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**PROCESS IMPROVEMENT STUDIES FOR THE SUBMERGED DEMINERALIZER SYSTEM (SDS)
AT THE THREE MILE ISLAND NUCLEAR POWER STATION, UNIT 2**

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PROCESS IMPROVEMENT STUDIES FOR THE SUBMERGED DEMINERALIZER SYSTEM (SDS)
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ABSTRACT

Tests were made to investigate flowsheet modifications which might improve the expected performance of the reference Submerged Demineralizer System (SDS) flowsheet for decontaminating the high-activity-level water at the Three Mile Island Nuclear Power Station, Unit 2. The tests included one series designed to show the effects of aging time, temperature, and pH on reduction of the concentrations of residual ^{137}Cs and ^{90}Sr , and a second series designed to evaluate the physical sorption of ^{125}Sb on silica gel or other inorganic sorbents. Results of the tests indicated that the most promising method for reducing ^{137}Cs and ^{90}Sr concentrations below 10^{-4} $\mu\text{Ci/mL}$ is to age the effluent water from the zeolite columns for at least 2 h at 75°C prior to its passage through another zeolite column. Sorption of the ^{125}Sb on silica gel or other inorganic sorbents did not show sufficient promise to be considered for practical use. A previously identified method for removal of ^{125}Sb requires deionization of the water by removal of the sodium on a cation exchange resin prior to sorption of ^{125}Sb on anion exchange resin; however, this method would generate a relatively large amount of low-activity-level solid waste (ion exchange resin).

1. INTRODUCTION

The reference Submerged Demineralizer System (SDS) flowsheet for decontamination of the high-activity-level water (HALW) at the Three Mile Island Nuclear Power Station, Unit 2 (TMI-2) was evaluated in 10^{-5} scale tests at the Oak Ridge National Laboratory (ORNL) during early 1980.¹ The tests confirmed that the SDS flowsheet would meet the design objective of decontaminating the HALW sufficiently that, when the effluent was mixed with the normal plant discharges, the concentrations of all radionuclides (except tritium) in the decontaminated water would be <10% of those listed in 10 CFR 20.2,3

The tests showed that the bulk of the cesium and strontium was effectively adsorbed on the zeolite sorbent but that the subsequent treatment with organic-based polishing resins would not provide additional decontamination from cesium and strontium or removal of the minor contaminants, ^{125}Sb and ^{106}Ru , which are anionic in nature.

Therefore, further tests were made at ORNL during November and December 1980 to study potential improvements to the original SDS process. One series of tests was designed to show the effects of aging of the zeolite-bed effluent solution at ambient conditions, at elevated temperature, and/or at reduced pH on subsequent sorption of the residual cesium and strontium on zeolite (Ionsiv IE-95*) or a strong-acid, organic-based cation exchange resin (Nalcite HCRS**). These are the sorbents that were specified for the SDS reference flowsheet. A second series of tests was made to determine whether ^{125}Sb could be removed effectively by reduction of the solution pH followed by sorption on one of several inorganic sorbents. Silica gel was of particular interest because of reports^{4,5} that it had proved effective for sorbing antimony from slightly acidic ($\leq 0.5 \text{ M}$) solutions.

2. REMOVAL OF RESIDUAL CESIUM AND STRONTIUM

The variables (and their parameters) investigated in this study are summarized in Table 1. The feed solution for the tests was TMI-2 Containment Building (CB) sump water which had been stored for several months following the SDS flowsheet evaluation tests in early 1980. The solids contained in the water had settled during that time; thus clarified water for the tests was easily obtained by decantation.

The procedure used for conducting the tests is outlined in Fig. 1. At each pH level, aging tests were performed both at ambient temperature ($\sim 23^\circ\text{C}$) and at elevated temperature ($\sim 75^\circ\text{C}$). The filtration test was carried out only at ambient temperature ($\sim 23^\circ\text{C}$).

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Table 1. Conditions of treatment for removal of residual cesium and strontium

Feed	Effluent from passing TMI-2 CB sump water through an 8-mL zeolite column; collection time, 1-3 h
Aging time, h	2; 24; 168
Temperature, °C	23; 75
pH	6; 8.5
Sorbent	Linde Ionsiv IE-95 zeolite; Nalcite HCRS (organic-based strong-acid cation resin)
Sorbent contact time, h	2

An ion exchange column containing 8 mL of Ionsiv IE-95 zeolite (Na^+ form) was used in a hot cell to prepare the feed for the aging tests. This column simulated the first zeolite column of the SDS reference flowsheet. Flow through the column was controlled at 40 mL/h to provide a 12-min residence time such as that intended for each zeolite column in the SDS reference flowsheet. The operating temperature was the cell ambient temperature (i.e., $\sim 35^\circ\text{C}$). A batch of 80 or 90 mL of effluent from the zeolite column (about ten bed volumes) was accumulated and then transferred to a laboratory hood, where it was divided into the portions necessary for subsequent tests. The zeolite column performance, summarized in Table 2, was similar to that observed in previous tests.

In one test, 5 mL of effluent sample IX7-1A from the zeolite column in the hot cell was filtered through a 0.1- μm Millipore filter; the concentrations of the radioactive contaminants remaining in the filtrate were then compared with those in the feed to the filter. The data (Table 3) show that the strontium and cesium concentrations were decreased by a factor of 8 and a factor of 2, respectively. No reduction was observed in the antimony and ruthenium concentrations. These results indicate, as did those of previous tests, that the strontium is associated with colloid-size particulate matter in the water to a greater extent than are the other contaminants.

