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PROPERTIES OF RADIOACTIVE WASTES AND WASTE CONTAINERS

STATUS REPORT
OCTOBER 1980 — SEPTEMBER 1981

Nabil Morcos, Ramesh Dayal,
and Allen J. Weiss

NUCLEAR WASTE MANAGEMENT DIVISION
DEPARTMENT OF NUCLEAR ENERGY, BROOKHAVEN NATIONAL LABORATORY
UPTON, LONG ISLAND, NEW YORK 11973

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Principal Investigators:
Nabil Morcos, Ramesh Dayal,
and Allen J. Weiss

Contributors:

W.W. Becker	P.R. Hayde
S. Chen	M.P. Hope
A. Colavito	L.W. Milian

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DONALD G. SCHWEITZER, HEAD
NUCLEAR WASTE MANAGEMENT DIVISION
DEPARTMENT OF NUCLEAR ENERGY, BROOKHAVEN NATIONAL LABORATORY
UPTON, NEW YORK 11973

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EXECUTIVE SUMMARY

The objectives of this research program at BNL are to provide a technical basis that will assist the NRC's capability to predict low-level waste isolation performance, and to establish regulatory criteria. To meet these objectives, the work undertaken in this program was directed at characterizing low-level waste forms, their behavior and their performance. Research performed during the 1981 fiscal year is summarized below.

A study designed to evaluate the leachability of ^{137}Cs, ^{85}Sr, and ^{60}Co from simulated boric acid waste in Portland III cement and to measure the compressive strength of the ensuing waste forms was performed. The simulated waste forms were leached in deionized water using a modified IAEA leaching procedure for periods extending to 229 days. The compressive strength of the specimens was measured before and after their exposure to a leaching environment for 352 days. The waste forms were made with 3%, 6%, and 12% boric acid solutions; the waste-to-cement ratios studied were 0.5 and 0.7.

Increasing the waste-to-cement ratio from 0.5 to 0.7 caused an increase in the leachability of ^{137}Cs from the three boric acid/cement composite formulations. This effect is not noticeable for the leachability of ^{85}Sr.

For a waste-to-cement ratio of 0.7, increasing the boric acid solution concentration (from 3% to 6% and 12%) effectively decreased the leachability of both ^{137}Cs and ^{85}Sr. This trend is less noticeable for a waste-to-cement ratio of 0.5 when comparing composites made with 3% and 6% boric acid solutions, but becomes prominent between composites made with 3% and 12%. The reasons for this decrease in ^{137}Cs and ^{85}Sr leachability with increasing boric acid content are not presently understood.

The extent of ^{85}Sr release in a given time was approximately one-twentieth that of ^{137}Cs from these composites. Cobalt-60, on the other hand, was below the detection limit in the leachates from all the composites (3.0×10^{-2} μCi per 1.5 L samples).

For a waste/cement ratio of 0.5, leaching for 352 days caused a substantial decrease (approx. 50%) in the compressive strength of the specimens.

For a waste/cement ratio of 0.7, although initially the compressive strength of these specimens was approximately 40 to 50% lower than those for w/c ratio of 0.5, it did not decrease further after 352 days of leaching.

The compressive strength of the specimens at waste/cment ratios of both 0.5 and 0.7 was approximately 20 to 38 times higher than the lower acceptable limit (50 psi) set forth in the proposed Code of Federal Regulations, 10CFR Part 61.56.

An experiment was performed to determine the leachability of ^{137}Cs, ^{85}Sr, and ^{60}Co from organic ion exchange resin/cement composites. Portland II and Lumnite (high alumina) cements were used as binders, with waste-to-cement ratios of 1.0 and 1.8 for both cements. In addition, the displacement of the three radiotracers from the ion exchange resins upon mixing the loaded resins with the two cements (before solidification) was measured.

Lumnite cement showed a lower leachability of ^{137}Cs and ^{85}Sr than Portland II cement. Cobalt-60 was below the detection limit in leachates from both types of cement (3.0×10^{-2} μCi per 1.5 L sample). Strontium-85 was observed only in leachates from Portland II cement. These observations may be due to either an isotopic dilution of radiotracers with the cement components or to an actual chemical interaction of the radiotracers with the cement matrices.

The upper surfaces of the Portland II/organic ion exchange resin composites for waste/cement ratios of 1.8 crumbled before leaching was initiated, and the waste forms deteriorated further upon water immersion. The composites made with Lumnite cement, however, maintained their physical integrity throughout the leaching experiment.

The amounts of ^{137}Cs, ^{85}Sr, and ^{60}Co displaced from the ion exchange resins during the paste phase of the solidification process were comparable when mixed with Portland II cement (waste-to-cement ratio of 1.0 and 1.8), but not with Lumnite cement. The displacement of ^{85}Sr from the resins by Lumnite cement was approximately 10% less than that of ^{137}Cs and ^{60}Co at waste-to-cement ratios of 1.0 and 1.8. The displacement of ^{137}Cs and ^{60}Co by Lumnite cement was comparable to that with Portland II cement.

A study correlating the leachability of ^{137}Cs from small-scale to large-scale cement forms was performed. The waste forms consisted of organic ion exchange resins incorporated in Portland I cement, with a waste-to-cement ratio of 0.6, and a water-to-cement ratio of 0.4. The samples varied from 1 in. x 1 in. to 22 in. x 22 in. (diameter x height) in size. Leaching data extending over a period of 169 days was obtained. A method based on semi-infinite plane source diffusion model was applied to analyze the leaching data. An effective bulk diffusion coefficient was calculated from the leach data. A derived mathematical expression allows prediction of the amount of ^{137}Cs leached from the forms as a function of leaching time and waste form dimensions. A reasonably good agreement between the experimental and calculated data is obtained.

H^+, Na^+, Cs^+, and Sr^{+2} forms cationic resins, loaded with ^{137}Cs and ^{85}Sr, were mixed in varying ratios with anionic resins in SO_4^{-2} form, and subsequently solidified in bitumen. The leachability and physical integrity of the resulting composites were evaluated.

The presence of anionic resins in the sulfate form (50% by weight of resins) increased the leachability of ^{137}Cs from the waste forms by approximately two orders of magnitude (from 5×10^{-4} to approx. 3×10^{-2}), and by approximately one order of magnitude for ^{85}Sr (from 1.6×10^{-4} to 1.5×10^{-3}). The physical integrity of the forms deteriorated during leaching, as the amount of SO_4^{-2} resin increased.

A total of 30 DOT 17H drums from three different manufacturing lots were tested to determine if they met the specifications set forth in the Code of Federal Regulations 49 CFR Parts 178-118-66. Only one drum was found to meet the specifications.

Under a subcontract from BNL, a study was undertaken at the Georgia Institute of Technology to investigate the effects of radiation on the physical and chemical properties of organic ion exchange resins, and corrosion effects on the walls of the irradiation containers in contact with the resins and their radiolytic byproducts. Organic ion exchange resins were irradiated up to a dose of 5×10^9 rad. The irradiation containers were fabricated from stainless steel type 304.

Increasing the total absorbed dose of anionic resins from 10^8 to 5×10^9 caused an increase in liquids generated, whereas for cationic resins no noticeable increase in liquids was observed. The presence of amines was detectable at lower doses and ammonia at the higher doses for irradiated anionic resins. Both types of resins showed fracturing of the resin beads and layer shedding after irradiation. The color of the irradiated resins were darker. This effect was more pronounced in cationic resins.

The pH of the liquid phase resulting from irradiating anionic resins decreased with increasing absorbed dose. The pH data from the liquid phase obtained from cationic resins does not show a consistent trend. Analysis of aqueous extracts from irradiated anionic resins revealed that total dissolved carbon, nitrogen, ammonia, monoethyl, and dimethyl amines increased with the absorbed dose, but trimethyl amine decreased with increasing the dose. Extracts from cationic resins show an increasing trend of total dissolved carbon and sulfate with dose.

Gases detected following irradiation of resins were: H_2, O_2, N_2, CO_2, CO, CH_4, C_2H_6, C_3H_8, C_4H_{10}, and sulfur gas (SO_2, SO_3). Irradiated anion resins (0.8×10^9 rad) yielded a total volume of gases (cc/g) that was approximately twice that obtained from cation resins (2.5×10^9 rad).

Chemical analyses of irradiated cationic resins indicate that cationic resin solids maintain essentially the same chemical composition for total absorbed doses up to 5×10^9 rad, whereas for anionic resins, decomposition was observed.

Radiolytic by-products resulting from irradiating cationic resins did not significantly attack Type 304 stainless steel, whereas those from anionic resins caused significant localized corrosion. The attack was in the form of etching under surface deposits and pitting. This type of localized corrosion might cause perforation of the container walls at longer exposures.

PROPERTIES OF RADIOACTIVE WASTES AND WASTE CONTAINERS
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INTRODUCTION

This report summarizes work performed during the 1981 fiscal year at Brookhaven National Laboratory for the NRC-sponsored program, "Properties of Radioactive Wastes and Waste Containers," under contract DE-AC-02-76CH00016, FIN-A-3027.

Licensing of near surface low-level radioactive waste disposal sites and waste forms/containers requires the ability to predict the dispersibility of radionuclides from waste forms and waste containers disposed in burial sites. Basic concerns in licensing radioactive waste forms and containers are their dimensional stability and the potential for migration of the radionuclides enclosed therein in a near- and long-term predictable fashion. To assess these concerns, a data base is needed for evaluating the acceptability of solidified low-level radioactive waste packages for disposal. Furthermore, the need to develop test procedures and methodologies exists to enable the prediction of long-term performance of waste forms based on short-term laboratory tests.

The objectives of the research program at BNL are to provide an improved understanding of phenomena, testing methodology and data. This improves the NRC's capability to predict low-level waste isolation performance, and to provide a better technical basis for regulatory standards. The areas addressed to meet these objectives during the 1981 fiscal year were:

- Leachability and compressive strength of boric acid waste in Portland III cement. The tracers used for the study were ^{137}Cs, ^{85}Sr, and ^{60}Co.

- Leachability of ^{137}Cs, ^{85}Sr, and ^{60}Co from organic ion exchange resin/Portland III and Lumnite cements.

- Displacement of ^{137}Cs, ^{85}Sr, and ^{60}Co from organic ion exchange resins upon mixing with Portland II and Lumnite cements.

- Leachability of organic ion exchange resins/Bitumen composites using resins in the H^+, Na^+, Cs^+, Sr^{+2}, and SO_4^{-2} forms, and ^{137}Cs and ^{85}Sr tracers.

- Correlation of ^{137}Cs leachability from small-scale (laboratory) samples to large-scale waste forms.

- Hydrostatic testing of DOT 17H drums.

1. LEACHABILITY AND COMPRESSIVE STRENGTH OF BORIC ACID WASTE IN PORTLAND III CEMENT (P. Hayde and N. Morcos)

1.1 Introduction

Boric acid waste and its sodium salts are a major constituent (up to 12% by weight) of radioactive waste derived from pressurized water reactors. The interaction of this waste with the cement matrix, sometimes prevents solidification, resulting in waste forms with poor physical integrity. Work was performed earlier in our laboratory on optimized process parameters and the treatment of boric acid waste prior to solidification in Portland III cement.(1,2) This earlier work indicates that adjustment of the boric acid waste pH to a value of 10 to 12 assures proper solidification for waste to binder ratios of 0.5 and 0.7.

1.2 Experimental

Samples incorporating simulated boric acid waste in Portland III cement matrix were made at two different waste-to-cement ratios (w/c) of 0.5 and 0.7. Boric acid solutions of 3%, 6% and 12% (by weight) were solidified in Portland III cement at each of these w/c formulations. Control samples for compressive strength testing consisted of Portland III cement only. The leached samples were also evaluated for their compressive strength after a 352-day period of leaching. All samples were prepared in five replicates.

1.2.1 Specimen Preparation

Stock solutions containing approximately 3, 6, and 12 weight percent boric acid were prepared. The pH of these solutions was adjusted to approximately 12, by the addition of sodium hydroxide. The stock solution compositions are shown in Table 1.1.

Table 1.1

Boric Acid Stock Solutions Composition[a]

Waste Components	3% Solution	6% Solution	12% Solution
Deionized H_2O	95.2%	90.5%	81.7%
H_3BO_3	2.9%	5.8%	11.1%
NaOH	1.9%	3.7%	7.2%

[a]Composition expressed as weight percent.

These solutions, hereafter referred to as "simulated waste," were heated to 170°F prior to solidification in Portland III cement so as to simulate actual solidification conditions at power reactor sites. The dimensions of the solidified samples were 4.6 cm (diameter) by 9.1 to 9.5 cm (height). The leaching samples contained one microcurie each of ^{137}Cs, ^{85}Sr, and ^{60}Co. Control samples for compressive strength testing were made with Portland III cement and water. The compositions of simulated boric acid waste forms are summarized in Table 1.2. All samples were cured for a period of 35 days in sealed polyethylene containers.

Table 1.2

Composition of Boric Acid Waste Composites[a]

		H_3BO_3 Stock Solution		
Waste/Cement Ratio	Matrix Component	Set A 3% H_3BO_3 Solution	Set B 6% H_3BO_3 Solution	Set C 12% H_3BO_3 Solution
0.5	H_2O	93.6	89.0	83.1
	H_3BO_3	2.9	5.7	11.3
	NaOH	1.9	3.6	7.3
	Portland III	196.7	196.7	203.3
0.7	H_2O	111.2	107.0	98.7
	H_3BO_3	3.4	6.9	13.4
	NaOH	2.2	4.4	8.7
	Portland III	166.7	168.8	172.7

[a] Weight in grams.

The ratios by weight of the components (H_3BO_3 and NaOH) to cement (Portland III) used to make the waste forms are summarized in Table 1.3, together with ratios of NaOH to H_3BO_3.

Table 1.3

Ratios of NaOH, H_3BO_3, and Cement in the Waste Composites

% H_3BO_3 Solution Used	Waste/Cement	H_3BO_3/Cement x 100	NaOH/Cement x 100	NaOH/H_3BO_3
3%	0.5	1.5	1.0	0.7
	0.7	2.0	1.3	0.7
6%	0.5	2.9	1.8	0.6
	0.7	4.1	2.6	0.6
12%	0.5	5.6	3.6	0.7
	0.7	7.8	5.0	0.7

1.2.2 Specimen Leaching

The specimens were leached using a modified IAEA leaching procedure(3) consisting of 24-hour leaching periods for the first ten days. Thereafter, the leaching period was extended to a week, and later on to a month, based on the amount of activity observed in the leachates. The leachates were counted under identical geometries using a Ge(Li) detector system, and their radioactivity contents were determined.

1.2.3 Compressive Strength Testing

Compressive strength measurements were performed on specimens after the cure period and on the leached specimens at the end of the leaching experiment.

The compressive strength of the specimens was measured using a Soil-Test concrete tester in accordance with ASTM C 39-72. Prior to testing, the samples were capped on each end with a sulfur-based capping compound approved for use with concrete specimens.

1.3 Results and Discussion

1.3.1 Leaching Data

As shown in Table 1.2, three sets (A, B and C) of waste forms were evaluated, each set corresponding to one of the three boric acid solution concentrations (3%, 6% and 12% by weight) considered. In addition, in each set, two different waste-to-cement ratios (w/c) were studied (w/c = 0.5 and 0.7).

The cumulative fractional release data from these specimens are summarized in Tables A.1 through A.6 for ^{137}Cs and in Tables A.7 through A.12 for ^{85}Sr (Appendix A). The errors quoted represent only the statistical errors associated with the counting of each fraction. These data are also shown graphically in Figures 1.1 through 1.12 for ^{137}Cs and Figures 1.13 through 1.24 for ^{85}Sr. Each pair of figures shown on a page presents leach data of five replicate samples for a given formulation and the average cumulative fractional release of the five replicates. These average cumulative fractional releases were normalized for the volume to geometric surface area (V/S) of the waste forms. Cobalt-60 in the leachates from all samples was below the detection limit (3.0×10^{-2} µCi per 1.5 L sample or an incremental fractional release of less than 3.0×10^{-2}) of the experimental system. Based on this detection limit, and the amount of ^{60}Co present in the waste forms (1 µCi), this radiotracer may totally leach out of the forms without detection. However, earlier work(5) has shown that the leachability of ^{60}Co from BWR regenerative waste solidified in Portland II cement was approximately two orders of magnitude lower than that of ^{137}Cs.

The curves shown in Figures 1.1-1.24 were computer-generated, using a cubic spline interpolation between data points. In the absence of additional data between known points, any reasonable method may be employed to connect adjacent points. These curves do not imply any expected or known leaching behavior.

(Continued Page 18)

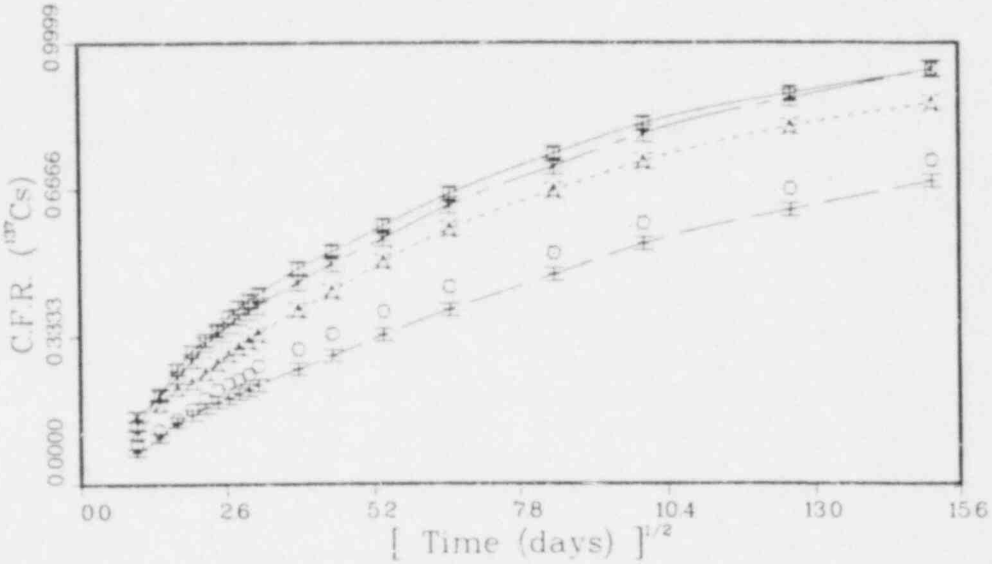

Figure 1.1 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm).

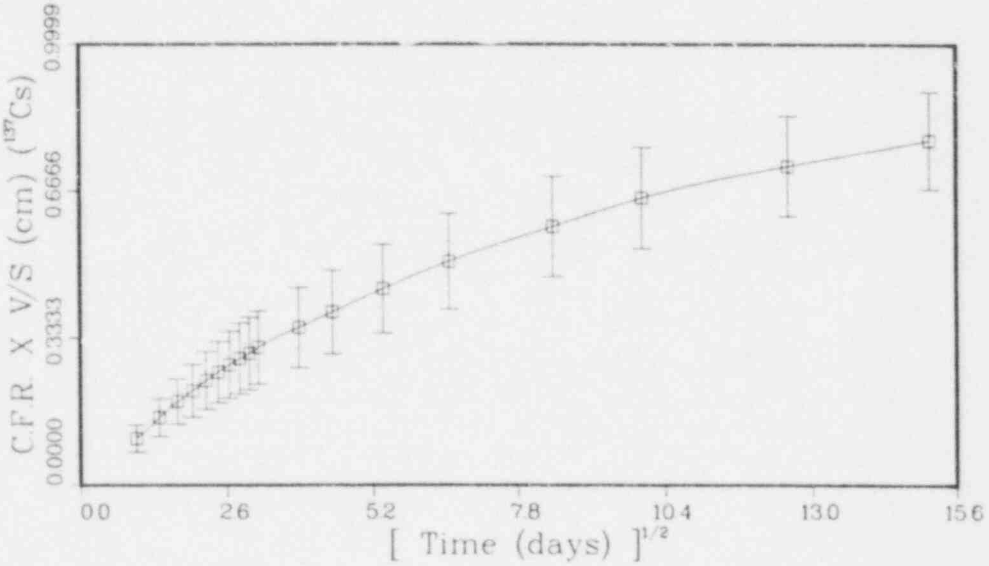

Figure 1.2 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm). The average CFR was normalized for V/S.

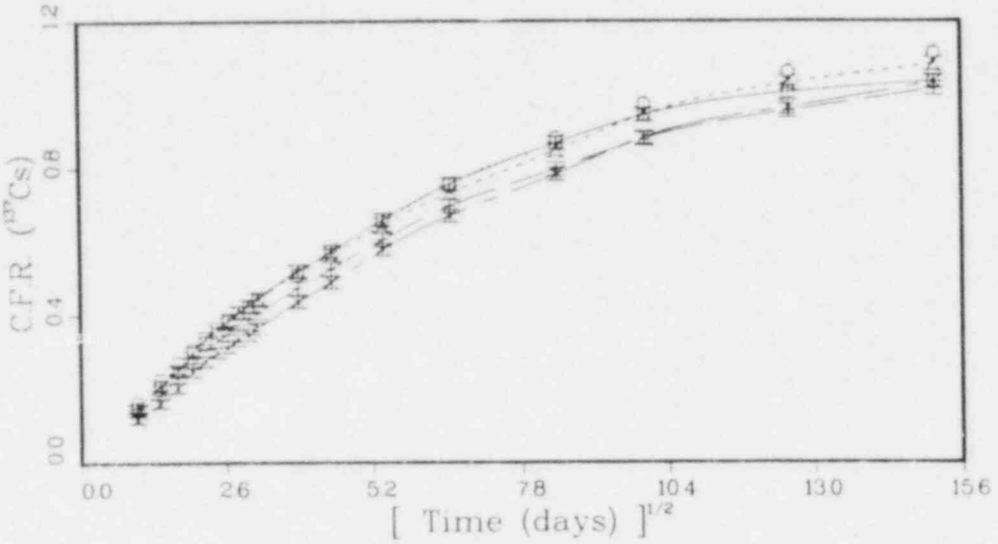

Figure 1.3 ^{137}Cs cumulative fractional release of vs $(\text{time})^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.7 ; V/S = 0.94 cm).

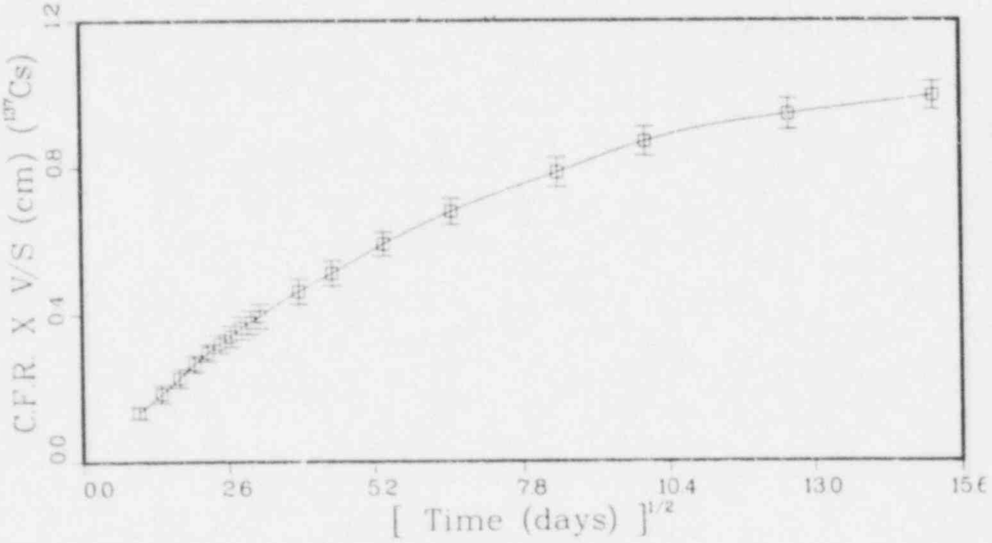

Figure 1.4 Average cumulative fractional release of ^{137}Cs vs $(\text{time})^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.7 ; V/S = 0.94 cm). The average CFR was normalized for V/S.

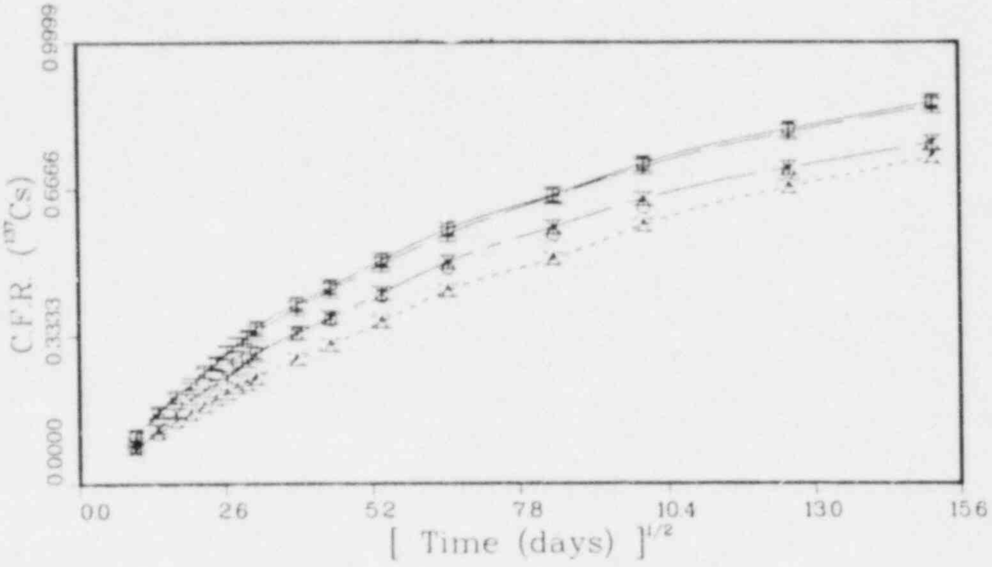

Figure 1.5 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm).

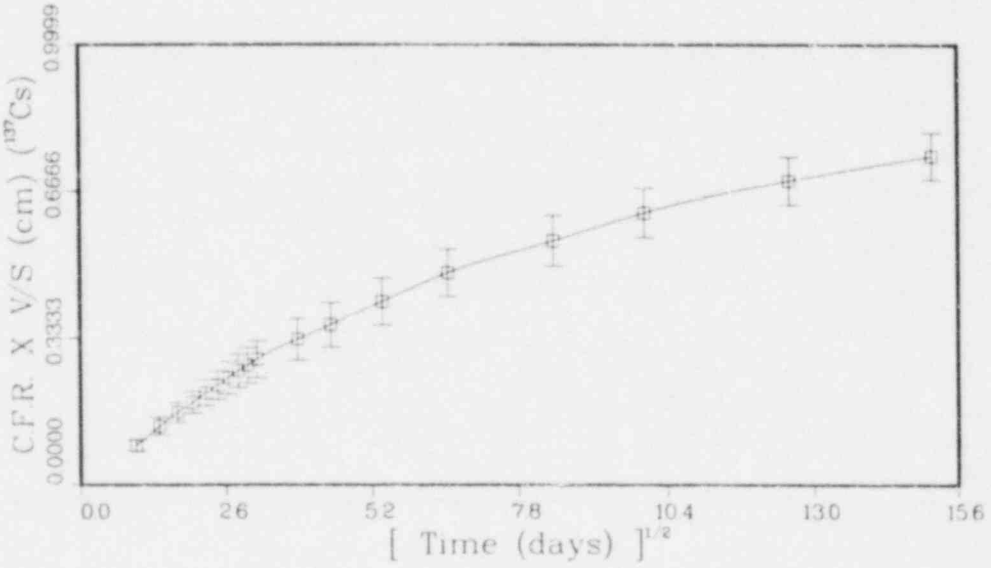

Figure 1.6 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm). The average CFR was normalized for V/S.

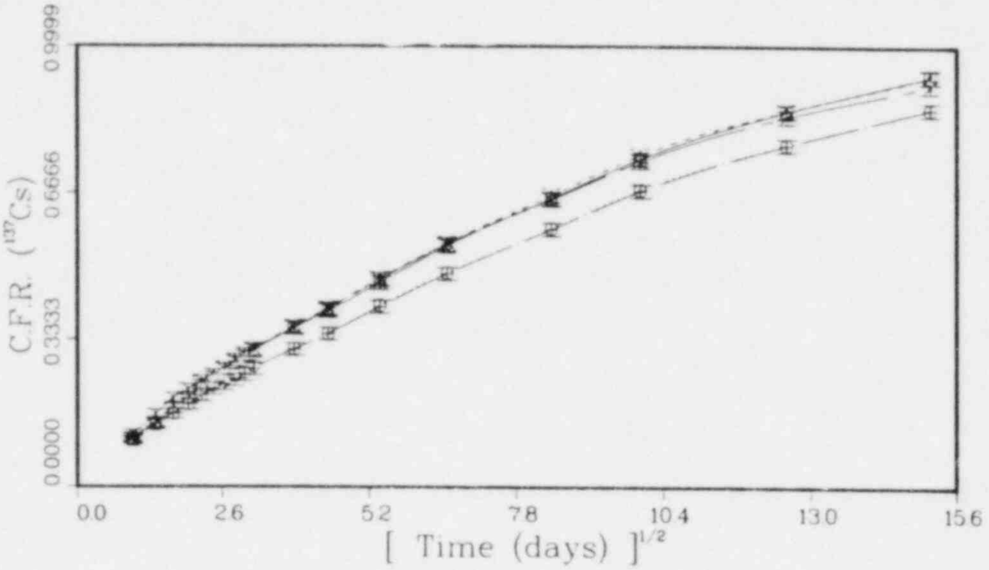

Figure 1.7 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm).

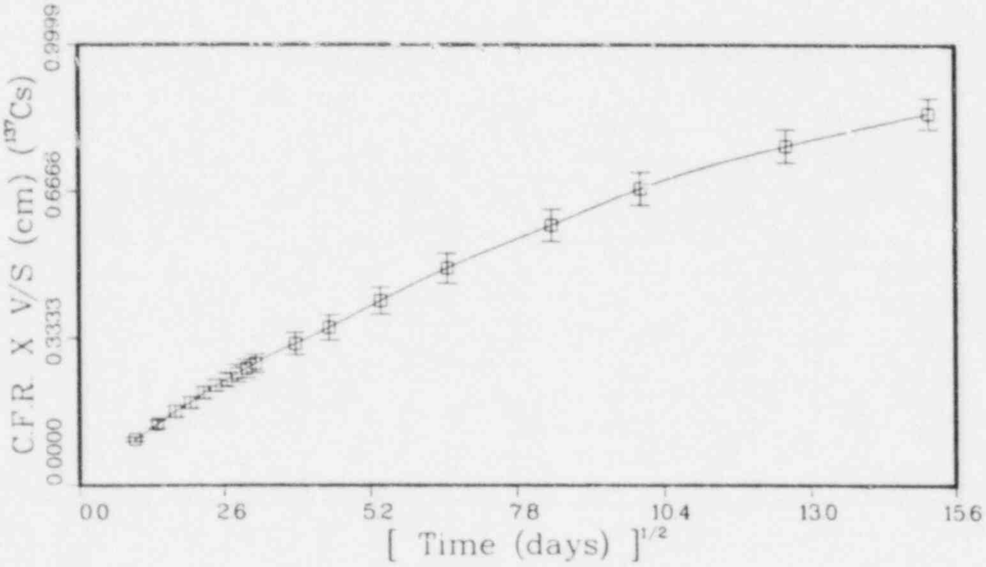

Figure 1.8 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm). The average CFR was normalized for V/S.

