

Chemical Technology Division

DEVELOPMENT OF A PROCESS FLOWSHEET FOR THE ELUTION OF RADIOCESIUM FROM
THE TMI-2 MAKEUP AND PURIFICATION DEMINERALIZERS

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CONTENTS

	<u>Page</u>
ABSTRACT	1
1. INTRODUCTION	2
2. LABGRATORY DEVELOPMENT STUDIES	7
2.1 DESCRIPTION AND ANALYSIS OF DEMINERALIZER SAMPLES	8
2.2 RESIN ELUTION	9
2.3 DECONTAMINATION FACTORS OF ^{137}Cs AND ^{90}Sr IN ZEOLITE BED TESTS	18
2.4 SORPTION OF SOLUBLE ORGANIC COMPOUNDS IN ZEOLITE AND IN CHARCOAL BED TESTS	22
2.5 LIQUID CLARIFICATION	29
3. PROCESS FLOWSHEET FOR DEMINERALIZER ELUTION	33
4. CONCLUSIONS AND SUMMARY	35
5. ACKNOWLEDGMENTS	36
6. REFERENCES	37
7. APPENDIXES	39

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ABSTRACT

A process was developed for removing radiocesium from the two makeup and purification demineralizers at TMI-2 which had been highly contaminated as a result of the accident in 1979. Process conditions were established in hot-cell experiments using relatively small samples of the contaminated and degraded resins from the TMI-2 demineralizers.

The process was designed to elute the radiocesium from the demineralizer resins and then resorb it onto the zeolite ion exchangers contained in the Submerged Demineralizer System (SDS). It was also required to limit the maximum cesium activities in the resin eluates (SDS feeds) so that the radiation field surrounding the pipelines would not be excessive. The process consisted of 17 stages of batch elution. In the initial stage, the resin was contacted with 0.18 M boric acid. Subsequent stages subjected the resin to increasing concentrations of sodium in $\text{NaH}_2\text{BO}_3\text{-H}_3\text{BO}_3$ solution (total boron = 0.35 M; and 1 M sodium hydroxide in the final stages).

Simulated SDS tests with small zeolite beds (2 mL) showed that the eluates were compatible with SDS processing. Cesium was effectively sorbed (99.99%), and bed performance was unaffected by the presence of relatively small quantities of soluble organic resin degradation products (100 to 200 ppm total C) in the eluates. Attempts to remove the soluble organic compounds by sorption on charcoal were unsuccessful. Clarification tests on the eluates showed that the small quantity of suspended particulates in the eluates could be effectively removed by gravity settling and then filtration through a filter disk (stainless steel) having a 10- μm rating. The final filtration polish of the eluates avoids the unnecessary introduction of fine particulate matter to the SDS and subsequent water management systems. However, it was also shown that unfiltered eluates could be processed in simulated SDS tests without any significant effect on performance. Also included in this report are radionuclide, elemental, and fissile isotopic analyses of the demineralizer samples, which may be of interest to investigators concerned with the phenomenological aspects of the TMI-2 accident.

1. INTRODUCTION

The primary goals of this investigation were to develop and to test an elution process flowsheet for the removal of the cesium activity from the two makeup and purification demineralizers (DA and DB) at the Three Mile Island Nuclear Power Station, Unit 2 (TMI-2). Process developmental studies were conducted using rather small samples (up to 40 mL) of the demineralizer resins. An additional objective in this investigation was to provide certain analyses on the demineralizer, samples that are of interest to other studies that are primarily concerned with the phenomenological aspects of the TMI-2 accident on March 28, 1979. Quantitative analyses were performed to determine the radionuclides and chemical contaminants in the resin and liquid phases of the demineralizers samples. Physical damage to the resin particles as a result of the accident was also qualitatively assessed by visual microscopy.

As a consequence of the accident at TMI-2, the two demineralizers in the water makeup and purification system were severely contaminated with fission product radionuclides. The resin beds in the demineralizers were significantly degraded both radiolytically and thermally by the decay of most of the radionuclides during the time period since the accident. The principal gamma-emitting radionuclides remaining on the resin beds when demineralizer cleanup activities began were the relatively long-lived ^{137}Cs ($t_{1/2} = 30.1$ y) and ^{134}Cs ($t_{1/2} = 2.06$ y). Prior to the inception of the present investigation, nondestructive assay (NDA) methods had been employed to estimate the quantity of cesium activity and material content of each demineralizer (Table 1). Comparison of the postaccident resin-bed volumes with that of preaccident volumes showed that severe shrinkage (~55%) of the resin beds had occurred and indicated that the beds had been significantly degraded. It was known that the beds had not only been subjected to high radiation dosages ($\sim 10^9$ rads) but also to high temperatures because of the radioactive decay heat. The necessity to isolate the demineralizers from the liquid flow at about 19 h after the accident prevented effective removal of the decay heat, and estimates¹ indicate that center-line bed temperatures may have been as high as about 540°C (1000°F). The demineralizers were sampled by GPU Nuclear personnel in early 1983, and it

Table 1. Estimated demineralizer vessel loadings based on
NDA characterizations

Loadings	Preaccident ^a	Postaccident	
		DA	DB
Resin			
Volume, ft ³	50	22	22
Weight, lb	2139	1025	1025
¹³⁷ Cs, Ci	0	3500	7000
¹³⁴ Cs, Ci	0	270	540
Liquid			
Volume, ft ³	44	3	3
Weight, lb	2746	193	193
Debris			
U, lb		5	1
Core debris, lb		95	19
¹³⁷ Cs, Ci		177	35
¹³⁴ Cs, Ci		16	3
¹⁰⁶ Ru, Ci		21	4
¹⁴⁴ Ce, Ci		28	5
¹²⁵ Sb, Ci		116	23
TRU, Ci		0.5 ^b	0.1 ^b

^aDA and DB vessel loadings are identical.

^bAlpha activity only.

Source: J. D. Thompson and T. R. Oseterhoudt, TMI-2 Purification
Demineralizer Resin Study, GEND-INF-013, May 1984.

was observed that the DA vessel contained only dry, caked, resin, whereas liquid was still present in DB. However, the caked bed was apparently deagglomerated after water addition and sparging so that resin samples were obtained in a later sampling effort. The absence of liquid in the DA vessel is contrary to the NDA estimate of 3 ft³.

Conceptual studies of the various alternative methods for cleanup of the demineralizers¹ indicated that the most desirable method was to elute the cesium and subsequently sorb it on the zeolites in the Submerged Demineralizer System (SDS).²⁻⁶ This concept alleviated the high-level radiation problems associated with the eventual removal of the degraded resin beds and with the management of the resin wastes. Since the effects

of the degradation of the resin bed were unknown with regard to the quantitative elution behavior of cesium and the quality of the eluates, this concept required experimental determination of its feasibility and the development of a satisfactory chemical flowsheet.

A program was established in early 1983 involving the collaborative efforts of the Oak Ridge National Laboratory (ORNL), General Public Utilities Nuclear Corporation (GPU Nuclear Corporation), and EG&G/Idaho, Inc./TMI (EG&G) to: (1) establish the technical feasibility of cesium elution, (2) develop a chemical flowsheet that met processing requirements for the SDS at TMI-2, and (3) clean up the demineralizers. ORNL was responsible for establishing the feasibility of cesium elution, for testing, and (where necessary) for modifying the proposed flowsheet,¹ using demineralizer samples provided by GPU Nuclear. GPU Nuclear, with some assistance from EG&G, was responsible for the installation of the process at TMI-2 and the subsequent cleanup of the demineralizers. It was realized that essentially complete elution of cesium from the degraded resins could not be effected. The goal in elution was to reduce the cesium activity to values that significantly alleviated the high-level radiation problems in the eventual removal of the resins from the demineralizers and subsequent management of the resin wastes.

Several requirements had to be met in the development of a satisfactory process flowsheet for the elution and fixation of cesium. In elution, it was necessary that chemical reagents be employed that were compatible with ionic solution chemistry of SDS feed solutions. This requirement dictated that cesium elution be accomplished by displacing the cesium with sodium ions using sodium borate or sodium hydroxide solutions. The elution process was also required to limit the cesium activity in eluates to levels that were no greater than about 1 mCi/mL to avoid excessive radiation fields in subsequent SDS operations. At the 1-mCi/mL concentration level, the eluates could still be conveniently diluted to levels (~50 μ Ci/mL) that do not produce an excessive radiation field around the pipelines to SDS. Even with compatible ionic solution chemistry, it was by no means certain that eluates could be satisfactorily processed in the SDS. Cesium loading of the SDS zeolite beds might be seriously impaired by the presence of resin degradation products in eluates, depending on their nature

and the quantity. Finely dispersed solids (or colloids) from resin particle breakage and degradation or from fuel debris might not be readily separable and cause plugging of the zeolite bed. Soluble organic compounds and emulsified oils might sorb on zeolites and foul or block the exchange sites for cesium. Batch elution was selected for cesium removal rather than column elution because of the presence of finely divided solids in the degraded resin beds. Batch contact operation permits the use of relatively long settling times in eluate clarification. The available free space in the demineralizers limited the volume ratio of the liquid-to-solid phase to 1.5 in the elution of the DA or DB resin.

At the beginning of the present work, a conceptual generic flowsheet study¹ was already well under way, assuming the cesium was elutable. Although the development of the elution system design was the responsibility of the Hanford Engineering and Development Laboratory (HEDL), individuals from other organizations (ORNL, GPU, and EG&G) provided information for input into the HEDL study at the meetings held at TMI-2. The conceptual flowsheet was used as a guide for the present experimental work. A schematic of this conceptual flowsheet is shown in Fig. 1. After removal of the existing liquid in the demineralizers, cesium is eluted in multi-stage batch contacts of the resin with eluent solutions which, at that time, were undefined. Contact of the resin with eluent solutions is accomplished by an upward flow of solution through the bed to promote mixing; after sufficient contact time, the suspended solids are allowed to settle. Removal of the eluates is then accomplished by using an eductor to lift the liquid, which also dilutes the eluates with process water. The diluted eluates are subsequently filtered through a backflushable, stainless steel filter frit to remove finely dispersed resin and/or fuel debris solids, and finally treated by a charcoal bed to remove soluble or emulsified organic compounds prior to SDS processing. It was believed that the charcoal bed treatment might be necessary to avoid plugging or fouling problems with the zeolite beds in subsequent processing if appreciable quantities of organic compounds (especially emulsified liquids or tars) were present in the filtered eluates. However, it turned out that the charcoal bed was not necessary because the experimental studies later showed that only modest amounts of soluble organics (100 to 300 ppm) were