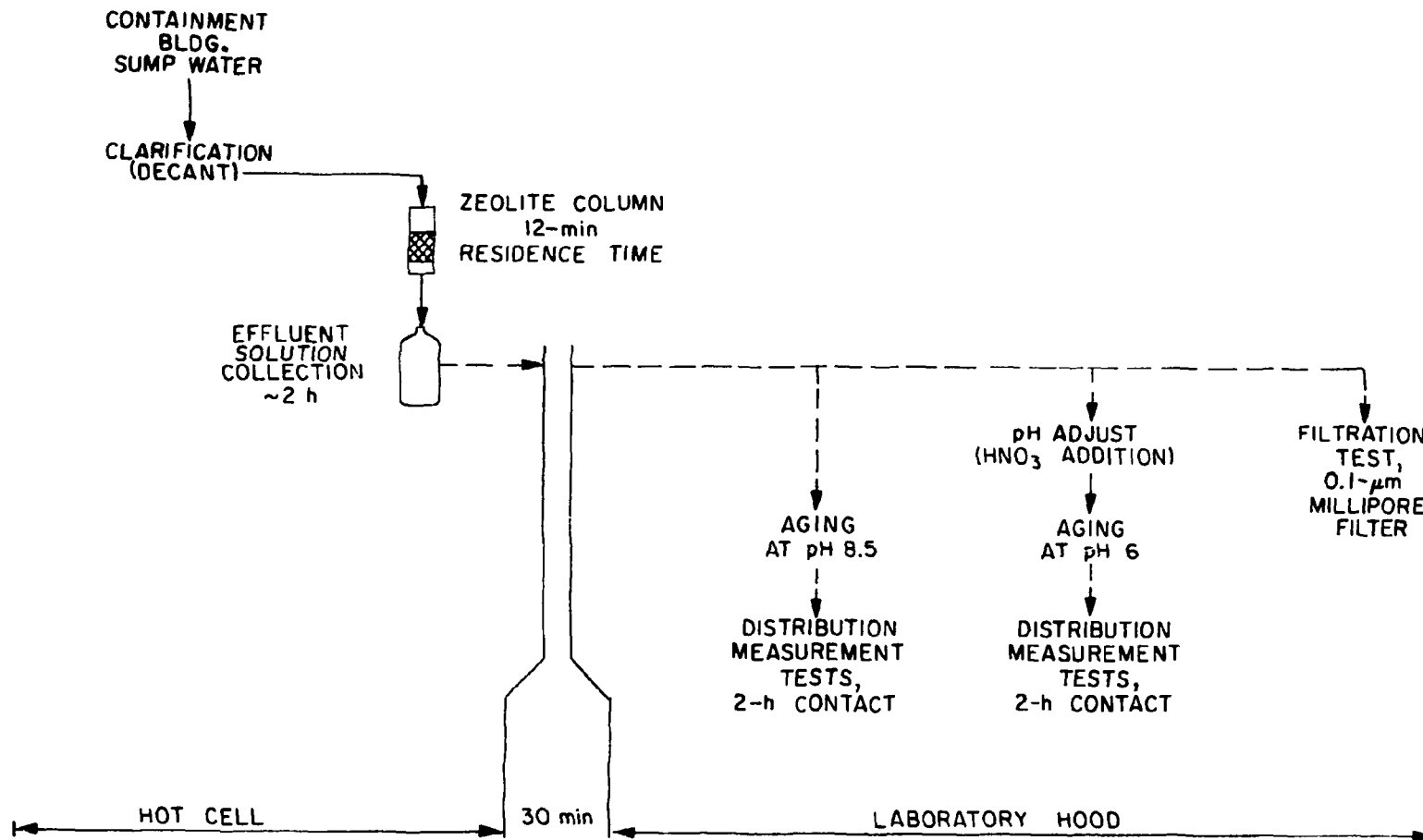


Fig. 1. Outline of residual cesium-strontium removal tests.

Table 2. Performance of the 8-mL zeolite column used in the hot cell to prepare feed for subsequent filtration and aging — distribution coefficient (K_d) measurement tests

		Bed volumes		
		1-11	12-22	23-32
Sample number	Feed	IX7-1	IX7-1A	IX8-1
pH	8.5	8.5	8.5	8.5
Gross γ , cpm/mL ^a	8.64E7	6.48E3	5.80E3	6.12E3
Gross β , cpm/mL ^a	3.94E7	1.18E4	7.32E3	Not measured
Key radionuclides, $\mu\text{Ci/mL}^a$				
⁹⁰ Sr	4.08	5.41E-4	7.03E-4	9.73E-4
¹⁰⁶ Ru	<1.8	4.14E-4	4.35E-4	3.95E-4
¹²⁵ Sb	<1.0	1.80E-2	1.81E-2	1.84E-2
¹³⁷ Cs	162	2.40E-3	1.32E-3	2.16E-3
Cumulative DFs				
⁹⁰ Sr		7.5E3	5.8E3	4.2E3
¹³⁷ Cs		6.8E4	1.2E5	7.5E4

^aConcentrations as of Dec. 1, 1980.

Table 3. Filtration of 5 mL of hot-cell zeolite column effluent at 23°C using a 0.1- μm Millipore filter

	Column effluent, IX7-1A	Filtrate	DF
pH	8.5	8.5	
Gross γ , cpm/mL ^a	5.80E3	6.12E3	—
Gross β , cpm/mL ^a	7.32E3	6.12E3	1.2
Key radionuclides, $\mu\text{Ci/mL}^a$			
⁹⁰ Sr	7.03E-4	8.92E-5	7.9
¹⁰⁶ Ru	4.35E-4	4.19E-4	1.0
¹²⁵ Sb	1.81E-2	2.18E-2	—
¹³⁷ Cs	1.32E-3	6.65E-4	2.0

^aConcentrations as of Dec. 1, 1980.

The aging-distribution measurement tests were made using the remainder of zeolite column effluent sample IX7-1A and all of sample IX8-1. Each of the samples was divided into two equal batches. One batch of each sample was used without treatment. The pH of the second batch was adjusted to 6.0 by the addition of dilute HNO_3 . The solutions were stored (aged) either at room temperature (23°C) or in a water bath at 75°C . After the desired aging period (2, 24, or 168 h), a 5-mL aliquot of the solution was added to 0.5 g of each sorbent (Ionsiv IE-95 and HCRS, both in the Na^+ form). The solution was added directly to the air-dried sorbent. The mixtures, at ambient temperature in 15-mL bottles, were agitated by rotating the bottles for 2 hr at 25 rpm. Each bottle was held by a support arm that located the bottle ~8 in. from the center of rotation. At the end of the 2-h period, the solutions contacted with zeolite were slightly hazy; therefore, they were lightly centrifuged prior to sampling. The larger HCRS resin settled rapidly, and centrifugation was not required before sampling.