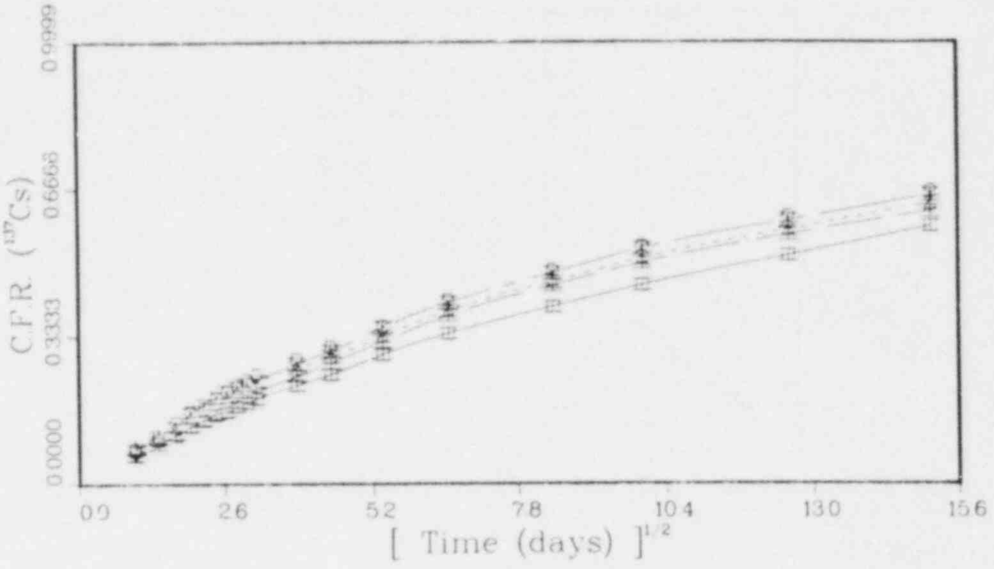

Figure 1.9 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm).

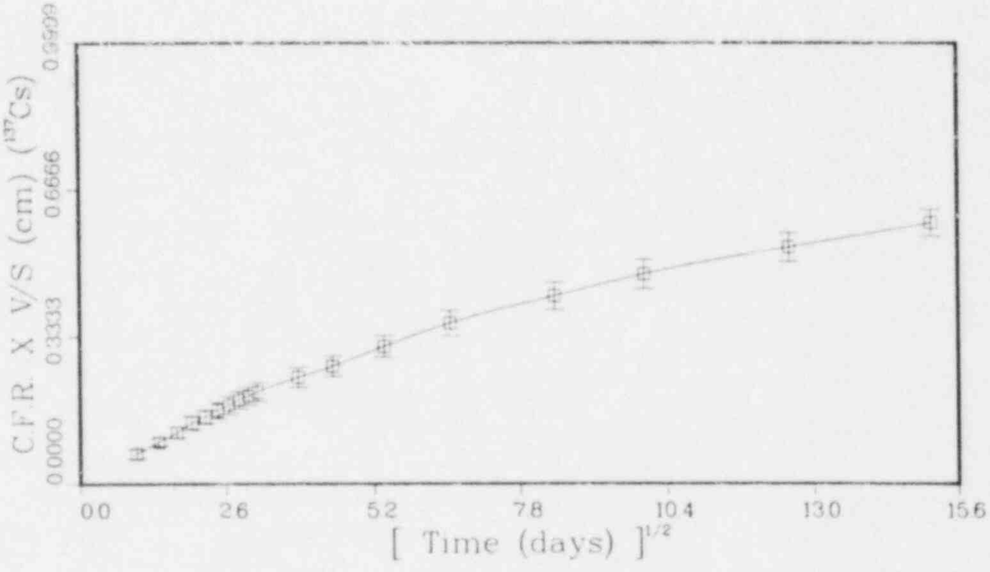

Figure 1.10 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm). The average CFR was normalized for V/S.

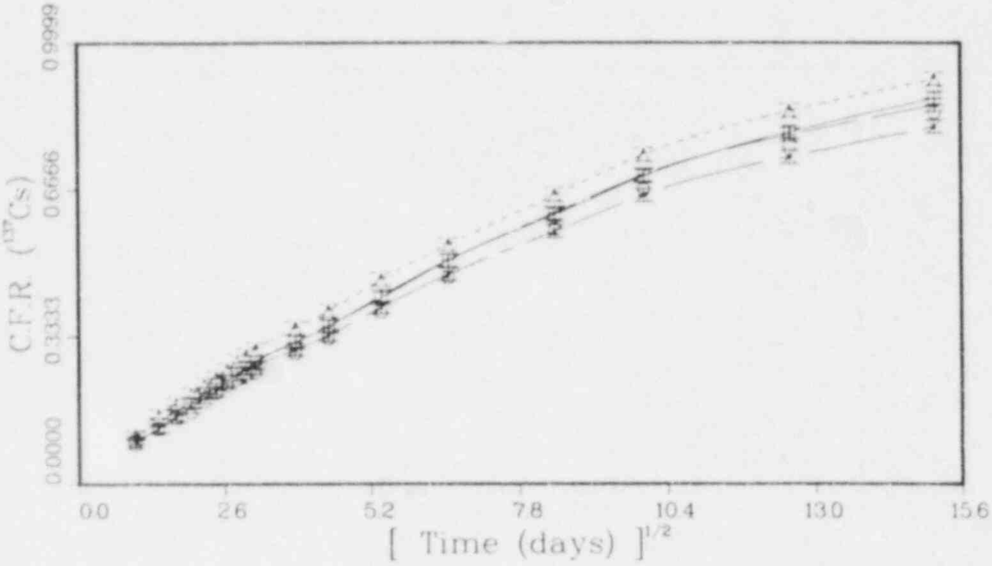

Figure 1.11 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm).

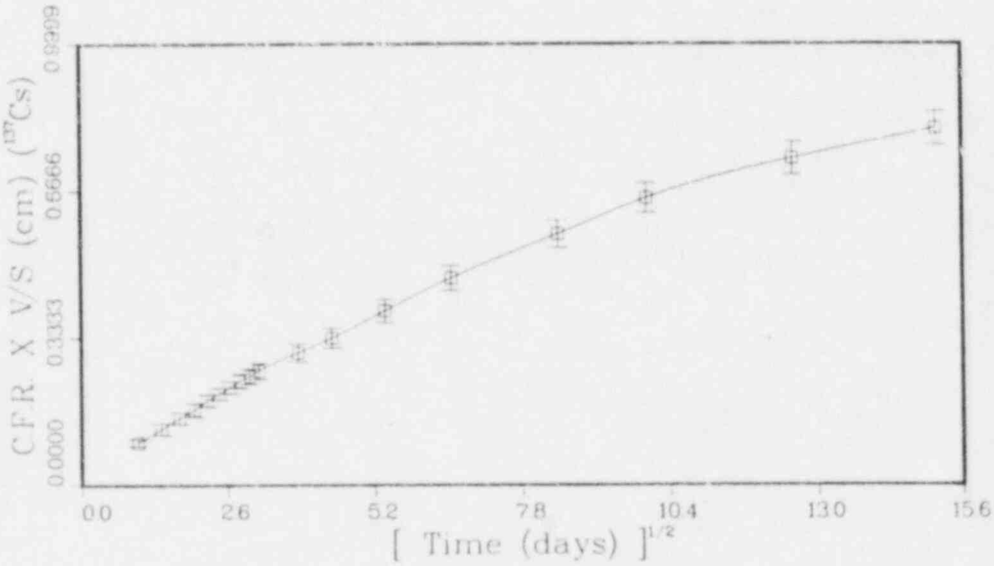

Figure 1.12 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm). The average CFR was normalized for V/S.

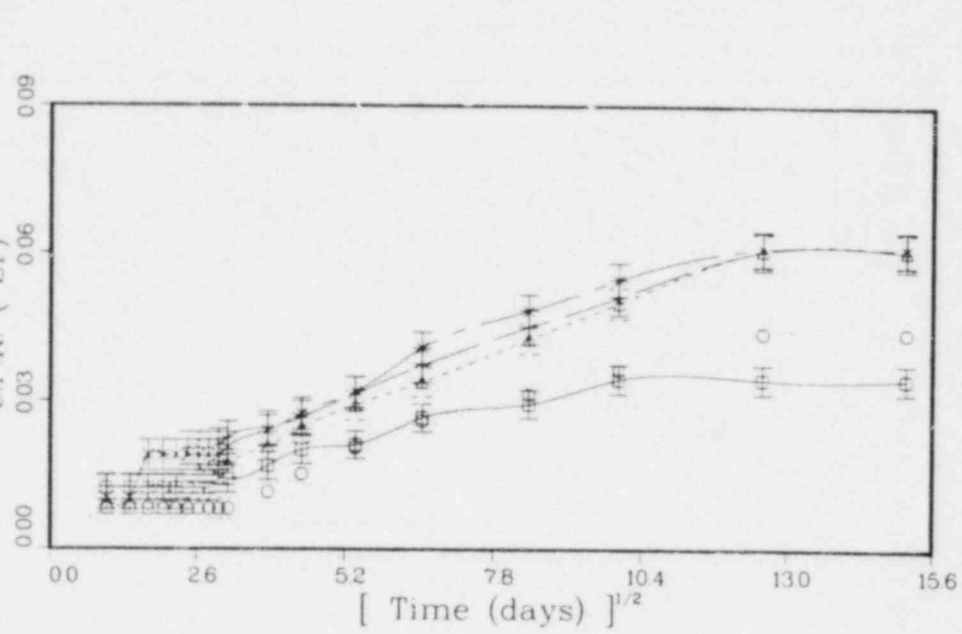

Figure 1.13 ^{85}Sr cumulative fractional release vs (time)$^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm).

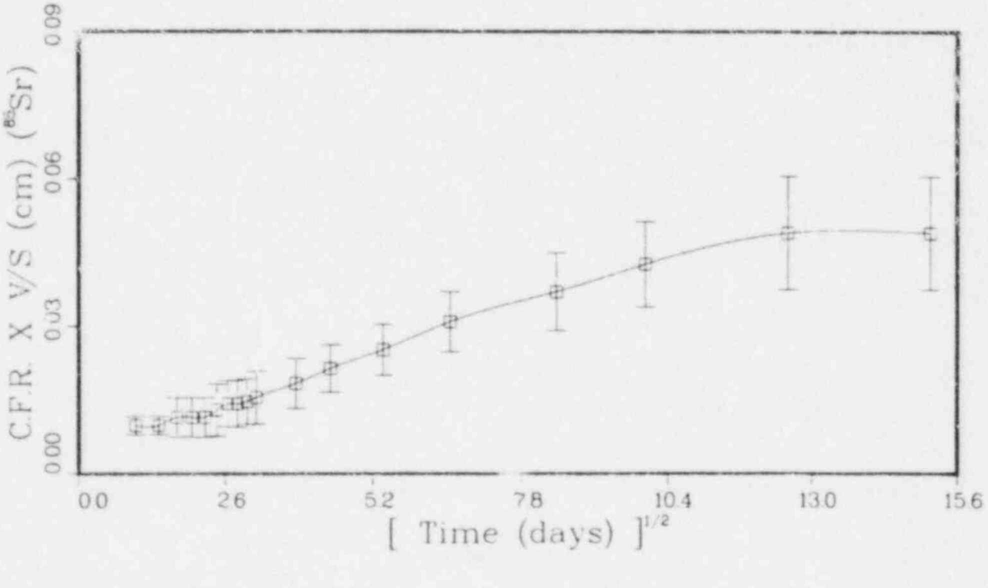

Figure 1.14 Average cumulative fractional release of ^{85}Sr vs (time)$^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm). The average CFR was normalized for V/S.

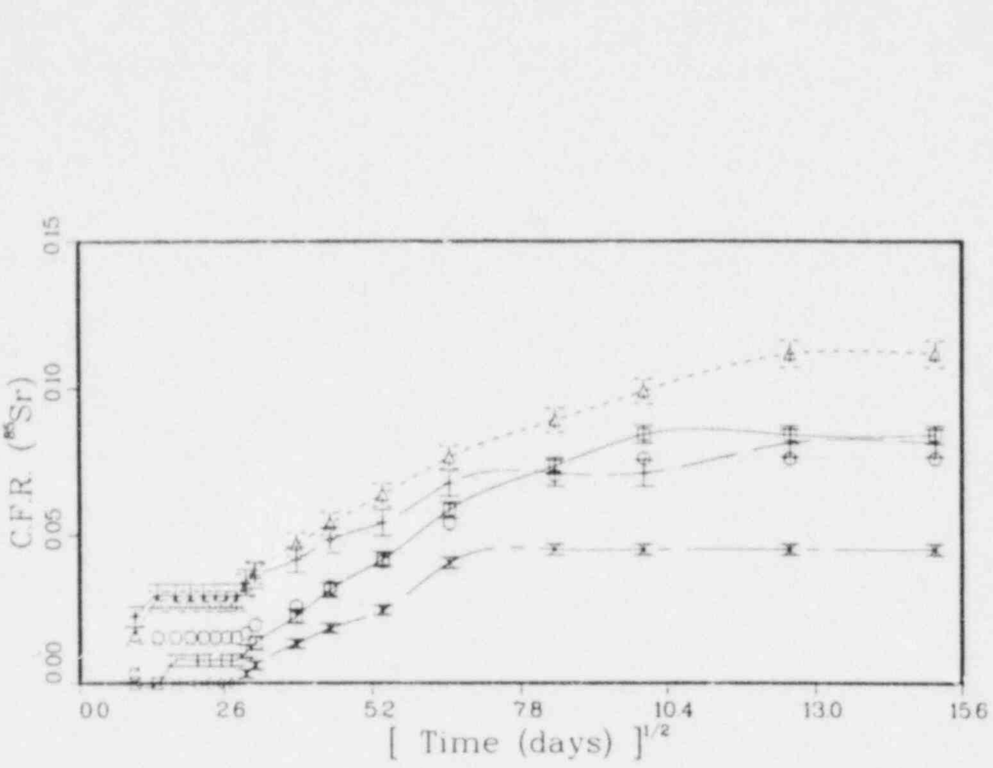

Figure 1.15 ^{85}Sr cumulative fractional release vs $(time)^{1/2}$ from 3% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm).

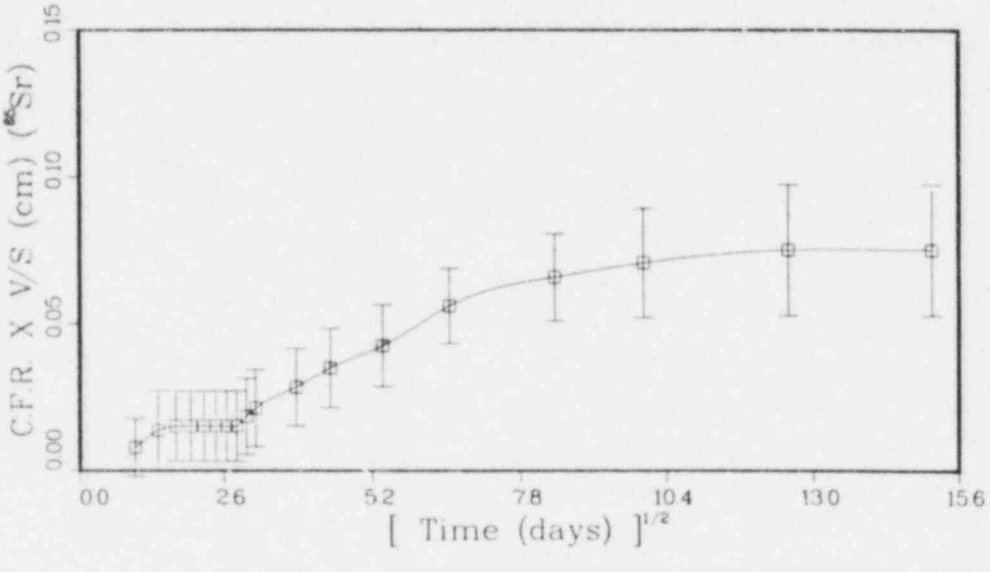

Figure 1.16 Average cumulative fractional release of ^{85}Sr vs $(time)^{1/2}$ from H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm). The average CFR was normalized for V/S.

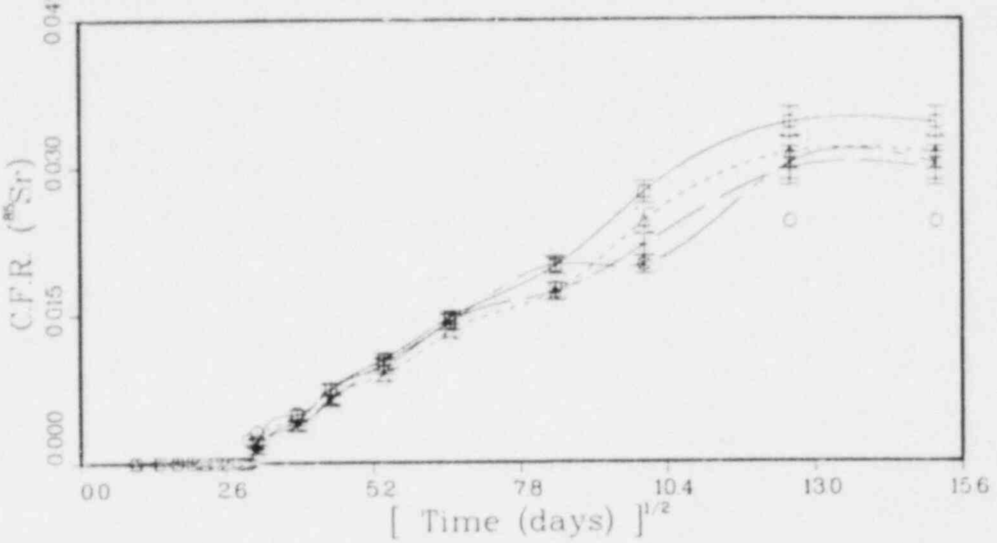

Figure 1.17 ^{85}Sr cumulative fractional release vs $(\text{time})^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm).

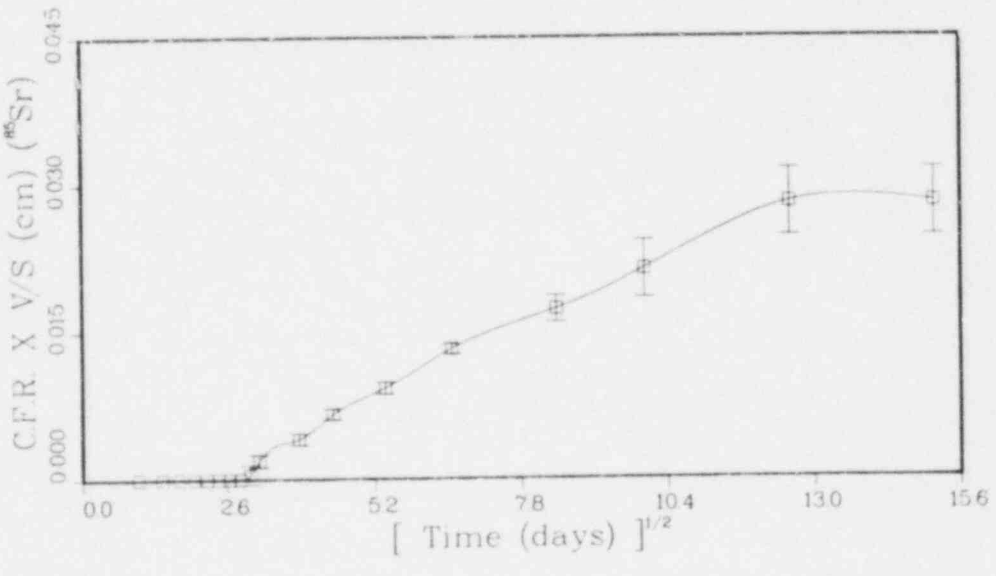

Figure 1.18 Average cumulative fractional release cf ^{85}Sr vs $(\text{time})^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm). The average CFR was normalized for V/S.

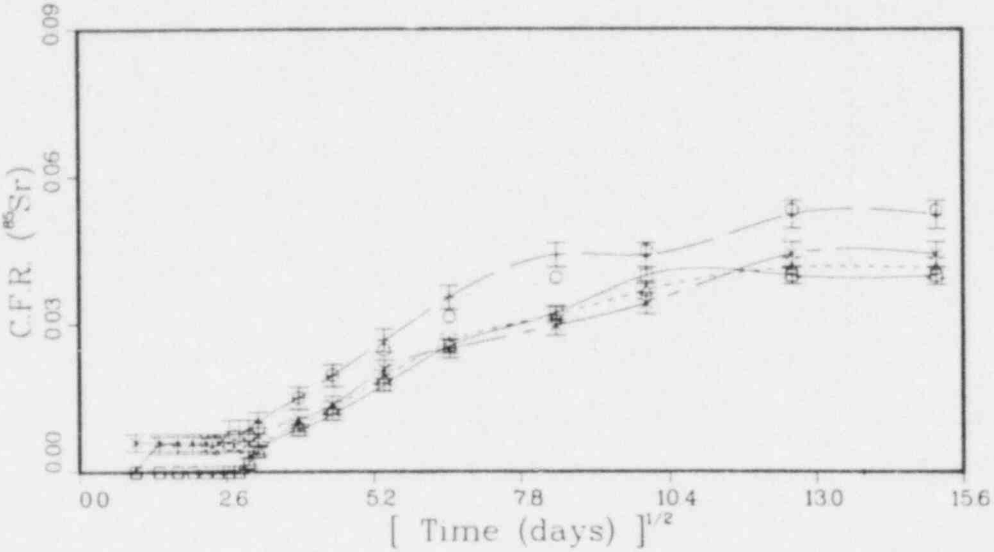

Figure 1.19 ^{85}Sr cumulative fractional release vs (time)$^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm).

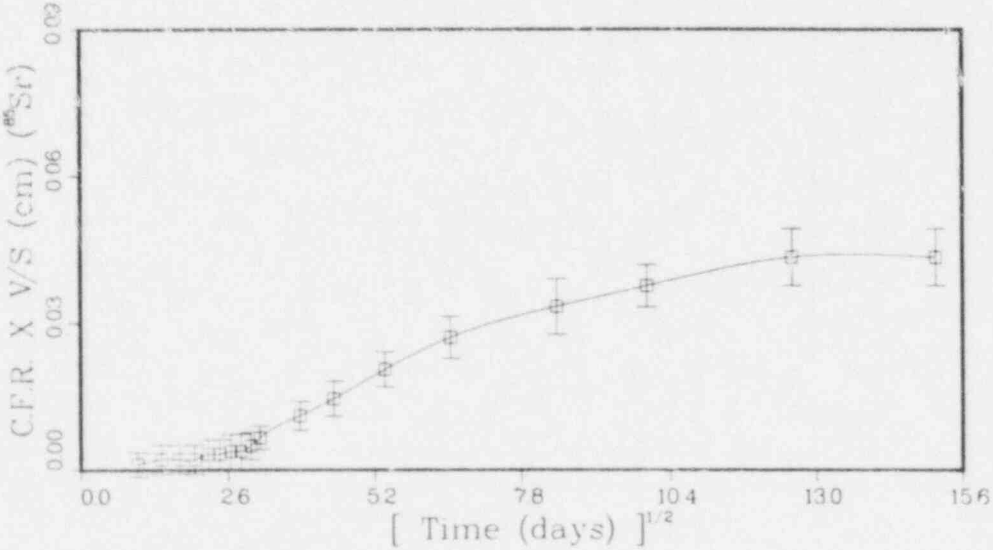

Figure 1.20 Average cumulative fractional release of ^{85}Sr vs (time)$^{1/2}$ from 6% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm). The average CFR was normalized for V/S.

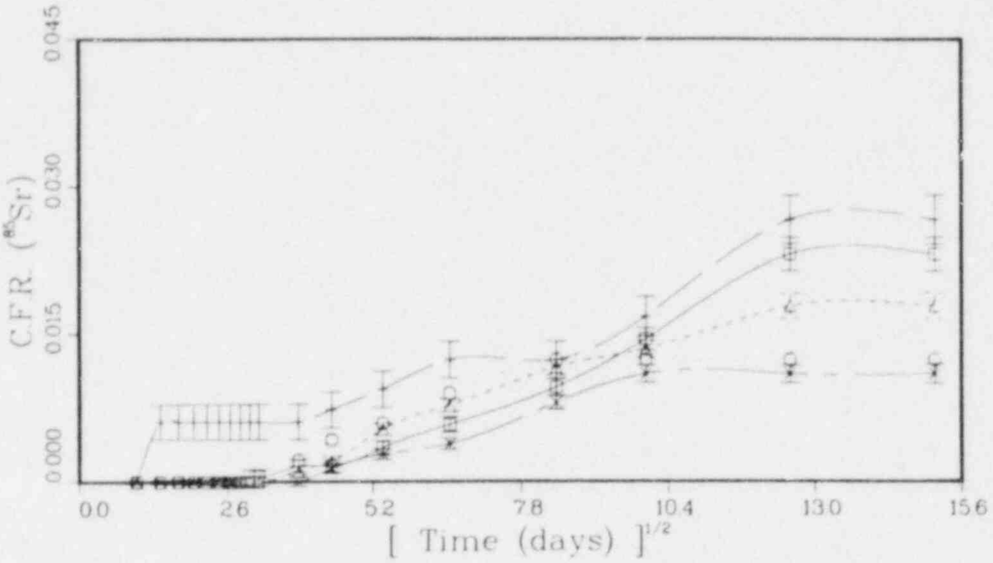

Figure 1.21 ^{85}Sr cumulative fractional release vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm).

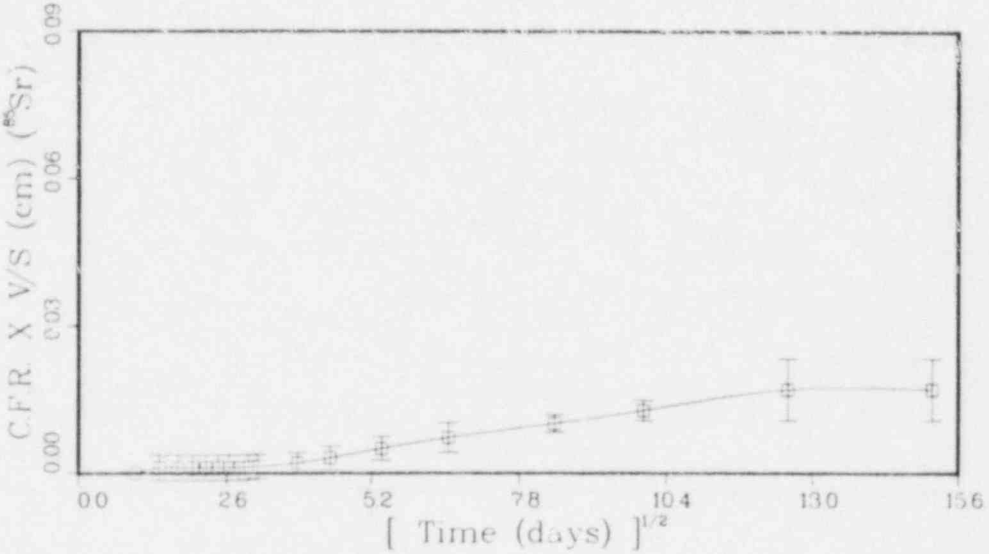

Figure 1.22 Average cumulative fractional release of ^{85}Sr vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.5; V/S = 0.94 cm). The average CFR was normalized for V/S.

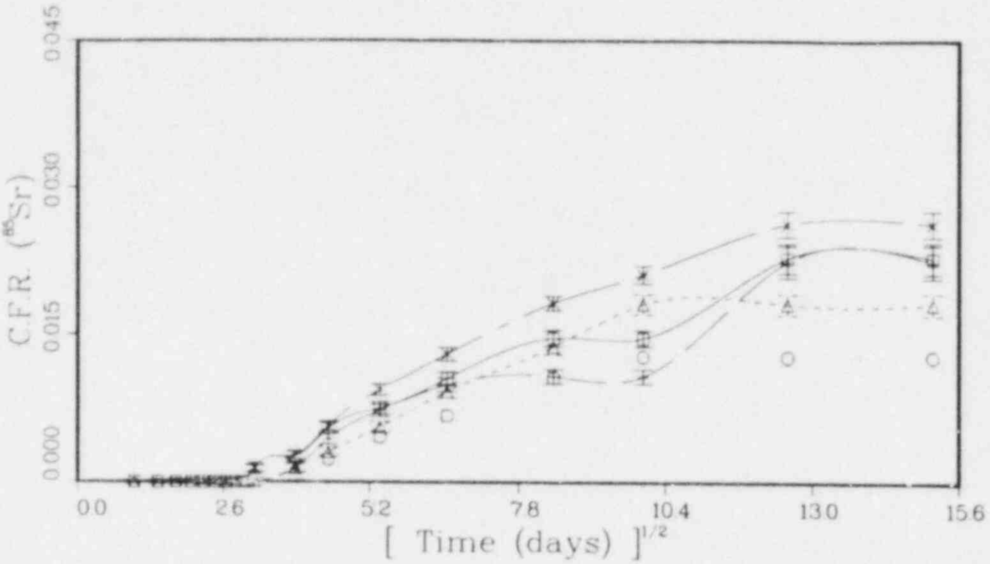

Figure 1.23 ^{85}Sr cumulative fractional release vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm).

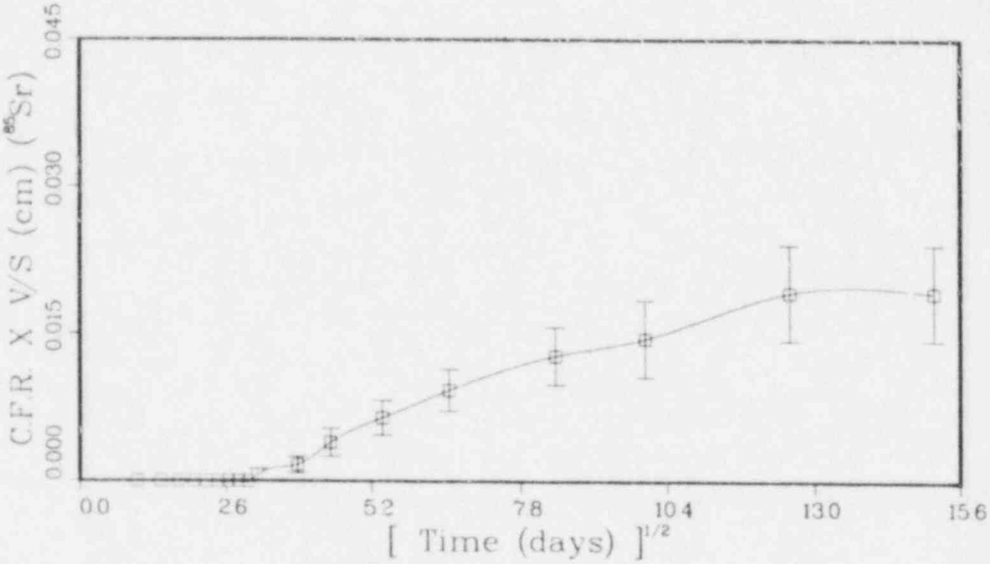

Figure 1.24 Average cumulative fractional release of ^{85}Sr vs (time)$^{1/2}$ from 12% H_3BO_3 waste composites (w/c = 0.7; V/S = 0.94 cm). The average CFR was normalized for V/S.