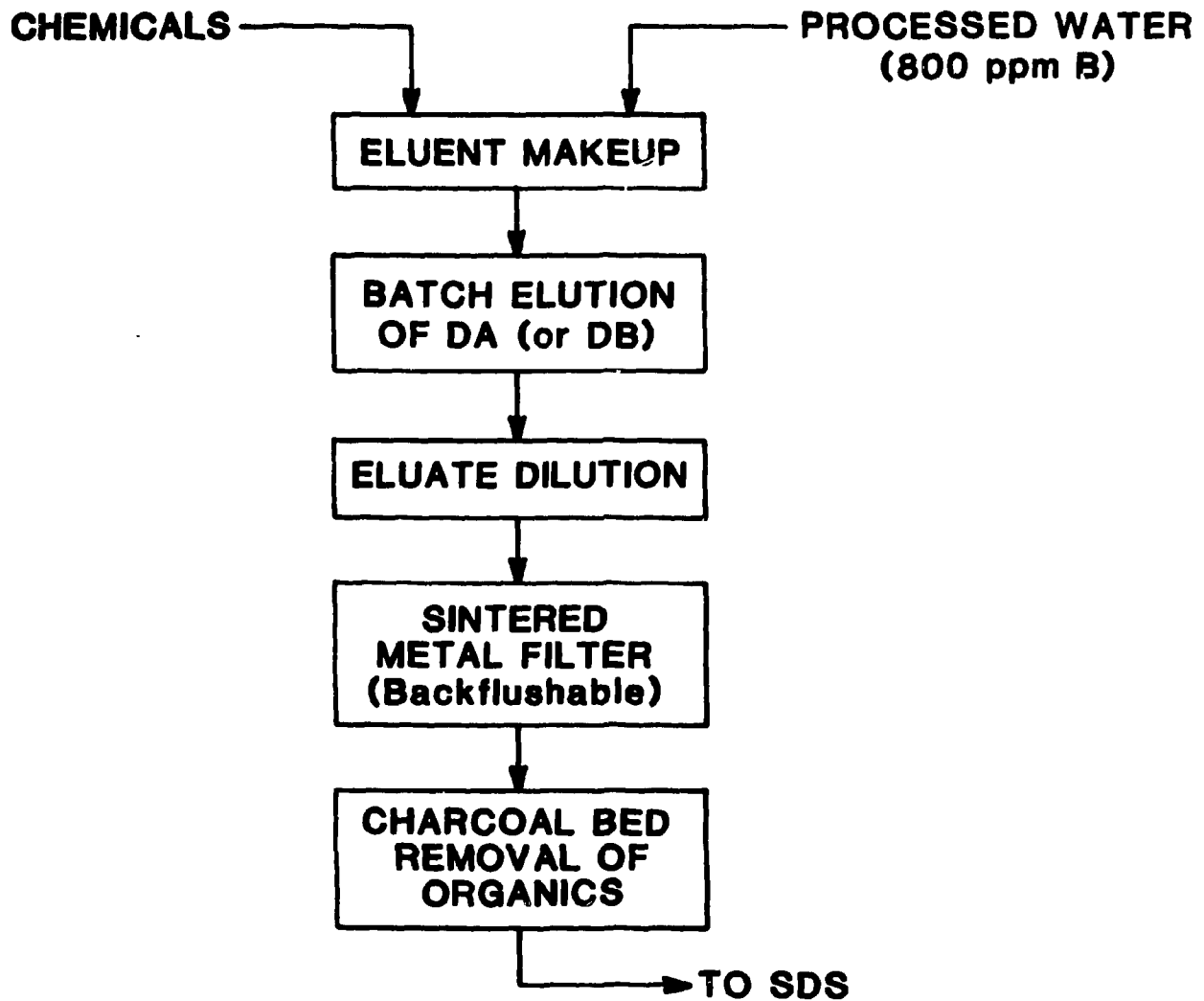


Fig. 1 Conceptual flowsheet for demineralizer elution.

present in the eluates. These organics were not effectively removed by charcoal and had no effects in SDS processing. Thus, the charcoal bed treatment was eliminated from the final process flowsheet for demineralizer cleanup.

2. LABORATORY DEVELOPMENTAL STUDIES

Small-scale experimental tests were conducted in a hot-cell facility using the conceptual flowsheet previously described (Fig. 1) as a guide. The results of these tests were used to develop a process flowsheet for cesium elution that was subsequently employed at TMI-2 in the cleanup of the demineralizers.⁶ Specific tests carried out included:

1. single-stage batch elution tests to demonstrate the feasibility of eluting cesium from the degraded demineralizer resins by sodium ion displacement;
2. multistage, batch elution tests to establish the sodium ion concentration elution gradient necessary for limiting the cesium activities in eluates to a maximum of 1 mCi/mL and to determine the number of stages necessary for removal of the elutable cesium;
3. zeolite bed tests on the ^{137}Cs and ^{90}Sr decontamination of the DA and DB eluates;
4. charcoal-bed treatment of eluates to determine the effects of the removal of organic compounds on subsequent SDS processing; and
5. clarification of eluates by settling and by filtration.

These tests were carried out in separate campaigns because the DB sample was available about 1 year sooner than the DA sample (May 1983 vs April 1984). Therefore, most of the experimental work was conducted using the DB sample. Because of the previous experience with the DB sample, the conditions for elution of the DA resin could be established more rapidly.

2.1 DESCRIPTION AND ANALYSIS OF DEMINERALIZERS SAMPLES

Only the samples used in the development of the chemical flowsheet are described here. Some early samples were taken from DA and DB which were of insufficient amounts to be used for flowsheet developmental studies, and results of their analysis are given in Appendix A.

Samples used in the flowsheet developmental studies contained both resin and liquid phases. The DA sample contained ~20 mL of resin and ~50 mL of liquid; the DB sample contained ~40 mL of resin and ~80 mL of liquid. The liquid was separated from the resin by settling and decantation. Most of the resin settled very rapidly. The resin was then dried in air at ambient conditions (25 to 30°C). In some cases the separated liquid was further clarified by centrifugation before analysis. The resin and liquid were analyzed by chemical and radiochemical methods. The resin was also examined by visual microscopy at ~20X magnification to assess its physical characteristics.

Chemical and radionuclide analyses of the resin and liquid phases of the DA and DB samples are given in Table 2. Only the principal constituents

Table 2. Radionuclide and chemical analyses of the demineralizer samples

Analyses	DA		DB	
	Liquid ^a	Resin	Liquid ^b	Resin
Radionuclides, $\mu\text{Ci/g}$				
^{137}Cs	209	5,520	1,480	21,800
^{134}Cs	11	285	101	1,458
^{90}Sr	6.72	3,060	9.46	890
Chemical, ppm				
C	164	NA ^c	950	NA
B	1,000	20	2,000	<200
Na	500	4,900	8,500	<1,000
SO_4^{d}	900	29,000	7,700	15,000
U	<1	2,400	<1	200
Fe	4	2,400	10	200
Ca	3	970	15	30
Ba	NA	240	NA	<1

^apH = 7.1.

^bpH = 5.7.

^cNA = not analyzed.

^dSulfur was determined as sulfate.

have been listed in Table 2. Small quantities (10 to 1000 ppm) of many other metal cations were also present in the resin but are not relevant to the work reported here. The complete analyses, including isotopic assay of uranium and plutonium, are given in Appendix B.

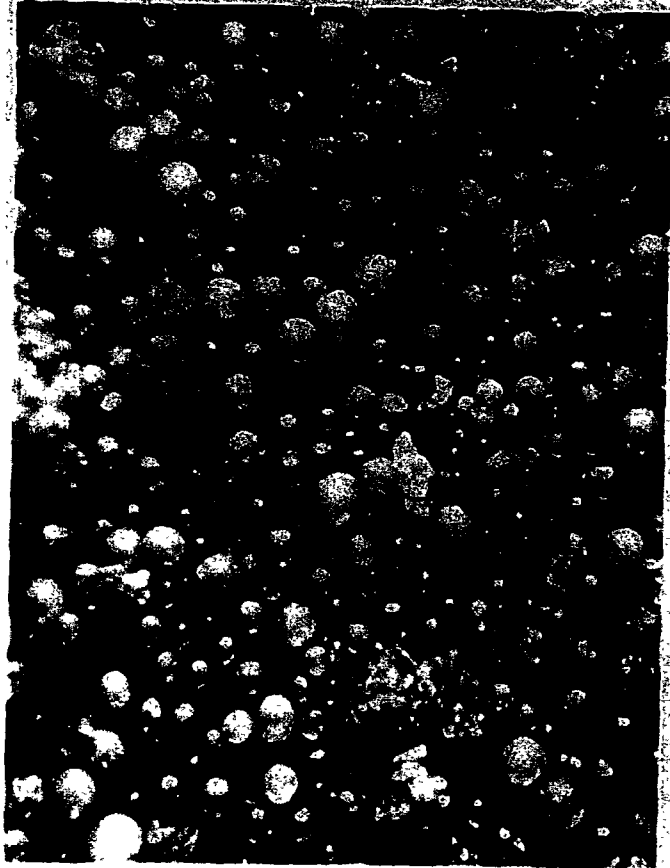
Microscopic examinations revealed that the DA resin was more severely damaged than the DB resin (Fig. 2). The DA resin contained large angular particles and clustered resin beads in significant amounts, perhaps 5 to 10 vol % of the sample. However, the remainder of the particles were clearly distinguishable as individual resin beads, with colors ranging from black (nontransparent) to amber (transparent). The angular particles had an appearance that is typical of pyrolytic carbons derived from tars, pitches, and polymeric resins. The angular particles were not observed in the DB resin.

The DB resin principally contained only resin beads that were black or amber colored. Very few bead clusters were observed. In some cases, partial spalling of the surface layer of the black beads exposed an amber-colored, transparent interior. The blackening only appeared to occur to a depth of a few micrometers.

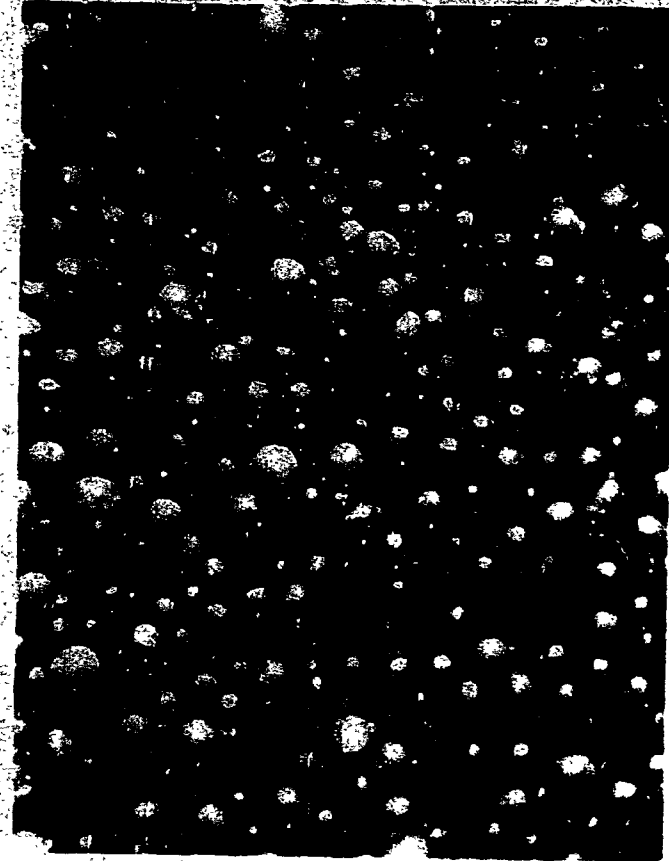
Liquids separated from the demineralizer samples were visually of good clarity but were yellowish-brown in color. It is later shown in further liquid clarification tests that the solutions were slightly turbid even after a settling time of 1 to 4 d.

2.2 RESIN ELUTION

Initial batch equilibrium tests with the DB resin showed that the ^{137}Cs was elutable with sodium ions (Table 3). The sharp dependence of ^{137}Cs distribution coefficient (K_d) values on sodium concentration of the eluents indicated that it would be relatively easy to develop a multistage batch elution procedure in which a sodium ion concentration gradient of eluents is employed to control the cesium activity levels of eluates. K_d is defined throughout this work for the distribution of radionuclides as: mCi per g of resin/mCi per mL of eluate. Subsequent multistage batch elution tests showed that the cesium activity levels of eluates could be limited to the desired levels.



DA Resin



DB Resin

Fig. 2. Photomicrographs of the demineralizer resins (20X magnification).

