redox, and Purex process high-level acidic wastes were made alkaline and thermally concentrated in underground tanks. It also comprises such diverse solids as $\text{ZrO}_2 \cdot \text{H}_2\text{O}$ formed by addition of NaOH to Zirflex deacidating wastes⁽⁴⁾ and $\text{Ni}_2[\text{Fe}(\text{CN})_6]$ and $\text{Sr}_5(\text{OH})(\text{PO}_4)_3$ precipitates

from early day scavenging of aged BiPO_4 process wastes. The latter solids contain high concentrations of ^{90}Sr and ^{137}Cs while the Redox and Purex process sludges contain large amounts of actinides and ^{90}Sr (3).

One alternative for long-term management(3,5,6) of high-level Hanford wastes involves retrieval, after a yet-to-be determined interim storage time, conversion to more immobile forms, and terminal storage in a suitable geologic repository. Technology for conversion of salt cake to immobile silicate-based glass or mineral forms is partially developed(3,7,8,). Similar technology for immobilization of sludge and residual liquid is being developed(9,10).

Another alternative for long-term management of salt cake and residual liquid involves removing most of the long-lived ($t_{1/2} \geq 10\text{y}$) radionuclides and many of the shorter-lived ones from these wastes; the objective of this Hanford Radionuclide Removal process (Fig. 1) is to partition the salt cake and residual liquid into: (1) a small volume of highly radioactive waste requiring further immobilization for long-term storage and (2) a large volume of low-level waste which can be safely stored or disposed of by inexpensive means. Cost data(3) indicate potential economic benefits may result from a reduction in the volume of high-level waste requiring immobilization and high-integrity containers and storage facilities. Criteria for acceptably decontaminated salt have not yet been established. Our current working guideline is to reduce the sum of the concentrations of actinides and other long-lived ($t_{1/2} > 10\text{y}$) radionuclides to $< 10 \text{ nCi/g}$ of dried salt.

Results of successful bench-scale radionuclide removal tests with various actual salt cakes and certain liquid waste solutions representative of residual liquids were reported earlier(11). This paper describes conditions and results of recent hot cell tests of the complete Hanford Radionuclide Removal Process. These advanced tests, made with actual residual liquid containing large concentrations of ethylenediaminetetracetic acid (EDTA) and other organic compounds, provided a rigorous and convincing proof of the process flowsheet.

HANFORD RADIONUCLIDE REMOVAL PROCESS

Figure 2 illustrates the sequence of steps currently proposed for removal of long-lived radionuclides from salt cake solutions, residual liquids, or mixtures of the two. Radiocesium is removed from alkaline (1-3M NaOH) solutions by passage through two or more beds of Duolite ARC-359 resin, a cation exchange resin made by the Diamond Shamrock Co. Subsequently, ^{90}Sr , actinides, fission

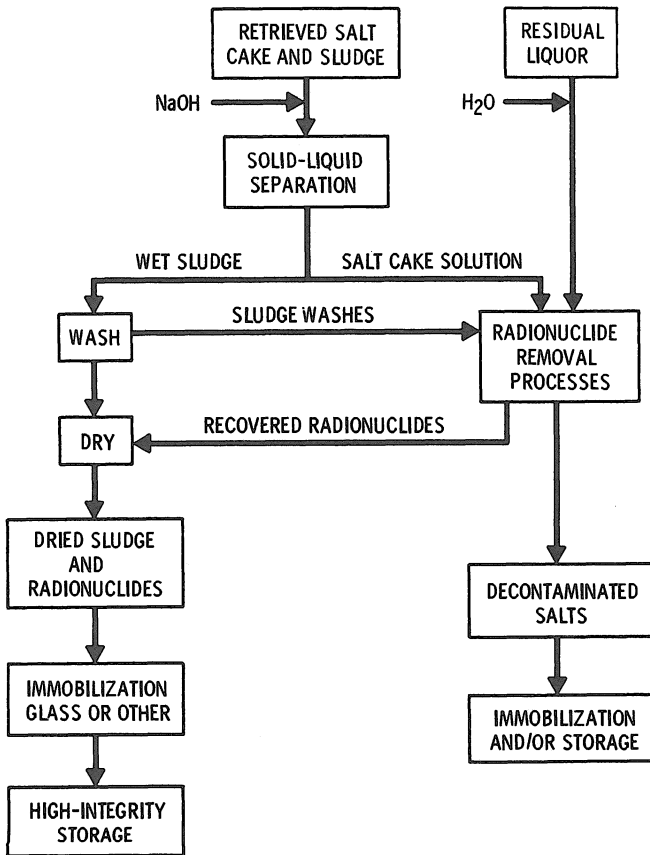


Fig. 1. Conceptual Radionuclide Removal Option for Long-Term Management of Hanford In-Tank Wastes.