The results of the aging-distribution measurement tests are tabulated in Tables A-1 through A-8 of the Appendix and are summarized in Fig. 2. This figure shows the concentrations of the residual ^{137}Cs and ^{90}Sr in the water after the distribution measurement tests as a function of aging time for each of the sorbents and for the various aging conditions. Concentrations, rather than distribution coefficients, were plotted to enable a direct comparison with the goal of " 10^{-4} $\mu\text{Ci/mL}$." These tests represent only one stage of contact with the sorbent, whereas more than one theoretical stage is obtained under normal conditions in an ion exchange column. In this case, however, the separation is believed to be limited by the rate of equilibration of the nonionic species with the ionic species instead of the number of stages of contact with the sorbent. Lower concentrations might be achievable by repeated sequences of aging followed by sorbent contact; however, this mode of operation was not investigated.

The minimum aging time evaluated, 2 h, might actually represent a period of 4 to 7 h in continuous operation. The conditions necessary to perform the tests included: (1) a period of 2.25 to 2.75 h to accumulate and transfer the column effluent solution and, in some cases, adjust the pH; (2) a 2-h aging period; and (3) a 2-h contact for the distribution measurement test.

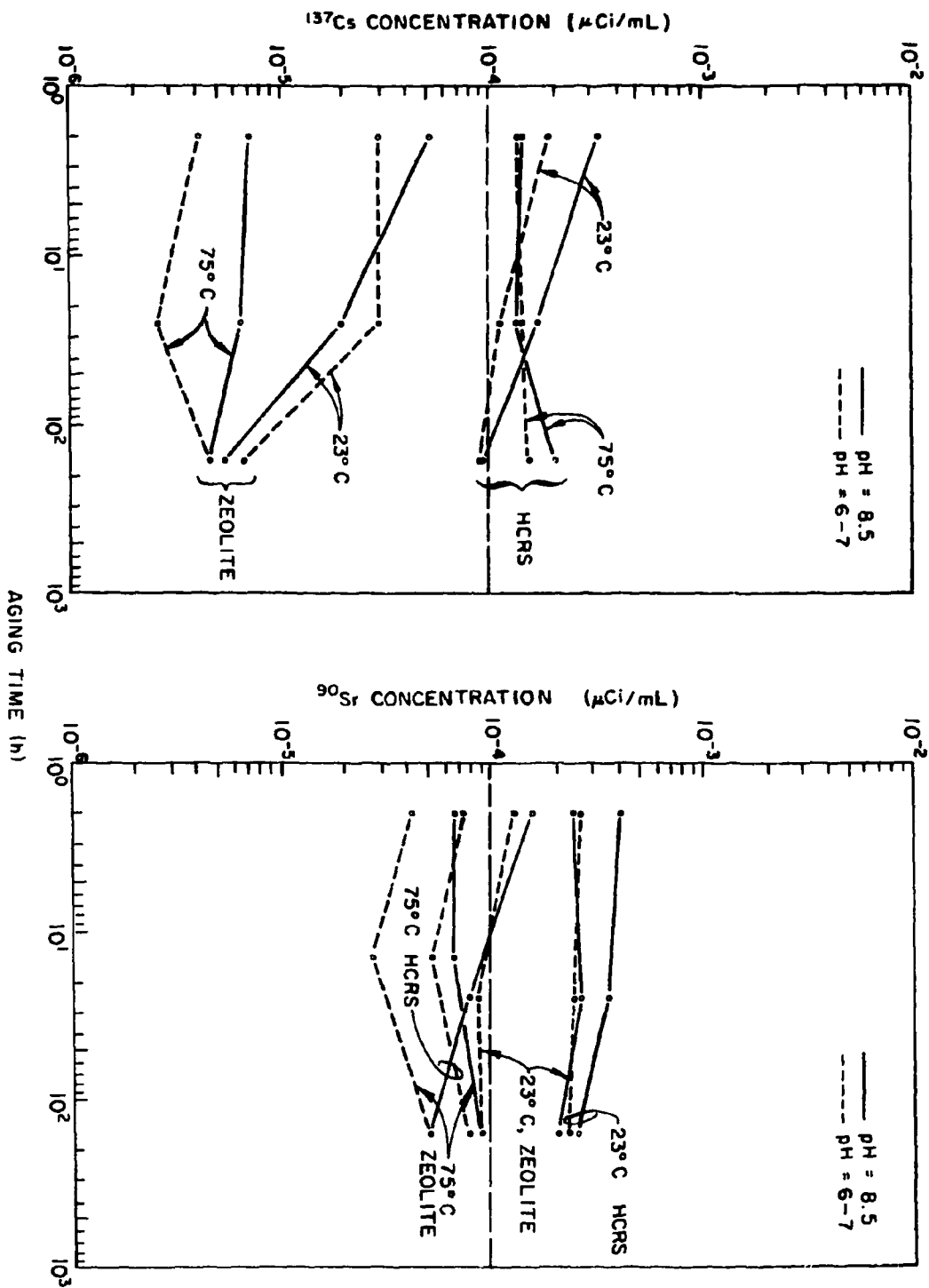


Fig. 2. Effects of aging time, temperature, and pH on subsequent sorption of cesium and strontium on zeolite or cation resin.

For cesium, the data shown in Fig. 2 indicate that increased aging times (1 or 7 d) were beneficial only at the lower aging temperature. For strontium, the longer aging time appeared to be beneficial only during the test in which the solution was aged at pH 8.5 and 75°C prior to contact with the HCRS resin.