Table 3. Distribution coefficient (K_d) of ^{137}Cs between the DB resin and the liquid phase for eluents of increasing sodium ion concentration

(Conditions: 20 mL of eluent contacted with 0.5 g of resin for 30 min; initial specific activity of resin = 21.8 mCi/g)

Test	Eluent	% ^{137}Cs eluted	K_d , $\frac{\text{mCi/g}}{\text{mCi/mL}}$
1	H_2O	10.4	3.77×10^2
2	0.035 <u>M</u> NaH_2BO_3 - 0.32 <u>M</u> H_3BO_3	41.6	5.38×10^1
3	0.1 <u>M</u> NaOH	53.0	3.47×10^1
4	0.5 <u>M</u> NaOH	87.3	5.68×10^0
5	1.0 <u>M</u> NaOH	98.5	6.06×10^{-1}

Two multistage batch elution tests were conducted with the DB resin (Runs E-1 and E-2), and one test was performed with the DA resin (Run E-3) under conditions that were identical to Run E-2. Experimental conditions and results are given in Tables 4, 5, and 6. Run E-1 consisted of 20 stages of batch contacts in which the sodium ion concentration of the eluent was maintained constant at 0.035 M in stages 1 to 15 using sodium borate solutions. The sodium ion concentration was increased to 1 M in stages 16 to 20, with sodium hydroxide as the eluent. Runs E-2 and E-3 were 17-stage tests in which the sodium concentration of the eluents was increased more rapidly using a three-step increase in the sodium ion concentration. After an initial rinse of the resin with 0.18 M H_3BO_3 in stage 1, the resin was eluted with 0.035 M NaH_2BO_3 -0.32 M H_3BO_3 in stages 2 and 3, 0.26 M NaH_2BO_3 -0.09 M H_3BO_3 in stages 4 through 12, and finally, 1 M NaOH in stages 13 through 17. Both elution procedures gave satisfactory control of the ^{137}Cs concentration of the eluates. However, the more rapidly increasing sodium ion concentration of eluents in Run E-2 eluted the ^{137}Cs from the DB resin more effectively (96 vs 93%) and required fewer stages (17 vs 20). Therefore, the elution conditions used for Runs E-2 and E-3 were selected as the elution conditions for the demineralizer cleanup flowsheet at TMI-2. The cumulative percentages of ^{137}Cs elution determined for the latter stages of elution in Runs E-2 and E-3 (Tables 5

Table 4. Multistage batch elution of ^{137}Cs from DB resin in Run E-1(Conditions: 7.48 g of resin (~10 mL); 21.8 mCi of ^{137}Cs /g; contacted with 15 mL of eluent for 30 min)

Stage No.	Eluent	Eluate concentration		% of initial ^{137}Cs eluted ^a		K_d , $\frac{(\text{mCi/g})}{(\text{mCi/mL})}$
		^{137}Cs (mCi/mL)	Total C ($\mu\text{g/mL}$)	Instan.	Cumul.	
1	0.035 M NaH_2BO_3 --0.32 M H_3BO_3	1.119	370	10.3	10.3	17.5
2		0.432	145	4.0	14.3	43.3
3		0.330	205	3.0	17.3	54.6
4		0.281	75	2.6	19.9	62.1
5		0.284	73	2.6	22.5	59.5
6		0.273	74	2.5	25.0	59.9
7		0.255	72	2.4	27.4	62.1
8		0.252	54	2.3	29.7	60.8
9		0.245	60	2.2	31.9	60.6
10		0.240	62	2.2	34.1	59.8
11		0.231	135	2.1	36.3	60.1
12		0.225	70	2.1	38.3	59.8
13		0.214	60	2.0	40.3	60.8
14		0.212	50	2.0	42.3	59.4
15	1 M NaOH	0.316	100	2.9	45.2	37.8
16		1.443	870	13.3	58.4	6.3
17		1.424	435	13.1	71.5	4.4
18		1.111	435	10.2	81.8	3.6
19		0.822	250	7.6	89.3	2.8
20		0.492	230	3.6	93.0	3.1

^aInstan. = instantaneous; Cumul. = cumulative.

Table 5. Multistage batch elution of ^{137}Cs from DB resin in Run E-2
(Conditions: 7.22 g of resin (~10 mL); 21.8 mCi of $^{137}\text{Cs/g}$; contacted with 15 mL of eluent for 30 min)

Stage No. ^a	Eluent	Eluate concentration		% of initial ^{137}Cs eluted ^b		K_d , $\frac{\text{mCi/g}}{\text{mCi/mL}}$
		^{137}Cs (mCi/mL)	Total C ($\mu\text{g/mL}$)	Instan.	Cumul.	
1	0.18 M H_3BO_3	1.090	485	10.4	10.4	17.9
2	0.035 M NaH_2BO_3 --0.32 M H_3BO_3	0.340	200	3.2	13.6	55.4
3		0.420	140	4.0	17.6	42.8
4	0.26 M NaH_2BO_3 --0.09 M H_3BO_3	1.230	292	11.7	29.4	12.5
5		1.140	305	10.8	40.2	11.4
6		1.050	260	10.0	50.2	10.3
7		0.870	235	7.7	58.0	10.5
8		0.757	335	7.2	65.2	10.0
9		0.632	143	6.0	71.2	9.9
10		0.516	128	4.9	76.1	10.1
11		0.405	88	3.9	80.0	10.8
12		0.343	88	2.8	82.8	10.9
13	1 M NaOH	0.432	560	4.1	86.9	6.6
14		0.395	340	3.8	90.7	5.1
15		0.264	260	2.5	93.2	5.6
16		0.175	240	1.7	94.9	6.4
17		0.097	320	0.8	95.7	9.7

^aEluent volumes in stages 7, 12, and 17 were 14, 13, and 13 mL, respectively.

^bInstan. = instantaneous; Cumul. = cumulative.

Table 6. Multistage batch elution of ^{137}Cs from DA resin in Run E-3

(Conditions: 6.56 g of resin (~10 mL); 5.52 mCi of $^{137}\text{Cs/g}$; contacted with 15 mL of eluent for 30 min)

Stage No.	Eluent	^{137}Cs in eluent ($\mu\text{Ci/mL}$)	% of initial ^{137}Cs eluted ^a		K_d , $\frac{(\text{mCi/g})}{(\text{mCi/mL})}$
			Instan.	Cumul.	
1	0.18 <u>M</u> H_3BO_3	96.8	4.0	4.0	54.8
2	0.035 <u>M</u> NaH_2BO_3 --0.32 <u>M</u> H_3BO_3	136	5.6	9.6	36.7
3		135	5.6	15.2	34.7
4		245	10.1	25.3	16.8
5	0.26 <u>M</u> NaH_2BO_3 --0.09 <u>M</u> H_3BO_3	185	7.7	33.0	20.0
6		122	5.0	38.0	28.0
7		78.1	3.2	41.2	41.5
8		67.3	2.8	44.0	46.0
9		51.9	2.2	46.2	57.2
10	1 <u>M</u> NaOH	37.0	1.6	47.7	78.0
11		25.5	1.1	48.8	111
12		18.1	0.7	49.5	154
13		64.3	2.7	52.2	41.0
14		42.7	1.8	54.0	59.5
15		25.7	1.1	55.0	96.5
16		16.0	0.7	55.7	153
17		12.1	0.5	56.2	200

^aInstan. = instantaneous; Cumul. = cumulative.

and 6) show that little benefit would be gained by carrying out any additional stages of elution. The residual ^{137}Cs activities remaining on the DA and DB resins after 17 elution stages were 2.4 and 0.9 mCi/g, respectively.

The ^{137}Cs was clearly much less elutable from the more-degraded DA resin than from the less-degraded DB resin (56% vs 96%). Also, the K_d values for the DB resin showed little change with the same sodium concentration of the eluent, whereas the K_d values for the DA resin showed significant increases (Fig. 3). Increasing K_d values as elution proceeds is typical of degraded resins because a variety of chemical sites are available which have different affinities for binding cations. To determine if the large clustered and carbonized particles in the DA resin sample contained most of the uneluted ^{137}Cs , the eluted resin bed was air dried and classified by screening into three fractions which were analyzed. Only ~12 to 14 particles were present in the >2000- μm fraction, about 10 vol % of the bed in the 850 to 2000- μm fraction, and ~90 vol % in the <850- μm fraction. The specific activities of ^{137}Cs were 4.1, 4.7, and 2.4 mCi/g for the >2000-, 850- to 2000-, and <850- μm fractions, respectively. Although the larger particles are of higher specific activity, most of the uneluted ^{137}Cs (~80%) is associated with the <850- μm fraction.

Organic compounds were solubilized throughout the series of batch contacts (Tables 4 and 5; Fig. 4). However, the carbon compounds were only solubilized to an extent of ~8 mg of C/g of resin in 17 stages of contact. The total carbon concentrations of individual batch eluates from the multistage test with DA resin were not determined, but the analysis of DA eluate composite (see Sect. 2.3, Table 9) indicated no significant difference in dissolved organic content. No attempt was made to identify the compounds. The higher sodium concentrations increased the solubilization of the organic compounds.

The clarity of eluates appeared generally good by visual observations. However, some cloudiness was observed in the first stages of elution with the DB resin and during the final treatment with 1 M NaOH with either the DA or DB resin. The eluates from stages 1 and 4 of the DB resin elution were filtered through a 0.5- μm -rated nylon filter disk, and the filter was subsequently scanned by γ spectroscopy (Table 7). Only small quantities

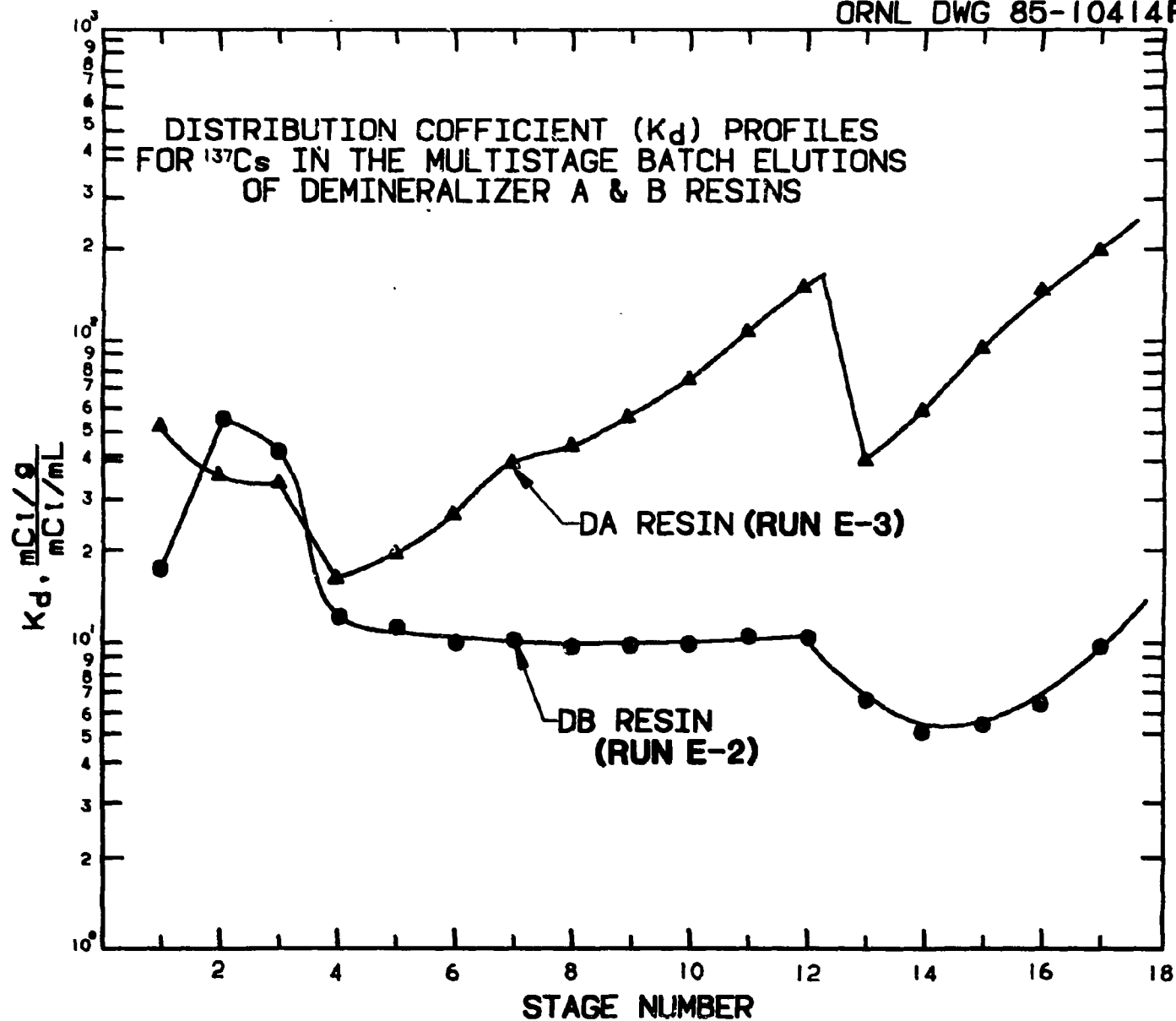


Fig. 3. Distribution coefficient (K_d) profiles for ^{137}Cs in the multistage batch elutions of demineralizer A and B resins.