The average cumulative fractional releases for a given formulation, based on five replicates after 229 days of leaching, are summarized in Table 1.4.

Table 1.4

Average ^{137}Cs and ^{85}Sr Cumulative Fractional Releases (CFR) After 227 Days of Leaching

Waste/ Cement	Radiotracer	Average CFR From Composites Incorporating:[a,b]		
		3% H_3BO_3 Solution	6% H_3BO_3 Solution	12% H_3BO_3 Solution
0.5	^{137}Cs	83.3 ± 11.8	79.9 ± 5.7	63.0 ± 3.2
0.7	^{137}Cs	105.6 ± 3.8	89.8 ± 3.8	86.1 ± 4.0
0.5	^{85}Sr	5.20 ± 1.2	4.74 ± 0.6	1.82 ± 0.7
0.7	^{85}Sr	7.98 ± 2.4	3.00 ± 0.4	2.05 ± 0.5
	^{60}Co	—[c]	—[c]	—[c]

[a] The error presented is the standard deviation from the mean of the five replicates for each formulation.

[b] CFR is expressed in percent.

[c] ^{60}Co was below the detection limit (3.0×10^{-2} μCi per 1.5 L).

The following observations and conclusions are noted:

- Increasing the waste-to-cement ratio from 0.5 to 0.7 resulted in an increase in the leachability of ^{137}Cs from the three boric acid/cement composite formulations (3%, 6% and 12% boric acid solutions as waste). This effect is not noticeable for ^{85}Sr release.

- For a waste-to-cement ratio of 0.7, increasing the boric acid solution concentration (from 3% to 6% and 12%) effectively decreased the leachability of both ^{137}Cs and ^{85}Sr. This trend is less pronounced for a waste-to-cement ratio of 0.5 when comparing composites made with 3% and 6% boric acid solutions, but becomes prominent when comparing the leachability of ^{137}Cs and ^{85}Sr from composites made with 3% to those made with 12% boric acid solutions. The reasons for the observed decrease in ^{137}Cs and ^{85}Sr leachability with increasing boric acid content of the composites are not presently understood, but may explain the low (below detection limit) cumulative fractional releases of ^{85}Sr in these forms.

- The leachability of ^{85}Sr was approximately one-twentieth that of ^{137}Cs from these composites. This may be attributed to the assimilation of the divalent strontium into the cement matrix, presumably substituting for calcium. Furthermore, the extent of displacement of the two cations from the resins to the matrix will be different. This factor will have to be taken into consideration for a more quantitative evaluation of the data.

- ^{60}Co in the leachates from all samples was below the detection limit (3.0×10^{-2} μCi per 1.5 L sample) of the experimental method. Earlier work(3) has shown that the leachability of ^{60}Co from B.W.R. regenerative waste solidified in Portland II cement was approximately two orders of magnitude lower than that of ^{137}Cs.

1.3.2 Compressive Strength Data

The compressive strength of some composites was measured immediately after curing. These samples included control samples (composed of Portland III cement and water), and samples of identical composition to those which were leached. All the composites, which were leached (for a duration of 352 days), were also evaluated for their compressive strength.

The compressive strength data from these measurements are summarized in Table 1.5 and graphically shown in Figure 1.25. All reported values represent an average value of five replicates.

Table 1.5

Average Compressive Strength of H_3BO_3/Portland III Cement Composites Before and After 352 days of Leaching.

	Compressive Strength (Psi)			
	Initially	After Leaching 352 (days)	Initially	After Leaching 352 (days)
Formulation	w/c = 0.5		w/c = 0.7	
Control[a]	3400 ± 2%	---	1400 ± 26%	---
3% Boric Acid	2600 ± 16%	1900 ± 16%	1100 ± 23%	1000 ± 12%
6% Boric Acid	2400 ± 35%	1200 ± 9%	1700 ± 18%	1400 ± 12%
12% Boric Acid	3300 ± 9%	1600 ± 14%	1900 ± 12%	1300 ± 17%

[a] Samples consisted of water and Portland III cement only.

The data indicate that, for a waste-to-cement ratio of 0.5, leaching for 352 days caused a substantial decrease (approx. 50%) in the compressive strength of the composites. The data derived from the composites with a waste-to-cement ratio of 0.7 indicate that, although their initial compressive strength is lower than those with a w/c ratio of 0.5 (by approx. 40 to 50%), their compressive strength did not decrease substantially after 352 days of leaching. It should be noted that the compressive strength of these composites is still approximately 20 to 38 times higher than the lower limit (50 psi) set forth in the proposed Code of Federal Regulations, 10CFR Part 61.56.

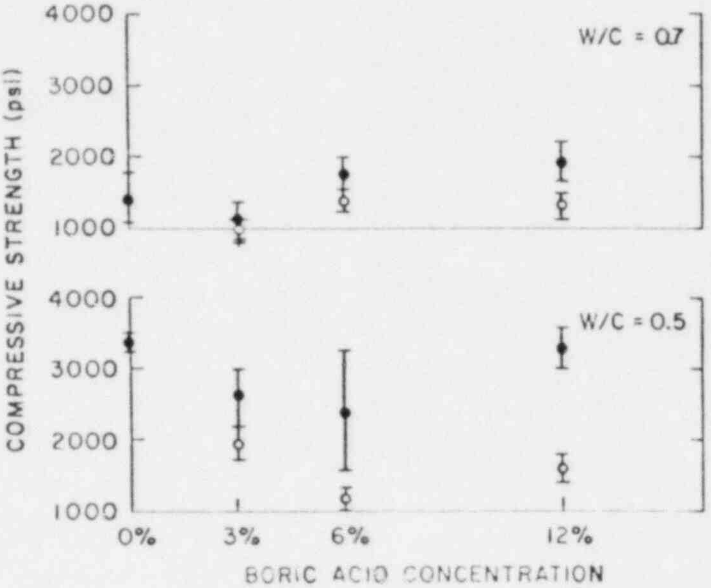

Figure 1.25 Average compressive strengths of boric acid waste forms before leaching (•) and after 352 days of leaching (o) for both formulations (w/c = 0.5 and 0.7) as a function of boric acid waste formulations.

1.3.3 Leachate pH Data

The leachate average pH data for each formulation (3%, 6% and 12% boric acid solutions), and both waste-to-cement ratios (0.5 and 0.7) are shown in Figure 1.26. The pH values varied between 7 and 12, and were consistently lower for those from composites incorporating 3% boric acid solutions than those from the composites incorporating 6% and 12% boric acid solutions.

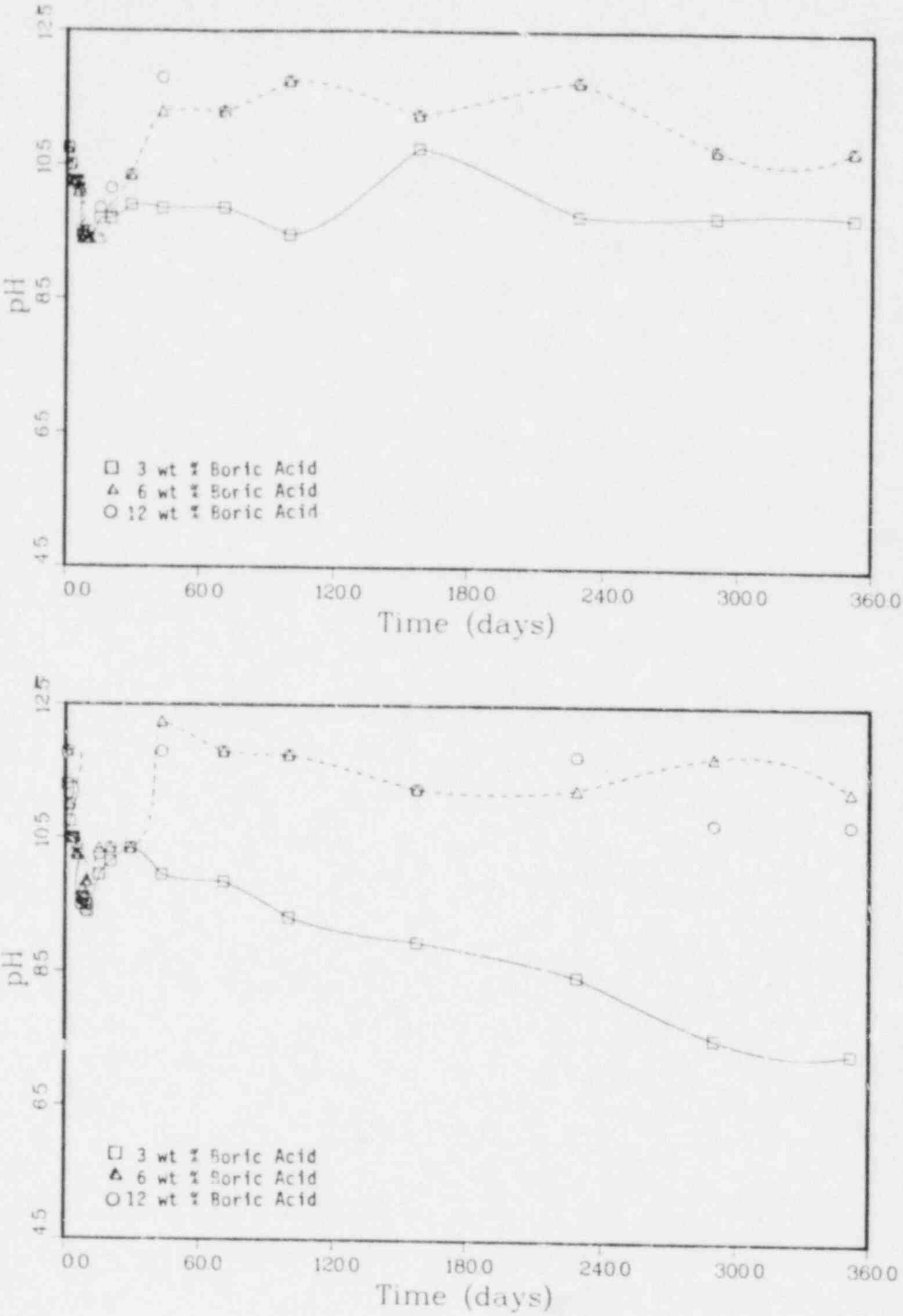

Figure 1.26 Average leachate pH values for 3%, 6%, and 12% H_3BO_3 waste composites at waste-to-cement ratios of 0.5 and 0.7. Upper figure is for w/c ratio of 0.5, and lower figure is for w/c ratio of 0.7.

2. DISPERSION OF ^{137}Cs, ^{85}Sr, and ^{60}Co FROM CEMENT/ORGANIC ION EXCHANGE RESIN COMPOSITES AND FROM ORGANIC ION EXCHANGE RESIN/CEMENT MIXES

(L. Milian and N. Morcos)

2.1 Introduction

Organic ion exchange resins from power plants are currently disposed in shallow land burial sites in the dewatered state. A preliminary draft of 10 CFR Part 61 stipulates that "ion exchange resins shall be immobilized by solidification."(4,5) In addition, the commercial burial site operators have indicated that after July 1981, some unsolidified dewatered organic resins will not be acceptable for burial in their facilities.(6)

Several solidification agents such as cement, bitumen, urea formaldehyde, and vinyl ester-styrene are compatible with organic ion exchange resins. However, certain difficulties associated with the use of cement have been reported.(7) These difficulties include poor integrity of the waste forms when immersed in water and the subsequent release of the contained radioactivity.

We have reported earlier on the displacement of adsorbed ^{137}Cs from ion exchange resins when they are mixed with cement.(8,9) The present study addresses the leachability of ^{137}Cs, ^{85}Sr, and ^{60}Co from organic ion exchange resin/cement composites. The observed leachability is the net result of (a) leaching the radionuclides that were initially displaced from the resin during the mixing with cement, and (b) leaching of the radionuclides remaining on the resins in the waste form after solidification.

2.2 Experimental

2.2.1 Cement/Organic Ion Exchange Resin Composities

Spent organic ion exchange resins are expected to change their initial form (H^+) to that of the most prevalent cations in the water entering the resin bed. To simulate the conditions found in spent resins, samples of Amberlite cation exchange resins (Rohm and Haas IRN-77, H^+ form) were converted to the Na^+ form, and then batch loaded with one microcurie each of ^{137}Cs, ^{85}Sr, and ^{60}Co. The loaded resins were slurried in water and mixed with either Portland II or Lumnite cements. The compositions of the mixtures used for each type of waste form are shown in Table 2.1. The specimens were cured for six weeks prior to leaching. The specimens were 4.7 cm (diameter) by 8.9 cm (height), and were leached using a modified IAEA leaching procedure.(3) The leaching volume was determined by the relationship: $V = 10\ cm \times S$, where V is the leachant volume, and S is the geometric surface of the sample being leached. The waste (water plus resin) to cement ratios used in fabricating the forms were 1.0 and 1.8. Five replicates of each formulation were made.

The activities in the leachates were determined using a Ge(Li) detector system, and are expressed as fractional (incremental fractional release), and as cumulative fractional release.

Table 2.1

Composition of Cement/Cation Exchange Resin Composites

Cement Type	Waste/Cement Ratio (by weight)	Component	Component Weight (grams)
Portland II	1.0	Cation exchange resin (IRN-77, Na^+ form)	65
		Water	65
		Cement	130
Portland II	1.8	Cation exchange resin (IRN-77, Na^+ form)	86
		Water	69
		Cement	86
Lumnite (HAC)	1.0	Cation exchange resin (IRN-77, Na^+ form)	63
		Water	63
		Cement	125
Lumnite (HAC)	1.8	Cation exchange resin (IRN-77, Na^+ form)	86
		Water	69
		Cement	86

2.2.2 Cement/Organic Ion Exchange Resin Mixtures

Samples of loaded cation organic ion exchange resins (described in the previous section) were slurried in water and mixed with Portland II or Lumnite cements. The ratios of waste/cement/water for both types of cements were 1/1/0.8. After contact times ranging from five minutes to two hours, the resins were separated from the cement mixtures and rinsed with deionized water. The amounts of ^{137}Cs, ^{85}Sr, and ^{60}Co radioactivity displaced from the resins into the cement were determined. Control samples of resins loaded with activity in a similar fashion (not mixed with cement) were rinsed with an equal volume of water as those contacted with cement.

The fractions of ^{85}Sr, ^{137}Cs, and ^{60}Co remaining on the cation exchange resins relative to those loaded onto the resins were measured as a function of contact time with Portland II and Lumnite cements. Averages of five samples for each contact time were computed.

2.3 Results and Discussion

2.3.1 Cement/Organic Ion Exchange Resin Composites

The average pH for each formulation for varying leaching periods is graphically shown in Figure 2.1. The pH of leachates from Portland II cement composites ranged from 11 to 12, and those from Lumnite cement composites ranged from 9.9 to 11.5. After approximately two weeks, the spread in pH narrowed and remained at 11-11.5.

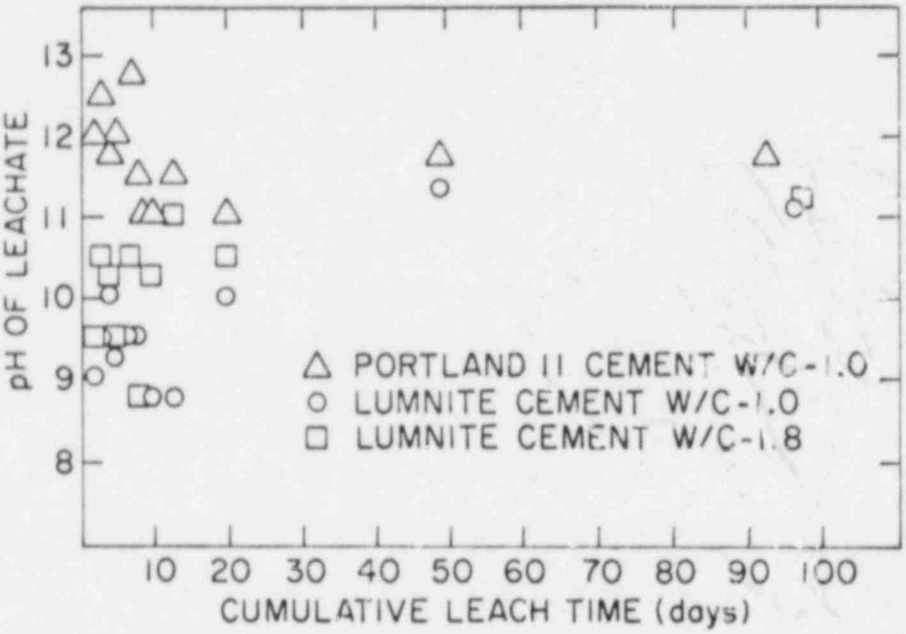

Figure 2.1 pH of leachates as a function of time.

Cumulative fractional release data for ^{137}Cs and ^{85}Sr from each of five replicate organic ion exchange resin/Portland II cement composites are shown in Figures 2.2 and 2.3, respectively plotted as functions of $\sqrt{t}$. Figures 2.4 and 2.5 show the ^{137}Cs cumulative fractional release of replicate composites made with organic ion exchange resins and Lumnite cement at waste-to-cement ratios of 1.0 and 1.8, respectively.

The mean cumulative fractional release obtained from five replicate samples for each formulation are shown in Figures 2.6 and 2.7. The error bars represent the standard deviation from the mean. Lumnite cement showed a lower leachability of ^{137}Cs and ^{85}Sr than Portland II cement. This effect was reported earlier$^{(9)}$ for the leachability of ^{137}Cs from both types of cements and was attributed to the lower permeability to water of Lumnite cement.$^{(10,11)}$ Cobalt-60 was below the detection limit in leachates from both types of cements (3.0 x 10^{-2} μCi per 1.5 L sample), and ^{85}Sr was not observed in leachates from Lumnite composites. These observations may be attributed to either an isotopic dilution of the radiotracers with the cement components or to an actual chemical interaction of the tracers with the matrices.

The upper surfaces of Portland II/organic ion exchange resin composites (waste/cement = 1.8) crumbled before leaching was initiated, and the waste form deteriorated further upon water immersion. The composites made with Lumnite cement maintained their physical integrity throughout the leaching experiment.

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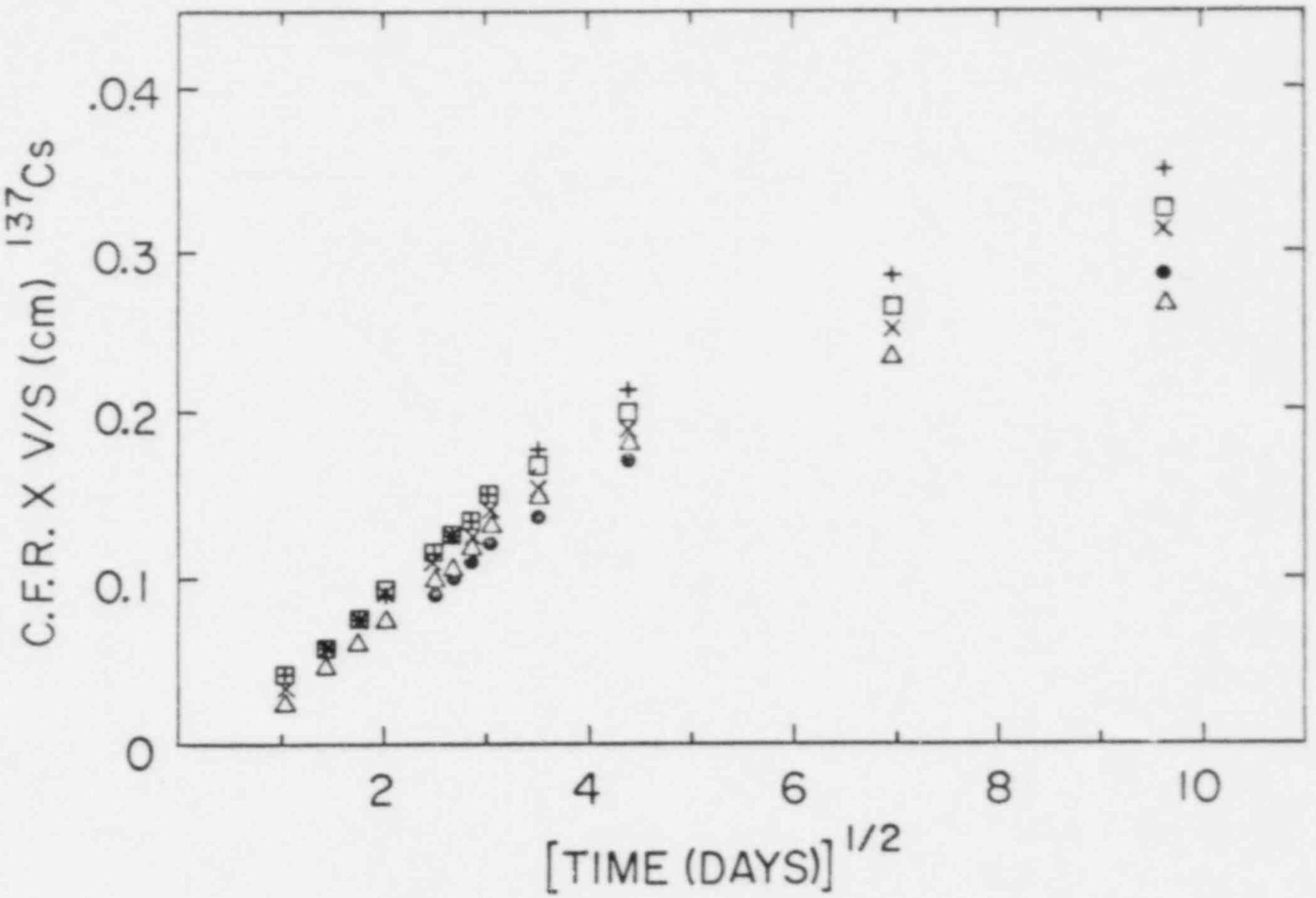

Figure 2.2 ^{137}Cs cumulative fraction release from Portland II cement/cation organic exchange resin composites vs $\sqrt{t}$, w/c = 1.0 by weight and V/S = 0.929 cm.

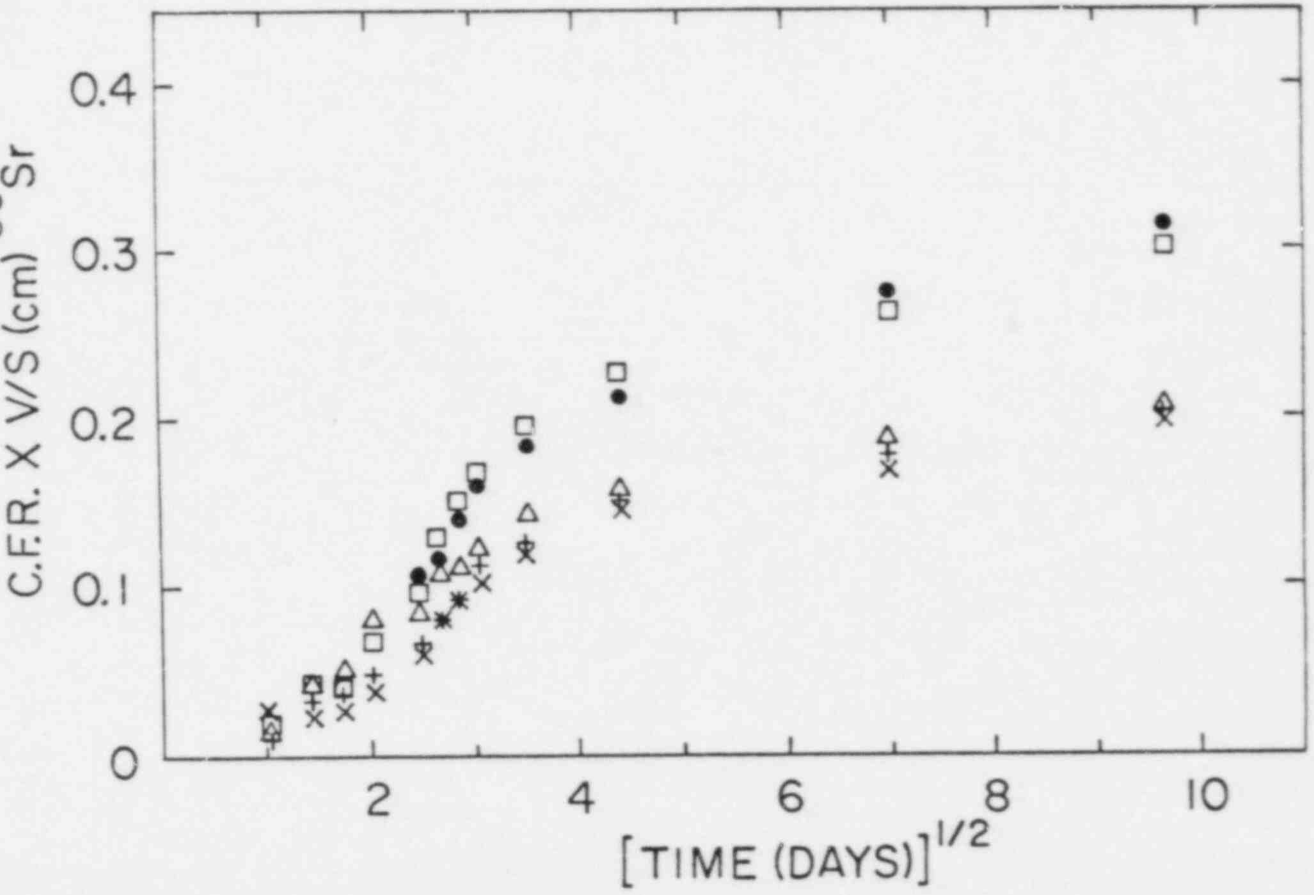

Figure 2.3 ^{85}Sr cumulative fraction release from Portland II cement/cation organic ion exchange resin composites vs $\sqrt{t}$, w/c = 1.0 by weight and V/S = 0.929 cm.

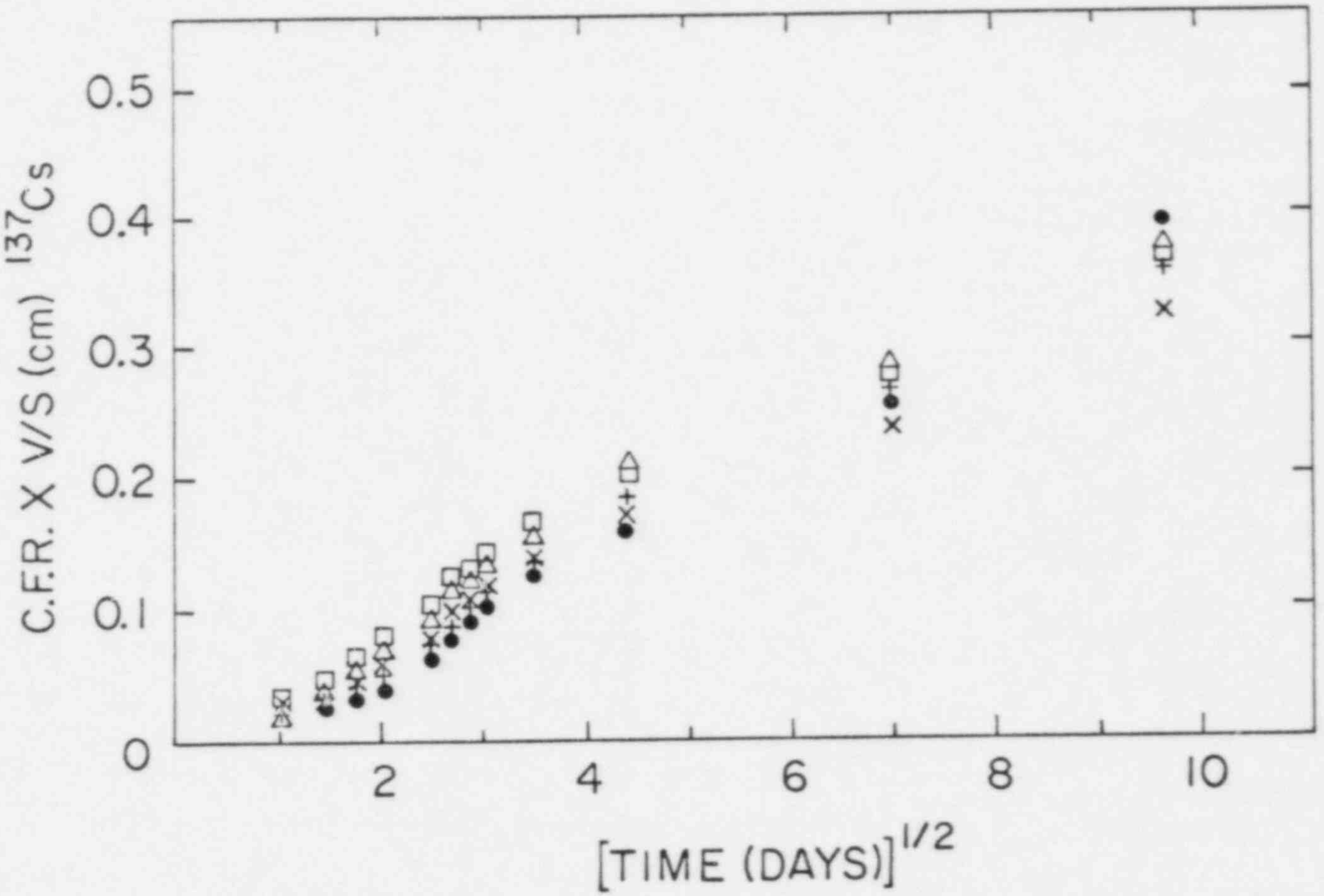

Figure 2.4 ^{137}Cs cumulative fraction release from Lumnite cement/cation organic ion exchange resin composites vs $\sqrt{t_s}$, w/c = 1.0 by weight and V/S = 0.927 cm.