The conclusions that can be drawn from these tests are as follows:

1. After aging, recontact was more effective with zeolite than with HCRS; the effects were greater for cesium than for strontium.
2. Aging at elevated temperature (75°C) resulted in more rapid and more effective removal of both cesium and strontium.
3. Aging at pH 6.0 gave better results for strontium than for cesium removal.
4. Reduction of the concentration of strontium to $<10^{-4}$ $\mu\text{Ci/mL}$ was more difficult than that for cesium. Adequate removal of strontium required aging at elevated temperature (75°C) or reduced pH (6.0) prior to sorption on the zeolite.

Although the test results suggest that aging at a reduced pH would be beneficial, particularly for subsequent sorption of strontium on zeolite, potential problems could be encountered if this method were attempted in plant-scale operations. Precise adjustment of the pH (by acid addition) to levels between 7.5 and 3.5 could be difficult, as indicated by the titration curve shown in Fig. 3, and degradation of the zeolite structure could occur if pH levels <4 were obtained.

In summary, the results of this series of tests indicate that the most promising of the methods tested for reducing the cesium and strontium concentrations below 10^{-4} $\mu\text{Ci/mL}$ is to age the zeolite column effluent for at least 2 h at $\sim 75^\circ\text{C}$ before passing it through another zeolite column.

3. REMOVAL OF ^{125}Sb

During the original SDS flowsheet evaluation tests,¹ ^{125}Sb was found to behave as an anionic species; however, it was poorly sorbed by the anion resin in the polishing column because of competition from the borate ion. Distribution tests made at that time indicated that the antimony can be sorbed effectively by anion resin after the water has been deionized by

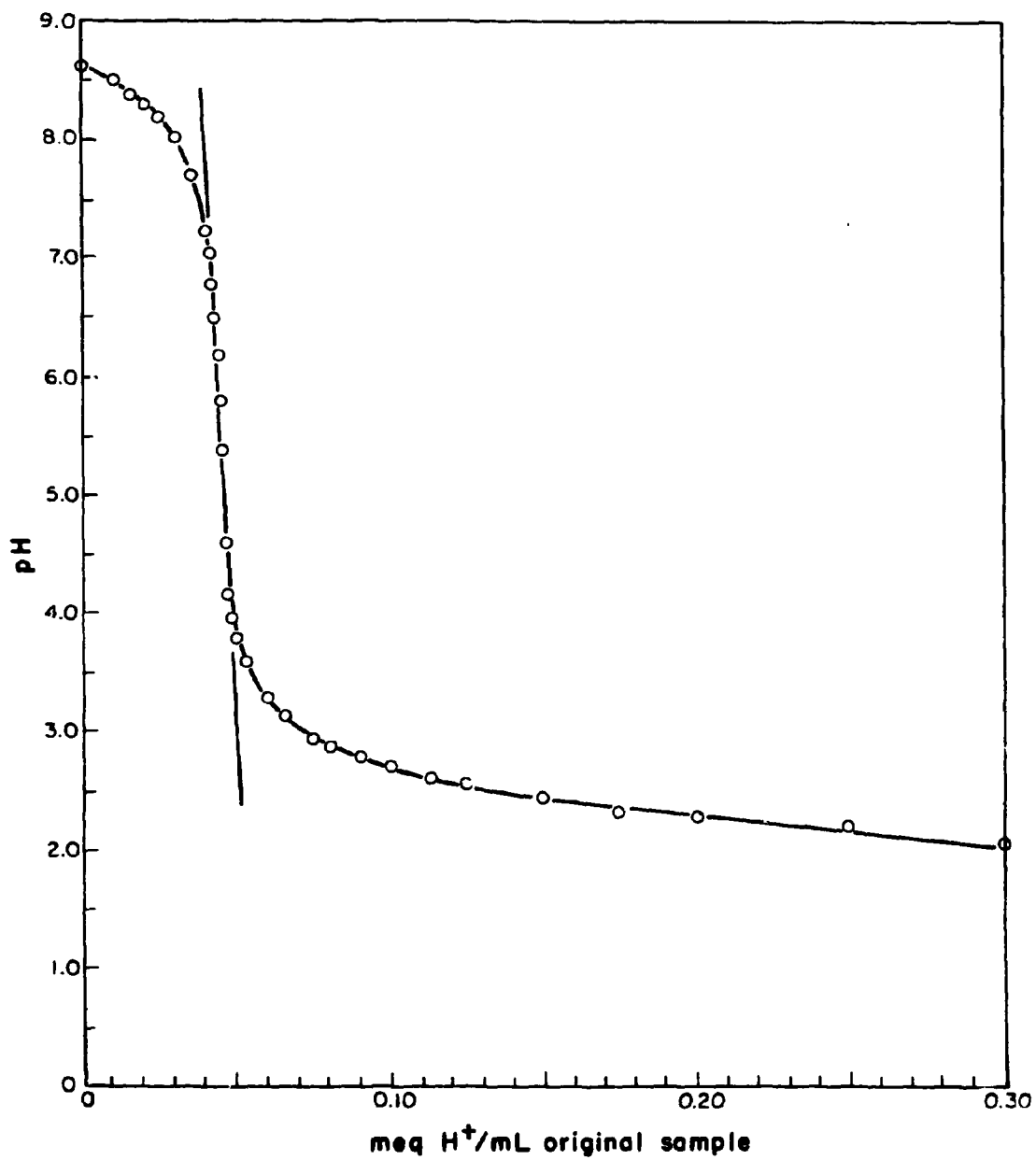


Fig. 3. Titration curve for zeolite column effluent (TMI-LX7-1A).

sorption of the Na^+ ion on H^+ -form cation exchange resin. This treatment converts the borate ion to weakly ionized boric acid, which does not compete with antimony and ruthenium for exchange sites on the anion resin. The penalty for including such a step would be the generation of a relatively large volume of radioactive waste. In comparison to the $\sim 14 \text{ m}^3$ (~ 3600 gal) of zeolite expected to be used for sorption of the bulk radioactive contaminants, cesium and strontium (based on the reference flowsheet loading to 200 bed volumes), $\sim 76 \text{ m}^3$ ($\sim 20,000$ gal) of HCRS ion exchange resin would be required for removal of the sodium. This latter volume would, however, be low-activity-level waste.