ORNL DWG 85-10415 R

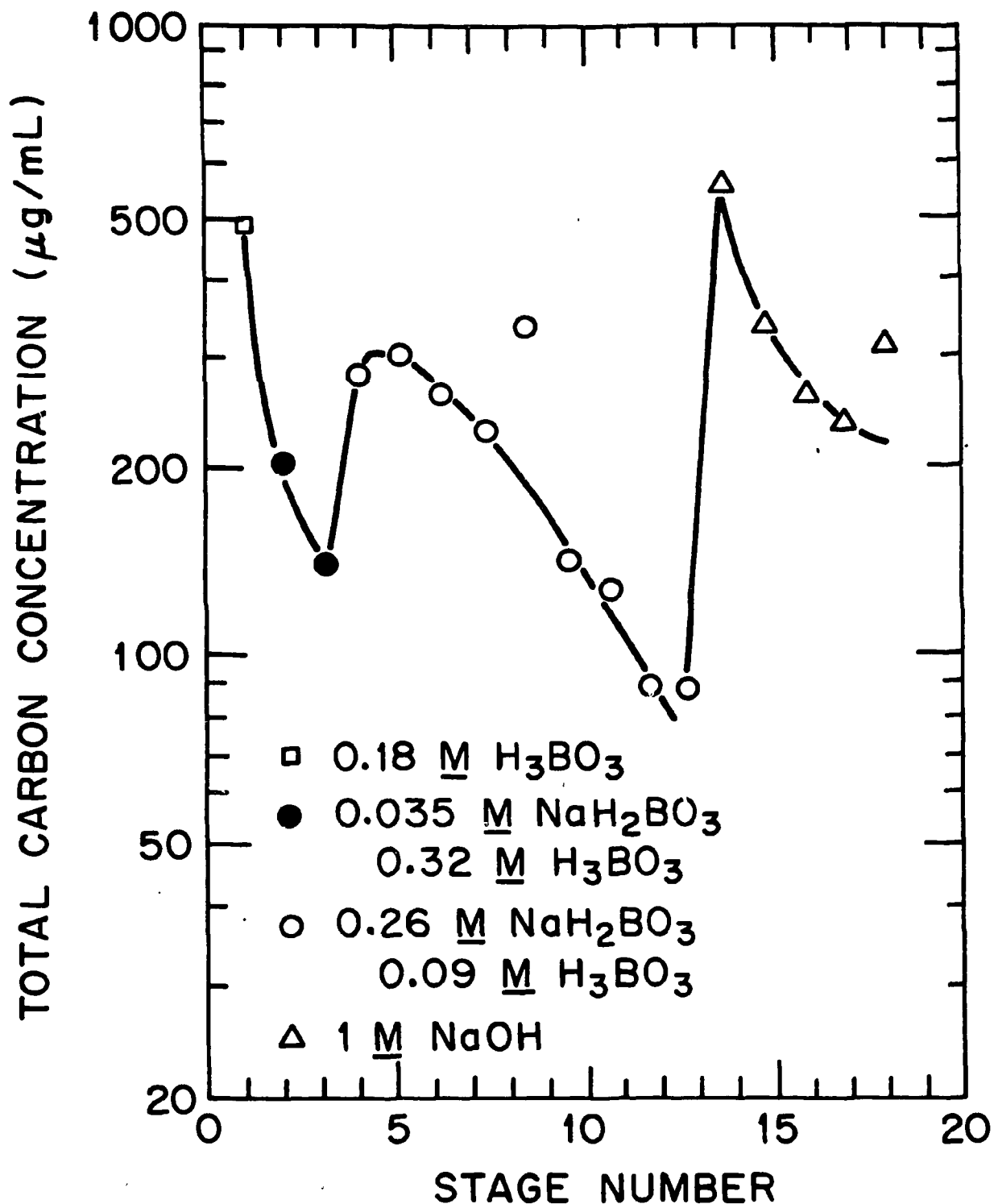


Fig. 4. Leaching of carbon compounds in multistage batch elution of the DB resin in Run E-2.

Table 7. Gamma scan of filter solids from a DB resin eluate

Radionuclide	Activity (Bq)
^{60}Co	27
^{125}Sb	310
^{134}Cs	1.66×10^3
^{137}Cs	2.69×10^4
^{144}Ce	52

of radioactivity were observed and were due principally to the presence of ^{137}Cs and much lesser amounts of ^{125}Sb , ^{144}Ce , and ^{60}Co . The observed activity of ^{137}Cs was ~0.01% that of the filtered solution (~400 MBq).

As expected, ^{90}Sr is partially eluted in the multistage batch tests (Table 8). However, it is not eluted as effectively as ^{137}Cs (Fig. 5). The elution of ^{90}Sr is not a major concern since the SDS system at TMI-2 is also designed to remove strontium. About 25% of the ^{90}Sr was eluted from either the DA or DB resins. Elution with 1 M NaOH was not as effective in increasing the ^{90}Sr removal as it was for ^{137}Cs removal. The 1 M NaOH may have decreased the solubility of ^{90}Sr because of the precipitation of the metal hydroxide. Since no difference was observed in the percentage of the ^{90}Sr that was eluted from the DA and DB resins, it seems likely that most of the ^{90}Sr eluted was associated with the least-damaged resin particles in the DA or DB resins.

2.3 DECONTAMINATION FACTORS OF ^{137}Cs AND ^{90}Sr IN ZEOLITE BED TESTS

Decontamination factors (DFs) for ^{137}Cs and ^{90}Sr were determined in the zeolite bed tests with resin eluate feeds and with the liquid separated from the as received DA sample that had been diluted 20:1 with 0.18 M H_3BO_3 (DA liquid). Tests consisted of passing 70 to 100 bed volumes of feed through a 2-mL bed of mixed zeolites (60 vol % Ionsiv-96 and 40 vol % Linde A-51). The superficial bed residence time was maintained constant at 8.4 min throughout each test — a value which is comparable to that (~10 min) used in SDS processing at TMI-2.

Table 8. Elution of ^{90}Sr in the multistage batch elution tests
of the DA resin (Run E-2) and DB resin (Run E-3)

Stage No.	Elution agent	Cumulative % of initial ^{90}Sr eluted		K_d , $\frac{\text{mCi/g}}{\text{mCi/mL}}$	
		DA	DB	DA	DB
1	0.135 <u>M</u> H_3BO_3	0.16	1.05	1400	200
2	0.035 <u>M</u> NaH_2BO_3 --0.32 <u>M</u> H_3BO_3	0.31	1.24	1600	1100
3		0.69	1.55	600	650
4	0.26 <u>M</u> NaH_2BO_3 --0.09 <u>M</u> H_3BO_3	3.87	3.31	69	110
5		7.42	5.85	60	77
6		10.3	8.56	72	70
7		12.8	10.4	80	94
8		15.0	12.8	86	75
9		17.3	15.2	85	74
10		18.9	17.4	110	77
11		20.6	19.7	110	76
12		22.2	21.5	120	78
13	1 <u>M</u> NaOH	22.5	22.1	570	270
14		22.7	23.1	930	160
15		22.9	24.3	880	130
16		23.1	25.6	880	120
17		23.3	26.7	850	120

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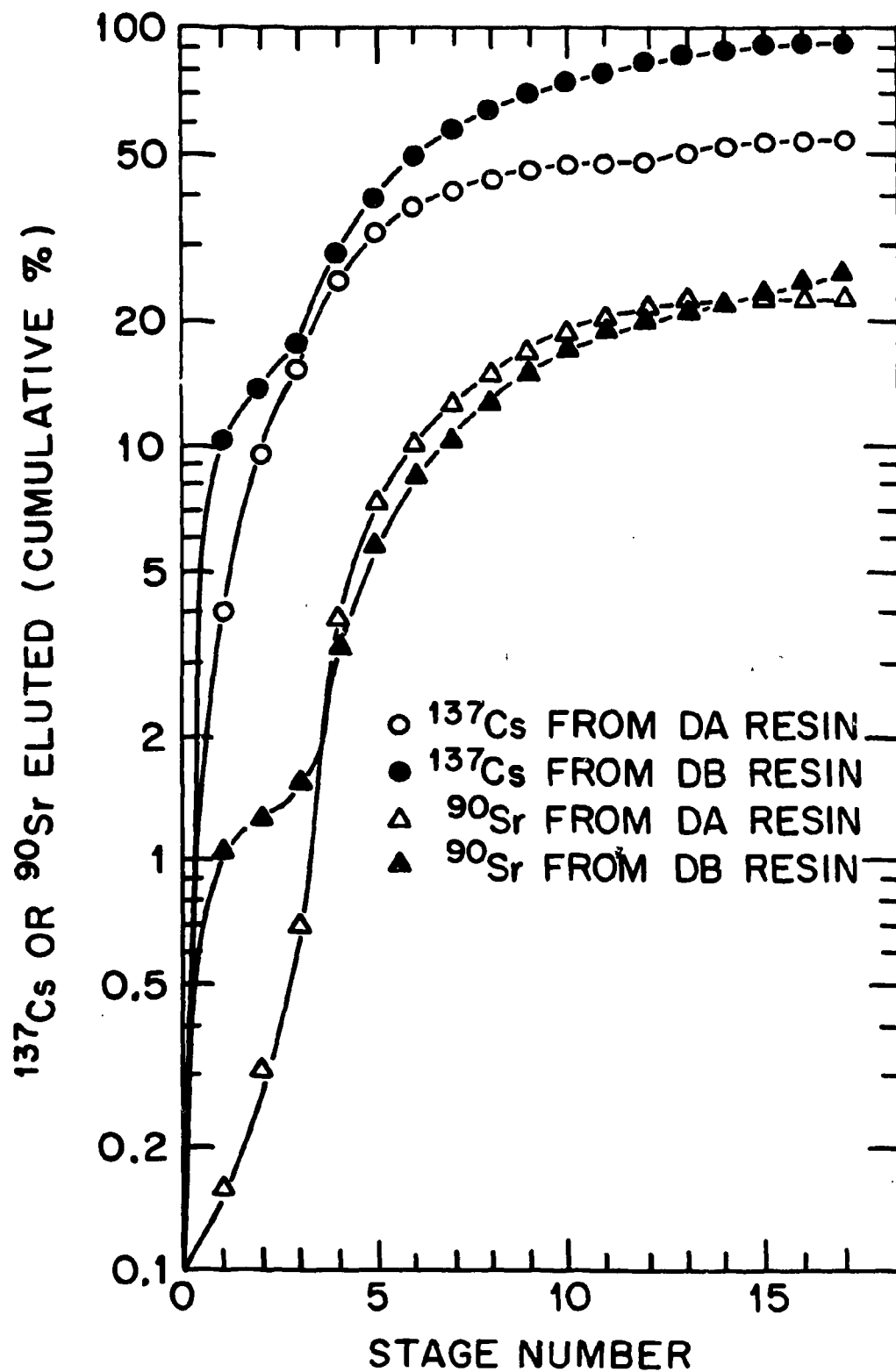


Fig. 5. Percentage of ^{137}Cs and ^{90}Sr eluted in multistage batch elution tests with the DA and DB resins.