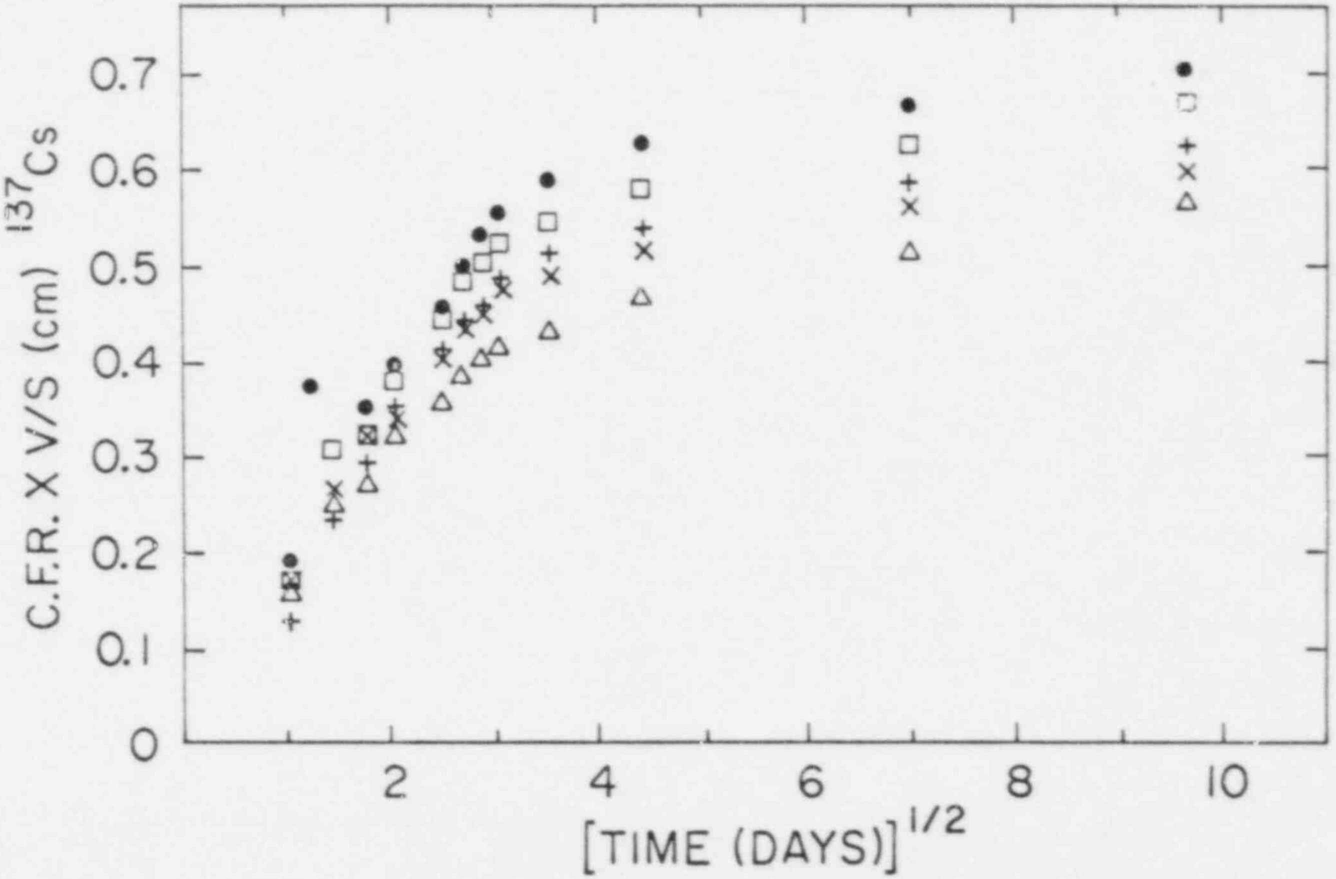

Figure 2.5 ^{137}Cs cumulative fraction release from Lumnite cement/cation organic ion exchange resin composites vs $\sqrt{t}$, w/c = 1.8 by weight and V/S = 0.930 cm.

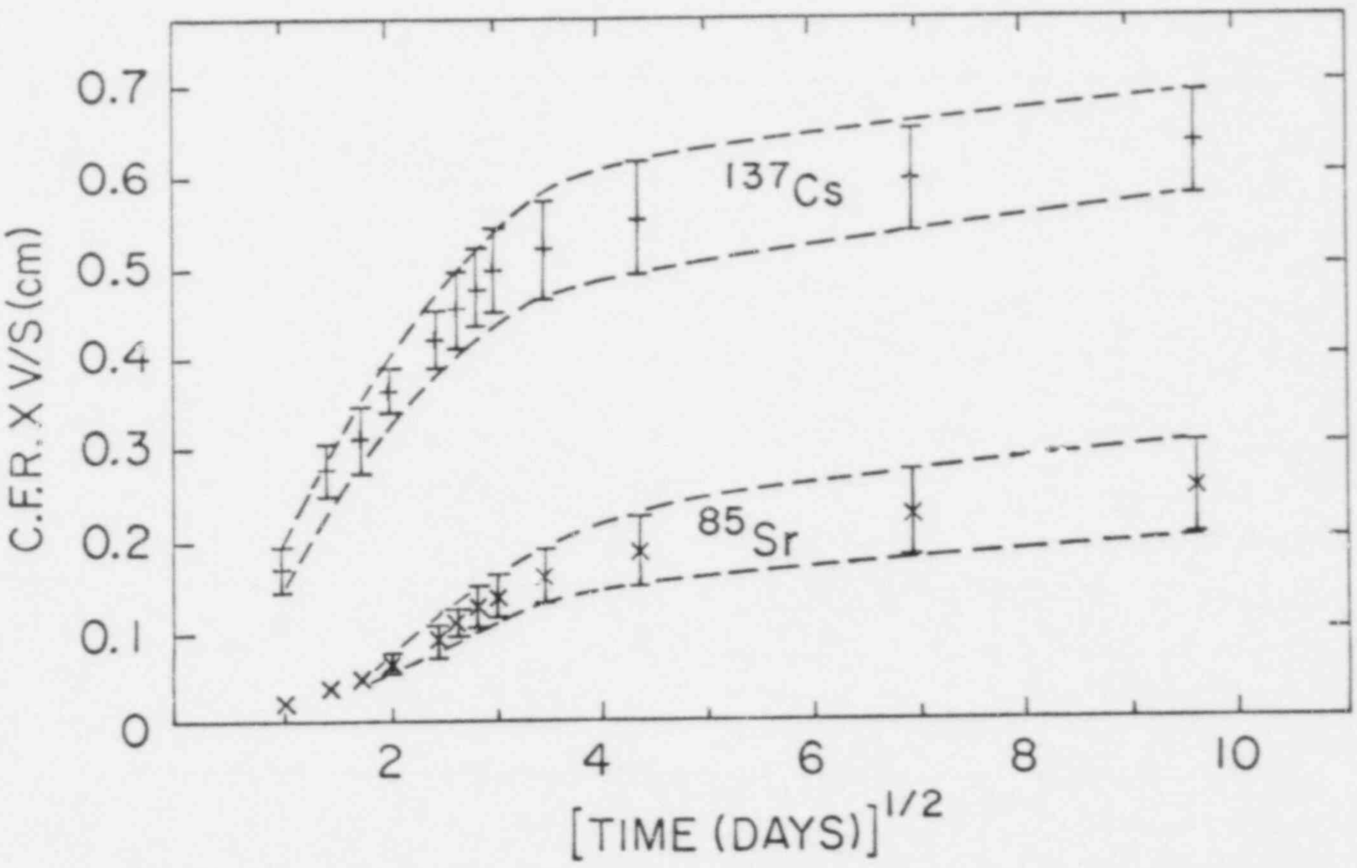

Figure 2.6 Mean cumulative fraction release of ^{137}Cs and ^{85}Sr from Portland II cement/cation organic ion exchange resin composites vs $\sqrt{t}$, w/c = 1.0 by weight and V/S = 0.929 cm.

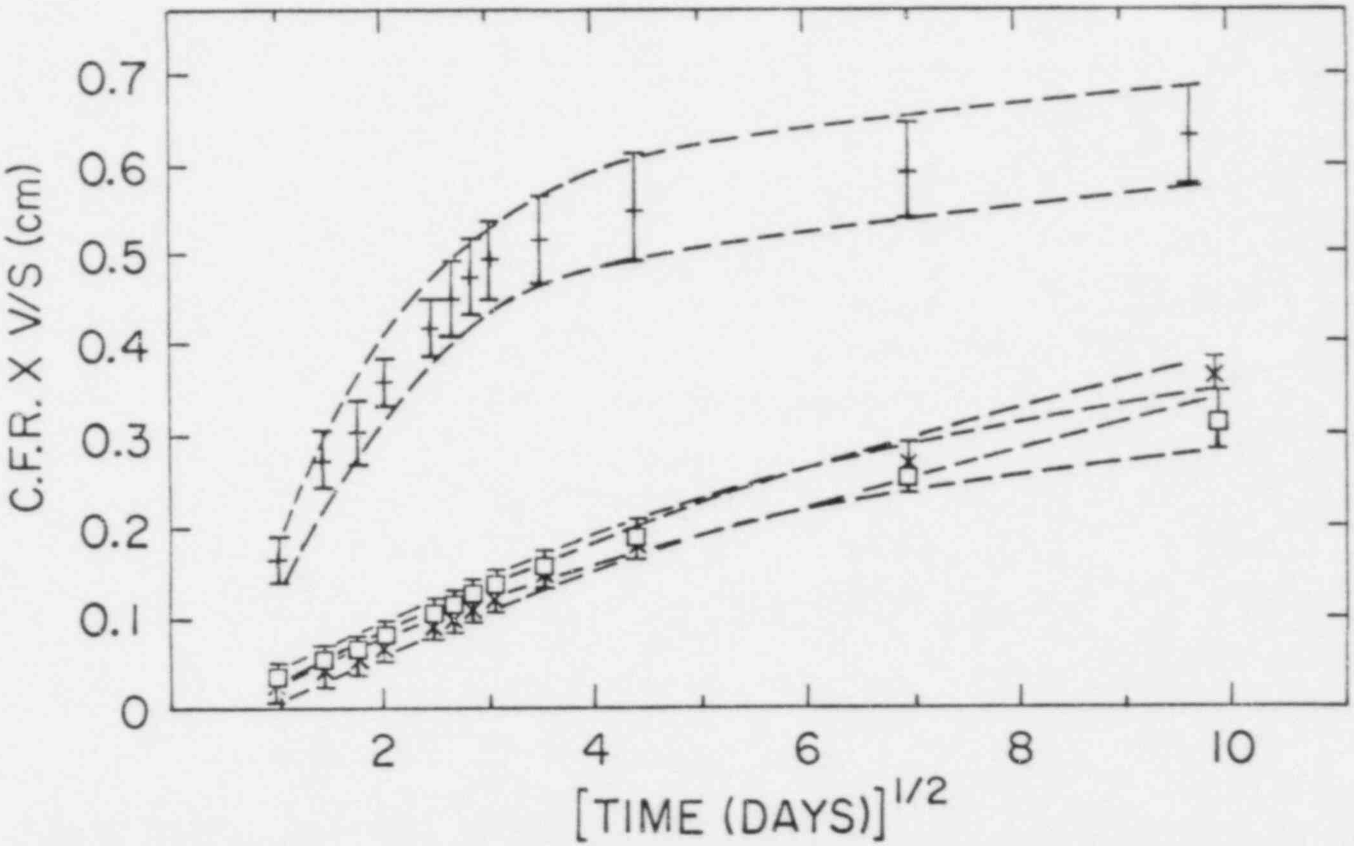

Figure 2.7 Mean cumulative fraction release of ^{137}Cs from Lumnite cement/cation organic ion exchange resin composites vs $\sqrt{t}$, w/c = 1.0 and 1.8 by weight and V/S = 0.927 and 0.930 cm. + Portland II; w/c = 1.0; V/S = 0.929 cm; X Lumnite; w/c 1.0; V/S = 0.927 cm; Lumnite; w/c = 1.8; V/S = 0.930 cm.

2.3.2 Cement/Organic Ion Exchange Resin Mixtures

The fractions of the ^{137}Cs, ^{85}Sr, and ^{60}Co remaining on the cation exchange resins relative to those loaded initially were measured as a function of contact time with Portland II and Lumnite cements. The averages of five samples for each contact time for a given cement formulation were computed and are summarized in Table 2.2. No detectable activity was displaced from the control resin samples.

Table 2.2

Activity Remaining on Cation Resins (IRN-77)
as a Function of Contact Time With Portland II and Lumnite Cements

Cement Type	w/c Ratio by Weight	Contact Time	Fraction of Radiotracer Remaining on Cation Exchange Resins ^{85}Sr	^{137}Cs	^{60}Co
Portland II	1.0	5 min	82.5 ± 9.4	73.6 ± 8.8	82.6 ± 8.0
		1 h	79.3 ± 3.0	73.0 ± 3.0	77.7 ± 4.3
		2 h	86.2 ± 2.6	78.1 ± 3.1	79.8 ± 2.8
	1.8	5 min	88.3 ± 6.1	81.9 ± 4.4	85.0 ± 4.4
		1 h	97.7 ± 5.6	88.1 ± 2.0	86.3 ± 3.1
		2 h	91.5 ± 5.7	83.2 ± 4.7	79.3 ± 4.5
Lumnite	1.0	5 min	99.9 ± 1.6	86.2 ± 1.5	82.8 ± 2.4
		1 h	99.0 ± 5.4	80.9 ± 4.3	81.8 ± 3.6
		2 h	91.0 ± 6.1	78.3 ± 5.6	80.0 ± 5.3
	1.8	5 min	97.6 ± 4.0	88.9 ± 3.9	79.8 ± 4.6
		1 h	97.5 ± 4.7	86.8 ± 4.8	81.4 ± 4.5
		2 h	94.8 ± 3.1	86.7 ± 4.0	77.7 ± 1.7

2.4 Conclusions

2.4.1 Cement/Organic Ion Exchange Resin Composites

Cobalt-60 in the leachates from both Portland II and Lumnite cements for the two waste formulations studied (waste-to-cement ratios of 1.0 and 1.8) was below the detection limit (3.0×10^{-2} µCi per 1.5 L sample). Strontium-85 was not observed in the leachates from Lumnite cement for both formulations. Earlier work(3) has shown that the leachability of ^{60}Co from Portland II cement was approximately two orders of magnitude lower than that of ^{137}Cs. These effects may be attributed to either an isotopic dilution of the radiotracers with cement components or to a chemical interaction of these radiotracers with the cement matrices, or to a combination of these two processes. The cumulative fractional release of ^{137}Cs from the resin/Lumnite cement forms was approximately one-half the release from the resin/Portland II cement forms. The cumulative fractional release of ^{85}Sr from the resin/Portland II cement forms was approximately one-third of the release of ^{137}Cs.

2.4.2 Cement/Organic Ion Exchange Resin Mixtures

Of the three radiotracers studied (^{137}Cs, ^{85}Sr, and ^{60}Co), ^{85}Sr was most tenaciously held by the organic resins when they were mixed with Lumnite cement (Table 2.2).

3. LEACHABILITY OF BITUMEN/ORGANIC ION EXCHANGE RESIN COMPOSITES

(S. Chan)

3.1 Introduction

Organic ion exchange resins from power plants may be currently disposed in shallow land burial sites in the dewatered state. However, in view of the existing regulations (49 CFR Parts 171-179) and proposed regulations (10 CFR Part 61)(4,5), it is anticipated that in the near future ion exchange resins will require solidification before disposal. In addition, the commercial burial sites operators have indicated that after July 1981, some unsolidified dewatered organic resins will not be acceptable for burial in their facilities.(6)

Bitumen is considered a viable material for the solidification of organic ion exchange resins. The following properties are attributed to bitumen: resistant to leaching, radiation, and bacterial attack; tolerant of oil contamination in the waste; requires no water or catalysts for the solidification process; and is readily available.(11,12)

Bitumen solidification systems claim the ability to handle a wide variety of reactor waste streams including acids, bases, organic liquids, evaporator bottom concentrates, decontamination liquids, sludges, and ion exchange resins. However, bitumen burns, and there is evidence that the incorporation of oxidizing agents increases the fire hazard.(13-16) Substances that decompose at the working temperature of a process should not be considered for solidification in bitumen.(8)

This study was undertaken to evaluate the integrity and leachability of bitumen/organic ion exchange resin composites. Anion and cation resins in the SO_4^{-2}, H^+, Na^+, Cs^+, and Sr^{+2} forms were evaluated.

3.2 Experimental

Anion and cation exchange resins were used in this study. The cation resins (IRN-77) were totally converted from their original H^+ form to the Na^+ form and were dried overnight in an oven at 100°C. Four hundred μCi of ^{137}Cs and 500 μCi of ^{85}Sr (in a total of 60 mL of solution) were slowly added, with stirring, to 90 grams (dry weight) of resins in the Na^+ form suspended in 200 mL of deionized water. After overnight equilibration, the supernate activity was found to be negligible. The resins were filtered by suction and dried again. The resins, containing 4.4 μCi of ^{137}Cs and 5.6 μCi of ^{85}Sr per gram, were mixed with nonradioactive resins in either the Na^+ or the SO_4^{-2} forms. The mixtures of resins were then folded into molten Pioneer 221 bitumen and allowed to solidify overnight. The waste forms were leached with deionized water in plastic containers. The volume of the leachant used for each form is given by the relationship: $V = 10\text{ cm} \times S$, where V is the volume of leachant, and S is the geometric surface of the form.

Initially, the leachate was sampled daily, followed by weekly samplings. The leachates were analyzed for ^{137}Cs and ^{85}Sr.

3.3 Results and Discussion

Figures 3.1-3.4 show cumulative fractional release data for bitumen waste forms plotted as a function of $\sqrt{t}$. The waste forms incorporated the following:

a) Na^+ form cation resins loaded with ^{137}Cs,

b) A mixture of 50% (by weight) Na^+ form cation resins, loaded with ^{137}Cs and 50% (by weight) $SO_4^=$ form anion resins,

c) Na^+ form cation resins loaded with ^{85}Sr, and

d) A mixture of 50% (by weight) Na^+ form cation resins, loaded with ^{85}Sr and 50% (by weight) $SO_4^=$ form anion resins.

The waste to bitumen ratio was maintained at 1 and the total amount of resins per waste form was constant.

The presence of sulfate form anionic resins (50% by weight of resins) in the bitumen waste forms increased the leachability of ^{137}Cs by approximately two orders of magnitude (from approximately 5×10^{-4} to approximately 3 and by approximately one order of magnitude for ^{85}Sr (from 1.6×10^{-4} to 1.5×10^{-3}). The data derived from this study with radiotracers show the same trends observed in a previous leaching study in which stable cesium was used.(9)

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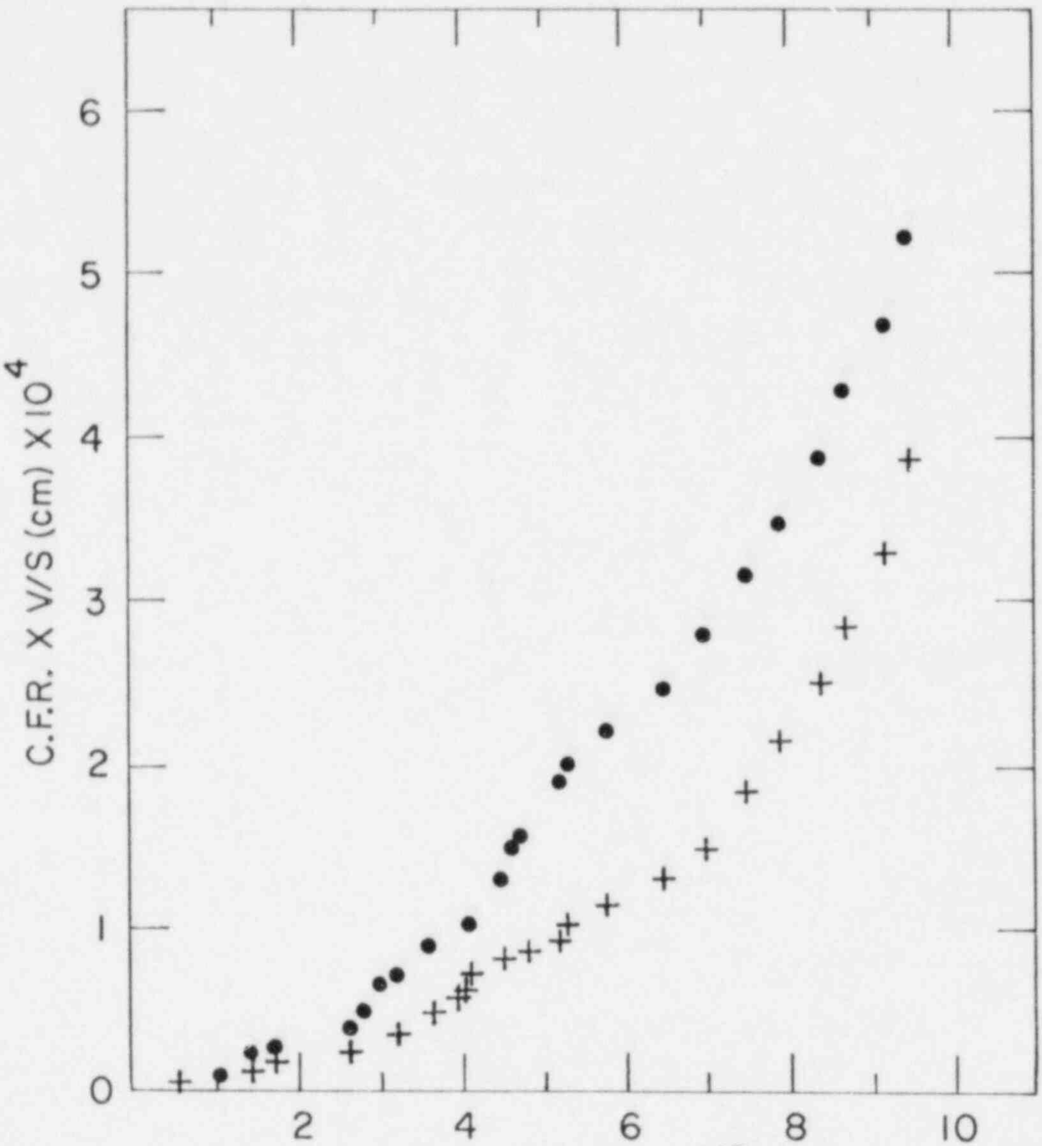


Figure 3.1 ^{137}Cs cumulative fraction release normalized for V/S as a function of $\sqrt{t}$ from bitumen plus resins in the Na^+ form. ●, w/b = 1.0; V/S = 3.1 cm; +, w/b = 0.67; V/S = 3.3 cm.

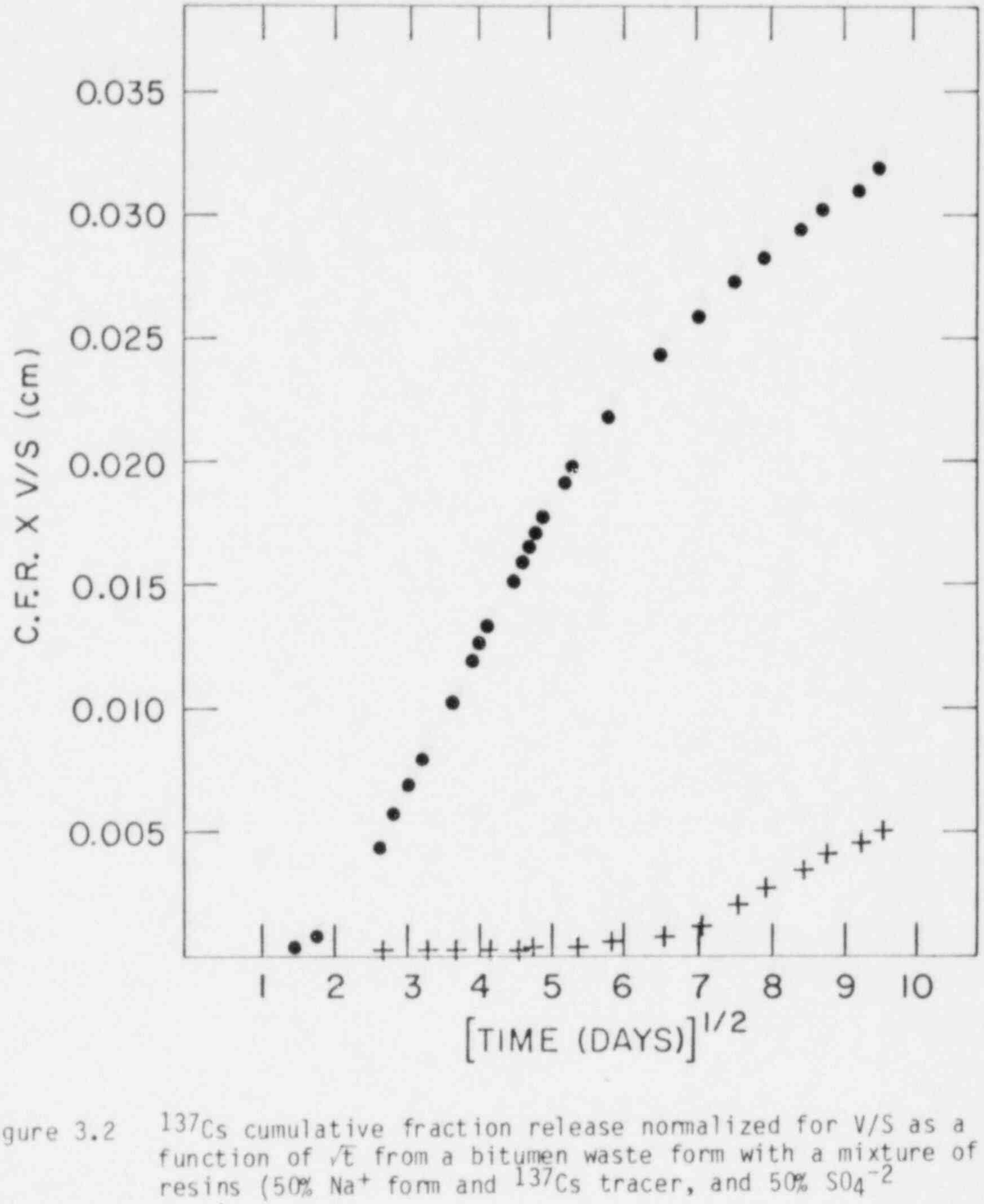
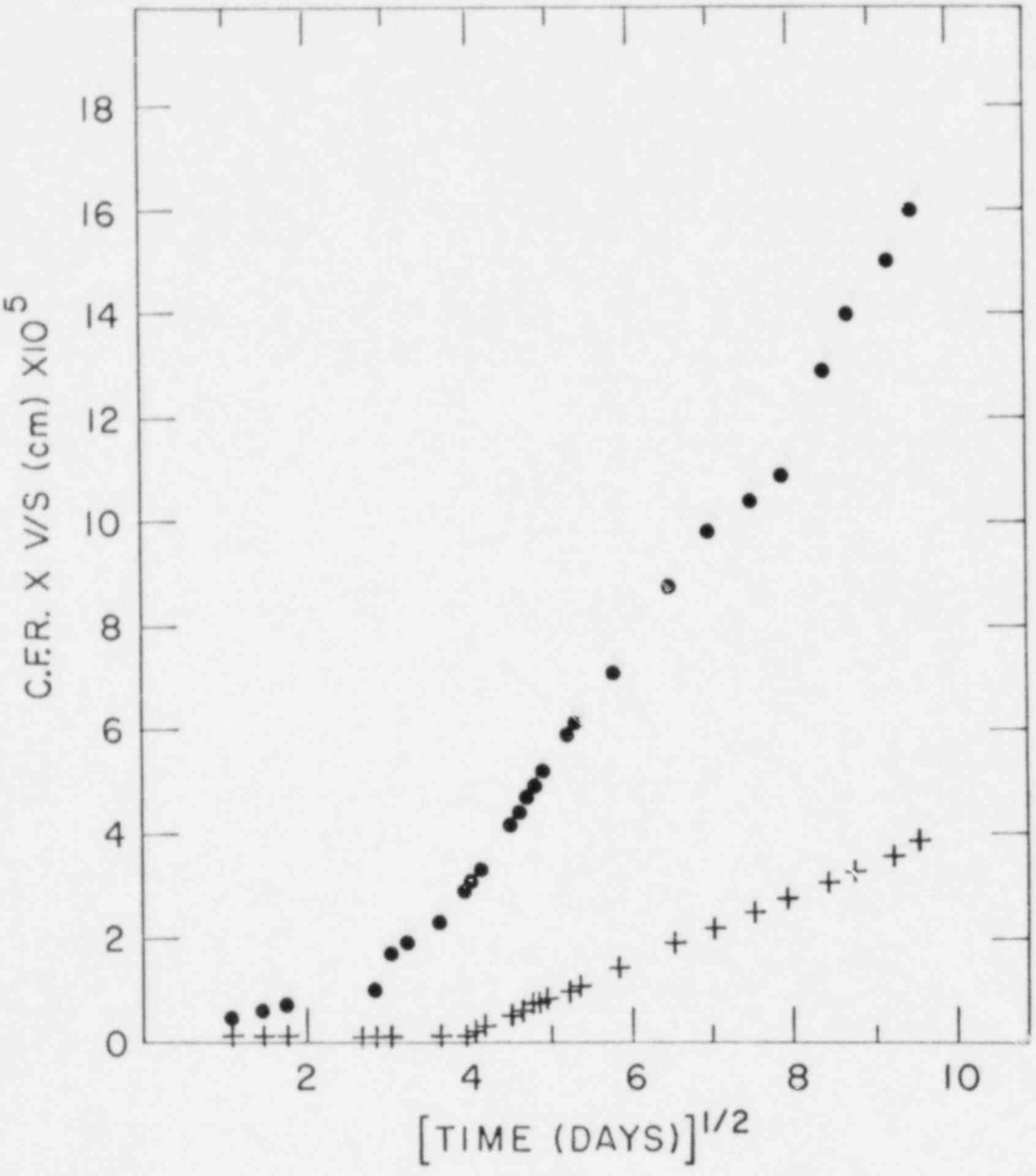


Figure 3.2 ^{137}Cs cumulative fraction release normalized for V/S as a function of $\sqrt{t}$ from a bitumen waste form with a mixture of resins (50% Na^+ form and ^{137}Cs tracer, and 50% SO_4^{-2} form).
●, w/c = 1.0; V/S = 3.3 cm; +, w/b = 0.67; V/S = 3.7 cm.



Figures 3.3 ^{85}Sr cumulative fraction release normalized for V/S as a function of $\sqrt{t}$ from bitumen plus resins in the Na^+ form. ●, w/b = 1.0; V/S = 3.3 cm; +, w/b = 0.67; V/S = 3.7 cm.

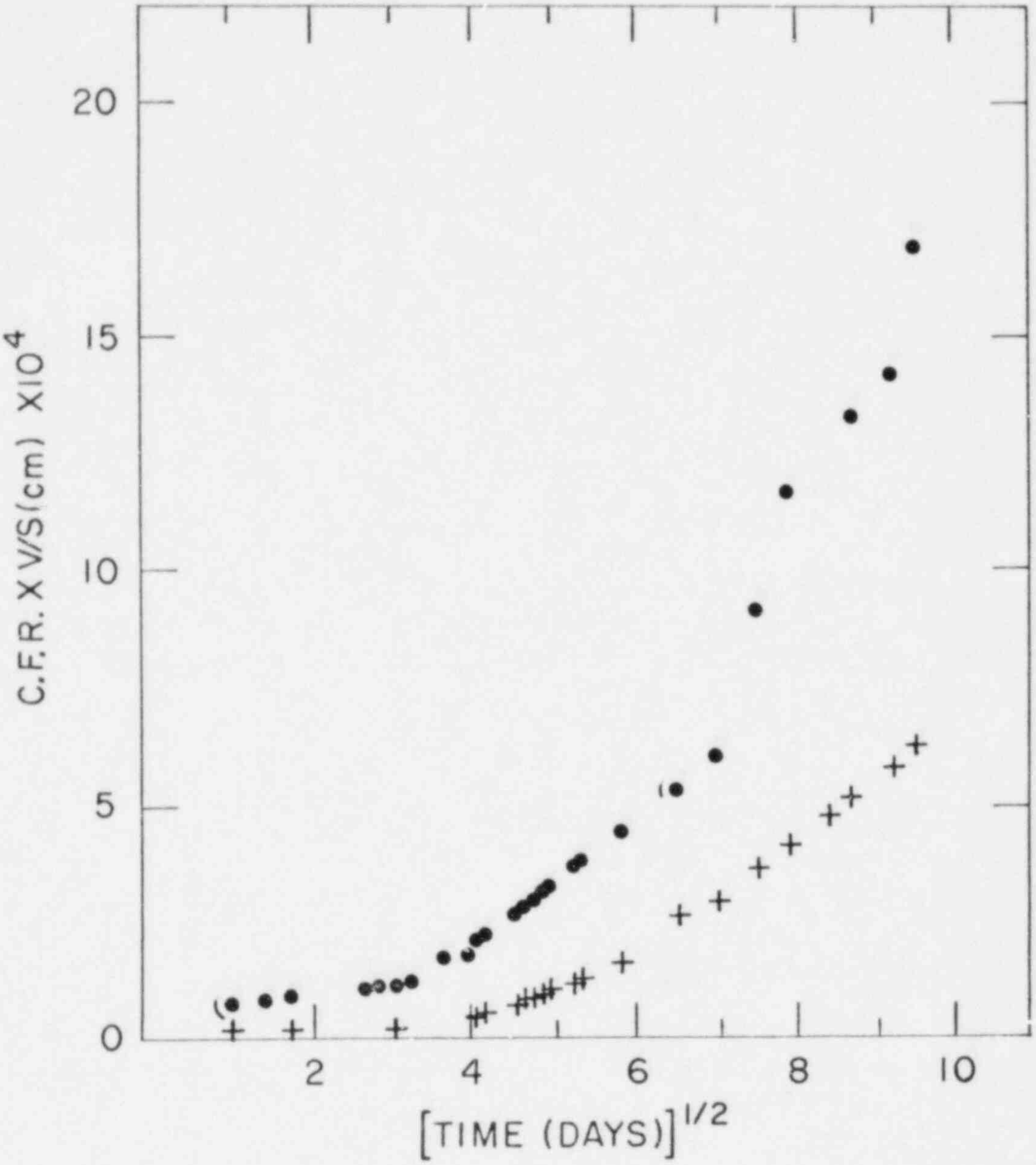


Figure 3.4 ^{85}Sr cumulative fraction release as a function of $\sqrt{t}$ from a bitumen waste form with a mixture of resins (50% Na^+ form and ^{85}Sr tracer, and 50% SO_4^{-2} form).
●, w/b = 1.0; V/S = 3.3 cm; +, w/b = 0.67; V/S = 3.7 cm.