A possible alternative to the use of cation exchange resin was also considered during the original evaluation tests. This approach involved neutralization of the sodium by the addition of an acid prior to sorption of the antimony on an anion resin. The acid was required to have an anion that would be less competitive for the sorption of antimony than the borate ion. Tests were made with formic, oxalic, and aminoacetic acids, but none of them improved the subsequent sorption of antimony by the anion resin.¹

After the original evaluation tests had been concluded, another relatively simple method for antimony removal that might be useful was identified in the literature.^{4,5} The basic steps consist of acidifying the water (to $< 0.5 \text{ M H}^+$) by the addition of nitric acid and then adsorbing the antimony onto a bed of silica gel. A series of tests, which are outlined in Table 4, was made to evaluate this method for adsorption of the antimony on silica gel, as well as other inorganic sorbents.

The feedwater used in this series of tests was a composite of effluent solutions from the earlier SDS flowsheet tests;¹ it had a pH of 8.1 and a ^{125}Sb concentration of $1 \times 10^{-2} \text{ } \mu\text{Ci/mL}$. During each test, the pH was adjusted to the desired level by addition of nitric acid; then 20 mL of the acidified feedwater was contacted with 0.2 g of the particular sorbent for either 15 or 60 min by slowly stirring the water-resin mixture in a beaker. Throughout the contact period, acid was added as required to maintain the pH at the desired level.

The results of this series of tests are presented in Tables A-9 and A-10 of the Appendix and are shown graphically in Figs. 4 and 5. For these tests, we defined an apparent distribution coefficient, K'_d , which

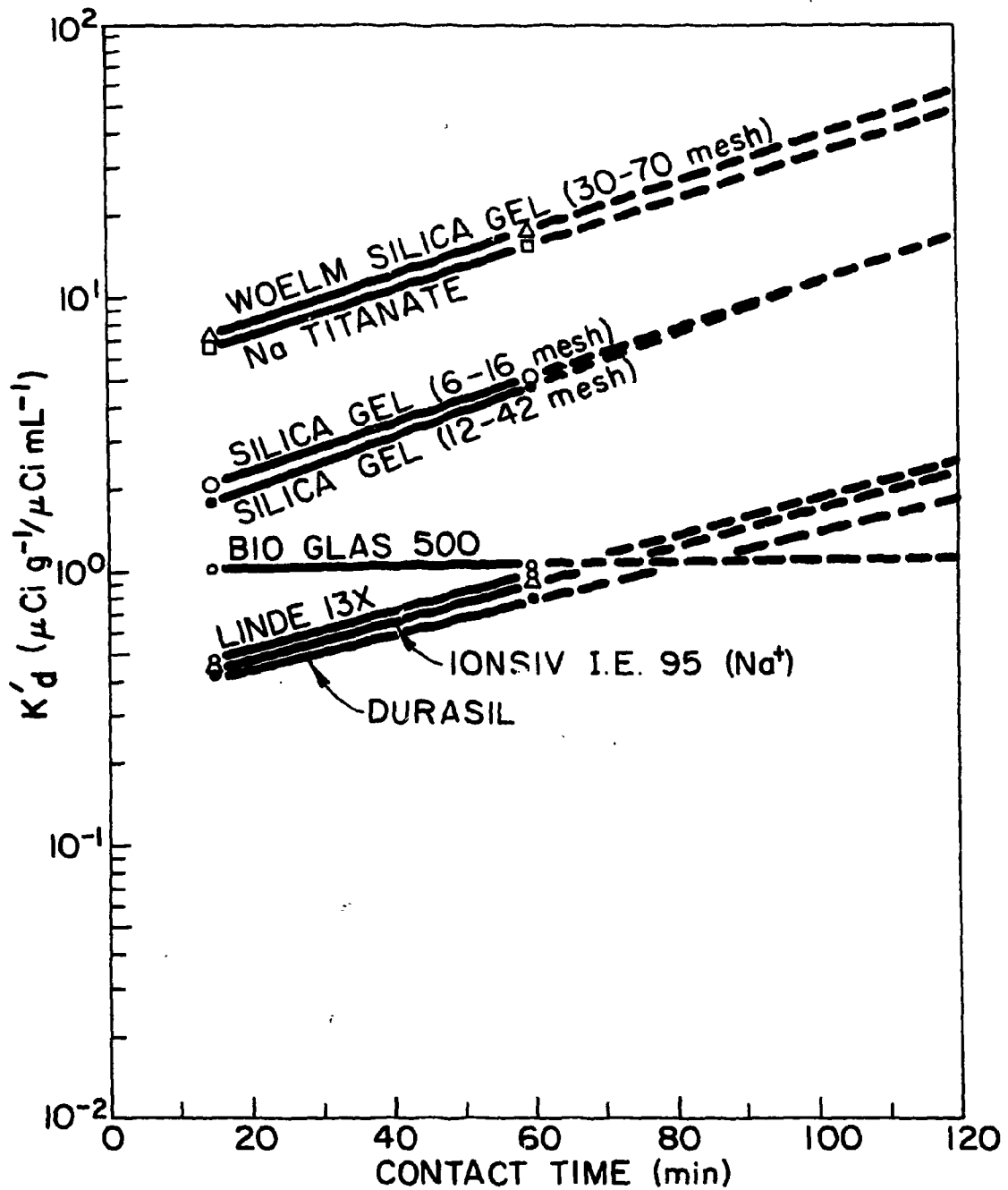


Fig. 4. Apparent distribution coefficient, K'_d , for ^{125}Sb as a function of time for various sorbents (pH = 6.0).