Results of the tests are shown in Table 9. The cumulative DFs for ^{137}Cs ranged from $\sim 5 \times 10^3$ to 3×10^5 and for ^{90}Sr from ~ 200 to 500. Instantaneous DFs as a function of bed volumes of feed are shown in Figs. 6-8. The values of the DFs and their variation with the feed concentrations of ^{137}Cs and ^{90}Sr are consistent with previous studies^{2,3} on the performance of the SDS zeolites. The DF of ^{137}Cs for the DA liquid feed was slightly improved by filtering it with a 10- μm -rated filter frit (Run Z-5), perhaps indicating a very small percentage of the ^{137}Cs was associated with dispersed solids that were present. No evidence of bed plugging was noted in any of the tests although the feeds for Runs Z-1, Z-2, Z-3, and Z-4 were not filtered. The zeolite bed effluents contained very low levels of ^{60}Co and ^{125}Sb activities in addition to the residual cesium and strontium activities (Fig. 9).

The results from Runs Z-1 and Z-2 with eluate composites from the DB resin elution may also indicate that very small quantities of ^{137}Cs (0.01%) are associated with finely dispersed solids or colloids in those feeds. The higher cumulative DF value of 1.5×10^5 in Run Z-2 (Table 10) was obtained with feed that had been previously treated by a charcoal bed (see Sect. 3-4, Table 11), whereas the lower DF (1.0×10^4) was obtained with the raw feed in Run Z-1. The charcoal bed may have acted as a filter or a sorbent for the finely dispersed cesium-bearing solids previously described in Table 7.

2.4 SORPTION OF SOLUBLE ORGANIC COMPOUNDS IN ZEOLITE AND IN CHARCOAL BED TESTS

The sorption of soluble organic compounds from demineralizer resin eluates was investigated in three of the zeolite bed tests previously described (Runs Z-1, Z-2, and Z-3) and in two charcoal bed tests with SK-4 charcoal.* Neither bed material was very effective for removing the organic compounds (Tables 10 and 11). Removals were poor even at relatively low bed loadings. No attempt was made to identify the specific organic compounds in the liquid feeds or those that remained in the effluents.

* Obtained from Alltech Associates, Applied Sciences Division, Deerfield, Illinois.

Table 9. Cumulative DFs for ^{137}Cs and ^{90}Sr in zeolite bed tests
 [Conditions: 2-mL mixed zeolite bed (60 vol % Ionsiv 96 and 40 vol % Linde A-51);
 residence time = 8.4 min]

Test	Source	Feed solutions			Effluent bed volumes	Cumulative DF	
		^{137}Cs , $\mu\text{Ci/mL}$	^{90}Sr , $\mu\text{Ci/mL}$	Total C, $\mu\text{g/mL}$		^{137}Cs	^{90}Sr
Z-1	DB resin eluates ^a Stages 1-12, Run E-1	349	1.07	120	90	1.04×10^4	2.5×10^2
Z-2	DB resin eluates ^b Stages 1-12, Run E-2	346	0.14	80	90	1.50×10^5	5.11×10^2
Z-3	DA resin eluates Stages 1-12, Run E-3	97	32.5	130	70	$>3.48 \times 10^5$	4.96×10^2
Z-4	DA Liquid	10	0.26	8	100	4.34×10^3	2.60×10^2
Z-5	DA liquid; filtered through 10 μm -rated filter	8	0.18	12	100	1.09×10^4	2.03×10^2

^aFrom elutions described in Table 4 prior to charcoal-bed treatment.

^bFrom elution described in Table 5 following charcoal-bed treatment.

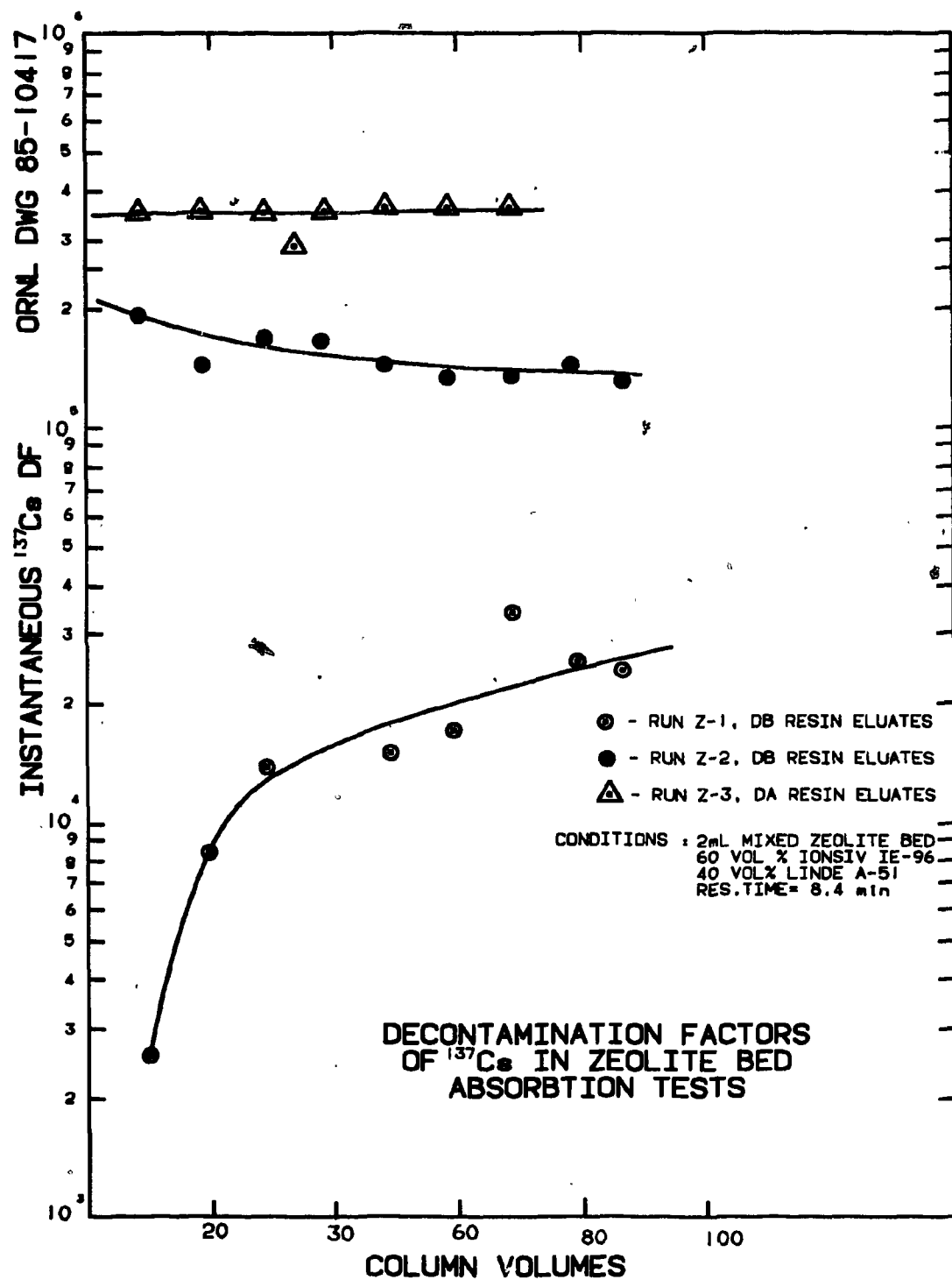


Fig. 6. Decontamination factors of ^{137}Cs in zeolite bed absorption tests with resin eluate feeds.

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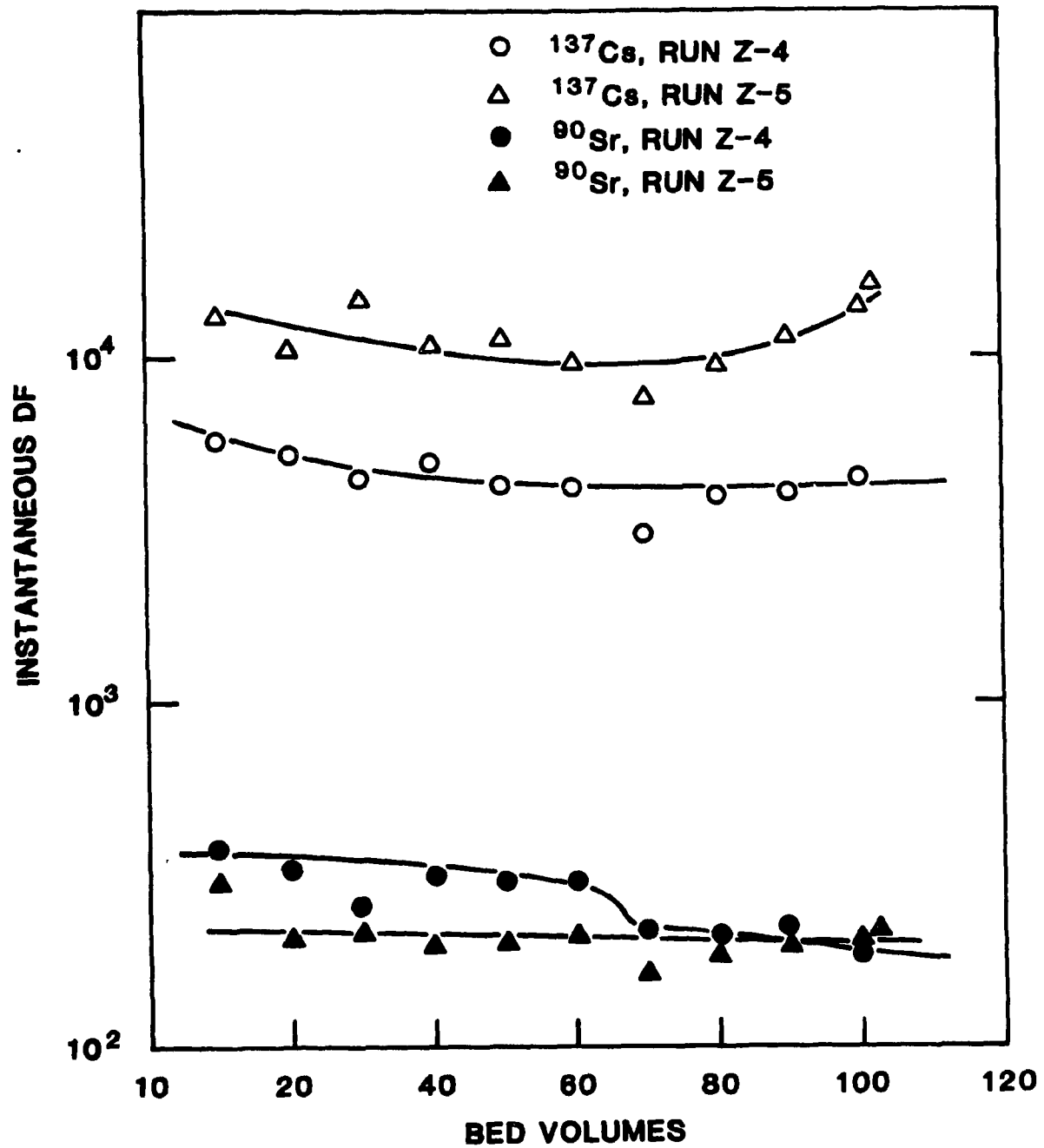


Fig. 7. Instantaneous DF values of ^{137}Cs and ^{90}Sr in zeolite bed tests with diluted DA liquid feed.