4. CORRELATION OF ^{137}Cs LEACHABILITY FROM SMALL-SCALE (LABORATORY) SAMPLES TO LARGE-SCALE WASTE FORMS (W. Becker, A. Colavito, P. Hayde, L. Milian, and N. Morcos)

4.1 Introduction

Several theoretical and empirical methods based on mass transport and diffusion theory have been developed to predict the leachability of radioisotopes from waste composites.(16-23) A method has been recommended earlier (1970) by the International Atomic Energy Agency (IAEA) for leaching samples and for the analysis and interpretation of leaching data.(24) Recently, a standard method, which has much in common with the earlier IAEA method, was proposed by the American Nuclear Society Standards Committee Working Group (ANS-16.1). This method suggests the accumulation of data over a short-term period (five days) to determine the "Leachability Index," a material parameter. This parameter characterizes the leaching of a radionuclide from the waste form under evaluation, and may be used for performance predictions under actual environmental conditions, if the type of material being tested was characterized through generic studies. A working group (ISO/TC 85/SC 5/WG 5) of the International Standards Organization (ISO) is also currently directing efforts toward the adoption of a uniform standard leach test.

The IAEA method assumes a semi-infinite plane source model of diffusion for radioisotopes from waste composites, and relates the amount of substance diffused out of a waste composite to the leaching time, the amount of that substance initially present, and a diffusion rate. The solution for the rate equation describing this diffusion mode can be written as(8,16):

$$f = \frac{S}{V} \times 2\left(\frac{D\ t}{\pi}\right)^{1/2} \tag{4.1}$$

where f = fraction of substance diffused out of the composite during time t,

S/V = ratio of the geometric surface of the sample to its volume,

D = effective bulk diffusion coefficient of the substance for the particular composite matrix,

The underlying assumptions dictated by Equation (4.1) are that the isotope under study is either stable or has a long half-life as compared to the duration of the experiment and that the initial isotope surface concentration of the waste form is zero. Furthermore, the relationship in Equation (4.1) implies that initially for t = 0, the fraction leached (f) is also zero. However, experimental leaching data deviates from this prediction for small values of t, and a more general relationship is suggested(16,25):

$$f = \frac{S}{V} \cdot 2\left(\frac{D\ t}{\pi}\right)^{1/2} + \alpha \qquad (4.2)$$

where the term (α) represents non-diffusive contributions from the surface of the waste form.(4,12) Furthermore, a linear relationship of the term (α) with the S/V ratio of the waste form was shown to exist,(26) indicating that it is indeed surface controlled.

Experiments were initiated to determine if the ^{137}Cs leach data on small-scale laboratory samples could be used in predicting the leaching behavior of larger samples. This report presents the experimental data on 1 x 1, 2 x 2, 2 x 4, 3 x 3, 6 x 6, 6 x 12, and 22 x 22 forms incorporating organic ion exchange resins loaded with ^{137}Cs and solidified in Portland I cement. A method is presented to correlate the leaching data from the small-scale samples to those from large samples. It also allows estimating the cumulative fractional release for a given waste form size and a given leaching time.

4.2 Experimental

The waste forms evaluated in this study are representative of those that are expected to be generated at nuclear power plants, e.g., organic ion exchange resins and boric acid waste solidified in Portland cements.

The amounts of ^{137}Cs added to the different size waste forms were chosen by scaling the amounts used in earlier leaching experiments where 2 x 4 waste forms were studied. This insured detection of activity in the leachates. The relationship $f_1(V/S)_1 = f_2(V/S)_2$ where f_1 and f_2 are the cumulative fractional releases leached from two different size waste forms during the same leaching time, and $(V/S)_1$ and $(V/S)_2$ are the geometric surface-to-volume ratios of these two forms. Using the leached fractions from 2 x 4 organic cation exchange resin/Portland II cement composites(27) as f_1, f_2 was calculated for the various sizes using the appropriate $(V/S)_2$ values.

A modified IAEA leaching procedure(3) was followed. The first leaching period was 100 minutes, and thereafter the leachant was changed daily, during the first 42 days, except for weekends, where the leaching periods extended from Friday to Monday. (However, the leachant was changed during the first weekend). After 42 days, the daily leaching period was extended to a week, and later to a month.

4.2.1 Organic Cation Exchange Resin Preparation

Organic cation exchange resins (Rohm and Haas IRN-77, H$^+$ form) were converted to the Na$^+$ form with 2 molar NaOH solution. The volume of NaOH solution was twice that of the resin and following the sorption period, it was decanted. The resins were then rinsed with deionized water until the pH of the rinse water was comparable to that of the initial deionized water, indicating that the excess NaOH was rinsed out completely. The resins were stored in deionized water.

4.2.2 Organic Cation Exchange Resin/Cement Composites

Organic cation exchange resin/cement composites were fabricated with a waste-to-cement (w/w) (Portland I) ratio of 0.6 and a water-to-cement (w/w) ratio of 0.4. The simulated waste composition consisted of 33 weight percent IRN-77 Na^+ form cation exchange resin loaded with ^{137}Cs, and 67 weight percent deionized water. This formulation was chosen because test samples maintained their physical integrity during a prolonged leaching period (4-5 weeks), and because it provided good workability of the mixture during the mixing stage. Earlier process parameter investigations(2) for the solidification of IRN-77 resins in cement had defined boundaries for the components of the waste form (resin, water, and cement) where a freestanding solid product was produced. However, the durability of the solidified product upon immersion in water had not been evaluated. Formulations corresponding to those shown in the area bounded by heavy lines in the table reproduced from Reference 2 were evaluated (see next page). Up to twelve two-inch-diameter by four-inch-high forms were made and immersed in deionized water to evaluate their physical integrity (no evidence of crumbling) under leaching conditions. Only two formulations, denoted by the triangle and circle in the table, passed the immersion and workability tests. However, some of the forms with the triangle formulation began to crumble after several days in water, whereas there were no failures of the formulation shown in the circle.

The following procedure was used in preparing the 1 x 1, 2 x 2, 2 x 4, and 3 x 3 forms: Appropriate amounts of resins were added to each mold and covered with deionized water to which a measured amount of ^{137}Cs radiotracer was added while stirring. After a 24-hour equilibration period, the water was sampled and assayed for ^{137}Cs content to assure uptake by the resin. The water was then removed by suction through a fritted filter and an amount of deionized water was added commensurate with the formulation for the composite. The larger samples (6 x 6, 6 x 12, 12 x 12, and 22 x 22) were prepared in a slightly different manner. The amount of water added to the resins prior to the addition of ^{137}Cs was the amount needed for solidification, and therefore was not decanted after equilibration and ^{137}Cs assay. The amount of ^{137}Cs remaining in the aqueous phase (after sorption) for all samples was less than 0.1% of the initially added activity, indicating that greater than 99.9% of the ^{137}Cs was sorbed onto the resins.

The mixtures of cement, water, and resins in individual molds were hand stirred with polyethylene rods (for samples up to 3 x 3), and mechanically stirred with a stainless steel mixer attached to a motor for the larger forms for five minutes and capped to prevent water evaporation during the 28-29-day curing period. Earlier work has shown that ion exchange resin/cement composites cured in air or left open to air after curing, disintegrated after immersion in water.

Table (From Manaktala and Weiss - Reference 2)

Formulation of Ion Exchange Resin Test Samples
(all weights given in grams)

Water/Cement ↓ / Waste/Cement →	0.4	0.6	0.8	1.0	1.2	1.4	1.6	1.8
0.3	15.7[a] 47.1 157.1	41.2 41.2 137.6	61.1 36.7 122.2	77.0 33.0 110.0	90.0 30.0 100.0	100.8 27.5 91.7	110.0 25.4 84.6	117.9 23.6 78.6
0.4	0.0 62.8 157.2	27.5 55.0 137.5	48.9 48.9 122.2	66.0 44.0 110.0	80.0 40.0 100.0	91.7 36.7 91.7	101.5 33.9 84.6	110.0 31.4 78.6
0.5		13.8 68.7 137.5	36.7 61.1 122.2	55.0 55.0 110.0	70.0 50.0 100.0	82.5 45.8 91.7	93.1 42.3 84.6	102.1 39.3 78.6
0.6		0.0 82.5 137.5	24.4 73.3 122.2	44.0 66.0 110.0	60.0 60.0 100.0	73.3 55.0 91.7	84.6 50.8 84.6	94.3 47.1 78.6
0.7			12.2 85.6 122.2	33.0 77.0 110.0	50.0 70.0 100.0	64.2 64.2 91.7	76.2 59.2 84.6	86.4 55.0 78.6
0.8			0.0 97.8 122.2	22.0 88.0 110.0	40.0 80.0 100.0	55.0 73.3 91.7	67.7 67.7 84.6	78.6 62.8 78.6
0.9				110.0 99.0 110.0	30.0 90.0 100.0	45.8 82.5 91.7	59.2 76.2 84.6	70.7 70.7 78.6
1.0				0.0 110.0 110.0	20.0 100.0 100.0	36.6 91.7 91.7	50.8 84.6 84.6	62.8 78.6 78.6

[a] Each entry is composed of three parts, viz., resin (15.7), water (47.1), and cement (157.1).

Table 4.1 summarizes the waste composite sizes made to date, together with their contents, volume-to-surface ratios, and leachant volumes. Each sample size was prepared in triplicate.

Table 4.1

Composite Dimensions, Components, and Leachant Volumes

Sample	Composite				Components (g)			^{137}Cs Added to Composite (μCi)	Volume of Leachant (mL)
	Diameter (in.)	Height (in.)	V/S (cm)	Weight (g)	Cement	Waste			
					Portland I	IRN-77[a]	H_2O		
1 x 1	0.93	0.94	0.396	20	12.5	2.5	5.0	1	265
	0.93	0.94	0.396	b	12.5	2.5	5.0	1	265
	0.93	0.94	0.396	b	12.5	2.5	5.0	1	265
2 x 2	1.83	1.89	0.784	150	93.8	18.8	37.5	10	1,050
	1.83	1.89	0.784	150	93.8	18.8	37.5	10	1,050
	1.83	1.89	0.784	150	93.8	18.8	37.5	10	1,050
2 x 4	1.85	3.62	0.936	290	181.3	36.3	72.5	10	1,700
	1.85	3.62	0.936	290	181.3	36.3	72.5	10	1,700
	1.85	3.62	0.936	290	181.3	36.3	72.5	10	1,700
3 x 3	3.00	3.34	1.32	734	450	92.0	184	10	2,950
	3.00	3.34	1.32	735	460	92.0	184	10	2,950
	3.00	3.34	1.32	735	460	92.0	184	10	2,950
6 x 6	6.06	5.79	2.53	b	3,250	650	1,300	500	11,000
	6.06	5.73	2.52	b	3,250	650	1,300	500	11,000
	6.06	5.71	2.51	b	3,250	650	1,300	500	11,000
6 x 12	6.00	11.6	3.03	9,620	6,139	1,228	2,456	1,000	18,200
	6.00	12.5	3.07	9,250	6,139	1,228	2,456	1,000	18,200
	6.00	11.1	3.00	9,430	6,139	1,228	2,456	1,000	18,200
12 x 12	12.5	11.5	5.14	40,000	24,900	4,990	9,980	10,000	44,000
	12.3	11.5	5.09	40,100	24,900	4,990	9,930	10,000	44,000
	12.3	11.5	5.09	39,800	24,900	4,990	9,990	10,000	44,000
22 x 22	21.5	21.5	9.10	228,340	143,700	28,740	51,490	20,000	136,200

[a] Rohm and Haas Amberlite organic cation exchange resin.
[b] Not weighed.

4.2.3 Leaching

The composites were leached in deionized water using a modified IAEA leaching procedure(3) described earlier. The leaching volume was determined by the relationship: V = 10 cm x S, where V is the leachant volume and S is the geometric surface of the composite being leached.

Leaching was carried out in two sets of containers. The samples were placed in a fresh leachant and the leachate from the previous period was acidified with HNO_3 (volume of conc. acid 1% volume of leachate). Ten-milliliter aliquots of the acidified leachate were withdrawn in a plastic test tube and assayed for ^{137}Cs content using a 3 in. x 3 in. NaI well crystal. The remaining liquid was removed, the container was washed, and fresh leachant was added to it for the next leaching period. The leachant was allowed to equilibrate to room temperature overnight before transferring the waste form from the other container.

All samples were counted until a minimum of 1,000 counts were accumulated in the "window" set around the ^{137}Cs photopeak (661.6 keV). Data reduction was performed using a computer program developed at BNL.(28) Incremental fractional and cumulative fractional releases from the forms were calculated. Cumulative fractional release data are presented as a function of time.

4.3 Results and Discussion

4.3.1 Leaching

The calculated incremental and cumulative fractional releases from each waste form are given in Tables B.1 through B.7. These tables are included in Appendix B. The errors quoted represent only the statistical errors associated with the counting.

These data are also graphically shown in Figures 4.1-4.18. Each pair of figures shown on a page presents the leaching data of three replicate samples and the average cumulative fractional release of the three replicates except for the 22 x 22 samples. The average cumulative fractional release curves have been normalized for V/S variation in the waste forms.

Two of the 1 x 1, and one of the 6 x 6 composites partially disintegrated during the first three weeks of leaching. The cause of the deterioration of the 1 x 1 samples is not known. However, in the case of the 6 x 6 sample, the deterioration occurred mainly along a line on the sample that was inadvertently exposed to ambient air during the curing process. This line corresponds to a crack in the mold used in the fabrication of this sample.

The cumulative fractional releases from the composites that deteriorated were higher than those from the composites that remained intact. However, the actual geometric surfaces of the deteriorated composites could not be measured, and their geometric surfaces prior to leaching were used to calculate the normalizing V/S ratio.

(Continued Page 56)

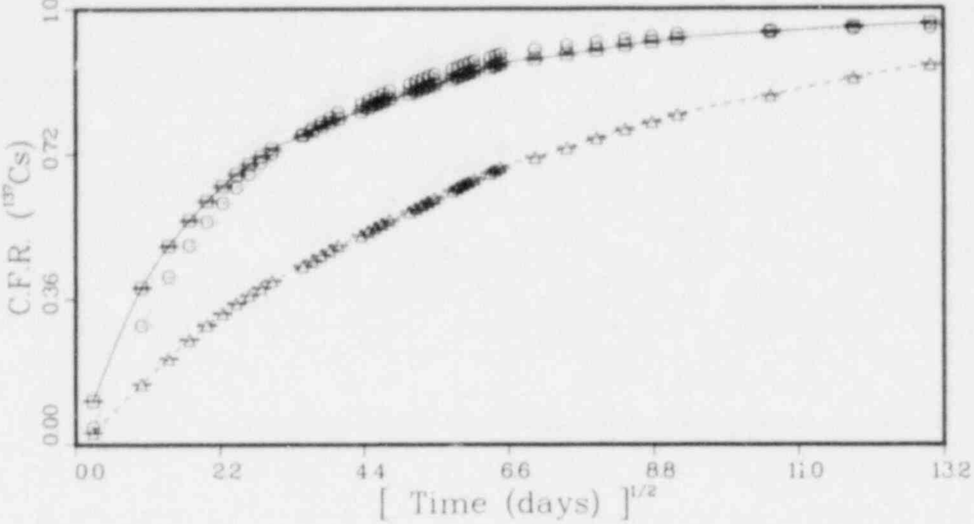


Figure 4.1 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 1-inch-diameter x 1-inch-high waste composites (w/c = 0.6; V/S = 0.396 cm). (The two forms denoted by □ and o partially disintegrated during the first three weeks of leaching.)

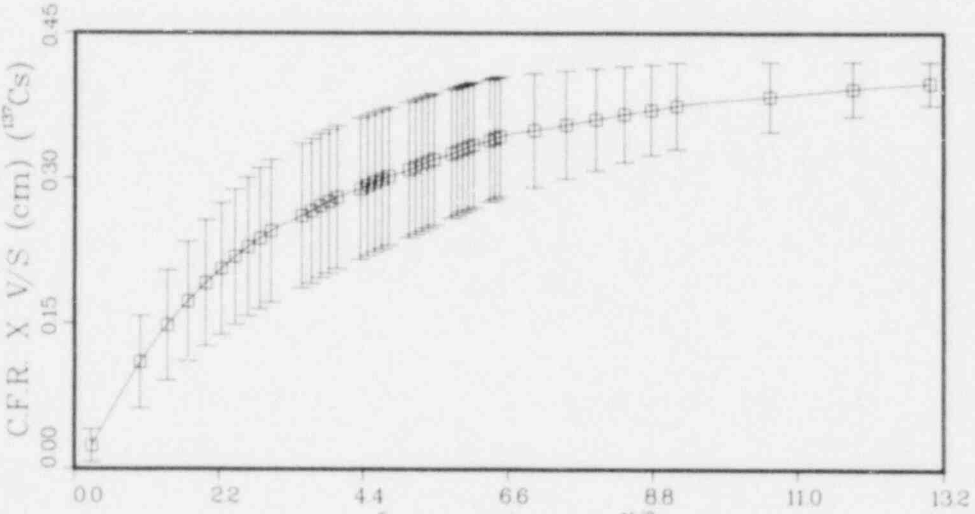


Figure 4.2 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 1-inch-diameter x 1-inch-high waste composites (w/c = 0.6; V/S = 0.396 cm). The average CFR was normalized for V/S.

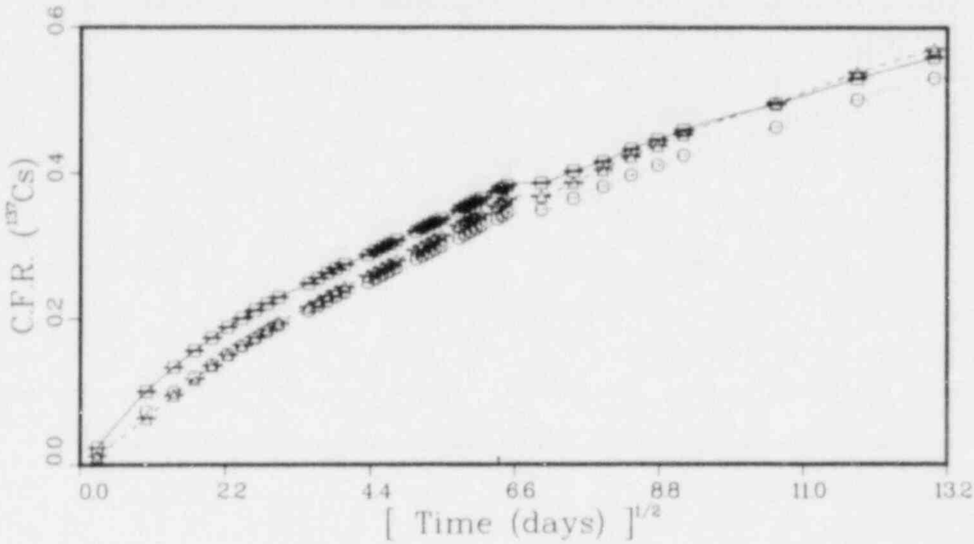


Figure 4.3 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 2-inch-diameter x 2-inch-high waste composites (w/c = 0.6; V/S = 0.784 cm).

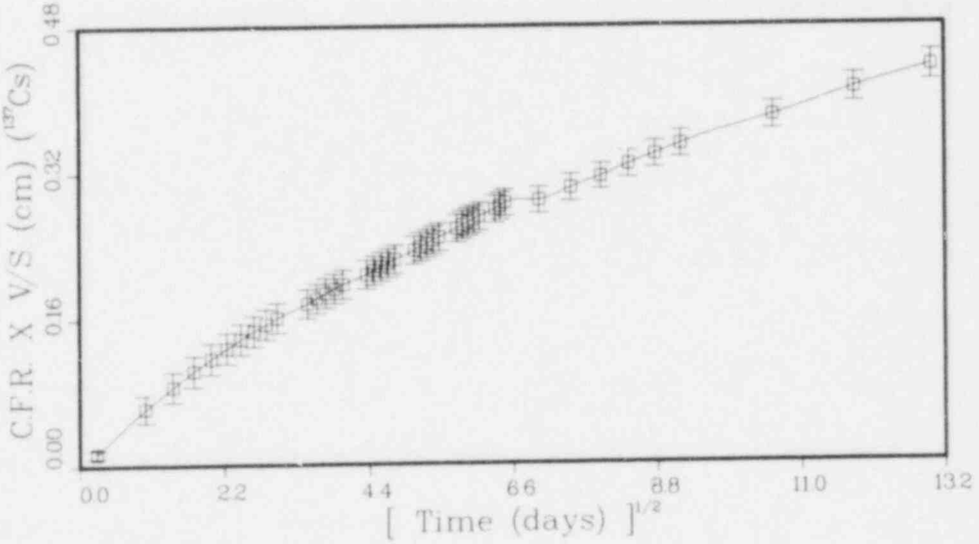


Figure 4.4 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 2-inch-diameter x 2-inch-high waste composites (w/c = 0.6; V/S = 0.784 cm). The average CFR was normalized for V/S.

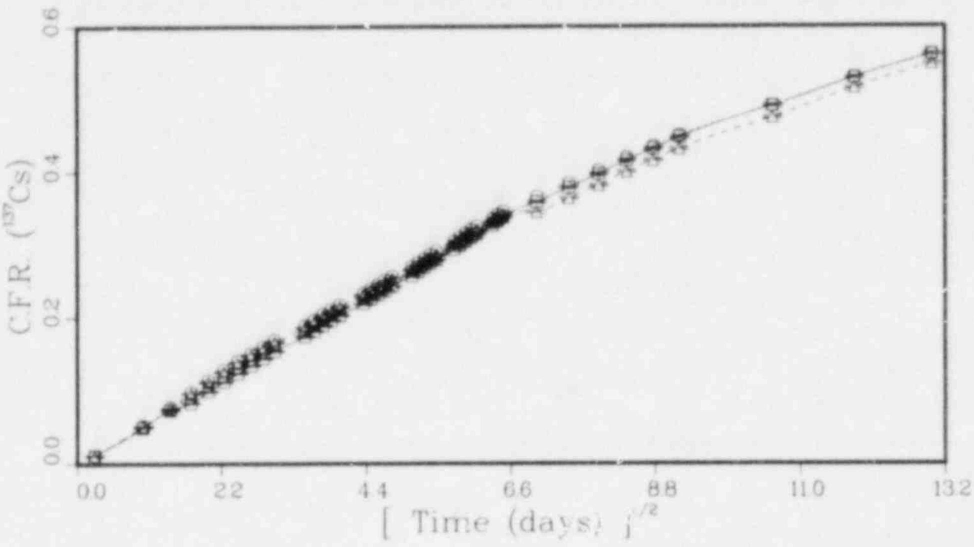


Figure 4.5 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 2-inch-diameter x 4-inch-high waste composites (w/c = 0.6; V/S = 0.936 cm).

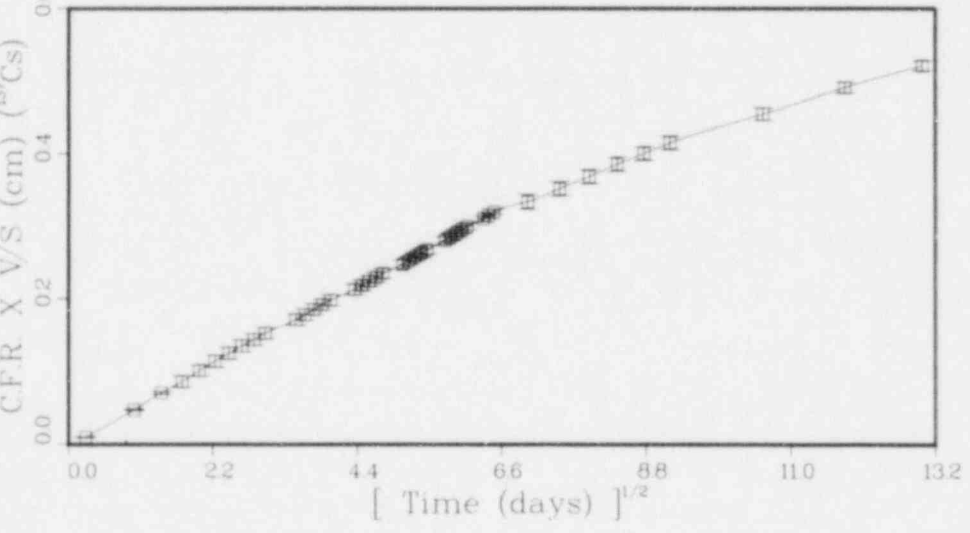


Figure 4.6 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 2-inch-diameter x 4-inch-high waste composites (w/c = 0.6; V/S = 0.936 cm). The average CFR was normalized for V/S.

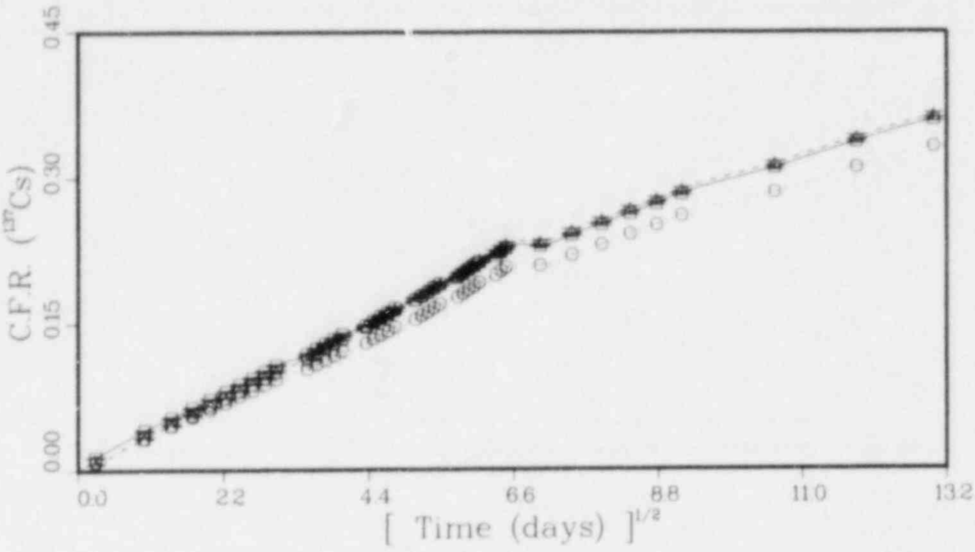


Figure 4.7 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 3-inch-diameter x 3-inch-high waste composites (w/c = 0.6; V/S = 1.32 cm).

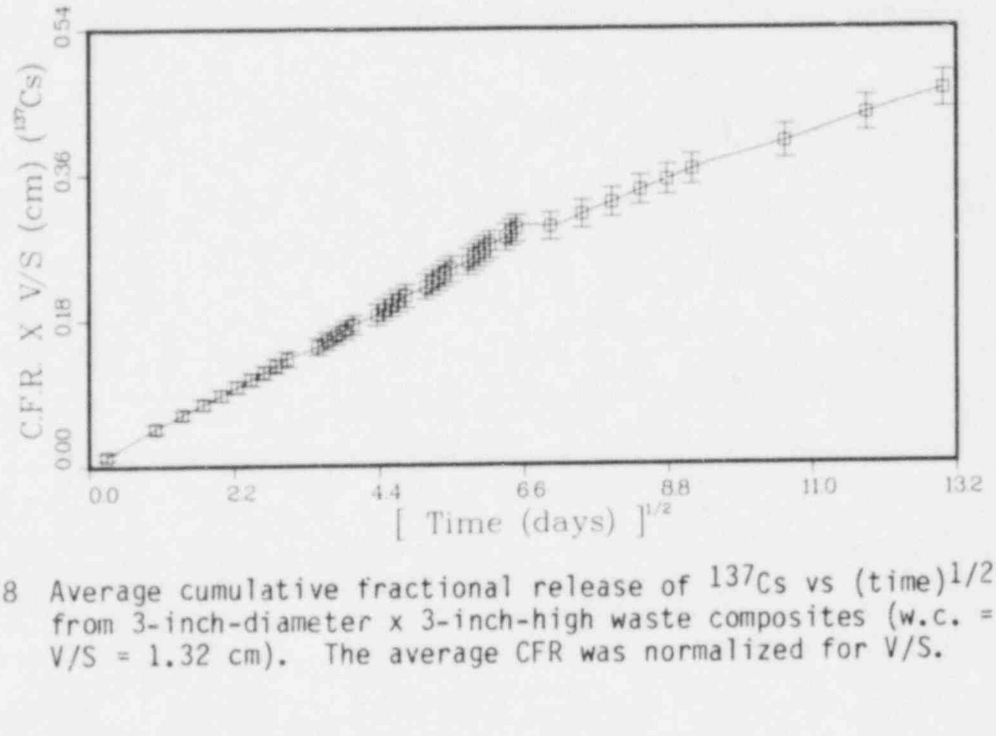


Figure 4.8 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 3-inch-diameter x 3-inch-high waste composites (w.c. = 0.6; V/S = 1.32 cm). The average CFR was normalized for V/S.