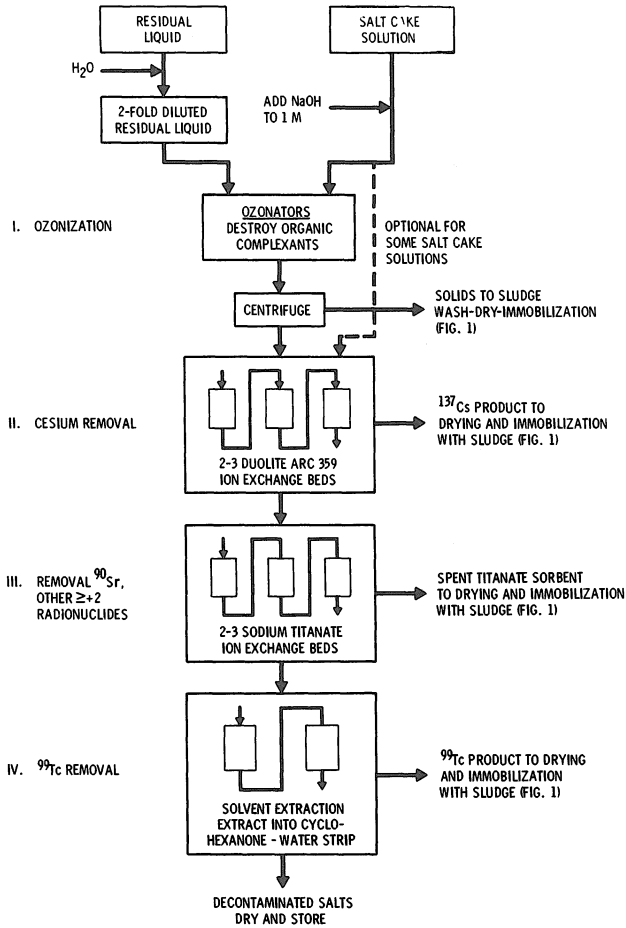


Fig. 2. Process Sequence for Removal of Radionuclides from Hanford Salt Cake and Residual Liquid.

product rare earths, and other multivalent cationic radionuclides are removed by passage of the effluent from the Duolite ARC-359 resin columns through a series of interconnected beds of sodium titanate ($\text{Na}[\text{Ti}_2\text{O}_5\text{H}]$) a newly developed inorganic cation exchanger(12). Finally, ^{99}Tc is removed by a cyclohexanone solvent extraction process. Details of each of the individual process steps are provided elsewhere(11,13).

Ozonolytic destruction of EDTA, HEDTA (2-hydroxyethylethylenediaminetriacetic acid), and other organic complexing agents present in most residual liquids and in some salt cake solutions is a key headend step in the Hanford Radionuclide Removal process. EDTA and other organic complexants form anionic and/or neutral species with ^{90}Sr and other important radiocations and prevent their uptake by sodium titanate. The goal of producing decontaminated salt containing 10 nCi/g or less of long-lived radionuclide simply cannot be achieved without providing for oxidative destruction of EDTA and other deleterious organic complexants. A paper describing relevant ozone chemistry and application of ozone technology to destroy organic materials in alkaline radioactive waste solutions has recently been published(14).

Reference 11 provides a list of all principal radionuclides expected to be present to the extent of 1 nCi/g in existing Hanford salt cake and residual liquid. Some suggestions for techniques which might be applied, if necessary, to further immobilize decontaminated salt are also presented in Reference 11.

Savannah River Laboratory workers have also previously developed technology for removal of ^{137}Cs , ^{134}Cs , ^{90}Sr , and actinides from stored Savannah River Plant high-level wastes. Their process(15) employs Duolite ARC-359 resin for removal of radiocesium and plutonium and the cation exchange resin Amberlite IRC-718 (Rohm and Haas Co.) for removal of ^{90}Sr . No provision has been made in their flowsheet for removal of ^{99}Tc . The Savannah River Plant wastes do not contain deleterious amounts of organic complexants.