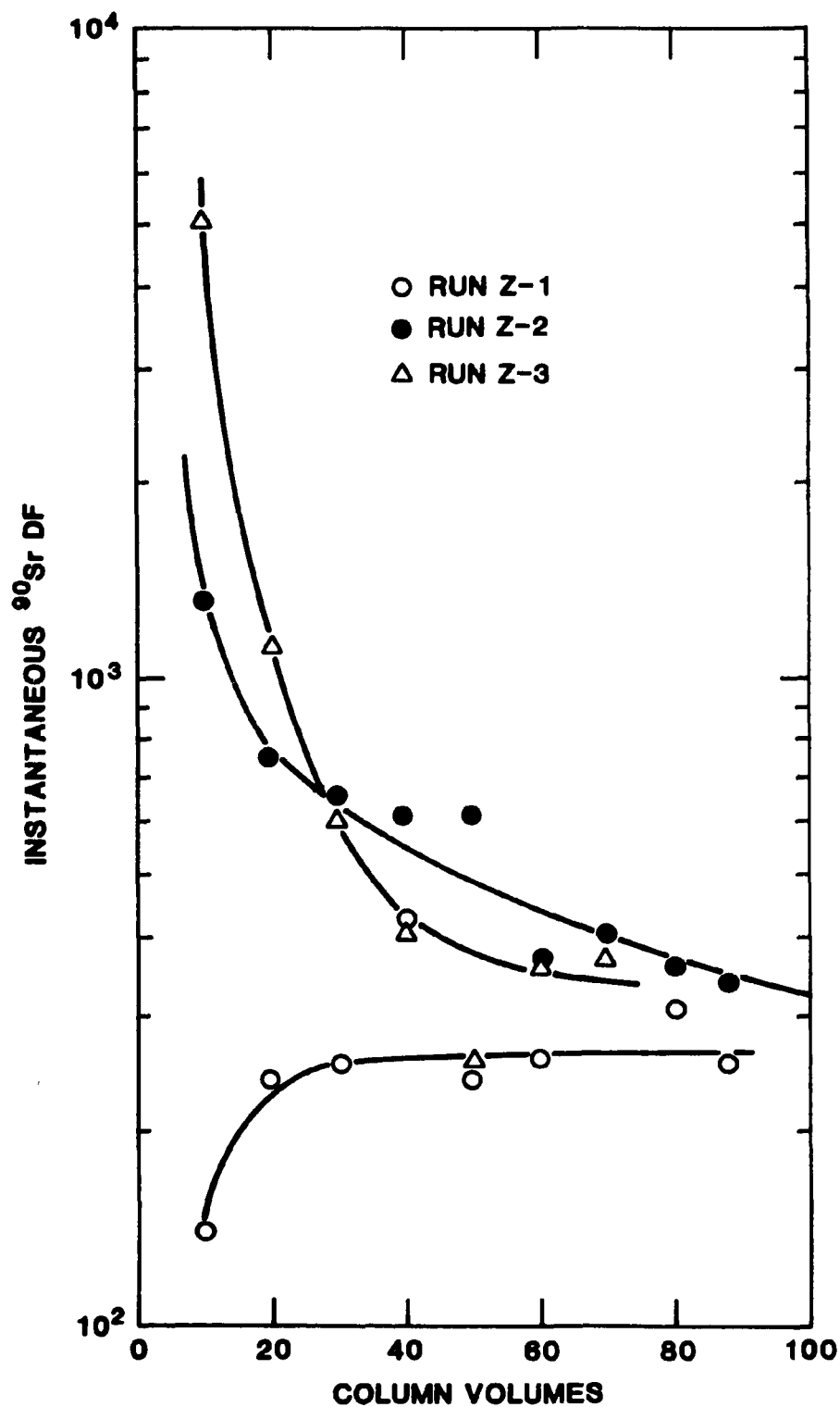


Fig. 8. Instantaneous DF values of ^{90}Sr in zeolite bed tests with resin eluate feeds.

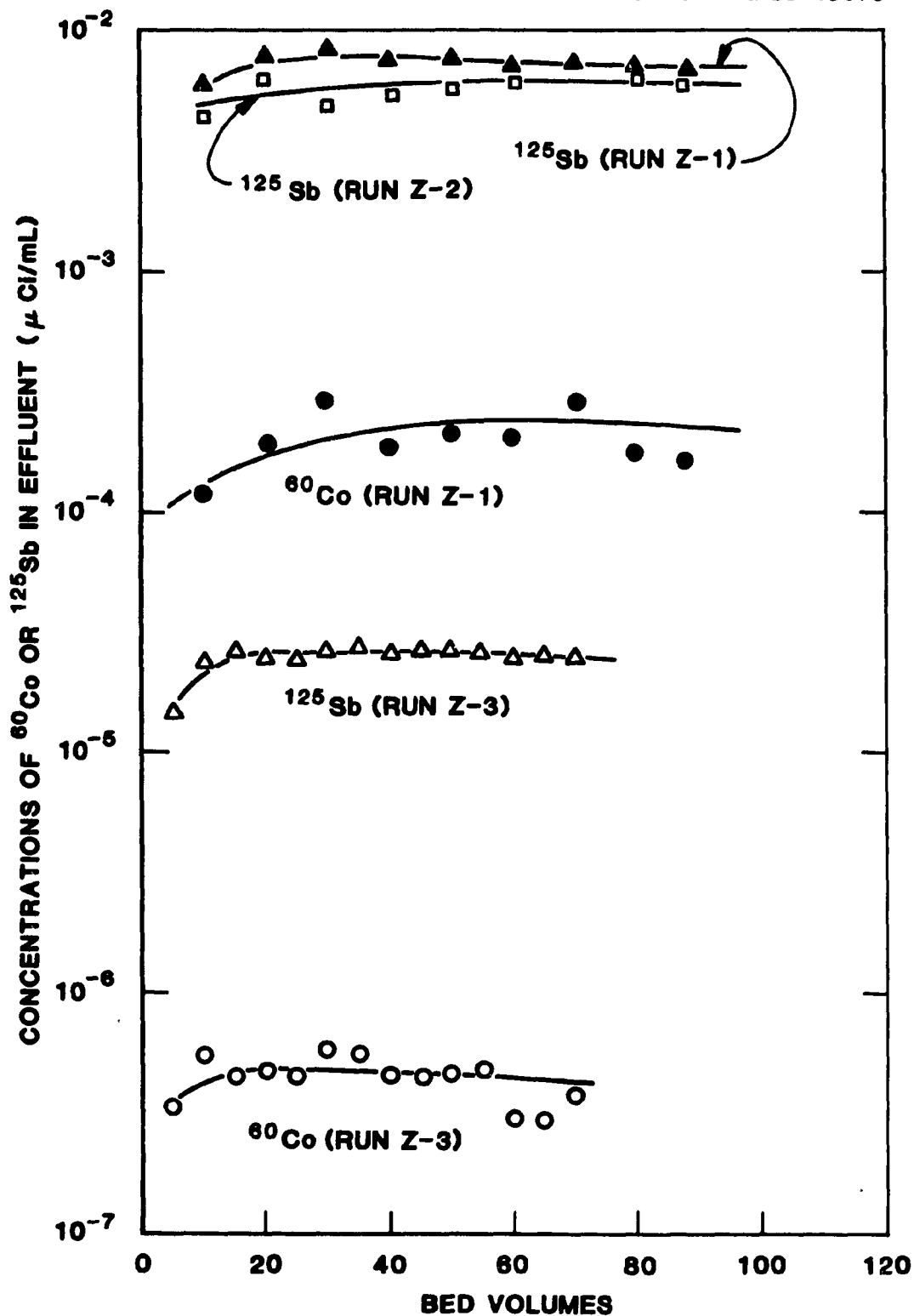


Fig. 9. Concentrations of ^{60}Co and ^{125}Sb in effluents of zeolite bed tests.

Table 10. Instantaneous breakthrough and loading of carbon compounds in zeolite bed tests with composite eluate feeds

(Conditions: 2-mL mixed zeolite bed; superficial bed residence time = 8.4 min)

Bed volumes of eluent	Instantaneous breakthrough (%)			Cumulative bed loading, $\frac{\text{mg of C}}{\text{g of zeolite}}$		
	Run Z-1 ^a	Run Z-2 ^{a,b}	Run Z-3 ^c	Run Z-1	Run Z-2	Run Z-3
5			41.5			0.19
10	39.6	42.5		0.72	0.46	
15			43.1			0.56
20	62.5	71.3		1.2	0.69	
25			45.4			0.92
30	70.8	76.3		1.5	0.88	
35			43.9			1.3
40	66.7	71.3		1.9	1.1	
45			43.9			1.6
50	70.8	73.8		2.3	1.3	
55			46.2			2.0
60	72.9	78.8		2.6	1.5	
70	75.0	80.0	43.1	2.9	1.7	2.5 ^d
80	84.4	85.0		3.3	1.8	
88	78.2	92.5		3.5 ^d	1.8 ^d	

^aFeed for Runs Z-1 and Z-2 were composites of eluates from multistage batch elution Run E-1 and E-2 with DB resin, stages 1-12.

^bComposite resin eluate feed had been treated with SK-4 charcoal (see Table 12, Run C-2) prior to Run Z-2.

^cFeed was a composite of eluates from the multistage batch elution Run E-3 with DA resin, stages 1-12.

^dCumulative percentage breakthrough of organic compound for Runs Z-1, Z-2, and Z-3 were 67.1, 74.1 and 43.3%, respectively.

Table 11. Breakthrough of carbon compounds and beds loadings
in charcoal bed tests

(Conditions: Superficial bed residence time = 10.4 min; SK-4 charcoal,
80-100 mesh)

Bed volumes	Instantaneous breakthrough, (%)		Bed loading, $\frac{\text{mg of C}}{\text{g of charcoal}}$	
	Run C-1 ^a	Run C-2 ^b	Run C-1	Run C-2
2.5		35		0.52
5.0	38	36	0.51	1.0
7.5		42		1.5
10.0	46	46	0.96	1.9
12.5		51		2.3
15.0	59	61	1.3	2.6
17.5		62		2.9
20.0	83	75	1.4	3.2
22.5		63		3.4
25.0	85	66	1.6	3.7
27.5		66		4.0
30.0	84	69	1.7	4.2
32.5		72		4.5
35.0	91	69	1.8	4.7
37.5		71		4.9
40.0	80	76	1.9	5.1 ^c
42.5	79		2.0 ^c	

^aA 2-mL (0.91-g) bed; feed was a composite of effluent cuts from zeolite bed test Run Z-1; feed concentration = 75 $\mu\text{g c/mL}$.

^bA 6-mL (2.72-g) bed; feed was a composite of eluate cuts (stages 1-12) from DB resin elution, Run E-2; feed concentration = 145 $\mu\text{g c/mL}$.

^cCumulative breakthroughs for Runs C-1 and C-2 were 71.1 and 59.7%, respectively.