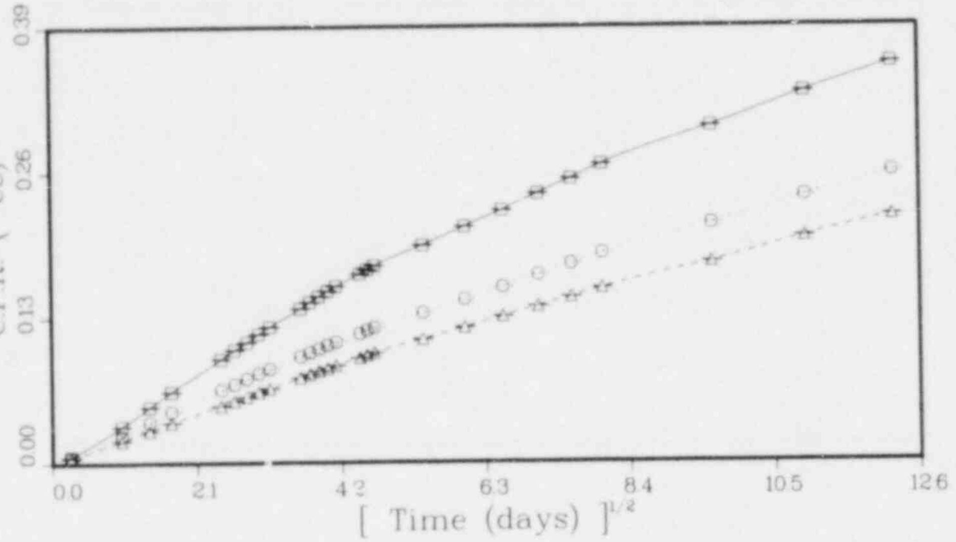


Figure 4.9 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 6-inch-diameter x 6-inch-high waste composites (w/c = 0.6; V/S = 2.52 cm). (The form denoted by ☐ partially disintegrated during the first four weeks of leaching.)

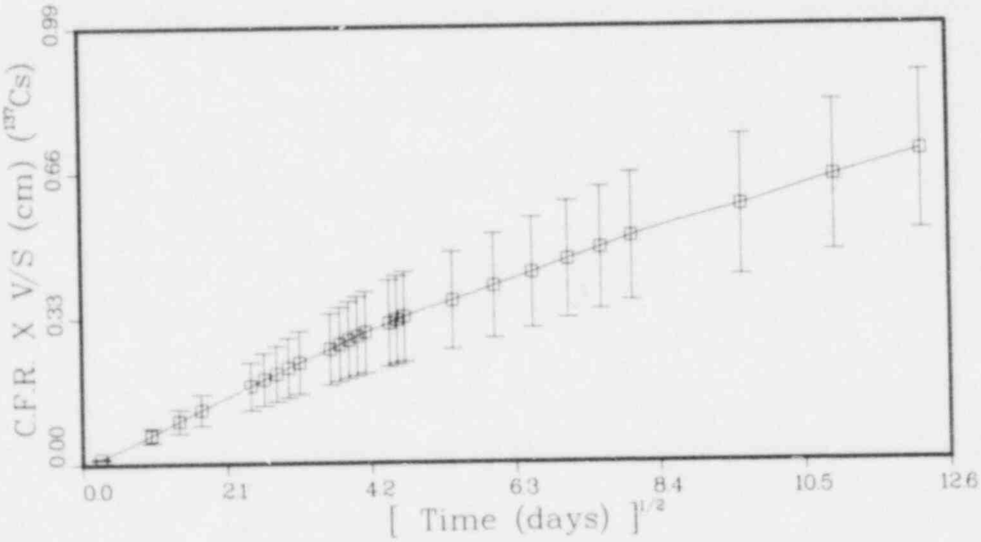


Figure 4.10 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 6-inch-diameter x 6-inch-high waste composites (w/c = 0.6; V/S = 2.52 cm). The average CFR was normalized for V/S.

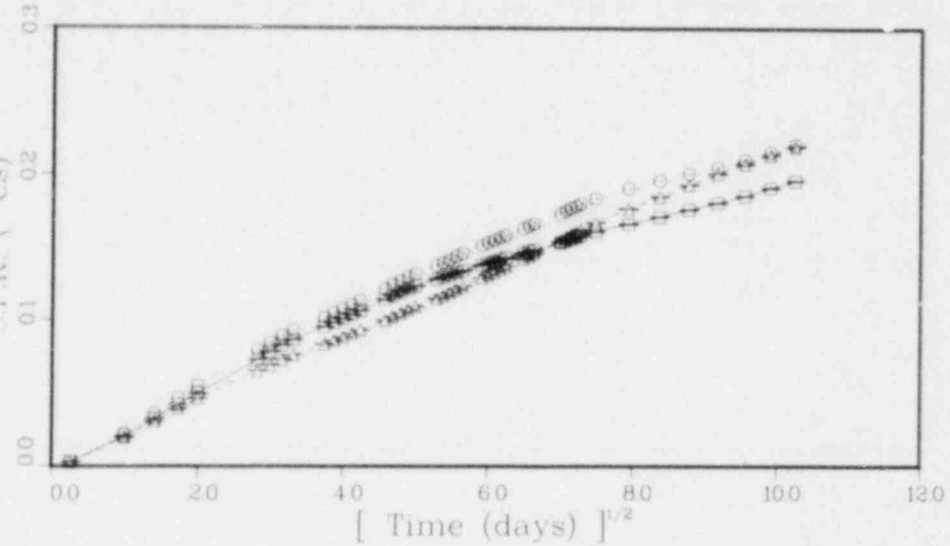


Figure 4.11 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 6-inch-diameter x 12-inch high waste composites (w/c = 0.6; V/S = 3.30 cm).

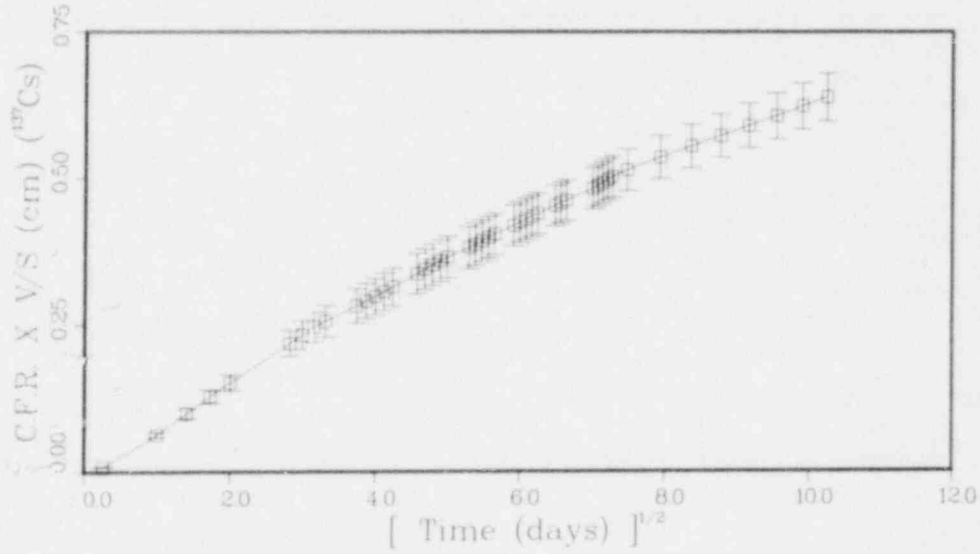


Figure 4.12 Average cumulative fractional release of ^{137}Cs vs (time)$^{1/2}$ from 6-inch-diameter x 12-inch-high waste composites (w/c = 0.6; V/S = 3.30 cm). The average CFR was normalized for V/S.

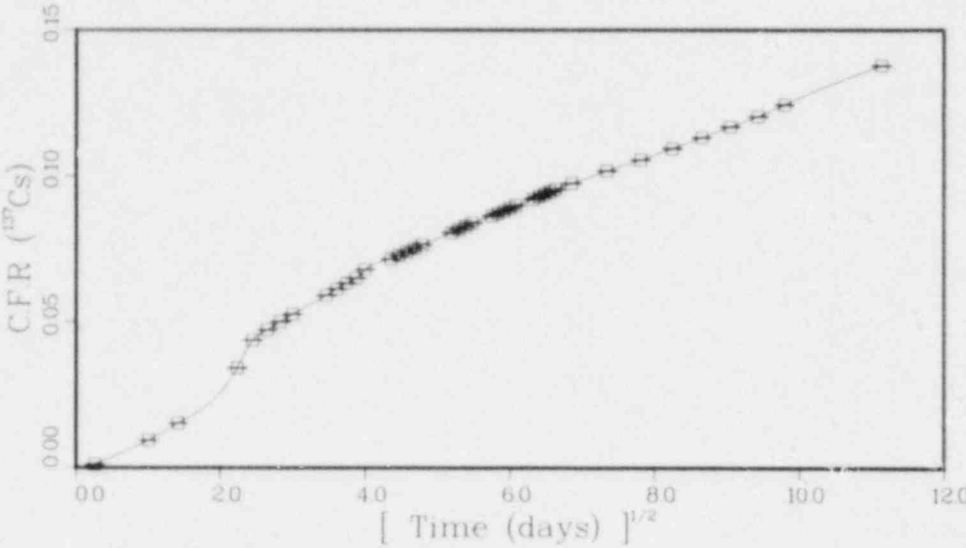


Figure 4.13 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 12-inch-diameter x 12-inch-high waste composite #1 (w/c = 0.6; V/S = 5.11 cm).

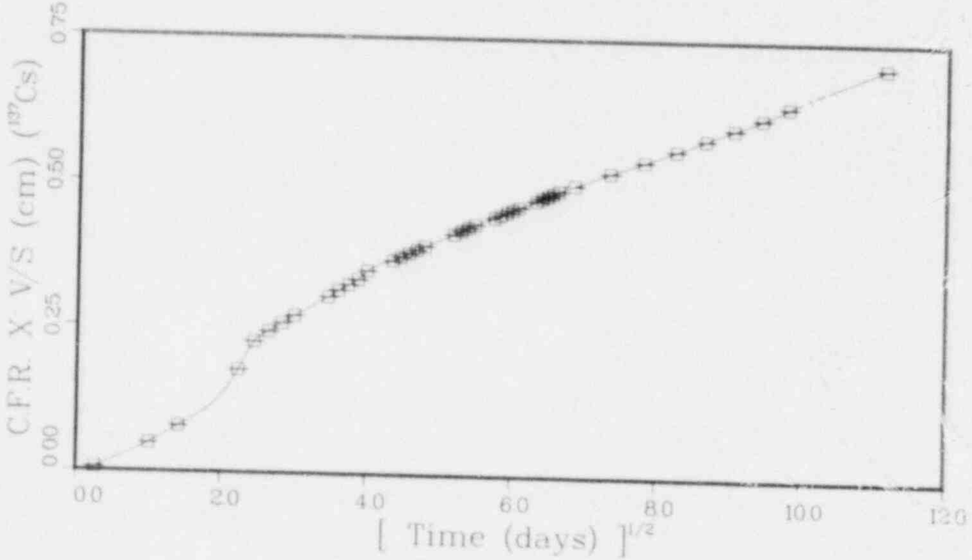


Figure 4.14 Cumulative fractional release of ^{137}Cs x V/S vs (time)$^{1/2}$ from 12-inch-diameter x 12-inch-high waste composite #1 (w/c = 0.6; V/S = 5.11 cm). CFR was normalized for V/S/.

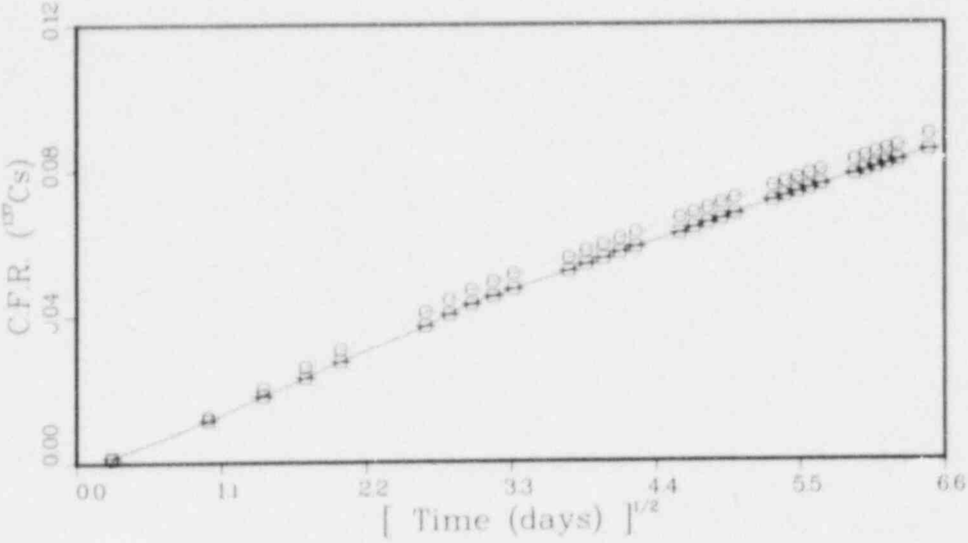


Figure 4.15 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 12-inch-diameter x 12-inch-high waste composites #2 and 3 (w/c = 0.6; V/S = 5.11 cm).

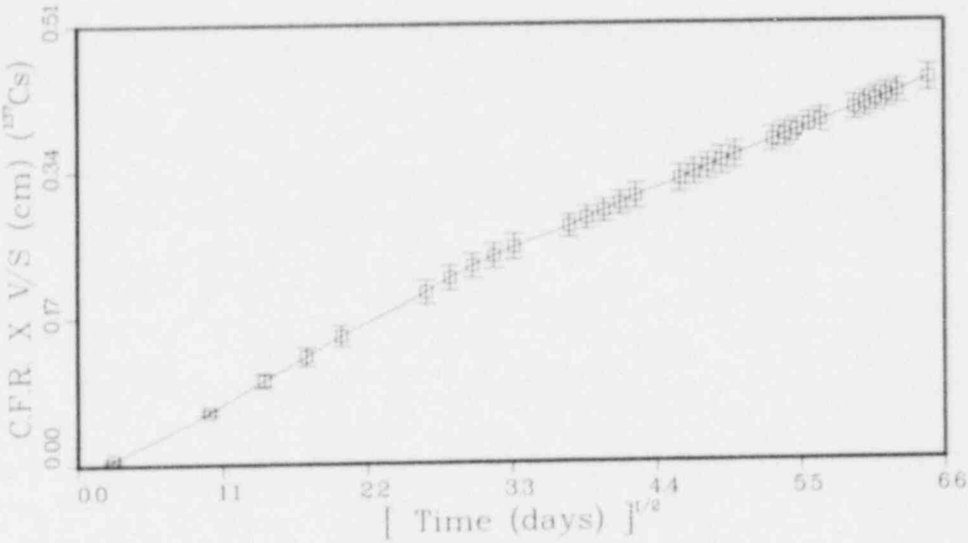


Figure 4.16 Average Cumulative fractional release of ^{137}Cs x V/S vs (time) $^{1/2}$ from 12-inch-diameter x 12-inch-high waste composites #2 and 3 (w/c = 0.6; V/S = 5.11 cm). The average CFR was normalized for V/S.

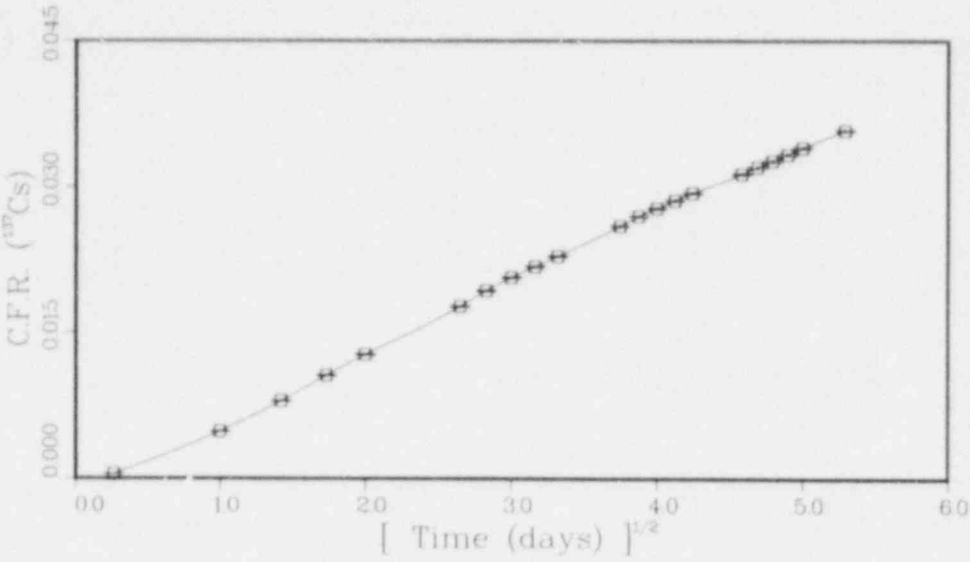


Figure 4.17 ^{137}Cs cumulative fractional release vs (time)$^{1/2}$ from 22-inch-diameter x 22-inch-high waste composite (w/c = 0.6; V/S = 9.10 cm).

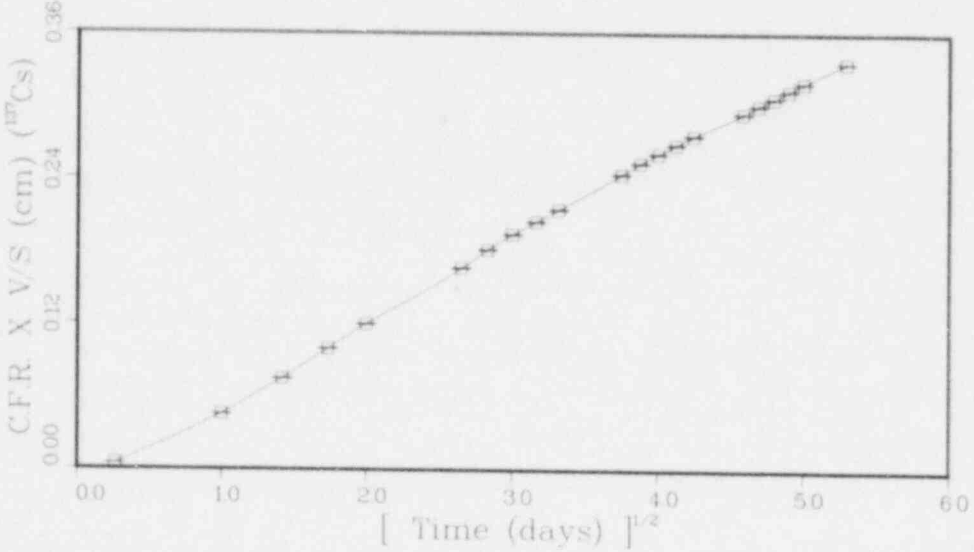


Figure 4.18 Cumulative fractional release of ^{137}Cs x V/S vs (time)$^{1/2}$ from 22-inch-diameter x 22-inch-high waste composite (w/c = 0.6; V/S = 9.10 cm). CFR was normalzied for V/S.

4.3.2 Analysis of Leaching Data

A semi-empirical approach based on the semi-infinite plane source model of diffusion is presented to analyze the leaching data. Rewriting Equation (4.2) as Equation (4.3) gives:

$$f = [S/V \cdot 2(D/\pi)^{1/2}] \times t^{1/2} + \alpha \tag{4.3}$$

This relationship describes a straight line with slope $[S/V \cdot 2(D/\pi^{1/2}]$, and intercept α when f is plotted vs $t^{1/2}$. The linear regions of the data in Figures 4.1-4.18 were determined visually and are summarized in Table 4.2.

Table 4.2

Linear Regions of CFR vs $t^{1/2}$ Data

Size	S/V	Linear Region[a] of CFR vs $t^{1/2}$
2x2	1.28	Beyond 36 days of leaching
2x4	1.07	" " " " "
3x3	0.76	" " " " "
6x6	0.40	" 16 " " "
6x12	0.33	" 49 " " "
12x12	0.19	" 9 " " "
22x22	0.11	" 7 " " "

[a] Linear regions of CFR vs $t^{1/2}$ in Figures 4.1-4.18

The average CFR's from the intact forms for each size studied were calculated over the regions outlined in Table 4.2, and were plotted vs $t^{1/2}$ as shown in Figure 4.19.

A least squares linear regression was performed on these lines to determine the best fit, slopes and intercepts. The results of these calculations are summarized in Table 4.3, together with the coefficients of determination as defined by:

$$R^2_{(S/V)} = \left(\frac{\alpha\Sigma(CFR)_i + b\Sigma(t^{1/2})_i \times (CFR)_i - 1/n(\Sigma(CFR)_i)^2}{\Sigma(CFR)_i^2 - 1/n(\Sigma(CFR)_i)^2} \right)_{(S/V)} \tag{4.4}$$

where the coeficients α and b are derived coefficients from the fitting: $(CFR)_{(S/V)} = [\alpha + b \times t^{1/2}]_{(S/V)}$ for each sample size studied.

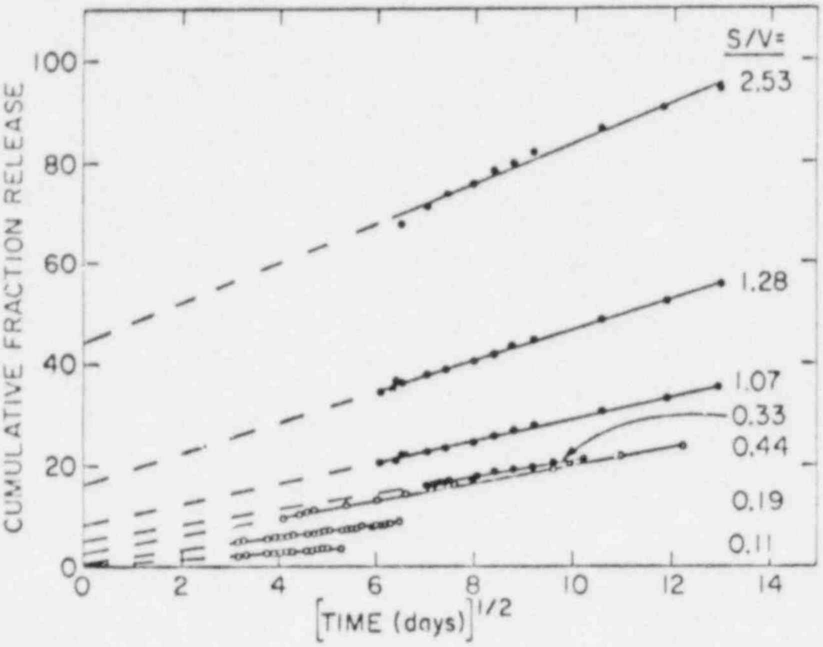


Figure 4.19 Experimental cumulative fractional release data vs $t^{1/2}$ and their least squares linear regression fits.

Table 4.3

Summary of Slopes and Intercepts of CFR vs $t^{1/2}$

Size	S/V (cm^{-1})	Slope[a] (b)	Intercept[a] (α)	Coefficient of Determination[c]
2 x 2	1.28	3.03	16.4	1.0
2 x 4	1.07	3.45	11.8	0.99
3 x 3	0.76	2.10	8.15	1.0
6 x 6	0.40	1.77	2.73	1.0
6 x 12	0.33	1.58	1.58	1.0
12 x 12	0.19	1.20	0.95	1.0
22 x 22	0.11	0.66	0.11	1.0

[a] Slopes and intercepts are obtained for the general relationship CFR = $\alpha + b(t)^{1/2}$ over the linear region of the data as explained in the text.

[b] b are the slopes of the lines in Figure 4.19.

[c] The coefficient of determination is defined in Equation 4.4.

The data from the 1 x 1 forms were excluded from further treatment since they did not appear to be linear with $t^{1/2}$ with (Figs. 4.1 and 4.2). The surface-to-volume ratio was the largest (2.53 cm^{-1}) for this size, and any surface effects will be accentuated in its leaching behavior relative to the other sample sizes.

Since the slopes of the lines (Table 4.2) are equal to $[S/V \times 2(D/\pi)]^{1/2}$, plotting these slopes as a function of S/V would yield a line with a slope equal to: $2(D/\pi)^{1/2}$, i.e., for $z = (S/V)$, then $d(\text{slope})/dz = 2(D/\pi)^{1/2} + \alpha$.

A plot of these slopes vs S/V is shown in Figure 4.20, indicating a linear relationship. A least squares linear regression on the points (slopes, S/V) (Table 4.3) yields an expression of the form: Slope = a + bx, with:

$R^2 = 0.90$,
$a = 0.75$,
$b = 2.07$

where $b = 2(D/\pi)^{1/2}$.

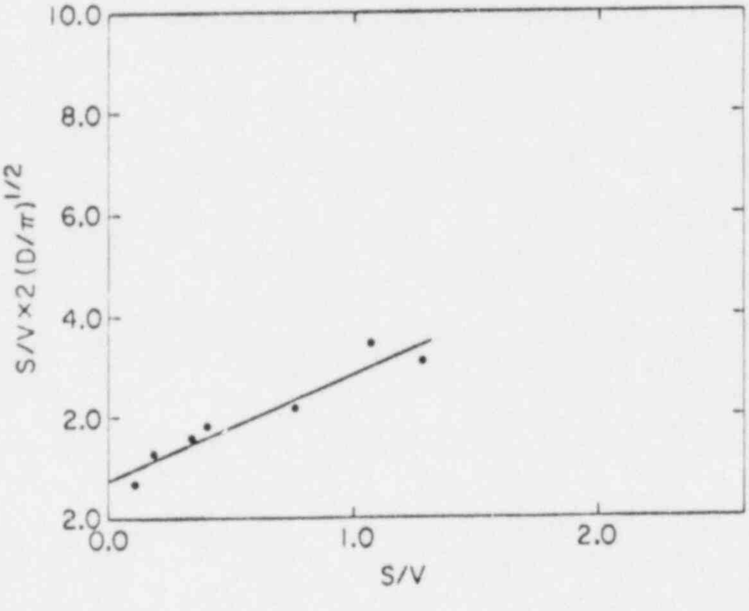


Figure 4.20 Plot of $[S/V \cdot 2(D/\pi)^{1/2}]$ or slopes of the lines shown in Figure 4.19 vs (S/V). Note: the straight line is a least squares linear regression on the points.

Therefore, the slope or $[S/V \cdot d'D/\pi)^{1/2}]$ could be calculated for any S/V ratio using the relationship:

$$(\text{slope})_{(S/V)} = 2.07 \times \frac{S}{V} + 0.75 \tag{4.5}$$

A similar fit was performed on the intercepts (α) of the lines shown in Figure 4.19, and the S/V ratios, resulting in Equation (4.6), with a coefficient of determination $R^2 = 0.99$.

$$(\alpha)_{S/V} = 1.80 + 12.6\,\frac{S}{V} \tag{4.6}$$

A plot of these intercepts $[(\alpha)_{S/V}]$ versus their corresponding S/V ratios is shown in Figure 4.21.

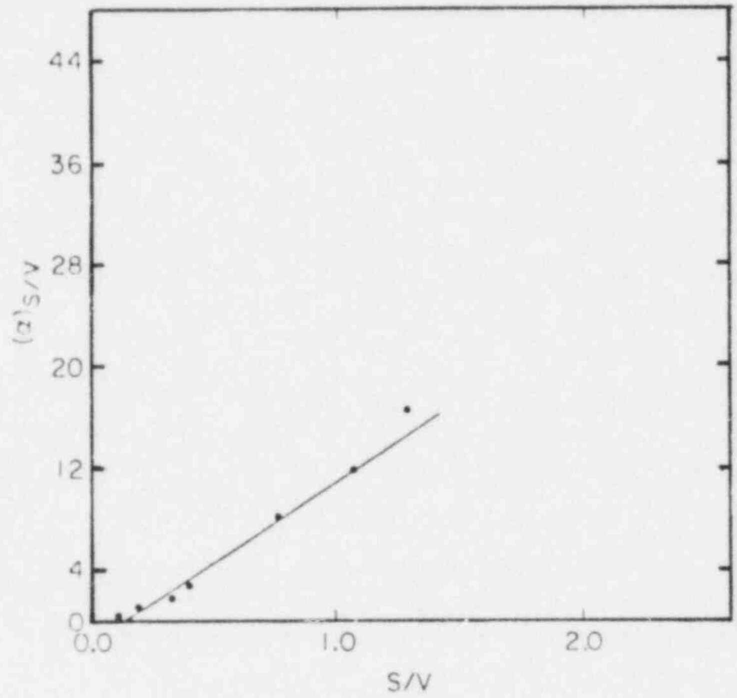


Figure 4.21 Plot of the intercepts $[(\alpha)_{S/V}]$ of the lines shown in Figure 4.19 vs S/V. Note: The straight line is a least-squares linear regression on the points for S/V less than 2.53 (i.e., forms larger than 1 x 1).

Combining Equations (4.5) and (4.6) yields

$$(\text{CFR}) = (2.07 \cdot S/V + 0.75)t^{1/2} + (12.6 \cdot S/V - 1.80) \tag{4.7}$$

Thus, the cumulative fractional release at a given time t from a sample with a geometric surface-to-volume ratio of S/V, can be calculated using Equation (4.7). The cumulative fractional release calculated for several S/V values at different time intervals are summarized in Table 4.4 together with the experimental data obtained at these values. A good agreement is observed between the calculated values and the experimental data measured over the linear region of CFR vs $t^{1/2}$.

In the derivation of the slope of Equation 4.5, it was found that:

$$\text{Slope} = 2.07 = 2(D/\pi)^{1/2}$$

Solving for D in Equation (4.8), we obtain the value of the effective bulk diffusion coefficient:

$$D = 3.37 \times 10^{-4} \ \frac{\text{cm}^2}{\text{day}} \text{ or } 3.9 \times 10^{-9} \ \text{cm}^2/\text{sec.}$$

It should be emphasized that this value for D corresponds only to the specific matrix and waste form formulation used in this experiment.

In order to estimate the radioisotopes containment period, we arbitrarily chose a limit of 95% release. The time required for 95% of radionuclide activity to be released from a waste form with a given S/V ratio can be determined from:

$$t_{0.95} = \left[\frac{(95 - 12.6 \times S/V + 1.8)}{2.07 \times S/V + 0.75} \right] \qquad (4.9)$$

The calculated times for the various waste form sizes used in this study are summarized in Table 4.5. Radioactive decay of ^{137}Cs is not considered in these calculations.

Table 4.4

Cumulative Fractional Release Experimental and Calculated Data

Dimension	S/V	Mode	Cumulative Fractional Release Time (Days)												
			4	5	9	15	21	30	42	47	56	88d	105	112	169
1 x 1	2.53	Expt.					50.7	56.8	68.6		73.5			86.4	94.0
		Calc.					57.6	62.9	68.9		74.9			93.5	108.0
2 x 2	1.28	Expt.				24.5	27.6	31.7	36.3		38.6			48.5	55.4
		Calc.				27.5	29.9	32.9	36.4		39.8			50.3	58.6
2 x 4	1.07	Expt.				20.5	23.9	28.6	34.2		37.6			48.6	55.7
		Calc.				23.2	25.3	27.9	30.9		33.9			43.1	50.3
3 x 3	0.76	Expt.				12.8	15.0	18.4	22.2		23.6			30.3	35.2
		Calc.				16.8	18.4	20.5	22.9		25.2			32.4	38.0
6 x 6	0.40	Expt.			7.20	9.20	10.7	12.2	14.4		16.3			19.6	24.0
		Calc.			7.98	9.36	10.5	11.9	13.5		15.1			19.9	23.8
6 x 12	0.33	Expt.		5.0	7.70	9.60	11.2	13.0	15.0		17.0		21.3		
		Calc.		5.57	6.66	7.91	8.93	10.2	11.7		13.1		17.1		
12 x 12	0.19	Expt.		3.40	5.30	6.50	7.38	8.40	9.37	9.73					
		Calc.		3.15	4.03	5.03	5.84	6.86	8.01	8.44					
22 x 22	0.11	Expt.	1.28		2.07	2.70	3.13	3.70	4.49			8.30			
		Calc.	1.54		2.52	3.37	4.7	4.94	5.92			8.76			

Table 4.5

Time Required for 95% Release of Radionuclide Activity From Waste Forms Having Varying Sizes

Size	S/V (cm^{-1})	Calculated Time (days)
1 x 1	2.53	120
2 x 2	1.28	560
2 x 4	1.07	790
3 x 3	0.76	1410
6 x 6	0.40	3380
6 x 12	0.33	4180
12 x 12	0.19	6820
22 x 22	0.11	9520
34 x 22.5[a]	0.09	10,440

[a] Nominal dimensions for a 55-gallon drum.