EXPERIMENTAL APPARATUS - PROCEDURES

Ozonation and radiocesium removal steps were carried out in a hot cell with equipment described in detail elsewhere(16). Subsequent process steps were performed outside the cell. Equipment and procedures used in miniature mixer-settler ^{99}Tc extraction tests are discussed in Ref. 11. Reference 11 also provides detailed coverage of the resins, other chemicals, and analytical procedures used in the present work.

DISCUSSION AND RESULTS

Feed Solution for Flowsheet Tests

Approximately 42 L of actual waste from Tank 102-AZ were loaded into a Rockwell hot cell in late 1978. The solution stored in Tank 102-AZ is waste produced from Hanford B Plant ^{90}Sr solvent extraction removal; it contains very high concentrations of organic complexants and their radiolytic decomposition products (Table I). This type of waste solution presents a severe challenge, perhaps the most rigorous, to the decontamination capabilities of the Hanford Radionuclide Removal process. Tank 102-AZ waste was used as feed in two flowsheet verification tests, results of which are discussed in this paper. For convenience, some of the simplified tables in the text provide only key composition data.

TABLE I. Partial Composition
of Tank 102-AZ Waste.

Component	Concentration
Na	8.0M
OH	0.97
CO_3	0.62M
NO_2	0.31M
Al	0.12M
Fe	0.12M
TOC(a)	7.4M
^{137}Cs	$3.2 \times 10^5 \text{ } \mu\text{Ci/L}$
^{90}Sr	$1.6 \times 10^5 \text{ } \mu\text{Ci/L}$
^{155}Eu	$2.6 \times 10^4 \text{ } \mu\text{Ci/L}$
^{99}Tc	10 $\mu\text{Ci/L}$
Pu	$7.9 \times 10^{-4} \text{ g/L}$
U	$2.8 \times 10^{-5} \text{ g/L}$

(a) Total organic carbon.

Ozonation

Trial runs showed that, to avoid excessive foaming and undesirable precipitation of Na_2CO_3 and $\text{Al}(\text{OH})_3$, it was necessary to dilute as-received Tank 102-AZ solution prior to ozonolysis. Trial ozonation runs (Table II) using 8:1, 4:1, and 2:1 (distilled

water to waste) dilutions were made. Satisfactory ozonations were performed at a 4:1 dilution; solids precipitated during ozonation of two-fold diluted waste.

TABLE II. Process Parameters for
Ozonation of Tank 102-AZ Waste.

Investigated

Solution Dilution - 2:1, 3:1, 4:1, 8:1
Diluent - H₂O
Temperature - 50°C

Selected

Solution Dilution - 4:1
Diluent - H₂O
Temperature - 50°C
Ozone Source - Air, O₂
Time - 72 hrs (air) & 8 hrs (O₂)
 - 48 hrs (O₂)
Ozone Content - 5 wt% in O₂
 3 wt% in Air

Standard conditions used to ozonate 1.5 liter batches of 4:1 diluted Tank 102-AZ waste are listed in the bottom half of Table II. As indicated, two different ozonation time schedules were followed depending on whether O₃ was generated from oxygen or air. Ozonation tests with solutions simulating 4:1 diluted Tank 102-AZ waste indicated satisfactory destruction of EDTA and other complexants was accomplished in about 24 hours when using O₃ generated from O₂. Since the primary purpose of these experiments was to demonstrate the overall capabilities of the Hanford Radioclide Removal process and not to make a detailed study of the ozonolysis step, ozonation was continued beyond that judged necessary by the tests with simulated waste.

Ozonation of Tank 102-AZ waste destroyed EDTA- and HEDTA-Fe complexes. The released iron precipitated as the hydrated oxide carrying down with it substantial amounts of ⁹⁰Sr, ¹⁵⁵Eu, plutonium, americium, and other multivalent cationic radionuclides (Table III). (Precipitated hydrated iron oxide clogged the metal frit used originally to disperse O₂-O₃ in the ozonator pot; this frit was replaced with a sparge ring.)

TABLE III. Ozonation of 4:1 Diluted Tank 102-AZ Waste: Typical Results.