The percentage breakthroughs and bed loadings were based entirely on total carbon analyses of feeds and effluents. No significant difference was observed in the effectiveness of zeolite and the charcoal for removing the organic compounds from DB resin eluates (compare Runs Z-1 and Z-2 in Table 10 with Runs C-1 and C-2 of Table 11). Charcoal bed tests were not made with the DA resin eluates. The breakthrough characteristics for carbon compounds as a function of bed loading in the zeolite bed tests were different for the DA and DB resin eluate feeds (Table 10). The breakthrough increased about twofold with increasing zeolite bed loading with DB resin eluate feeds (Run Z-1 and Z-2); whereas with the DA resin eluate feeds (Run Z-3), it remained about constant. The breakthrough characteristics of the SK-4 charcoal beds with DB resin eluate feeds were very similar to those for zeolite beds. Breakthrough increased from ~35% to ~80% as the feed volume increased from 5 to 40 bed volumes. No detectable quantity of ^{137}Cs was absorbed in the charcoal bed tests. The ^{137}Cs concentrations of feeds and effluents were identical.

Even when eluate feeds were treated sequentially by both charcoal and zeolite beds, the net cumulative removal of carbon compounds was still relatively poor. The composite of DB resin eluates from Run E-1 were treated first by a zeolite bed in Run Z-1 and then by a charcoal bed in Run C-1, and the composite of DB resin eluates from Run E-2 was treated first in Run C-2 with charcoal and then by zeolite in Run Z-2. The net cumulative breakthrough for carbon with either sequence of the zeolite and charcoal bed treatment was about 50%. On the basis of these tests and the fact that good DF values for ^{137}Cs and ^{90}Sr were obtained in the simulated SDS tests without prior charcoal bed treatment, the charcoal bed treatment was not included in the process flowsheet for demineralizer cleanup at TMI-2. The charcoal was simply ineffective for removing the organic compounds. The compounds, however, posed no detectable problems for SDS processing.

2.5 LIQUID CLARIFICATION

Clarification of the DA Liquid was investigated by filtration and by settling. Filtration tests were conducted with sintered metal (stainless

steel) filter disks with ratings of 0.5 and of 10 μm at filtration rates of 2.2 mL/cm²/min. This flow rate was the design basis for the filtration system to be used at TMI-2, which also provided for periodic backflushing when the filter pressure drop reached 20 psi. The liquid used for the filtration and settling tests was separated from the DA sample after 8 h of settling of the initially well-stirred sample and was subsequently diluted 20:1 with 0.18 M boric acid (2000 ppm B). Spark source mass spectrometric analysis of the diluted liquid is given in Table 12.

The filtration test results (Table 13) showed that good clarification was obtained using either the 0.5- or the 10- μm -rated filters. The turbidity was decreased from 12.6 to ~2 nephelometric turbidity units (NTUs) using either filter. The 0.5- μm -rated filter plugged rapidly (16 to 19 min), whereas the 10- μm -rated filter plugged more slowly (170 min). Pressure drop increase with filtrate volume for the 10- μm -rated filter is shown in Fig. 10. Onstream filtration times of 170 min were judged marginal for the clarification operations at TMI-2 with backflushing and a 20- μm -rated filter was installed at TMI-2 as an additional precaution.⁶ However, it remained to be demonstrated in the actual operations at TMI-2 that the filter could be satisfactorily restored by backflushing. The volume of samples available was too small to permit investigation of filter backflushing or of extensive studies of filtration.

Settling tests (Table 14) showed that clarification could be significantly improved by using longer settling times. Therefore, relatively long settling times after batch contacts of resin and eluents could be employed to reduce filter loadings in eluate clarifications and to increase liquid throughputs before filter plugging occurred. Relatively long settling times (24 to 72 h) were used in the processing of the eluates at TMI-2.⁶

An attempt was made to increase the settling rate by using fresh anion exchange resin (Amberlite IRA-400; 20-40 mesh) as a flocculation aid in one test. This resin is especially suited for silica removal in water treatment operations. However, it did not significantly improve the settling rate unless prohibitively large quantities were added. In that test, 1-g additions of the resin were made each day to the same 35-mL aliquot of liquid until a total of 4 g had been accumulated. After each

Table 12. Spark source mass spectrometry analysis of the diluted DA liquid

Element	Concentration (ppm)
B	~1000
Ca	10
Cr	0.3
Cs	0.08
K	0.7
Mg	2
Na	100
Ni	0.2
P	0.1
S	20
Si	≤5

Table 13. Clarification tests with sintered metal disk filters using DA liquid

(Conditions: 9.5-mm-diam filter disk, filtration rate = 2.2 mL/cm²/min; DA liquid diluted 20:1 with 0.18 M H₃BO₃)

Run No.	Filter rating (μm)	Time (min)	Final pressure drop (psi)	Volume (mL)	Turbidity ^a (NTUs)
1	0.5	19	20	30	2.1
2	0.5	19	20	25	2.3
3	10	170	14 ^b	270	2.2

^aTurbidity of feed was 12.6 NTUs (nephelometric turbidity units).

^bFeed supply was used up. Extrapolation of pressure-drop-vs-time curve indicated that 20 psi would be reached after 190 min and a filtrate volume of ~300 mL.

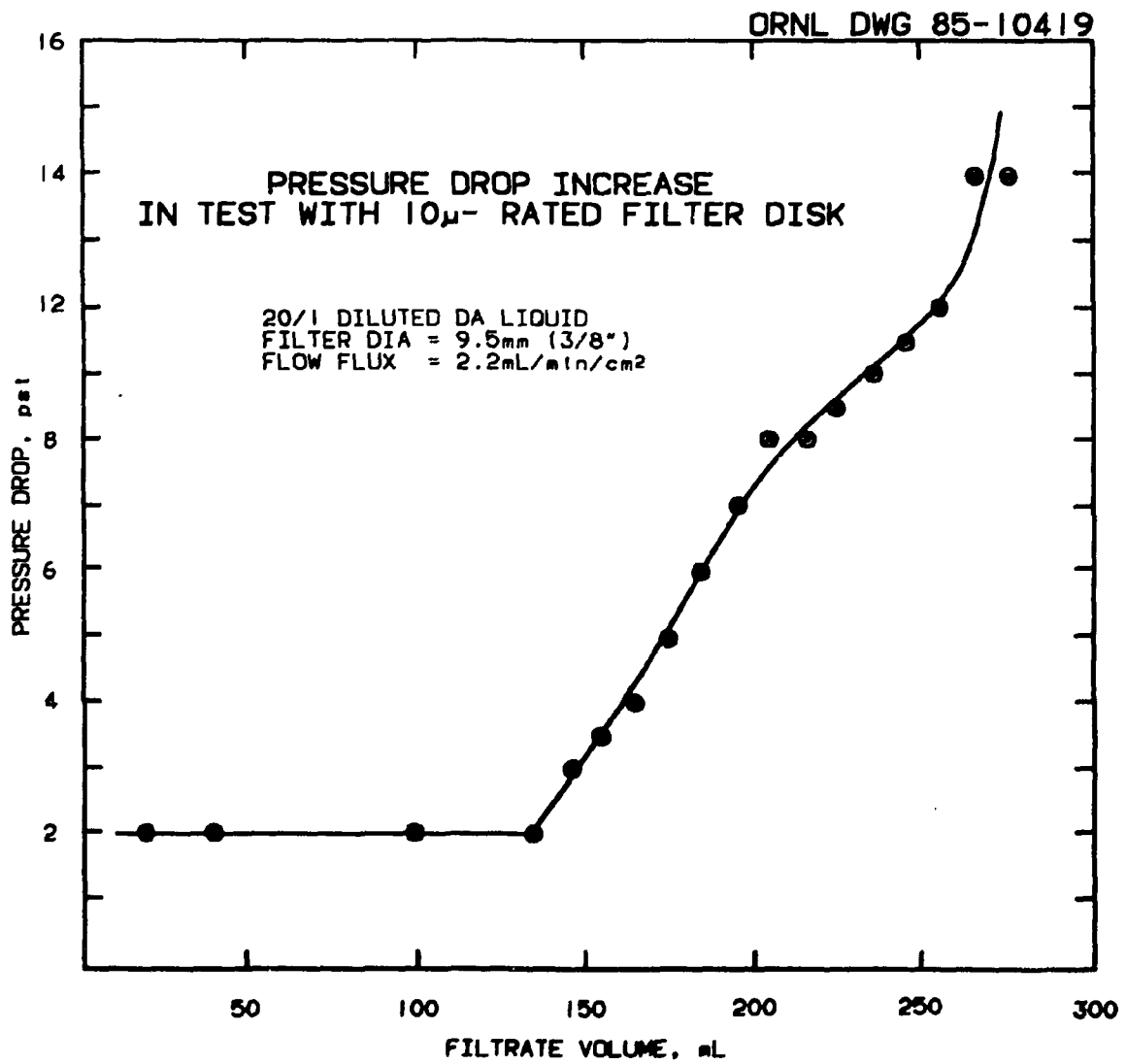


Fig. 10. Pressure drop increase in test with 10- μ -rated filter disk.

Table 14. Effect of settling time on DA liquid turbidity

Settling time (d)	Turbidity (NTUs) ^a
0	12.6
1	6.1
2	5.5
3	4.6
4	4.0

^aNTUs = nephelometric turbidity units.

l-g addition, the resin was contacted with the liquid by shaking at 15-min intervals over a period of 4 h and subsequently settled for 20 h. The resultant turbidities of the liquid resin were 5.2, 4.4, 3.5, and 2.8 NTUs, respectively.

3. PROCESS FLOWSHEET FOR DEMINERALIZER ELUTION

The experimental results from the previously described small-scale tests in hot cells provided the technical basis for the cleanup of the demineralizers at TMI-2.⁶ The schematic process flowsheet is shown in Fig. 11. After the removal of the existing liquid in the demineralizers, cesium is eluted in multistage batch contacts of the resin with the eluent solutions. As elution proceeds, the sodium concentrations of the eluent solution are increased from 0 to 1 M (0 to 23,000 ppm Na) to facilitate elution of the residual cesium on the resin. Liquid-to-solid phase volumetric ratios of ~1.5 are used in each stage, and the ratio is governed by the available free volume in the demineralizer vessels. Batch contacts of the eluent solutions with the resin are accomplished by upflowing the liquid into the demineralizer.