4.4 Conclusions

A relationship has been developed from the leaching data obtained to date in this study. This relationship can be used to estimate the cumulative fractional releases from forms varying in size from 1 x 1 to 22 x 22 (in inches) for a given leaching time. The effective bulk diffusion coefficient of ^{137}Cs in the waste form matrix has been estimated to be 3.9×10^{-9} cm^2/s. This value corresponds only to the specific matrix (Portland II cement) and waste form formulation used in this study. However, the method of arriving at this value can be applied to other waste forms. The calculated times required for 95% of radionuclide activity to be released from the waste forms vary from 120 days for 1 x 1 to 10,440 days for a 55-gallon drum.

5. HYDROSTATIC TESTING OF 55-GALLON CARBON STEEL DRUMS

(W. Becker, M. Hope, and N. Morcos)

5.1 Introduction

DOT 17H 55-gallon carbon steel drums are used for type A packaging at many power reactors. These drums are available in both open head (removable clamp-on lids) and closed head forms. Hydrostatic testing was performed earlier on these drums(29) to determine the maximum sustainable pressure and mode of failure. Shipping container specifications in the Code of Federal Regulations 49 CFR Part 178-118-66, state that DOT 17H drums should sustain a hydrostatic pressure of 15 psi for five minutes. Earlier results indicated that these drums did not meet the imposed specifications. The present study was undertaken to confirm these earlier results with an acceptable statistical sample of drums. Ten drums from each of three different manufacturing lots were acquired and tested. The drums were tested with the as-supplied gaskets, which were either O-rings or foam rubber.

5.2 Experimental

In preparation for hydrostatic testing, an air bleed valve with a 3 inch calibrated pressure gauge (0-30 psi) and a filling connector were brazed to a side of each drum. The drums were filled with water and the pressure gauge was monitored as the water pressure was gradually increased. The failing pressure was the pressure at which water leakage was observed. In the case where pressure was observed to drop after a rise, the highest pressure observed was noted. The physical arrangement of a drum under testing is shown in Figures 5.1 and 5.2.

5.3 Results and Discussion

The maximum sustained pressures observed in a total of 30 drums tested are summarized in Table 5.1.

Failure occurred by water leakage at the butt weld on the edge at the top of the drums, as shown in Figure 5.3.

Table 5.1

Maximum Sustained Pressure by DOT 17H Open Head Drums (psi)

	Lot A	Lot B	Lot C
1	11	8	7
2	2.5	7	5
3	5	5	15[a]
4	14	6	7
5	14	6	0
6	8	14	4
7	4[b]	3	2
8	15[c]	11	2
9	10	7	6
10	5	7	5

[a] Pressure sustained for 5 min.

[b] Foam gasket.

[c] Pressure sustained for 2 min.

5.4 Conclusions

Out of 30 drums tested, (10 from each of three manufacturing lots) only one drum was observed to meet the specifications imposed by 49 CFR Part 178-118-66. All failures were due to a poor seal created by an improper mechanical alignment at the butt weld on the upper lip of the drum.

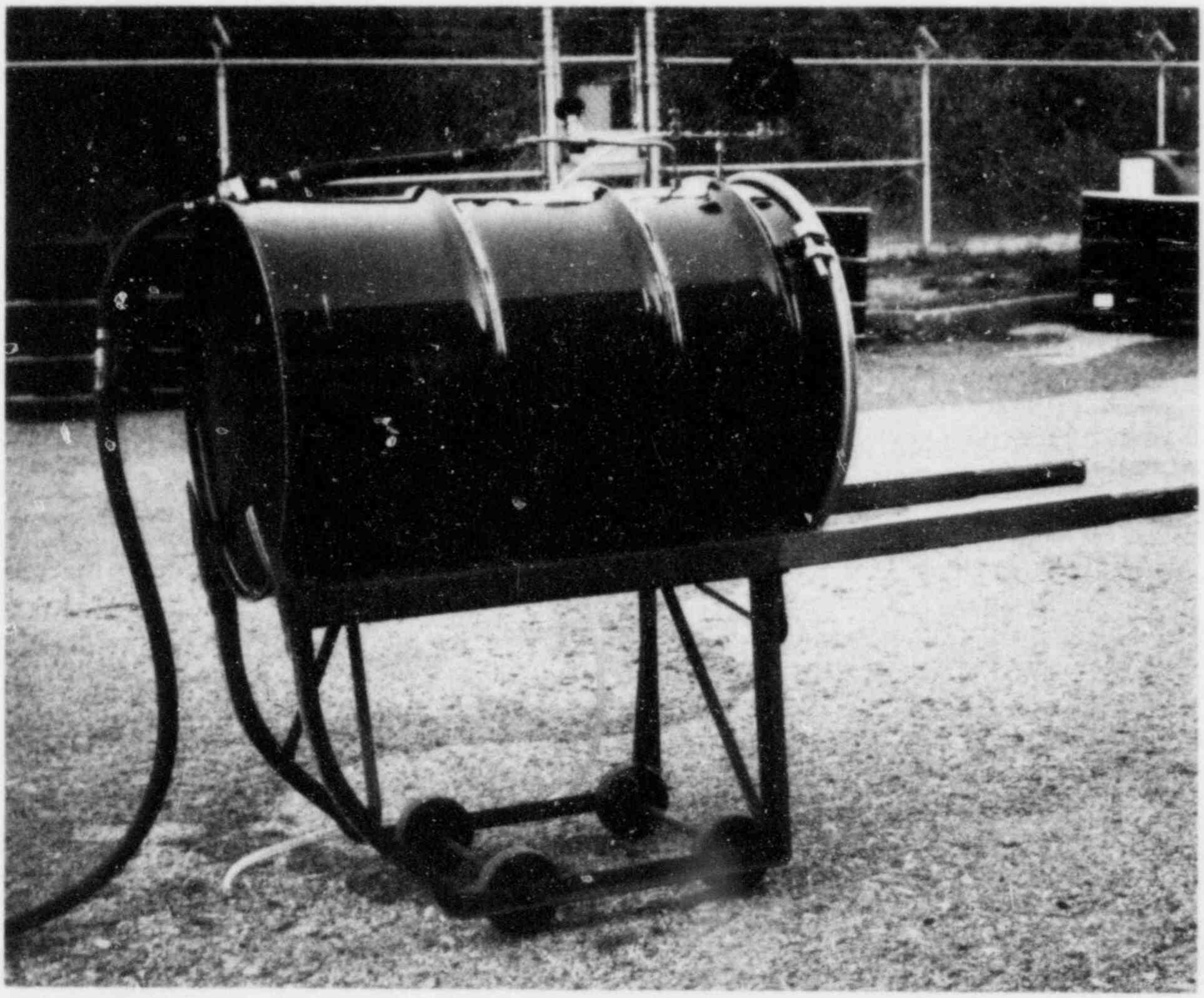


Figure 5.1 DOT 17H drum being filled with water prior to pressure testing.

Figure 5.2 DOT 17H drum undergoing pressure testing.

Figure 5.3 Butt weld on DOT 17H causing failure to sustained pressure.

6. ANALYSIS OF IRRADIATED ION EXCHANGE MATERIALS

6.1 Introduction

Organic ion exchange resins are routinely used in the nuclear industry to decontaminate water containing radionuclides. However, these resins are known to undergo radiation damage as the total absorbed dose exceeds 10^8 rad.(30)

Organic ion exchange resins were used in the decontamination of water in the auxiliary building at Three Mile Island (TMI-II) in an EPICOR decontamination system. As a consequence, some liners contain resins with high loadings of ^{137}Cs (approximately 40 Ci/ft^3 or 1300 curies per liner), and these resins could be expected to absorb a total dose as high as 10^8 rad within a two-year period.(31)

An earlier scoping study addressed some effects of external radiation (up to 2×10^9 rad) on organic ion exchange resins.(32) The effects due to radiation that were studied were: a) pH changes and agglomeration of resins during storage, c) generation of gases due to radiolysis, and d) corrosion of mild steel in contact with irradiated resins.

The present study was performed at the Georgia Institute of Technology (GIT), under a subcontract from BNL, to investigate three areas of concern regarding the effects of radiation on ion exchange resins currently used and proposed for use in the water decontamination processes at TMI-II. Organic ion exchange resins were irradiated up to a dose of 5×10^9 rad in stainless steel (Type 304) containers, and the areas addressed were:

a) The effects of radiation on the physical characteristics of the resins.

b) The chemical properties of materials resulting from radiation degradation of the resins (liquids, gases, and solids).

c) The corrosion effects on the walls of the irradiation containers in contact with the resins and their radiolytic byproducts.

The Georgia Institue of Technology research report is included as Appendix C and the results are summarized below.

6.2 Summary of Results and Conclusions From the GIT Report

6.2.1 Physical Characteristics of Irradiated Resins

When anion resion were exposed to increasing total doses from 10^8 to 10^9 rad, the following observations were made:

a) The amount of liquid generated increased from a small amount of liquid to a flowing slurry of liquid and resins.

b) The presence of amines was detected at the lower dose, whereas ammonia was detected at the higher dose.

c) The color of resins at both dose ranges was brown, and large fractured resin pieces were present and layer shedding was apparent.

Whereas, for cation resins, the amount of liquid generated did not increase with dose and no odors were detected, but the color of the resins increased in darkness with the dose up to a dark black color. The presence of fractured pieces and a floculent fine red-brown precipitate also increased with the absorbed dose. At the highest dose (5×10^9 rad), the resins tended to cling together.

6.2.2 Chemical Properties of Degradation By-Products of Irradiated Resins

6.2.2.1 Properties of Liquid Phase

The pH of the liquid phase resulting from irradiating anion exchange resins decreased from a range of 8.5 to 9.0 at 10^8 rad to a range of 8.5 to 7.5 when irradiated at a dose of 10^9 rad. The pH data from the liquid phase resulting from cation resin irradiation does not show a consistent trend with absorbed dose.

Analysis of aqueous extracts from irradiated anion resins revealed that total dissolved carbon, nitrogen, ammonia, monomethyl amine, and dimethyl amine increase with the absorbed dose; however, trimethyl amine decreased with increasing the dose. Extracts from cation resins revealed that the total dissolved carbon and sulfate increased with increasing absorbed dose. Organic sulfur was not found in extracts from irradiated cation resins.

6.2.2.2 Properties of Gas Phase

Gases generated by irradiation of resins were: H_2, O_2, N_2, CO_2, CO, CH_4, C_2H_2, C_3H_8, C_4H_{10}, and sulfur gases (SO_2, SO_3). Irradiated anion resins (0.8×10^9 rad) yielded a total volume of gases (cc/g of resin) that was approximately twice that obtained from cation resins (2.5×10^9 rad).

Hydrogen and methane production peaked at a total absorbed dose of 10^9 rad, then gradually decreased with increasing dose, whereas ethane, propane and butane production gradually increased with increasing dose up to 5×10^9 rad.

6.2.2.3 Properties of Solid Phase

Chemical analyses of irradiated ion exchange resins indicate that cation resin solids maintain essentially the same composition for total absorbed doses up to 5×10^9 rad. The carbon to hydrogen and carbon to sulfur ratios change by less than 10%, whereas for anion resins, decomposition was observed as the carbon to hydrogen ratio increased by 25% (10^9 rad), and the carbon to nitrogen ratio increased by 60% over the unirradiated resins.

6.2.3 Corrosion Effects of Radiolytic By-Products

The effects of radiolytic by-products resulting from irradiating anionic resins did not significantly attack Type 304 stainless steel, whereas the by-products from irradiated cationic resins caused significant localized corrosion of Type 304 stainless steel. The attack was in the form of etching under surface deposits and pitting. Although the corrosion attack did not cause loss of strength in the stainless steel, its localized nature might cause perforation of the container walls at longer exposures.

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APPENDIX A

LEACHING DATA FOR SECTION 1 OF THIS REPORT:

LEACHABILITY AND COMPRESSIVE STRENGTH OF BORIC ACID WASTE IN PORTLAND III CEMENT

Table A.1

^{137}Cs Incremental and Cumulative Fractions Released From Boric Acid/Portland III Cement Composites (3% Boric Acid Solution and w/c Ratio of 0.5)

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	13.34 (7.0)[a]	13.3 ± 0.9	9.01 (9.8)[a]	9.01 ± 0.88	11.2 (8.8)[a]	11.3 ± 0.96
2	7.00 (9.5)	20.4 ± 1.1	3.32 (16.0)	12.3 ± 1.0	6.64 (11.1)	17.9 ± 1.2
3	5.68 (10.6)	26.1 ± 1.3	2.47 (18.6)	14.8 ± 1.1	3.65 (14.9)	21.6 ± 1.3
4	4.02 (12.6)	30.1 ± 1.4	2.30 (19.3)	17.1 ± 1.2	1.38 (24.3)	22.9 ± 1.4
5	2.49 (16.0)	32.6 ± 1.5	2.38 (18.9)	19.5 ± 1.3	2.11 (19.6)	25.0 ± 1.4
6	2.30 (12.9)	34.9 ± 1.5	1.96 (20.9)	21.4 ± 1.4	2.10 (19.3)	27.2 ± 1.5
7	2.94 (14.8)	37.9 ± 1.6	1.28 (25.9)	22.7 ± 1.4	2.03 (20.0)	29.3 ± 1.5
8	2.11 (17.4)	40.0 ± 1.6	0.986 (29.3)	23.7 ± 1.4	1.54 (23.0)	30.8 ± 1.6
9	1.51 (20.5)	41.6 ± 1.6	1.11 (27.7)	24.8 ± 1.5	1.38 (24.3)	32.2 ± 1.6
10	1.38 (5.61)	42.8 ± 1.6	1.84 (5.6)	26.6 ± 1.5	1.57 (5.9)	33.7 ± 1.6
15	5.83 (2.80)	48.7 ± 1.6	3.86 (4.2)	30.5 ± 1.5	5.54 (3.2)	39.3 ± 1.6
20	3.87 (3.44)	52.5 ± 1.6	3.38 (4.5)	33.9 ± 1.5	4.23 (3.7)	43.5 ± 1.6
29	5.86 (2.81)	58.4 ± 1.7	5.08 (3.7)	39.0 ± 1.5	6.79 (2.9)	50.3 ± 1.7
43	7.20 (2.55)	65.6 ± 1.7	5.56 (3.5)	44.5 ± 1.5	7.29 (2.8)	57.6 ± 1.7
71	9.04 (2.31)	74.6 ± 1.7	7.65 (3.1)	52.2 ± 1.5	8.75 (2.6)	66.3 ± 1.7
100	6.87 (2.60)	81.5 ± 1.7	6.74 (3.3)	58.9 ± 1.5	6.57 (3.0)	72.9 ± 1.7
158	6.88 (2.60)	88.4 ± 1.7	7.84 (3.0)	66.7 ± 1.5	7.95 (2.8)	80.9 ± 1.7
229	5.66 (2.23)	94.1 ± 1.7	6.65 (2.4)	73.4 ± 1.6	5.56 (2.5)	86.4 ± 1.7

	Composite #4		Composite #5	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	7.28 (10.9)[a]	7.28 ± 0.80	15.4 (7.4)[a]	15.4 ± 1.0
2	3.30 (16.2)	10.6 ± 0.96	4.66 (13.4)	20.1 ± 1.3
3	3.45 (15.8)	14.1 ± 1.1	4.24 (14.0)	24.3 ± 1.4
4	1.65 (23.0)	15.7 ± 1.2	3.66 (15.1)	28.0 ± 1.5
5	1.56 (23.6)	17.3 ± 1.2	3.74 (14.9)	31.7 ± 1.6
6	1.27 (26.0)	18.5 ± 1.3	2.41 (18.6)	34.1 ± 1.7
7	0.867 (31.6)	19.4 ± 1.3	1.83 (21.4)	35.9 ± 1.7
8	1.04 (28.9)	20.4 ± 1.3	1.91 (20.9)	37.9 ± 1.8
9	0.884 (31.4)	21.3 ± 1.4	1.15 (27.0)	39.0 ± 1.8
10	0.954 (7.9)	22.3 ± 1.4	1.26 (6.6)	40.3 ± 1.8
15	3.71 (4.3)	26.0 ± 1.4	5.27 (3.4)	45.5 ± 1.8
20	2.93 (4.9)	28.9 ± 1.4	4.28 (3.7)	49.8 ± 1.8
29	4.77 (3.9)	33.7 ± 1.4	5.74 (3.2)	55.6 ± 1.8
43	5.65 (3.6)	39.3 ± 1.4	7.44 (2.9)	63.0 ± 1.8
71	7.96 (3.0)	47.3 ± 1.4	8.75 (2.7)	71.7 ± 1.9
100	7.00 (3.2)	54.3 ± 1.4	7.45 (2.9)	79.2 ± 1.9
158	7.65 (3.1)	61.9 ± 1.5	8.08 (2.8)	87.3 ± 1.9
229	6.75 (2.4)	68.7 ± 1.5	6.81 (2.3)	94.1 ± 1.9

[a] Number in ( ) = 1σ percent counting uncertainty.

Table A.2

^{137}Cs Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (3% Boric Acid Solution and w/c Ratio of 0.7)

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	15.0 (6.9)[a]	15.0 ± 1.0	16.4 (7.0)[a]	16.4 ± 1.1	14.7 (7.2)[a]	14.7 ± 1.1
2	6.04 (10.8)	21.0 ± 1.2	6.14 (11.3)	22.7 ± 1.3	5.41 (11.8)	20.1 ± 1.2
3	3.94 (13.4)	25.0 ± 1.3	4.50 (13.3)	27.2 ± 1.5	5.26 (12.0)	25.3 ± 1.4
4	5.06 (11.8)	30.0 ± 1.5	3.55 (14.9)	30.7 ± 1.6	3.46 (14.8)	28.8 ± 1.5
5	3.94 (13.4)	34.0 ± 1.6	2.05 (19.6)	32.8 ± 1.6	3.68 (14.3)	32.5 ± 1.6
6	2.60 (16.5)	36.6 ± 1.6	2.45 (18.0)	35.2 ± 1.7	3.16 (15.5)	35.6 ± 1.6
7	2.25 (17.7)	38.8 ± 1.7	3.16 (15.8)	38.4 ± 1.7	2.10 (18.9)	37.7 ± 1.7
8	2.11 (18.3)	40.9 ± 1.7	2.45 (18.0)	40.8 ± 1.8	2.93 (16.0)	40.6 ± 1.8
9	1.60 (20.9)	42.5 ± 1.7	1.44 (23.4)	42.3 ± 1.8	1.88 (20.0)	42.5 ± 1.8
10	1.97 (5.3)	44.5 ± 1.7	1.93 (5.3)	44.2 ± 1.8	1.80 (2.9)	44.3 ± 1.8
15	7.59 (2.8)	52.1 ± 1.8	6.92 (2.9)	51.1 ± 1.8	7.39 (2.9)	51.7 ± 1.8
20	5.33 (3.3)	57.4 ± 1.8	5.49 (3.2)	56.6 ± 1.9	4.84 (3.6)	56.6 ± 1.8
29	8.53 (2.6)	65.9 ± 1.8	8.68 (2.6)	65.3 ± 1.9	7.98 (2.8)	64.5 ± 1.8
43	9.64 (2.5)	75.6 ± 1.8	10.4 (2.4)	75.6 ± 1.9	8.67 (2.7)	73.2 ± 1.8
71	11.2 (2.7)	86.7 ± 1.8	12.4 (2.2)	88.0 ± 1.9	11.6 (2.4)	84.8 ± 1.9
100	7.64 (2.8)	94.4 ± 1.8	9.30 (2.5)	97.3 ± 1.9	9.66 (2.6)	94.4 ± 1.9
158	6.13 (3.1)	101.0 ± 1.8	8.75 (2.6)	1[illegible].0 ± 1.9	8.57 (2.7)	103.0 ± 1.9
229	2.99 (3.1)	104.0 ± 1.8	5.26 (2.6)	111.0 ± 1.9	5.17 (2.5)	108.0 ± 1.9

Time Days	Composite #4		Composite #5	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	13.7 (7.5)[a]	13.7 ± 1.0	11.8 (9.2)[a]	11.8 ± 1.0
2	4.89 (12.4)	18.6 ± 1.2	4.35 (13.5)	16.1 ± 1.1
3	4.51 (12.9)	23.1 ± 1.3	4.19 (13.8)	20.3 ± 1.2
4	3.91 (13.9)	27.0 ± 1.4	4.43 (13.4)	24.8 ± 1.4
5	3.61 (14.4)	30.6 ± 1.5	2.85 (16.7)	27.6 ± 1.5
6	2.33 (18.0)	33.0 ± 1.6	2.37 (18.3)	30.0 ± 1.5
7	1.96 (19.6)	34.9 ± 1.6	1.42 (23.6)	31.4 ± 1.6
8	2.63 (16.9)	37.5 ± 1.7	1.98 (20.0)	33.4 ± 1.6
9	1.75 (20.8)	39.3 ± 1.7	1.36 (24.1)	34.8 ± 1.7
10	1.41 (6.0)	40.7 ± 1.7	1.66 (6.2)	36.4 ± 1.7
15	6.61 (2.9)	47.3 ± 1.7	7.22 (3.0)	43.6 ± 1.7
20	5.18 (3.2)	52.5 ± 1.7	5.30 (3.5)	48.9 ± 1.7
29	7.99 (2.6)	60.5 ± 1.8	8.89 (2.8)	57.8 ± 1.7
43	9.13 (2.4)	69.6 ± 1.8	9.21 (2.7)	67.0 ± 1.7
71	10.0 (2.4)	79.6 ± 1.8	11.2 (2.5)	78.2 ± 1.7
100	8.25 (2.6)	87.9 ± 1.8	9.99 (2.6)	88.2 ± 1.8
158	7.59 (2.7)	95.5 ± 1.8	8.10 (2.9)	96.3 ± 1.8
229	6.09 (2.4)	102.0 ± 1.8	7.03 (2.3)	103.0 ± 1.0

[a] Number in ( ) = 1σ percent counting uncertainty.

Table A.3

^{137}Cs Incremental and Cumulative Fractions Released From Boric Acid/Portland III Cement Composites (6% Boric Acid Solution and w/c Ratio of 0.5)

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	11.05 (10.0)[a]	11.1 ± 1.0	10.58 (8.9)[a]	10.6 ± 1.0	8.59 (9.7)[a]	8.59 ± 0.83
2	5.28 (8.7)	16.3 ± 1.2	4.10 (14.2)	14.7 ± 1.1	2.89 (16.7)	11.5 ± 0.96
3	3.46 (12.5)	19.8 ± 1.3	3.77 (14.8)	18.5 ± 1.2	2.72 (17.2)	14.2 ± 1.1
4	2.80 (15.5)	22.6 ± 1.4	2.05 (20.0)	20.5 ± 1.3	1.60 (22.4)	15.8 ± 1.1
5	2.80 (15.5)	25.4 ± 1.5	2.38 (18.6)	22.9 ± 1.4	1.77 (21.4)	17.6 ± 1.2
6	1.98 (20.4)	27.4 ± 1.5	1.97 (20.4)	24.9 ± 1.4	1.61 (22.4)	19.2 ± 1.2
7	2.39 (18.6)	29.8 ± 1.6	1.15 (26.7)	26.0 ± 1.5	1.28 (25.0)	20.5 ± 1.3
8	1.81 (21.4)	31.6 ± 1.6	1.39 (24.3)	27.4 ± 1.5	1.36 (24.3)	21.8 ± 1.3
9	1.63 (22.4)	33.2 ± 1.7	1.10 (27.3)	28.5 ± 1.5	0.738 (32.8)	22.6 ± 1.3
10	1.88 (5.5)	35.1 ± 1.7	1.26 (6.7)	29.8 ± 1.5	1.25 (6.6)	23.8 ± 1.4
15	5.44 (3.5)	40.5 ± 1.7	4.13 (3.8)	33.9 ± 1.5	4.30 (3.9)	28.1 ± 1.4
20	3.93 (4.1)	44.5 ± 1.7	3.23 (4.2)	37.1 ± 1.5	3.19 (4.5)	31.3 ± 1.4
29	6.05 (3.4)	50.5 ± 1.7	5.32 (3.3)	42.4 ± 1.6	5.27 (3.6)	36.6 ± 1.4
43	7.14 (3.1)	57.7 ± 1.7	6.43 (3.0)	48.9 ± 1.6	7.14 (3.1)	43.7 ± 1.4
71	7.80 (3.0)	65.5 ± 1.7	7.56 (2.8)	56.4 ± 1.6	7.22 (3.1)	51.0 ± 1.4
100	7.05 (3.1)	72.5 ± 1.7	6.20 (3.1)	62.6 ± 1.6	7.66 (3.0)	58.6 ± 1.4
158	7.75 (3.0)	80.3 ± 1.8	7.83 (2.8)	70.5 ± 1.6	8.54 (2.8)	67.1 ± 1.5
229	6.08 (2.5)	86.4 ± 1.8	5.94 (2.5)	76.4 ± 1.6	6.72 (2.3)	73.9 ± 1.5

	Composite #4		Composite #5	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	9.80 (8.9)a	9.80 ± 0.88	7.67 (10.1)a	7.67 ± 0.78
2	6.14 (11.3)	15.9 ± 1.1	4.77 (12.8)	12.4 ± 0.99
3	3.34 (15.3)	19.3 ± 1.2	3.52 (14.9)	16.0 ± 1.1
4	2.33 (18.3)	21.6 ± 1.3	3.29 (15.5)	19.2 ± 1.2
5	3.19 (15.6)	24.8 ± 1.4	1.80 (20.9)	21.0 ± 1.3
6	2.18 (18.9)	27.0 ± 1.5	1.46 (23.1)	22.5 ± 1.3
7	2.57 (17.4)	29.5 ± 1.5	1.49 (23.0)	24.0 ± 1.4
8	1.87 (20.4)	31.4 ± 1.6	2.11 (19.3)	26.1 ± 1.4
9	1.26 (24.8)	32.7 ± 1.6	1.77 (21.1)	27.9 ± 1.5
10	1.48 (6.0)	34.1 ± 1.6	1.42 (6.1)	29.3 ± 1.5
15	5.52 (3.2)	39.7 ± 1.6	4.82 (3.6)	34.1 ± 1.5
20	4.01 (3.7)	43.7 ± 1.6	3.47 (4.3)	3.67 ± 1.5
29	5.88 (3.1)	49.6 ± 1.6	5.48 (3.4)	43.1 ± 1.5
43	6.97 (2.9)	56.5 ± 1.6	7.12 (3.0)	50.2 ± 1.5
71	8.57 (2.6)	65.1 ± 1.7	7.96 (2.9)	58.1 ± 1.5
100	6.69 (2.9)	71.8 ± 1.7	6.36 (3.2)	64.5 ± 1.6
158	7.78 (2.7)	79.5 ± 1.7	6.96 (3.1)	71.5 ± 1.6
229	5.93 (2.4)	85.5 ± 1.7	5.67 (2.5)	77.1 ± 1.6

[a] Number in ( ) = 1σ percent counting uncertainty.

Table A.4

^{137}Cs Incremental and Cumulative Fractions Released From Boric Acid/Portland III Cement Composites (6% Boric Acid Solution and w/c Ratio of 0.7)

Time Days	Composite #1 Incremental Fraction Released x 100	Composite #1 Cumulative Fraction Released x 100	Composite #2 Incremental Fraction Released x 100	Composite #2 Cumulative Fraction Released x 100	Composite #3 Incremental Fraction Released x 100	Composite #3 Cumulative Fraction Released x 100
1	10.5 (8.5)[a]	10.5 ± 0.90	10.9 (8.3)[a]	10.9 ± 0.91	10.8 (8.4)[a]	10.8 ± 0.91
2	3.61 (14.4)	14.2 ± 1.0	3.16 (15.5)	14.1 ± 1.0	3.86 (14.0)	14.7 ± 1.1
3	3.76 (14.2)	17.9 ± 1.2	2.49 (17.4)	16.6 ± 1.1	3.18 (15.5)	17.9 ± 1.2
4	2.11 (19.0)	20.0 ± 1.2	2.03 (19.3)	18.6 ± 1.2	2.35 (18.0)	20.2 ± 1.2
5	2.71 (16.7)	22.7 ± 1.3	2.11 (19.0)	20.7 ± 1.2	3.26 (15.3)	23.5 ± 1.3
6	1.81 (20.4)	24.5 ± 1.4	2.26 (18.3)	23.0 ± 1.3	1.82 (20.4)	25.3 ± 1.4
7	2.03 (19.3)	26.6 ± 1.4	0.828 (30.2)	23.8 ± 1.3	1.59 (21.8)	26.9 ± 1.4
8	1.51 (22.4)	28.1 ± 1.5	1.28 (24.3)	25.1 ± 1.4	1.74 (20.9)	28.6 ± 1.5
9	1.25 (24.6)	29.3 ± 1.5	0.858 (29.7)	25.9 ± 1.4	1.24 (24.6)	29.9 ± 1.5
10	1.33 (6.2)	30.7 ± 1.5	1.37 (6.1)	27.3 ± 1.4	1.29 (6.3)	31.2 ± 1.5
15	5.25 (3.2)	35.9 ± 1.5	3.90 (4.0)	31.2 ± 1.4	5.13 (3.2)	36.3 ± 1.5
20	3.90 (3.7)	39.8 ± 1.5	3.43 (4.2)	34.6 ± 1.4	4.15 (3.6)	40.4 ± 1.5
29	6.41 (2.9)	46.2 ± 1.5	6.4[illegible] (3.1)	41.1 ± 1.4	6.76 (2.9)	47.2 ± 1.5
43	8.38 (2.6)	54.6 ± 1.5	7.38 (2.9)	48.4 ± 1.5	8.01 (2.7)	55.2 ± 1.6
71	10.8 (2.3)	65.4 ± 1.6	10.1 (2.5)	58.6 ± 1.5	10.9 (2.3)	66.1 ± 1.6
100	8.94 (2.5)	74.3 ± 1.6	8.79 (1.2)	67.4 ± 1.5	9.12 (2.5)	75.2 ± 1.6
158	10.8 (2.3)	85.1 ± 1.6	10.2 (2.5)	77.5 ± 1.5	10.1 (2.4)	85.3 ± 1.6
229	8.13 (2.1)	93.3 ± 1.6	8.17 (2.1)	85.7 ± 1.5	7.78 (2.1)	93.1 ± 1.6

Time Days	Composite #4 Incremental Fraction Released x 100	Composite #4 Cumulative Fraction Released x 100	Composite #5 Incremental Fraction Released x 100	Composite #5 Cumulative Fraction Released x 100
1	11.3 (8.2)[a]	11.3 ± 0.93	11.9 (7.9)[a]	11.9 ± 0.93
2	3.02 (15.8)	14.3 ± 1.0	4.52 (12.7)	16.4 ± 1.1
3	2.27 (18.3)	16.5 ± 1.1	3.64 (14.2)	20.0 ± 1.2
4	2.12 (18.9)	18.7 ± 1.2	1.97 (19.3)	22.0 ± 1.3
5	1.96 (19.6)	20.6 ± 1.3	1.89 (19.6)	23.9 ± 1.3
6	1.59 (21.8)	22.2 ± 1.3	1.24 (24.2)	25.1 ± 1.4
7	1.03 (27.1)	23.2 ± 1.3	1.38 (23.0)	26.5 ± 1.4
8	0.982 (27.7)	24.2 ± 1.4	2.11 (18.6)	28.6 ± 1.4
9	1.21 (25.0)	25.4 ± 1.4	1.28 (23.9)	29.9 ± 1.5
10	1.27 (6.4)	26.7 ± 1.4	1.18 (6.5)	31.1 ± 1.5
15	4.40 (3.7)	31.1 ± 1.4	4.90 (3.3)	36.0 ± 1.5
20	3.57 (4.2)	34.7 ± 1.4	4.23 (3.5)	40.2 ± 1.5
29	6.09 (3.[illegible])	40.7 ± 1.4	6.65 (2.8)	46.9 ± 1.5
43	7.49 (2.[illegible])	48.2 ± 1.4	8.16 (2.6)	55.0 ± 1.5
71	10.1 (2.5)	58.4 ± 1.5	10.1 (2.3)	65.1 ± 1.5
100	8.69 (2.7)	67.1 ± 1.5	8.64 (2.5)	73.8 ± 1.6
158	10.4 (2.5)	77.5 ± 1.5	10.1 (2.3)	83.9 ± 1.6
229	8.28 (2.1)	85.8 ± 1.5	7.10 (2.2)	91.0 ± 1.6

[a] Number in ( ) = 1σ percent counting uncertainty.