Component	Before Ozonation	After Ozonation ^(a)
Na	1.8M	2.5M
OH	0.2M	0.89M
CO ₃	0.12M	0.11M
NO ₂	0.06M	$7.1 \times 10^{-4}M$
Al	0.02M	0.02M
Fe	0.02M	$4.9 \times 10^{-5} - 2.5 \times 10^{-4}M$
TOC	1.5M	0.16 - 0.32M
NH ₄		0.002
¹³⁷ Cs	$6.0 \times 10^4 \mu\text{Ci/L}$	$6.0 \times 10^4 \mu\text{Ci/L}$
⁹⁰ Sr	$5.0 \times 10^4 \mu\text{Ci/L}$	$4.6 \times 10^2 - 9.9 \times 10^2 \mu\text{Ci/L}^{(b)}$
¹⁵⁵ Eu	$5.0 \times 10^3 \mu\text{Ci/L}$	$3.0 \times 10^2 \mu\text{Ci/L}$
⁹⁹ Tc	$2 \mu\text{Ci/L}$	$5.3 \mu\text{Ci/L}$
Pu	$1.6 \times 10^{-4} \text{ g/L}$	$1.5 \times 10^{-6} - 6.5 \times 10^{-6} \text{ g/L}^{(b)}$
U	$6.0 \times 10^{-6} \text{ g/L}$	$2.7 \times 10^{-5} \text{ g/L}$

(a) After adjusting to $\sim 1.0M$ OH for subsequent removal of radiocesium.

(b) A second value represents concentration obtained as a result of addition of $\text{Fe}(\text{NO}_3)_3$ to ozonated solution.

Following completion of ozonation of each 1.5L batch of 4:1 diluted Tank 102-AZ waste, sufficient 18M NaOH was added to the solid-liquid mixture to adjust the solution to about 1.0M NaOH. In the first flowsheet test (Run 1) sufficient 1.5M $\text{Fe}(\text{NO}_3)_3$ was added to make the solution about 0.0015M Fe prior to its immediate precipitation as hydrated iron oxide. This iron addition was made to reduce further the concentration of ⁹⁰Sr, plutonium, and multi-valent cationic radionuclides prior to subsequent process steps. Iron was not added to ozonated batches of feed prepared in Run 2. Data in Table III show that the iron addition step did enhance, albeit slightly, removal of ⁹⁰Sr and plutonium.

A major program to determine the stoichiometry, kinetic, and other parameters of importance in ozonolytic oxidation of EDTA and HEDTA in typical Hanford waste solutions is currently in progress in Rockwell laboratories. Preliminary results⁽¹⁴⁾ of these tests with both radioactive and nonradioactive solutions indicate the reaction of O₃ with EDTA follows second-order reaction kinetics. Reaction products include CO₂, NH₃ and refractory aqueous soluble organic species such as oxalate ion.

Ozonated solutions from Runs 1 and 2, after adjustment to 1.0M NaOH, were allowed to stand several days at $\sim 25^{\circ}\text{C}$ for the iron precipitate to settle. Clear supernatant liquid was removed from each 1.5L batch and composited as feed to the cesium removal step.

Prior to the tests with ozonated solutions, approximately 9000 mL, 30 column volumes (CV), of diluted (1:1 with 1M NaOH) unozonated Tank 102-AZ solution were passed downflow at 25°C through two interconnected 0.3L beds of 20-50 mesh Duolite ARC-359 resin. (The resin bed experimental hook-up is described in Ref. 16). The ^{137}Cs concentration of the effluent from the final resin bed ranged from 46.8 $\mu\text{Ci/L}$ (3 CV) to 57.0 $\mu\text{Ci/L}$ (30 CV). Passage of the composite effluent solution through a third fresh 0.3L bed of Duolite ARC-359 resin only decreased its ^{137}Cs level to 36 $\mu\text{Ci/L}$. Apparently, the residual ^{137}Cs ($\sim 3 \times 10^{-9}\text{M}$) was complexed by the large amount of EDTA and other complexants present in the unozonated feed. The loaded Duolite resin bed system was washed with 3 CV of 0.01M NaOH and then eluted with 8.3 CV of 1.5M $(\text{NH}_4)_2\text{CO}_3$ - 1M NH_4OH . The elution step removed 84% of the ^{137}Cs loaded on the resin beds. The eluted resin beds were washed (upflow) with 1 CV of water and 3 CV of 1M NaOH.