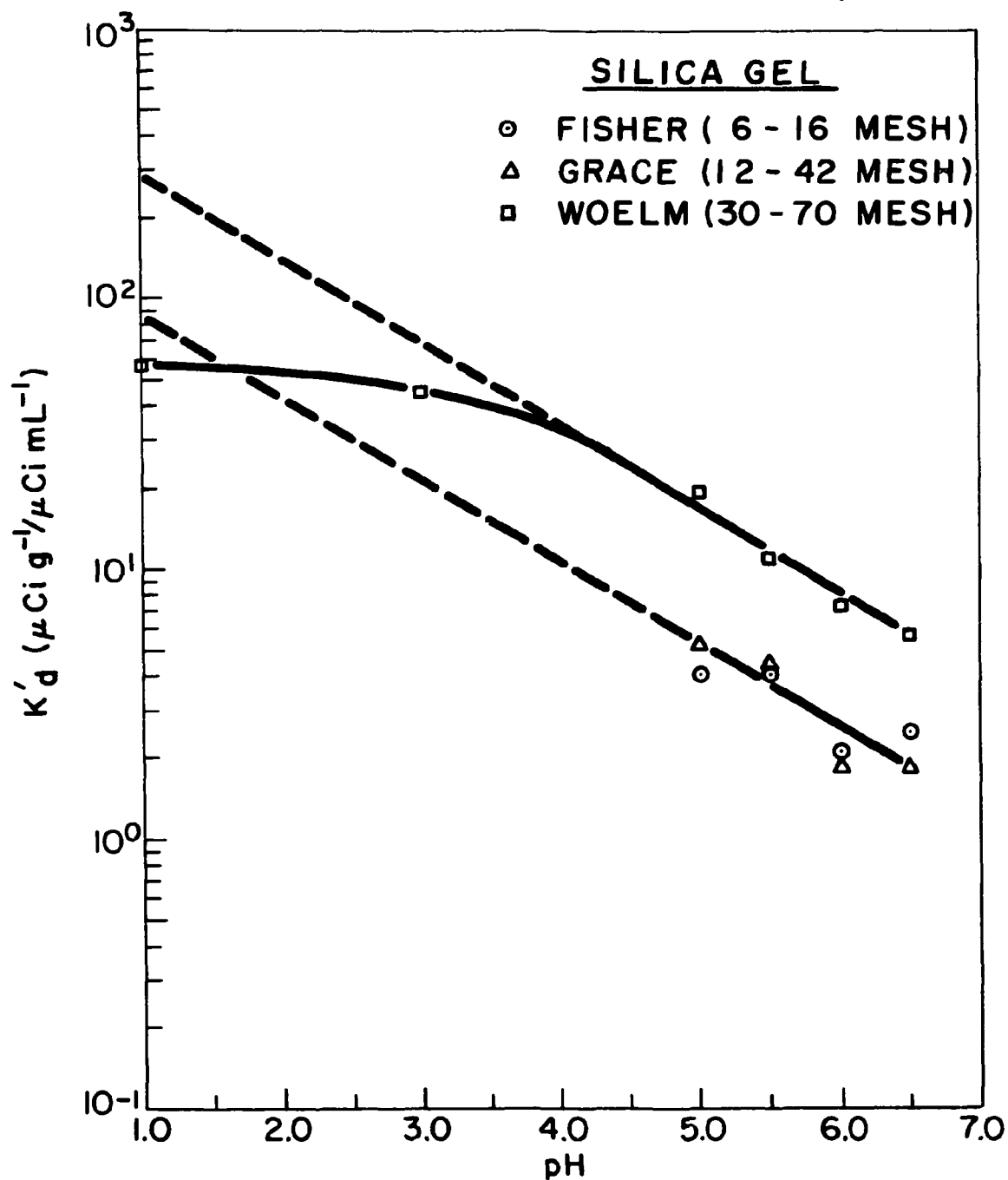


Fig. 5. Apparent distribution coefficient, K'_d , for ^{125}Sb as a function of pH for various sorbents (contact time = 15 min).

Table 4. Conditions for ^{125}Sb distribution measurement tests

Feed	Composite effluent water (pH 8.1) from SDS flowsheet tests
Temperature, °C	25-30
pH	Adjusted to 5.0, 5.5, 6.0, or 6.5 (via HNO_3 addition) ^a
Contact time, min	15; 60
Sorbents:	Woelm silica gel (30-70 mesh); W. R. Grace silica gel, grade 35 (12-42 mesh); Fisher Scientific silica gel, grade 05 (6-16 mesh); IE-95 zeolite (Na^+ form); Linde 13X molecular sieve; sodium titanate (pellets); Durasil sorbent glass; Bio-Glas-500 sorbent glass

^aTests using Woelm silica gel were also made at pH 1.0 and at pH 3.0.

represents the ratio of the concentration of the element in the sorbent ($\mu\text{Ci/g}$) to the residual concentration in the solution ($\mu\text{Ci/mL}$) following the indicated contact time. Woelm silica gel and sodium titanate (pellets) were superior to the other sorbents tested; however, even they did not appear adequate for practical use. Extrapolation of the original data points of Fig. 5, which were obtained at pH levels of 5.0 to 6.5, indicated that adequate values of K'_d ($>10^2$) might be obtained at pH levels of 1 to 2. Additional tests were made at pH levels of 1 and 3, but the maximum K'_d was only ~ 50 — a value not high enough for practical use.

4. CONCLUSIONS

In two series of SDS process improvement tests, we have identified modifications to the reference SDS flowsheet that would enable further reduction of the expected effluent concentrations. The most promising method tested for reducing the cesium and strontium concentrations below

10^{-4} $\mu\text{Ci/mL}$ is to age the zeolite column effluent for at least 2 h at 75°C before passing it through another zeolite column. No improved method was found for removing the anionic contaminants, ^{125}Sb and ^{106}Ru . The only satisfactory method, as identified in the previous tests,¹ would require deionization of the water by removal of sodium on a cation exchange resin prior to sorption of the ^{125}Sb and ^{106}Ru on anion exchange resin. Such treatment may be expected to generate a relatively large amount of low-activity-level solid waste (ion exchange resin).

5. REFERENCES

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4. E. Akatsu, T. Tomizawa, and Y. Aratono, "Separation of Antimony-125 in Fission Products," J. Nucl. Sci. Technol. II(12), 571-74 (1974).
5. H. Eschrich, M. Herrera-Huertas, and K. Tallberg, Sorption of Antimony by Silica Gel from Nitric Acid and Hydrochloric Acid Solutions, Eurochemic Technical Report No. 268 (October 1970).