BACKFLUSH TO OPPOSITE
DEMINERALIZER

• TIE-IN POINT

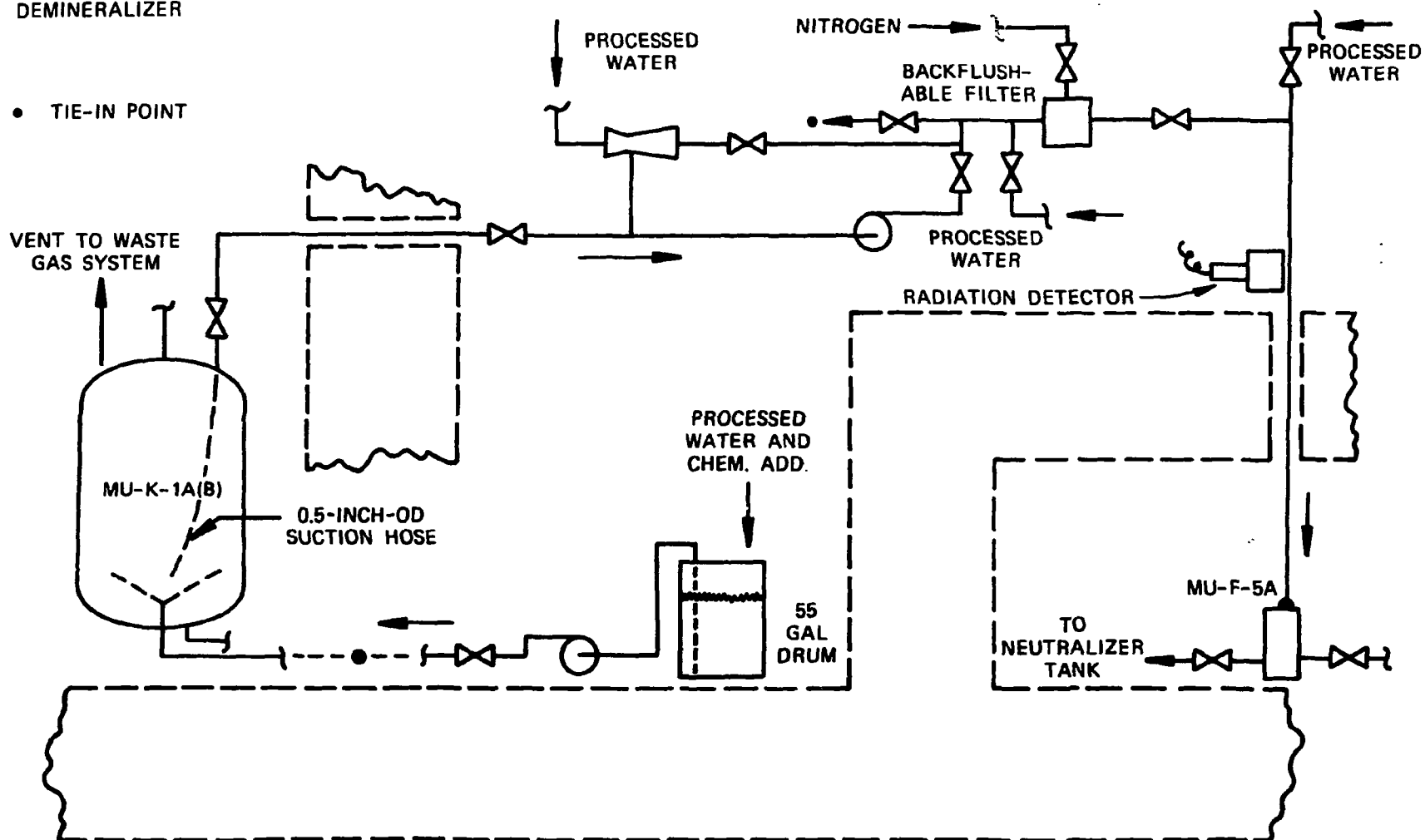


Fig. 11. Flowsheet for elution of the TMI-2 makeup and purification demineralizers.

After the batch additions, the bed is allowed to settle for 24 to 70 h. Typically, 3 to 4 drums, 210 L (55 gal) each, of eluent solution are added per batch contact. Eluates are withdrawn through a suction hose from a depth about 0.3 m (1 ft) below the liquid surface to ensure against the withdrawal of any solids that might be floating on the liquid. The removal is accomplished using either an eductor or a pump to lift the liquid. The eluates are then filtered through a sintered, stainless steel, filter frit (20- μ m rating) before being diluted with processed water (800 ppm B), transferred to in-plant neutralizer tank storage, and finally processed through the existing SDS. Dilution is necessary to minimize personnel exposure during transfer through piping to in-plant tanks. The SDS has been previously described.²⁻⁵

4. CONCLUSIONS AND SUMMARY

An elution process was developed for removal of radiocesium from the TMI-2 demineralizers on the basis of test results from hot-cell experiments with rather small samples of the degraded demineralizer resins. The process was designed to elute the cesium and then resorb it on the zeolites contained in the SDS. The elution conditions were chosen so that the cesium activity of eluates would not produce excessive radiation fields surrounding the pipelines to the SDS system. The process consisted of 17 stages of batch elution. In the initial stage, the resin is contacted with 0.18 M H_3BO_3 . Subsequent stages subject the resin to increasing concentrations of sodium in NaH_2BO_3 -- H_3BO_3 solutions (total B = 0.35 M) and then sodium hydroxide in the final stages. About 96% of the cesium was eluted from the DB resin, and about 56% was eluted from the more highly degraded DA resin.

Small-scale zeolite bed tests showed that the eluates were compatible with SDS processing. Cesium was effectively sorbed (99.99%), and bed performance was unaffected by the presence of relatively small quantities of soluble organic resin degradation products (100 to 200 ppm total C) in the eluates. Attempts to remove the soluble organic compounds by sorption on charcoal were unsuccessful. Clarification tests on the eluates showed that the small quantity of suspended particulates in the eluates could be

effectively removed by gravity settling and then filtration through a stainless steel filter frit having a 10- μ m rating. The final filtration polish of the eluates avoids the unnecessary introduction of fine particulate matter to the SDS and subsequent water management systems. However, it was also shown that unfiltered eluates could be processed by simulated SDS tests without any effects on SDS performance.

5. ACKNOWLEDGMENTS

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7. APPENDIXES

APPENDIX A

ANALYTICAL RESULTS ON EARLY SAMPLES FROM DEMINERALIZERS A AND E

Table A-1 gives the analytical results for (1) a DB sample obtained in April 1983 which consisted primarily of a liquid phase containing very little resin, and (2) a solids sample from the dry and caked resin bed in DA which was obtained in May of 1983. Elution studies could not be carried out on these samples because the quantity of resin was too small. However, the analytical results are reported here because they may be of interest to other workers concerned with the phenomenological aspects of the TMI-2 accident, particularly the migratory pathways of the various fission product radionuclides.

Table A-1. Analyses obtained on early DA and DB samples

Analysis	DB Sample (April 1983)		DA Sample (May 1983)
	Liquid phase	Solid phase	Solid phase
<u>Radionuclides, $\mu\text{Ci/g}$</u>			
^{134}Cs	181	778	15
^{137}Cs	2640	11,200	220
^{90}Sr	14	490	200
^{60}Co		NA ^a	2.0
^{125}Sm		NA	7.4
^{144}Ce		NA	9.9
pH	5.7		
<u>Elemental, ppm</u>			
B	3000		>200
C	1000		>10
Na	7000		>1000
Mg	<1		5
Al	10		50
Si	<3		<5
P	0.1		4
SO_4	9600		10,000
Cl	5		20
K	3		4
Ca	20		50
V			<0.1
Cr	0.3		<1
Mn	0.2		10
La			~1
Ba			40
Cs	30		100
I			100
Te	<1		10
In	0.2		300
Cd	<1		1000
Ag	0.4		600
Rh			<3
Mo			200
Nb	<.1		1
Zr	1		500
Sr	1		4

Table A-1. (continued)

Analysis	DB Sample (April 1983)		DA Sample (May 1983)
	Liquid phase	Resin phase	Resin phase
Rb	6		15
As	<.5		2
Zn	<.2		<1
Cu	1		2
Ni	0.5		100
Co	<.1		3
Fe	10		700
U	0.064	1620	1250
Pu	0.0007	3.6	3520
Fission product isotopic assay, at. %			
⁸⁵ Sr	30	NA	30
⁸⁷ Sr	70	NA	70
		NA	
¹³³ Cs	43	NA	45
¹³⁴ Cs	0.4	NA	0.6
¹³⁵ Cs	14	NA	14
¹³⁷ Cs	42	NA	40
¹²⁷ I	<10	NA	8
¹²⁹ I	<10	NA	84
¹³¹ Ia	<80	NA	8
Fissile isotopic assay, at. %			
²³⁴ U	0.022	0.023	0.021
²³⁵ U	2.23	2.46	2.27
²³⁶ U	0.13	0.072	0.075
²³⁸ U	97.62	97.45	97.64
²³⁸ Pu	<0.07	<0.05	<0.1
²³⁹ Pu	87.85	91.0	90.52
²⁴⁰ Pu	10.29	7.6	8.02
²⁴¹ Pu	1.79	1.4	1.45
²⁴² Pu	<0.05	<0.05	<0.2

^aNA = not analyzed.

APPENDIX B

ANALYTICAL RESULTS OBTAINED ON DA AND DB SAMPLES THAT WERE USED IN FLOWSHEET DEVELOPMENTAL STUDIES

All analyses obtained for these samples are listed in Table B-1. The initial results on the resin phase in the DA sample were suspect in that certain elements appeared unusually high (calcium, aluminum, etc.) and certain of the elemental analysis were rechecked. The repeat analysis of the DA resins are shown in parentheses next to the initial analyses values in Table B-1. The variance in values could be related to the difficulty of taking small analytical samples from the bulk resin phase. The macroscopic examination of the DA resin (see Fig. 2) showed that the resin phase was heterogeneous with regard to resin particle types. It also appears that the DB liquid became contaminated with plutonium from high-burnup fast reactor fuel that had previously been processed in the hot cell where the experimental work was carried out on the demineralizer samples. Thus, conclusions should not be drawn regarding the isotopic composition of the DB liquid phase. The isotopic assays of plutonium in the DB resin and the DA liquid and resin phases appear normal for LWR-produced plutonium.

Table B-1. Analytical results for the DA and DB samples used in batch elution flowsheet studies

Analysis	DA		DB	
	Liquid phase	Resin phase	Liquid phase	Resin phase
<u>Radionuclide, $\mu\text{Ci/g}$</u>				
^{134}Cs	11.03	285	101	1458
^{137}Cs	209	5520	1480	21800
^{90}Sr	6.72	3060	9.46	890
^{60}Co		4.89	NA ^a	NA
pH	7.1		5.7	
<u>Elemental, ppm</u>				
C	164	100,000	950	100,000
B	1000	$\approx 20(1500)$	2000	>200
Na	500	4900(2000)	8500	>1000
Mg	<1	<2400(5)	<1	2
Al	3	970(60)	10	70
Si	<20	<500	<3	<5
P	0.3	50 (3)	0.1	<1
SO_4	900	29,000(4000)	7700	15,000
Cl		20(40)	13	30
K	3	240(6)	2	4
Ca	3	970(50)	15	30
V		<50(<2)		1
Cr	0.6	190(50)	0.5	5
Mn	0.3	70(20)	0.2	5
La				3
Ba		240(10)		<1
Cs		100	30	100
Sn		300	NA	~ 2
In	0.9	240(400)	0.2	30
Cd		730(800)	<1	60
Ag	<3	<500	1.7	30
Rh				<3
Mo		(50)		NA
Sr			4	5
Nb			<0.1	<1
Zr		730(700)	1	6

Table B-1. (continued)

Analysis	DA		DB	
	Liquid phase	Resin phase	Liquid phase	Resin phase
Rb			4	15
As			<0.5	<1
Zn			<0.2	<1
Cu	0.8	170(4)	~1	<1
Ni	1	1700(300)	0.6	40
Co	NA	<20(4)	<0.1	<1
Fe	4	2400(500)	10	200
Y	0.7			NA
U	0.19	2420(2000)	0.11	241
Pu (ppb)	1.3	4600	0.7	440
<u>Long-lived fission products</u>				
¹²⁹ I, ppm		68.5		14.0
⁹⁹ Tc, Bq		<1200		<3900
<u>Fission-product isotopic assay, at. %</u>				
⁸⁵ Rb			30	30
⁸⁷ Rb			70	70
¹³³ Cs	43	49	47	44
¹³⁴ Cs	<1	<1	<1	0.7
¹³⁵ Cs	13	<1	11	13
¹³⁷ Cs	43	49	41	42
¹²⁷ I				<8
¹²⁹ I				<84
¹³¹ I				<8
²³⁴ U	0.021	0.022	0.027	0.024
²³⁵ U	2.19	2.36	2.38	2.24
²³⁶ U	0.072	0.070	0.095	0.086
²³⁸ U	97.71	97.55	97.56	97.64
²³⁸ Pu	<0.1	<0.1	<0.1	0.06
²³⁹ Pu	92.5	91.30	84.5	90.86
²⁴⁰ Pu	6.9	7.4	13.8	7.38
²⁴¹ Pu	0.57	1.3	1.60	1.49
²⁴² Pu	~0.06	~0.1	<0.2	<0.2

^aNA = not analyzed.

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