Subsequently, about 33L of ozonated feed from Run 1 were passed downflow at $\sim 25^{\circ}\text{C}$ and 1 CV/hr through two interconnected 0.3L beds of ARC-359 resin. The first bed in this system was the second ARC-359 resin used with the unozonated feed while the second bed was unused resin. The experimental set-up permitted periodic sampling of the effluent from both the resin columns. Cesium breakthrough curves for both the resin beds in Run 1 are plotted in Fig. 3. The composite of the first 72 CV of solution which passed through both resin beds contained 22 $\mu\text{Ci/L}$ ^{137}Cs . Passage of this composite through a third, fresh 0.3L bed of Duolite ARC-359 resin reduced the ^{137}Cs concentration to $< 0.15 \mu\text{Ci/L}$.

Following the first cesium removal run, the two Cs-loaded Duolite resin beds were washed with 4 CV of water and then the cesium was eluted with 17 CV of 3M $(\text{NH}_4)_2\text{CO}_3$ - 2M NH_4OH solution. This amount of eluent removed 97% of the ^{137}Cs loaded onto the bed. After the elution step the resin beds were washed with one CV of water and 6 CV of 1M NaOH. All washing and elution steps were performed at $\sim 25^{\circ}\text{C}$ under downflow conditions.

About 23L of ozonated feed from Run 2 were passed through the same two Duolite resin bed systems used in the first cesium removal run. Cesium breakthrough curves for the two Duolite resin beds in

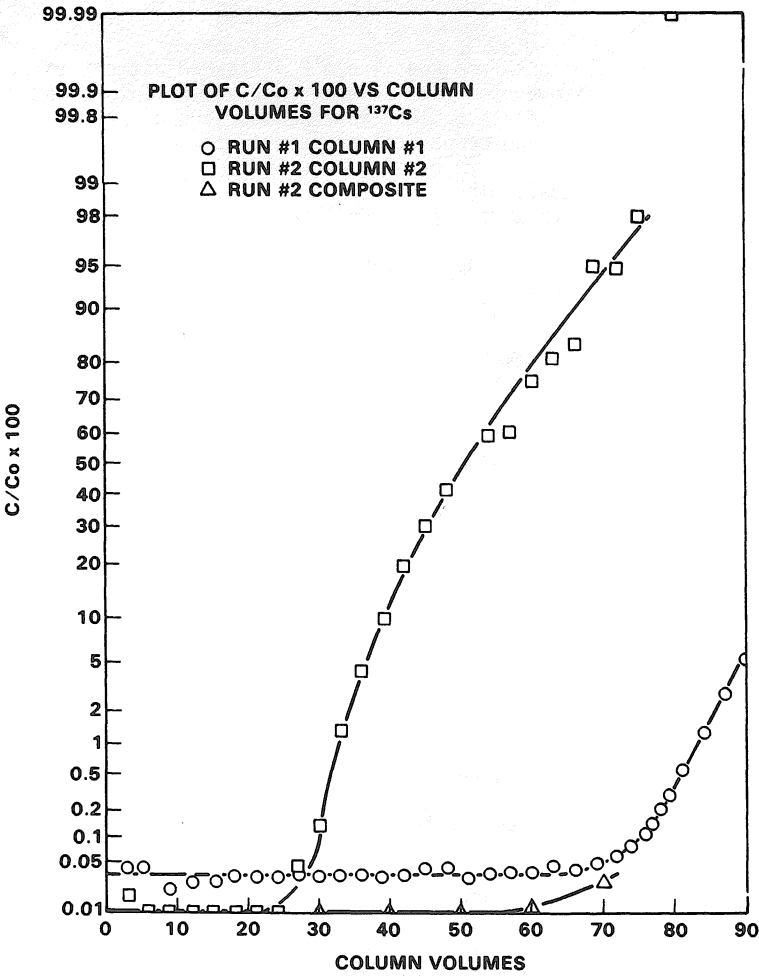


Fig. 3. Duolite ARC-359 Resin Sorption of ^{137}Cs .

this second run are plotted in Fig. 3. The composite of the 70 CV of solution which passed through both resin beds contained $4.6 \mu\text{Ci/L } ^{137}\text{Cs}$. Passage of a portion of this composite solution through a third fresh bed of Duolite ARC-359 resin reduced its ^{137}Cs concentration to $<0.11 \mu\text{Ci/L}$.