Table A-1. Effect of aging of zeolite column effluent at ambient temperature and existing pH on further sorption^a by zeolite

Test ZA							
Sorbent	IE-95 zeolite						
Initial form	Na ⁺						
Weight, g	0.5						
Feed source	Sample IX7-1A						
pH adjustment	None						
Volume, mL	5.0						
Aging temperature, °C	23						
Aging time, h ^b	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	7.03E-4	2.24E-4	2.27E-4	2.00E-4	21	21	25
¹⁰⁶ Ru	4.35E-4	4.08E-4	3.57E-4	3.54E-4	0.7	2.2	2.3
¹²⁵ Sb	1.81E-2	1.63E-2	1.78E-2	1.75E-2	1.1	0.2	0.3
¹³⁷ Cs	1.32E-3	5.14E-5	2.97E-5	5.68E-6	247	434	2310
pH	8.5	8.5	8.5	8.5			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.25-h period required to collect effluent from the zeolite column.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-2. Effect of aging of zeolite column effluent at ambient temperature and reduced pH on further sorption^a by zeolite

Test ZC							
Sorbent	IE-95 zeolite						
Initial form	Na ⁺						
Weight, g	0.5						
Feed source	Sample IX7-1A						
pH adjustment	To 6.0 with HNO ₃						
Volume, mL	5.0						
Aging temperature, °C	23						
	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
Aging time, h ^b		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	7.03E-4	1.27E-4	8.11E-5	8.38E-5	45	77	74
¹⁰⁶ Ru	4.35E-4	3.54E-4	5.30E-4	3.57E-4	2.3	-	2.2
¹²⁵ Sb	1.81E-2	1.69E-2	1.55E-2	1.53E-2	0.7	1.7	1.8
¹³⁷ Cs	1.32E-3	2.97E-5	2.97E-5	6.76E-6	434	430	1940
pH	6.0	7.2	7.2	7.2			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.75-h period required to collect effluent from the zeolite column.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-3. Effect of aging of zeolite column effluent at ambient temperature and existing pH on further sorption^a by cation resin

		Test RA					
Sorbent		HCRS					
Initial form		Na ⁺					
Weight, g		0.5					
Feed source		Sample IX7-1A					
pH adjustment		None					
Volume, mL		5.0					
Aging temperature, °C		23					
Aging time, h ^b	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	7.03E-4	4.05E-4	3.51E-4	2.54E-4	7.3	10	18
¹⁰⁶ Ru	4.35E-4	1.38E-4	4.35E-4	4.05E-4	22	-	0.7
¹²⁵ Sb	1.81E-2	1.85E-2	1.85E-2	1.88E-2	-	-	-
¹³⁷ Cs	1.32E-3	3.24E-4	1.64E-4	9.14E-5	31	70	134
pH	8.5	8.5	8.5	8.5			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.25-h period required to collect effluent from the zeolite column.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-4. Effect of aging of zeolite column effluent at ambient temperature and reduced pH on further sorption^a by cation resin

				Test RC			
Sorbent		HCRS					
Initial form		Na ⁺					
Weight, g		0.5					
Feed source		Sample IX7-1A					
pH adjustment		To 6.0 with HNO ₃					
Volume, mL		5.0					
Aging temperature, °C		23					
	Feed (μCi/mL)	Supernates (μCi/mL)			K' _d ^c		
Aging time, h ^b		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	7.03E-4	2.70E-4	2.38E-4	2.38E-4	16	20	20
¹⁰⁶ Ru	4.35E-4	1.49E-4	1.11E-4	3.73E-4	19	29	1.7
¹²⁵ Sb	1.81E-2	1.68E-2	1.81E-2	1.82E-2	0.8	-	-
¹³⁷ Cs	1.32E-3	1.88E-4	1.13E-4	8.78E-5	60	107	140
pH	6.0	6.7	6.7	6.8			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.75-h period required to collect effluent from the zeolite column and adjust its pH.

^cK'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-5. Effect of aging of zeolite column effluent at elevated temperature and existing pH on further sorption^a by zeolite

Test ZB							
Sorbent	IE-95 zeolite						
Initial form	Na ⁺						
Weight, g	0.5						
Feed source	Sample IX8-1						
pH adjustment	None						
Volume, mL	5.0						
Aging temperature, °C	75						
	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
Aging time, h ^b		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	9.73E-4	6.49E-5	6.22E-5	8.92E-5	140	147	99
¹⁰⁶ Ru	3.95E-4	3.89E-4	3.43E-4	3.95E-4	0.2	1.5	-
¹²⁵ Sb	1.64E-2	1.74E-2	1.77E-2	1.89E-2	-	-	-
¹³⁷ Cs	2.16E-3	7.03E-6	6.22E-6	5.14E-6	3060	3460	4200
pH	8.5	8.5	8.5	8.5			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.5-h period required to collect effluent from the zeolite column and heat it to 75°C.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-6. Effect of aging of zeolite column effluent at elevated temperature and reduced pH on further sorption^a by zeolite

Test ZD							
Sorbent	IE-95 zeolite						
Initial form	Na ⁺						
Weight, g	0.5						
Feed source	Sample IX8-1						
pH adjustment	To 6.0 with HNO ₃						
Volume, mL	5.0						
Aging temperature, °C	75						
	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
Aging time, h ^b		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	9.73E-4	4.05E-5	2.57E-5	4.86E-5	230	370	190
¹⁰⁶ Ru	3.95E-4	3.65E-4	3.41E-4	4.54E-4	0.8	1.6	-
¹²⁵ Sb	1.64E-2	1.62E-2	1.66E-2	2.06E-2	0.1	-	-
¹³⁷ Cs	2.16E-3	4.05E-6	2.70E-6	5.14E-6	5320	7980	4200
pH	6.0	7.0	7.0	7.0			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.75-h period required to collect effluent from the zeolite column, adjust the pH, and heat the solution to 75°C.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-7. Effect of aging of zeolite column effluent at elevated temperature and existing pH on further sorption^a by cation resin