As evidenced by the plots in Fig. 3, in both Runs 1 and 2 breakthrough of ^{137}Cs in the 2-column Duolite ARC-359 resin bed system did not exceed 0.05% until after passage of about 70 CV of feed. These capacity data are in agreement with results obtained earlier by Wiley(15). A significant and disappointing feature of our results is that even the very first portion of loading cycle effluent in both runs contained 2 to $4.6 \mu\text{Ci/L } ^{137}\text{Cs}$, a radio-cesium concentration substantially higher than the desired $<0.1 \mu\text{Ci/L}$. Various reasons for the failure of the 2-column resin bed system to provide the desired cesium removal have been suggested. These range from use of non-representative Duolite ARC-359 resin to "bleed through" of ^{137}Cs not eluted from a previous loading cycle. Desired ^{137}Cs concentrations were achieved in both Runs 1 and 2 by flowing the composite loading cycle effluent from the first two Duolite resin beds through a third resin bed. Repetitive load-elution cycle tests with $1\text{M NaOH} - 2\text{M NaNO}_3$ feed spiked to $20 \mu\text{Ci/L } ^{137}\text{Cs}$ to determine if one Duolite ARC-359 resin bed will continue to provide adequate cesium decontamination are in progress. Additional hot cell cesium load-elution tests are also planned to determine optimum conditions for operation with ozonated feeds.

Duolite ARC-359 resin is, of course, specifically useful for sorbing Cs^+ from highly alkaline solutions. Because it is a cation exchanger, the ARC-359 resin also sorbs other cationic radionuclides from such media. Thus, the 3-bed Duolite resin system used in Run 1 provided a ^{90}Sr decontamination factor (DF) of 9 while in Run 2 the 2-bed system provided a ^{90}Sr DF of about 6. Some sorption of plutonium, uranium, and europium was also observed.

Sodium Titanate Sorption Step

Composite effluent resulting from the third Duolite ARC-359 resin bed in both Runs 1 and 2 was passed through a series of three interconnected beds of titanate-loaded ($37 \text{ wt\% Na[Ti}_2\text{O}_5\text{H]})$ macroreticular anion exchange beds to remove ^{90}Sr (Table IV). (Procedures used to prepare this sorbent are described in Ref. 11). Three fresh resin beds were used to treat Run 1 feed; prior to use resins were washed free of chloride with 1M NaOH and then classified by upflow of water. Following Run 1, the first

bed in the series was removed; the second bed was moved to the first position, the third bed was moved to the second position, and a fresh bed of titanate-loaded resin was installed as the last bed in the series. In each run the effluent from the last bed in the series was periodically analyzed.

In accord with earlier bench-scale tests⁽¹¹⁾ the titanate-loaded resin provided both excellent affinity and capacity for sorbing ^{90}Sr ; breakthrough of ^{90}Sr did not occur in either Run 1 or 2. Because it retains its anion exchange properties the titanate-loaded resin sorbed $^{99}\text{TcO}_4^-$ as well as ^{90}Sr , particularly in the early portion of Run 1. The capacity of the titanate-loaded resin for sorbing ^{90}Sr is much higher than that for ^{99}Tc . In Run 1 ^{99}Tc breakthrough occurred after passage of about 350 CV of feed while in Run 2 with only one unused bed of resin technetium broke through after passage of only about 182 CV of feed.

In addition to ^{90}Sr and ^{99}Tc , the only gamma emitting radionuclides present to any major extent in the feeds to the titanate beds were ^{106}Rh , ^{125}Sb , ^{155}Eu , and ^{60}Co . As noted in Table IV, none of these latter radionuclides was sorbed to any significant degree. Nonsorption of ^{106}Rh and ^{125}Sb is in agreement with other evidence which suggests ruthenium and antimony are present largely as neutral and/or anionic species in alkaline Hanford defense waste solutions. Abundant data are available to show that sodium titanate strongly sorbs radiocobalt and radioeuropium from synthetic salt cake solutions. Failure of the titanate-loaded resin to remove ^{60}Co and ^{155}Eu from ozonated Tank 102-AZ solution was unexpected.

Ozonated Tank 102-AZ solution contains about 0.002M NH_4^+ (Table III); ^{60}Co could be present in this solution as a non-sorbable amine complex. There is also evidence that ^{155}Eu and possibly other rare earth ions (e.g. ^{147}Pm and ^{151}Sm) in ozonated Tank 102-AZ solution were complexed, at least to some extent, by the residual organic material. Thus, simple reozonation of 1.5L of the solution used as feed in the second (Run 2) titanate sorption step for 6 hours, using the same conditions as in the original ozonation step, reduced the ^{155}Eu and ^{60}Co concentrations to $<2.9 \mu\text{Ci/L}$ and $9.4 \mu\text{Ci/L}$, respectively.

Titanate-loaded macroreticular strong base anion exchange resin is considered the reference sorbent for ^{90}Sr and other multivalent cationic radionuclides in the Hanford Radionuclide Removal process. This material is available in kilogram amounts from the Cerac Corporation, Milwaukee, Wisconsin. Kilogram amounts of pure sodium titanate powder (40-140 mesh) are also available from the Cerac Corporation; the hydraulic properties of this finely-divided powder are not, however, suited for large

scale column operation. Sandia Laboratory scientists, funded in part by Rockwell Hanford Operations, have negotiated with the Norton Co. to use their proprietary technology to produce consolidated forms of sodium titanate which have satisfactory mechanical and chemical properties. The Norton Co. has recently succeeded in making small amounts of extruded titanate cylinders which appear to meet all requirements.

TABLE IV. Sodium Titanate Removal of ^{90}Sr .

Sorption System: Three interconnected 18.7 mL (1.4 cm dia.) beds of 50-100 mesh sodium titanate loaded macroreticular (Dowex AG-MP-1) anion exchange resin.

Flow Conditions: 2 CV/hr; 25°C, downflow.

Feed ⁹⁰ Sr μCi/L	Cumulative CV	Effluent ^(c) , μCi/L			
		⁹⁰ Sr	⁹⁹ Tc	¹⁵⁵ Eu	⁶⁰ Co
<u>Run No. 1^(a)</u>					
1.49	347	0.77	0.62	5.9	21.0
37.1	651	0.42	3.9	6.1	24.0
56.2	970	0.65	4.1	6.0	21.0
150.0	1277	0.59	4.3	5.6	26.0
233.0	1478	<0.1	3.9	6.5	24.0
<u>Run No. 2^(b)</u>					
	283	1.49	3.0		
	477	1.48	4.5		
	205	1.20	4.1		
	298	1.41	4.1		
	983	1.34	4.2		

(a) Feed also contained about 620 $\mu\text{Ci/L}$ ^{106}Rh , 100 $\mu\text{Ci/L}$ ^{125}Sb , 6 $\mu\text{Ci/L}$ ^{155}Eu , 36 $\mu\text{Ci/L}$ ^{60}Co , and 4.5 $\mu\text{Ci/L}$ ^{99}Tc .

(b) Feed contained 120 $\mu\text{Ci/L}$ ^{90}Sr and approximately the same concentrations of ^{106}Rh , ^{125}Sb , ^{155}Eu , ^{60}Co , and ^{99}Tc as the feed in Run 1.

(c) Concentrations of ^{106}Rh and ^{125}Sb were about the same as in feed.

Technetium Removal

In both Runs 1 and 2 the bulk of the effluent from the three interconnected titanate-loaded resin beds contained 4-5 $\mu\text{Ci/L}$ ^{99}Tc . The technetium concentration of this solution was reduced to 0.039 $\mu\text{Ci/L}$ by countercurrent (12 stage mixer-settler) extraction with an equal flow of cyclohexanone. No gamma emitting radionuclides were present in the organic extract. Results of these tests were in excellent agreement with previous work⁽¹¹⁾.

Decontaminated Salt

The concentration of all the long-lived ($t_{1/2} > 10\text{y}$) radionuclides (for which analytical methods are in place) in the dried salts produced from the final decontaminated solutions in Runs 1 and 2 are listed in Table V. For both runs, the dried salts contained less than the working guideline of a total of 10 nCi/g of long-lived radionuclides. Methods for analyzing for several long-lived weak beta emitters (^{151}Sm , ^{93}Zr , ^{79}Se , ^{59}Ni , ^{63}Ni) which might have been present in the decontaminated salts were not available when this work was done. Except for ^{79}Se , the Hanford Radionuclide Removal process should provide adequate removal of all these long-lived radionuclides.

TABLE V. Long-Lived Radionuclide Content of Decontaminated Salt.