Test RB							
Sorbent	HCRS						
Initial form	Na ⁺						
Weight, g	0.5						
Feed source	Sample IX8-1						
pH adjustment	None						
Volume, mL	5.0						
Aging temperature, °C	75						
	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
Aging time, h ^b		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	9.73E-4	1.51E-4	7.57E-5	6.49E-5	54	119	140
¹⁰⁶ Ru	3.95E-4	4.16E-4	4.32E-4	4.89E-4	-	-	-
¹²⁵ Sb	1.64E-2	1.82E-2	1.72E-2	2.15E-2	-	-	-
¹³⁷ Cs	2.16E-3	1.36E-4	1.30E-4	2.07E-4	149	156	94
pH	8.5	8.6	8.6	8.6			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.5-h period required to collect effluent from the zeolite column and heat it to 75°C.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-8. Effect of aging of zeolite column effluent at elevated temperature and reduced pH on further sorption^a by cation exchange

		Test RD					
Sorbent		HCRS					
Initial form		Na ⁺					
Weight, g		0.5					
Feed source		Sample IX8-1					
pH adjustment		To 6.0 with HNO ₃					
Volume, mL		5.0					
Aging temperature, °C		75					
Aging time, h ^b	Feed ($\mu\text{Ci/mL}$)	Supernates ($\mu\text{Ci/mL}$)			K'_d ^c		
		<u>2</u>	<u>24</u>	<u>168</u>	<u>2</u>	<u>24</u>	<u>168</u>
Key radionuclides							
⁹⁰ Sr	9.73E-4	7.03E-5	5.14E-5	7.57E-5	128	179	119
¹⁰⁶ Ru	3.95E-4	3.92E-4	4.00E-4	4.49E-4	0.1	-	-
¹²⁵ Sb	1.64E-2	1.75E-2	1.80E-2	1.92E-2	-	-	-
¹³⁷ Cs	2.16E-3	1.33E-4	1.41E-4	1.51E-4	153	143	133
pH	6.0	6.8	6.8	6.9			

^a2-h contact period.

^bDoes not include the 2-h contact and the 2.75-h period required to collect effluent from the zeolite column, adjust the pH, and heat the solution to 75°C.

^c K'_d is the apparent distribution coefficient following the 2-h contact period.

Table A-9. Results of ^{125}Sb sorption tests^a using various sorbents

Sorbent	Contact time (h)	^{125}Sb concentration		K'_d ^b
		Feed solution ($\mu\text{Ci/mL}$)	After equilibration Solution ($\mu\text{Ci/mL}$) Sorbent ($\mu\text{Ci/g}$)	
Silica gel (Woelm, 30-70 mesh)	0.25	1.11E-2	1.04E-2	7.3
	1.0	1.11E-2	9.59E-3	18
Silica gel (W. R. Grace, grade 35, 12-42 mesh)	0.25	1.11E-2	1.12E-2	1.8
	1.0	1.11E-2	1.11E-2	4.8
Silica gel (Fisher Scientific, grade 05, 6-16 mesh)	0.25	1.11E-2	9.89E-3	2.1
	1.0	1.11E-2	1.07E-2	5.2
Ionsiv IE-95 (Na^+ form)	0.25	1.11E-2	1.11E-2	0.4
	1.0	1.11E-2	1.14E-2	0.9
Linde 13X	0.25	1.11E-2	1.11E-2	0.5
	1.0	1.11E-2	1.12E-2	0.9
Sodium titanate	0.25	1.11E-2	1.05E-2	6.8
	1.0	1.11E-2	9.92E-3	16
Durasil glass	0.25	1.11E-2	1.11E-2	0.4
	1.0	1.11E-2	1.11E-2	0.8
Bio-Glas-500	0.25	1.11E-2	1.11E-2	1.1
	1.0	1.11E-2	1.11E-2	1.1

^aPrior to each test, the solution pH was reduced to 6.0 via addition of nitric acid. During each test, 20 mL of feed solution was mixed with 0.2 g of the sorbent at room temperature ($\sim 23^\circ\text{C}$) for the desired contact time. During the contact, the pH was maintained at 6.0 by addition of nitric acid.

^b K'_d is the apparent distribution coefficient for the indicated contact time.

Table A.10. Results of ^{125}Sb sorption tests^a at various pH levels

Sorbent	pH	^{125}Sb concentration			K'_d ^b
		Feed solution ($\mu\text{Ci/mL}$)	Solution ($\mu\text{Ci/mL}$)	Sorbent ($\mu\text{Ci/g}$)	
Silica gel (Woelm) 30-70 mesh	6.5	1.11E-2	1.05E-2	6.16E-2	5.9
	6.0	1.11E-2	1.04E-2	7.55E-2	7.3
	5.5	1.11E-2	1.00E-2	1.09E-1	10.9
	5.0	1.11E-2	9.16E-3	1.76E-1	19.2
	3.0	1.11E-2	7.35E-3	3.34E-1	45.4
	1.0	1.11E-2	6.62E-3	3.74E-1	56.5
Silica gel (W. R. Grace, grade 35, 12-42 mesh)	6.5	1.11E-2	1.10E-2	2.01E-2	1.8
	6.0	1.11E-2	1.12E-2	2.02E-2	1.8
	5.5	1.11E-2	1.07E-2	4.69E-2	4.4
	5.0	1.11E-2	1.08E-2	5.64E-2	5.2
Silica gel (Fisher Scientific, grade 05, 6-16 mesh)	6.5	1.11E-2	1.09E-2	2.69E-2	2.5
	6.0	1.11E-2	9.89E-3	2.06E-2	2.1
	5.5	1.11E-2	1.08E-2	4.45E-2	4.1
	5.0	1.11E-2	1.07E-2	4.28E-2	4.0

^aFor each test, 20 mL of feed solution was mixed with 0.2 g of the sorbent at room temperature (~23°C) for 15 min. During each test, the desired pH was maintained by addition of nitric acid.

^b K'_d is the apparent distribution coefficient for the indicated contact time.