Isotope	Concentration, nCi/g	
	Run 1	Run 2
^{90}Sr	3.1	0.92
^{99}Tc	<0.2	<0.17
^{137}Cs	<0.85	0.058
^{152}Eu	<0.81	0.023
^{238}U	0.009	0.006
^{239}Pu	0.32	0.064
^{241}Am	0.81	0.43
	<6.1	<1.6

(a) Reozonated, see page

Conclusions - Future Work

Results of the hot cell runs reported here coupled with those of earlier bench-scale tests continue to demonstrate the viability and applicability of the Hanford Radionuclide Removal process. Laboratory work now underway will provide much needed further understanding and development of the headend ozonation step. Further hot cell load-elution cycle tests of the Duolite ARC-359 resin ^{137}Cs decontamination process are needed to determine and optimize performance of this step with ozonated waste solutions. Alternative cesium removal processes, e.g. dipicrylamine solvent extraction, also need to be studied. Pilot plant-scale tests of each process step with both simulated and actual waste solutions are also required.

Acknowledgements

We are indebted to Paul Hammitt, Kathy Moss, and Judy Watts for their expert performance of the experimental work.

REFERENCES

1. W. W. Schulz and M. J. Kupfer, "Solidification and Storage of Hanford's High-Level Radioactive Liquid Wastes," in High-Level Radioactive Waste Management, M. H. Campbell, ed., Advances in Chemistry Series 153, American Chemical Society, Washington, D. C., 1976.
2. D. E. Larson, "Radioactive Waste Management Program," ARH-2185, Atlantic Richfield Hanford Co., Richland, WA, 99352, January 1971.
3. ERDA-77-44, Energy Research and Development Administration Report, "Alternatives for Long-Term Management of Defense High-Level Radioactive Waste-Hanford Reservation," September 1977.
4. J. L. Swanson, "The Zirflex Process," in Process Chemistry, R. R. Bruce, J. M. Fletcher, and H. H. Hyman, eds., Progress in Nuclear Energy Series III, Pergamon Press, New York, Vol. 3, p 289. 1961.
5. D. D. Wodrich, "Hanford Long-Term High-level Waste Management Program Plan - Phase I: Technology Development," ARH-LD-143, Atlantic Richfield Hanford Co., Richland, WA., 99352, June 1976.
6. R. Y. Lyon, "Status of Technology for Long-Term Management of Hanford High-Level Waste," ARH-LD-143, Atlantic Richfield Hanford Co., Richland, WA, 99352, June 1976.
7. M. J. Kupfer and W. W. Schulz, "The Endothermic Process -- Application to Immobilization of Hanford In-Tank Solidified Waste," ARH-2800. Atlantic Richfield Hanford Co., Richland, WA, 99352, July 1973.
8. W. W. Schulz, A. L. Dressen, C. W. Hobbick, and M. J. Kupfer, "Conversion of Hanford Salt Cake to Glass: Laboratory Studies," ARH-ST-135, Atlantic Richfield Hanford Co., Richland, WA, 99352, May 1976.
9. M. J. Kupfer and W. W. Schulz, "Fixation of Hanford Sludge by Conversion to Glass," ARH-SA-285, Atlantic Richfield Hanford Co., Richland, WA, 99352, March 1977.

10. M. J. Kupfer, "Fixation of Hanford Waste Liquids by Conversion to Glass," RHO-SA-20, Rockwell Hanford Operations, Richland, WA, 99352, April 1978.
11. W. W. Schulz, "Removal of Radionuclides from Hanford Defense Waste Solutions," RHO-SA-51, Rockwell Hanford Operations, Richland, WA, 99352, January 1980.
12. Chem. Eng. News, 54 (No. 2), 1976.
13. M. M. Beary, "Preliminary Flowsheet for the Conversion of Hanford High-Level Waste to Glass," RHO-F-4, Rockwell Hanford Operations, Richland, WA, 99352, October 1978.
14. T. W. Lutton, D. M. Strachan, W. W. Schulz, and L. J. Bollyky, "Ozonation of Hanford Nuclear Defense Waste," Ozone: Sci. and Eng., 1, 133, 1979.
15. J. R. Wiley, "Decontamination of Savannah River Plant Waste Supernate," DP-1436, E. I. du Pont de Nemours & Co. Savannah River Laboratory, Aiken, SC, August 1976.
16. A. P. Hammitt and W. W. Schulz, "Hot Cell Facility and Equipment for Tests of the Hanford Radionuclide Removal Process," RHO-SA-52, Rockwell Hanford Operations, Richland, WA, September 1978.