Table A.5

^{137}Cs Incremental and Cumulative Fractions Released From Boric Acid/Portland III Cement Composites (12% Boric Acid Solution and w/c Ratio of 0.5)

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	7.75 (10.0)[a]	7.75 ± .78	8.05 (10.0)[a]	8.05 ± 0.81	8.37 (9.9)[a]	8.37 ± 0.83
2	2.25 (18.6)	10.0 ± 0.88	2.90 (16.7)	11.0 ± 0.94	2.62 (17.7)	11.0 ± 0.95
3	0.931 (28.8)	10.9 ± 0.92	3.06 (16.2)	14.0 ± 1.1	1.97 (20.4)	13.0 ± 1.0
4	2.02 (19.6)	13.0 ± 1.0	2.42 (18.3)	16.4 ± 1.2	2.05 (20.0)	15.0 ± 1.1
5	1.40 (23.6)	14.4 ± 1.1	0.805 (31.6)	17.2 ± 1.2	1.64 (22.4)	16.7 ± 1.2
6	1.29 (24.5)	15.6 ± 1.1	2.17 (19.3)	19.4 ± 1.3	1.39 (24.3)	18.0 ± 1.2
7	0.698 (33.3)	16.3 ± 1.1	1.61 (22.4)	21.0 ± 1.3	1.39 (24.3)	19.4 ± 1.3
8	1.24 (25.0)	17.6 ± 1.2	1.37 (24.2)	22.4 ± 1.3	1.56 (23.0)	21.0 ± 1.3
9	0.807 (30.8)	18.4 ± 1.2	1.16 (26.4)	23.5 ± 1.4	0.869 (30.8)	21.9 ± 1.3
10	0.869 (7.7)	19.3 ± 1.2	1.13 (6.9)	24.7 ± 1.4	0.968 (7.5)	22.8 ± 1.3
15	3.06 (4.2)	22.3 ± 1.2	3.53 (4.3)	28.2 ± 1.4	3.41 (4.1)	26.2 ± 1.4
20	2.44 (4.7)	24.7 ± 1.2	2.80 (4.8)	31.0 ± 1.4	2.46 (4.8)	28.7 ± 1.4
29	4.53 (3.5)	29.3 ± 1.2	4.67 (3.8)	35.7 ± 1.4	4.71 (3.5)	33.4 ± 1.4
43	4.76 (3.4)	34.0 ± 1.2	5.96 (3.4)	41.6 ± 1.4	5.74 (3.2)	39.2 ± 1.4
71	6.02 (3.1)	40.0 ± 1.2	6.72 (3.2)	48.3 ± 1.4	6.50 (3.0)	45.7 ± 1.4
100	4.89 (3.4)	44.9 ± 1.3	5.46 (3.5)	53.8 ± 1.5	5.25 (3.3)	50.9 ± 1.4
158	6.71 (2.9)	51.7 ± 1.3	6.70 (3.2)	60.5 ± 1.5	6.56 (3.0)	57.5 ± 1.4
229	6.67 (2.3)	58.3 ± 1.3	5.70 (2.5)	66.2 ± 1.5	6.06 (2.5)	63.5 ± 1.4

Time Days	Composite #4		Composite #5	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
1	6.05 (11.4)[a]	6.04 ± 0.69	5.94 (11.7)[a]	5.94 ± 0.67
2	2.56 (17.4)	8.61 ± 0.82	3.90 (14.4)	9.84 ± 0.90
3	2.56 (17.4)	11.2 ± 0.93	3.17 (16.0)	13.0 ± 1.0
4	1.78 (20.9)	13.0 ± 1.0	3.17 (16.0)	16.1 ± 1.1
5	1.47 (23.0)	14.4 ± 1.1	1.87 (20.9)	18.1 ± 1.2
6	1.55 (22.4)	16.0 ± 1.1	1.55 (23.0)	19.6 ± 1.3
7	1.40 (23.6)	17.4 ± 1.2	1.22 (25.9)	20.8 ± 1.3
8	1.13 (26.1)	18.5 ± 1.2	0.895 (30.2)	21.7 ± 1.3
9	1.29 (24.6)	19.8 ± 1.2	0.830 (31.4)	22.5 ± 1.4
10	1.01 (7.3)	20.8 ± 1.2	0.813 (8.2)	23.4 ± 1.4
15	3.54 (4.2)	24.3 ± 1.3	3.87 (3.8)	27.2 ± 1.4
20	3.04 (4.5)	27.4 ± 1.3	2.83 (4.5)	30.1 ± 1.4
29	4.82 (3.6)	32.2 ± 1.3	5.29 (3.3)	35.3 ± 1.4
43	5.85 (3.3)	38.1 ± 1.3	5.61 (3.2)	41.0 ± 1.4
71	6.81 (3.1)	44.9 ± 1.3	6.73 (3.0)	47.7 ± 1.4
100	5.09 (3.6)	50.0 ± 1.3	5.54 (3.3)	53.2 ± 1.4
158	6.41 (3.2)	56.4 ± 1.3	6.34 (3.0)	59.6 ± 1.4
229	5.31 (2.5)	61.7 ± 1.3	5.80 (2.5)	65.4 ± 1.4

[a] Number in ( ) = 1σ percent counting uncertainty.

Table A.6

^{137}Cs Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (12% Boric Acid Solution and w/c Ratio of 0.7)

Time Days	Composite #1 Incremental Fraction Released x 100	Composite #1 Cumulative Fraction Released x 100	Composite #2 Incremental Fraction Released x 100	Composite #2 Cumulative Fraction Released x 100	Composite #3 Incremental Fraction Released x 100	Composite #3 Cumulative Fraction Released x 100
1	9.14 (9.2)[a]	9.14 ± 0.84	9.85 (8.6)[a]	9.85 ± 0.85	11.3 (8.2)[a]	11.3 ± 0.92
2	3.46 (14.9)	12.6 ± 1.0	2.55 (16.9)	12.4 ± 0.96	4.56 (12.8)	15.9 ± 1.1
3	2.38 (18.0)	15.0 ± 1.1	2.77 (16.2)	15.2 ± 1.1	2.47 (17.4)	18.3 ± 1.2
4	3.38 (15.1)	18.4 ± 1.2	1.02 (26.7)	16.2 ± 1.1	2.25 (18.3)	20.6 ± 1.2
5	2.23 (18.6)	20.6 ± 1.3	2.48 (17.2)	18.7 ± 1.2	2.02 (19.3)	22.6 ± 1.3
6	2.07 (19.3)	22.7 ± 1.3	1.97 (19.3)	20.6 ± 1.2	1.57 (21.8)	24.2 ± 1.3
7	0.845 (30.2)	23.5 ± 1.3	1.68 (20.9)	22.3 ± 1.3	1.87 (20.0)	26.0 ± 1.4
8	1.54 (22.4)	25.1 ± 1.4	1.24 (24.3)	23.6 ± 1.3	1.95 (19.6)	28.0 ± 1.5
9	1.14 (26.0)	26.2 ± 1.4	0.890 (28.7)	24.5 ± 1.3	1.32 (23.9)	29.3 ± 1.5
10	1.46 (6.0)	27.6 ± 1.4	0.992 (7.1)	25.4 ± 1.3	1.26 (6.4)	30.6 ± 1.5
15	4.53 (3.7)	32.2 ± 1.4	4.16 (3.5)	29.6 ± 1.3	4.82 (3.6)	35.4 ± 1.5
20	3.60 (4.1)	35.8 ± 1.4	3.46 (3.8)	33.1 ± 1.4	3.80 (4.0)	39.2 ± 1.5
29	6.33 (3.2)	42.1 ± 1.4	6.00 (3.0)	39.1 ± 1.4	7.11 (3.0)	46.3 ± 1.5
43	8.44 (2.8)	50.6 ± 1.5	7.78 (2.6)	46.8 ± 1.4	7.87 (2.8)	54.2 ± 1.5
71	10.4 (2.5)	61.0 ± 1.5	10.9 (2.3)	57.8 ± 1.4	11.3 (2.4)	65.5 ± 1.6
100	8.73 (2.7)	69.7 ± 1.5	8.90 (2.5)	66.7 ± 1.4	9.12 (2.7)	74.6 ± 1.6
158	10.0 (2.6)	79.7 ± 1.5	10.30 (2.3)	76.9 ± 1.4	9.97 (2.6)	84.5 ± 1.6
229	7.77 (1.8)	87.6 ± 1.5	7.30 (2.1)	84.2 ± 1.5	7.26 (2.2)	91.8 ± 1.6

Time Days	Composite #4 Incremental Fraction Released x 100	Composite #4 Cumulative Fraction Released x 100	Composite #5 Incremental Fraction Released x 100	Composite #5 Cumulative Fraction Released x 100
1	10.9 (8.3)[a]	10.9 ± 0.90	9.98 (8.8)[a]	9.98 ± 0.88
2	3.13 (15.5)	14.0 ± 1.0	2.71 (16.9)	12.7 ± 1.0
3	2.76 (16.5)	16.8 ± 1.1	2.86 (16.5)	15.6 ± 1.1
4	1.71 (20.9)	18.5 ± 1.2	1.78 (20.9)	17.3 ± 1.2
5	2.16 (18.6)	20.6 ± 1.2	2.24 (18.6)	19.6 ± 1.2
6	1.04 (26.7)	21.7 ± 1.3	1.47 (23.0)	21.0 ± 1.3
7	1.34 (23.6)	23.0 ± 1.3	1.86 (20.4)	22.9 ± 1.3
8	1.86 (20.0)	24.9 ± 1.4	1.24 (25.0)	24.1 ± 1.4
9	1.22 (24.7)	26.1 ± 1.4	1.15 (26.0)	25.3 ± 1.4
10	1.37 (6.0)	27.5 ± 1.4	1.18 (6.6)	26.5 ± 1.4
15	4.65 (3.4)	32.1 ± 1.4	4.01 (4.0)	30.5 ± 1.4
20	3.61 (3.8)	35.7 ± 1.4	3.28 (4.4)	33.8 ± 1.4
29	6.87 (2.8)	42.6 ± 1.4	6.25 (3.2)	40.0 ± 1.4
43	8.09 (2.6)	50.7 ± 1.4	7.46 (3.0)	47.5 ± 1.5
71	10.8 (2.3)	61.4 ± 1.5	9.75 (2.6)	57.2 ± 1.5
100	8.65 (2.5)	70.1 ± 1.5	8.24 (2.8)	65.4 ± 1.5
158	9.02 (2.5)	79.1 ± 1.5	8.67 (2.8)	74.1 ± 1.5
229	6.86 (2.2)	86.0 ± 1.5	6.92 (2.3)	81.0 ± 1.5

[a] Number in ( ) = 1σ percent counting uncertainty.

Table A.7

^{85}Sr Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (3% Boric Acid Solution and w/c Ratio of 0.5)

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	1.23 (20.7)	1.23 ± 0.25	0.809 (25.0)	0.809 ± 0.20	0.919 (23.4)	0.919 ± 0.22
2	0.000 ± 0.01	1.23 ± 0.25	0.000 ± 0.01	0.809 ± 0.20	0.000 ± 0.01	0.919 ± 0.22
3	0.000 ± 0.01	1.23 ± 0.25	0.000 ± 0.01	0.809 ± 0.20	0.000 ± 0.01	0.919 ± 0.22
4	0.000 ± 0.01	1.23 ± 0.25	0.000 ± 0.01	0.809 ± 0.20	0.000 ± 0.01	0.919 ± 0.22
5	0.000 ± 0.01	1.23 ± 0.25	0.000 ± 0.01	0.809 ± 0.20	0.000 ± 0.01	0.919 ± 0.22
6	0.000 ± 0.01	1.23 ± 0.25	0.000 ± 0.01	0.809 ± 0.20	0.000 ± 0.01	0.919 ± 0.22
7	0.000 ± 0.01	1.23 ± 0.25	0.000 ± 0.01	0.809 ± 0.20	0.687 (27.1)	1.61 ± 0.29
8	0.000 ± 0.01	1.23 ± 0.26	0.000 ± 0.01	0.809 ± 0.20	0.000 ± 0.01	1.61 ± 0.29
9	0.179 (54.1)	1.41 ± 0.28	0.000 ± 0.01	0.809 ± 0.21	0.000 ± 0.01	1.61 ± 0.29
10	0.000 ± 0.01	1.41 ± 0.28	0.000 ± 0.01	0.809 ± 0.21	0.121 (16.7)	1.73 ± 0.29
15	0.274 (11.2)	1.69 ± 0.28	0.344 (10.0)	1.15 ± 0.21	0.353 (9.7)	2.08 ± 0.29
20	0.326 (10.3)	2.01 ± 0.28	0.364 (9.7)	1.52 ± 0.21	0.374 (9.5)	2.45 ± 0.29
29	0.105 (18.0)	2.12 ± 0.28	0.546 (8.0)	2.06 ± 0.21	0.464 (8.5)	2.92 ± 0.29
43	0.548 (8.1)	2.66 ± 0.28	0.536 (6.1)	2.60 ± 0.22	0.475 (8.5)	3.39 ± 0.30
71	0.284 (11.1)	2.95 ± 0.29	0.546 (8.0)	3.14 ± 0.22	0.878 (6.2)	4.27 ± 0.30
100	0.502 (14.7)	3.45 ± 0.29	0.488 (14.9)	3.63 ± 0.23	0.733 (12.0)	5.00 ± 0.31
158	0.000 ± 0.01	3.45 ± 0.29	0.763 (16.3)	4.40 ± 0.27	1.07 (13.6)	6.08 ± 0.35
229	0.000 ± 0.01	3.45 ± 0.30	0.000 ± 0.01	4.40 ± 0.27	0.000 ± 0.01	6.08 ± 0.35

	Composite #4		Composite #5	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	1.25 (20.7)	1.25 ± 0.26	1.02 (22.3)	1.02 ± 0.23
2	0.000 ± 0.01	1.25 ± 0.26	0.000 ± 0.01	1.02 ± 0.23
3	0.000 ± 0.01	1.25 ± 0.26	0.879 (24.0)	1.90 ± 0.31
4	0.000 ± 0.01	1.25 ± 0.26	0.000 ± 0.01	1.90 ± 0.31
5	0.000 ± 0.01	1.25 ± 0.26	0.000 ± 0.01	1.90 ± 0.31
6	0.782 (26.2)	2.04 ± 0.33	0.000 ± 0.01	1.90 ± 0.31
7	0.000 ± 0.01	2.04 ± 0.33	0.000 ± 0.01	1.90 ± 0.31
8	0.000 ± 0.01	2.04 ± 0.33	0.000 ± 0.01	1.90 ± 0.31
9	0.054 (91.7)	2.10 ± 0.34	0.000 ± 0.01	1.90 ± 0.31
10	0.150 (15.7)	2.25 ± 0.34	0.152 (14.7)	2.05 ± 0.31
15	0.204 (13.2)	2.45 ± 0.34	0.344 (10.0)	2.40 ± 0.32
20	0.214 (13.0)	2.67 ± 0.34	0.354 (9.7)	2.75 ± 0.32
29	0.482 (8.7)	3.15 ± 0.34	0.435 (8.9)	3.18 ± 0.32
43	0.578 (8.0)	3.73 ± 0.35	0.900 (6.1)	4.08 ± 0.32
71	0.782 (6.9)	4.51 ± 0.30	0.758 (6.7)	4.84 ± 0.33
100	0.612 (13.7)	5.12 ± 0.36	0.637 (12.9)	5.48 ± 0.34
158	0.944 (15.0)	6.06 ± 0.39	0.638 (17.6)	6.12 ± 0.36
229	0.000 ± 0.01	6.06 ± 0.39	0.000 ± 0.01	6.12 ± 0.36

[a] Number in ( ) = 1σ percent counting uncertainty.
[b] For incremental fraction releases equal to zero, the error is based on the minimum detectable limit.

Table A.8

^{85}Sr Incremental and Cumulative Fractions Released From Boric Acid/Portland III Cement Composites (3% Boric Acid Solution and w/c Ratio of 0.7)

Time Days	Composite #1 Incremental Fraction Released x 100[a,b]	Composite #1 Cumulative Fraction Released x 100	Composite #2 Incremental Fraction Released x 100[a,b]	Composite #2 Cumulative Fraction Released x 100	Composite #3 Incremental Fraction Released x 100[a,b]	Composite #3 Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.298 (43.0)	0.298 ± 0.13	1.63 (18.0)	1.63 ± 0.29
2	0.000 ± 0.01	0.000 ± 0.02	1.25 (14.9)	1.54 ± 0.23	1.14 (15.2)	2.77 ± 0.34
3	0.773 ± (25.7)	0.773 ± 0.20	0.000 ± 0.01	1.54 ± 0.23	0.000 ± 0.01	2.77 ± 0.34
4	0.000 ± 0.01	0.773 ± 0.20	0.000 ± 0.01	1.54 ± 0.23	0.000 ± 0.01	2.77 ± 0.34
5	0.000 ± 0.01	0.773 ± 0.20	0.000 ± 0.01	1.54 ± 0.23	0.000 ± 0.01	2.77 ± 0.34
6	0.000 ± 0.01	0.773 ± 0.20	0.000 ± 0.01	1.54 ± 0.23	0.000 ± 0.01	2.77 ± 0.34
7	0.000 ± 0.01	0.773 ± 0.20	0.000 ± 0.01	1.54 ± 0.23	0.000 ± 0.01	2.77 ± 0.34
8	0.000 ± 0.01	0.773 ± 0.20	0.000 ± 0.01	1.54 ± 0.23	0.000 ± 0.01	2.77 ± 0.34
9	0.295 (41.4)	1.07 ± 0.24	0.132 (64.2)	1.68 ± 0.24	0.548 (30.1)	3.32 ± 0.38
10	0.285 (11.1)	1.35 ± 0.24	0.287 (11.2)	1.96 ± 0.25	0.422 (9.3)	3.74 ± 0.38
15	0.906 (6.2)	2.26 ± 0.24	0.628 (7.8)	2.59 ± 0.25	0.969 (6.1)	4.71 ± 0.39
20	0.895 (6.3)	3.15 ± 0.25	0.661 (7.5)	3.25 ± 0.26	0.696 (7.2)	5.41 ± 0.39
29	1.04 (5.8)	4.19 ± 0.26	0.849 (6.7)	4.10 ± 0.26	0.969 (6.1)	6.38 ± 0.40
43	1.67 (4.6)	5.86 ± 0.27	1.30 (5.4)	5.40 ± 0.27	1.28 (5.3)	7.65 ± 0.40
71	1.50 (4.9)	7.36 ± 0.28	1.63 (4.8)	7.03 ± 0.28	1.26 (5.4)	8.92 ± 0.41
100	1.07 (10.0)	8.43 ± 0.30	0.574 (14.2)	7.61 ± 0.29	0.991 (10.7)	9.91 ± 0.42
158	0.000 ± 0.01	8.43 ± 0.30	0.000 ± 0.01	7.61 ± 0.29	1.27 (12.9)	11.17 ± 0.45
229	0.000 ± 0.01	8.43 ± 0.30	0.000 ± 0.01	7.61 ± 0.30	0.000 ± 0.01	11.17 ± 0.45

Time Days	Composite #4 Incremental Fraction Released x 100[a,b]	Composite #4 Cumulative Fraction Released x 100	Composite #5 Incremental Fraction Released x 100[a,b]	Composite #5 Cumulative Fraction Released x 100
1	2.25 (15.5)	2.25 ± 0.34	0.000 ± 0.01	0.000 ± 0.01
2	0.704 (27.7)	2.96 ± 0.40	0.000 ± 0.01	0.000 ± 0.02
3	0.000 ± 0.01	2.96 ± 3.40	0.000 ± 0.01	0.000 ± 0.02
4	0.000 ± 0.01	2.96 ± 0.40	0.000 ± 0.01	0.000 ± 0.03
5	0.000 ± 0.01	2.96 ± 0.40	0.000 ± 0.01	0.000 ± 0.03
6	0.000 ± 0.01	2.96 ± 0.40	0.000 ± 0.01	0.000 ± 0.03
7	0.000 ± 0.01	2.96 ± 0.40	0.000 ± 0.01	0.000 ± 0.04
8	0.000 ± 0.01	2.96 ± 0.40	0.000 ± 0.01	0.000 ± 0.04
9	0.455 (34.5)	3.41 ± 0.43	0.309 (43.0)	0.309 ± 0.14
10	0.228 (12.4)	3.64 ± 0.43	0.286 (11.6)	0.595 ± 0.14
15	0.542 (8.2)	4.18 ± 0.43	0.732 (7.38)	1.33 ± 0.15
20	0.672 (7.3)	4.85 ± 0.44	0.526 (8.72)	1.85 ± 0.16
29	0.574 (8.0)	5.43 ± 0.44	0.618 (8.00)	2.47 ± 0.17
43	1.34 (5.2)	6.77 ± 0.45	1.60 (4.98)	4.07 ± 0.18
71	0.347 (10.3)	7.12 ± 0.45	0.457 (8.28)	4.53 ± 0.19
100	0.000 ± 0.01	7.12 ± 0.45	0.000 ± 0.01	4.53 ± 0.19
158	1.03 (14.3)	8.15 ± 0.47	0.000 ± 0.01	4.53 ± 0.19
229	0.000 ± 0.01	8.15 ± 0.47	0.000 ± 0.01	4.53 ± 0.20

[a] Number in ( ) = 1σ percent counting uncertainty.

[b] For incremental fraction releases equal to zero, the error is based on the minimum detectable limit.

Table A.9

^{85}Sr Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (6% Boric Acid Solution and w/c Ratio of 0.5)

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.00 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.000 ± 0.01	0.00 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
3	0.000 ± 0.01	0.00 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.03	0.000 ± 0.03
4	0.000 ± 0.01	0.00 ± 0.03	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.03
5	0.000 ± 0.01	0.00 ± 0.03	0.572 (29.8)	0.572 ± 0.17	0.000 ± 0.01	0.000 ± 0.03
6	0.000 ± 0.01	0.00 ± 0.03	0.000 ± 0.01	0.572 ± 0.17	0.000 ± 0.01	0.000 ± 0.04
7	0.000 ± 0.01	0.00 ± 0.04	0.000 ± 0.01	0.572 ± 0.17	0.000 ± 0.01	0.000 ± 0.04
8	0.000 ± 0.01	0.00 ± 0.04	0.000 ± 0.01	0.572 ± 0.17	0.000 ± 0.01	0.000 ± 0.04
9	0.175 (54.1)	0.175 ± 0.10	0.174 (54.1)	0.746 ± 0.20	0.256 (46.5)	0.256 ± 0.13
10	0.227 (12.3)	0.402 ± 0.11	0.153 (14.7)	0.899 ± 0.20	0.223 (13.0)	0.479 ± 0.13
15	0.443 (8.86)	0.845 ± 0.11	0.603 (7.66)	1.50 ± 0.20	0.468 (8.84)	0.947 ± 0.14
20	0.319 (10.3)	1.16 ± 0.12	0.460 (8.69)	1.96 ± 0.21	0.346 (10.3)	1.29 ± 0.14
29	0.598 (7.62)	1.76 ± 0.13	0.510 (8.23)	2.47 ± 0.21	0.624 (7.71)	1.92 ± 0.15
43	0.783 (6.61)	2.56 ± 0.14	0.674 (7.15)	3.15 ± 0.22	0.713 (7.22)	2.63 ± 0.16
71	0.680 (7.15)	3.23 ± 0.14	0.797 (6.70)	3.94 ± 0.22	0.557 (8.23)	3.19 ± 0.16
100	0.758 (11.8)	3.98 ± 0.17	0.542 (14.1)	4.49 ± 0.24	0.497 (15.3)	3.69 ± 0.18
158	0.000 ± 0.01	3.98 ± 0.17	0.836 (15.7)	5.32 ± 0.27		
229	0.000 ± 0.01	3.98 ± 0.18	0.000 ± 0.04	5.32 ± 9.27		

	Composite #4		Composite #5	
Time Days	Incremental Fractinn Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.596 (29.2)	0.596 ± 0.17
2	0.543 (32.1)	0.543 ± 0.17	0.000 ± 0.01	0.596 ± 0.17
3	0.000 ± 0.02	0.543 ± 0.18	0.000 ± 0.01	0.596 ± 0.17
4	0.000 ± 0.01	0.543 ± 0.18	0.000 ± 0.01	0.596 ± 0.18
5	0.000 ± 0.01	0.543 ± 0.18	0.000 ± 0.01	0.596 ± 0.18
6	0.000 ± 0.01	0.543 ± 0.18	0.000 ± 0.01	0.596 ± 0.18
7	0.288 (43.8)	0.832 ± 0.22	0.000 ± 0.01	0.596 ± 0.18
8	0.000 ± 0.01	0.832 ± 0.22	0.000 ± 0.01	0.596 ± 0.18
9	0.000 ± 0.02	0.832 ± 0.22	0.000 ± 0.01	0.596 ± 0.18
10	0.155 (15.7)	0.987 ± 0.22	0.121 (16.7)	0.717 ± 0.18
15	0.510 (8.72)	1.50 ± 0.22	0.353 (9.74)	1.07 ± 0.18
20	0.455 (9.05)	1.95 ± 0.23	0.283 (11.1)	1.35 ± 0.18
29	0.710 (7.38)	2.66 ± 0.23	0.72[illegible] (6.84)	2.08 ± 0.19
43	0.876 (6.62)	3.54 ± 0.24	0.424 (8.84)	2.50 ± 0.20
71	0.865 (6.71)	4.40 ± 0.25	0.464 (8.51)	2.97 ± 0.20
100	0.000 ± 0.01	4.40 ± 0.25	0.447 (15.4)	3.42 ± 0.21
158	0.824 (16.4)	5.23 ± 0.28	1.00 (14.0)	4.42 ± 0.25
229	0.000 ± 0.04	5.23 ± 0.29	0.000 ± 0.04	4.42 ± 0.26

[a] Number in ( ) = 1σ percent counting uncertainty.

[b] For incremental fraction releases equal to zero, the error is based on the minimum detectable limit.

Table A.10

^{85}Sr Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (6% Boric Acid Solution and w/c Ratio of 0.7)

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.01
3	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.300 ± 0.01	0.000 ± 0.01
4	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.01
5	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.02
6	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.02
7	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
8	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.03
9	0.000 ± 0.01	0.000 ± 0.03	0.232 (32.1)	0.232 ± 0.08	0.000 ± 0.01	0.000 ± 0.04
10	0.219 (11.1)	0.219 ± 0.04	0.077 (20.0)	0.309 ± 0.08	0.192 (13.2)	0.192 ± 0.04
15	0.266 (10.0)	0.485 ± 0.05	0.193 (13.0)	0.503 ± 0.09	0.192 (13.2)	0.383 ± 0.05
20	0.258 (10.3)	0.743 ± 0.05	0.242 (11.6)	0.745 ± 0.99	0.272 (11.5)	0.655 ± 0.06
29	0.305 (9.5)	1.05 ± 0.06	0.290 (10.7)	1.04 ± 0.10	0.232 (12.2)	0.887 ± 0.07
43	0.422 (8.0)	1.47 ± 0.07	0.387 (9.3)	1.42 ± 0.10	0.444 (8.9)	1.33 ± 0.08
71	0.524 (7.2)	1.99 ± 0.08	0.343 (9.7)	1.77 ± 0.11	0.403 (9.3)	1.73 ± 0.09
100	0.748 (10.6)	2.74 ± 0.11	0.238 (20.7)	2.01 ± 0.12	0.714 (12.3)	2.45 ± 0.12
158	0.702 (14.9)	3.45 ± 0.15	0.429 (21.0)	2.44 ± 0.15	0.692 (17.1)	3.14 ± 0.17
229	0.000 ± 0.01	3.45 ± 0.16	0.000 ± 0.05	2.44 ± 0.15	0.000 ± 0.01	3.14 ± 0.17

	Composite #4		Composite #5	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
3	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
4	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.01
5	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
6	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
7	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
8	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
9	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.04
10	0.141 (15.7)	0.141 ± 0.05	0.136 (14.6)	0.136 ± 0.04
15	0.292 (10.7)	0.433 ± 0.05	0.234 (12.1)	0.370 ± 0.05
20	0.312 (10.3)	0.745 ± 0.06	0.263 (11.5)	0.633 ± 0.06
29	0.232 (12.2)	0.977 ± 0.07	0.370 (9.5)	1.00 ± 0.07
43	0.453 (8.7)	1.43 ± 0.08	0.419 (8.9)	1.42 ± 0.09
71	0.302 (10.7)	1.73 ± 0.09	0.584 (7.5)	2.01 ± 0.09
100	0.471 (15.0)	2.20 ± 0.11	0.000 ± 0.08	2.01 ± 0.09
158	0.770 (16.0)	2.97 ± 0.17	1.03 (13.8)	3.03 ± 0.17
229	0.000 ± 0.01	2.97 ± 0.17	0.000 ± 0.01	3.03 ± 0.17

[a] Number in ( ) = 1σ percent counting uncertainty.

[b] For incremental fraction releases equal to zero, the error is based on the minimum detectable limit.

Table A.11

^{85}Sr Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (12% Boric Acid Solution and w/c Ratio of 0.5)

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
3	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
4	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
5	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
6	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
7	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
8	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.04
9	0.059 (91.7)	0.059 ± 0.06	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.04
10	0.000 ± 0.01	0.059 ± 0.07	0.062 (23.3)	0.062 ± 0.04	0.000 ± 0.01	0.000 ± 0.04
15	0.098 (18.0)	0.157 ± 0.07	0.156 (14.7)	0.218 ± 0.05	0.110 (17.3)	0.110 ± 0.05
20	0.000 ± 0.01	0.157 ± 0.07	0.208 (13.0)	0.425 ± 0.05	0.100 (18.0)	0.210 ± 0.05
29	0.186 (13.2)	0.342 ± 0.07	0.166 (14.4)	0.591 ± 0.06	0.320 (10.3)	0.530 ± 0.06
43	0.235 (11.7)	0.577 ± 0.08	0.301 (11.1)	0.892 ± 0.07	0.240 (11.7)	0.770 ± 0.07
71	0.381 (9.3)	0.958 ± 0.09	0.343 (10.3)	1.24 ± 0.08	0.390 (9.3)	0.116 ± 0.09
100	0.438 (14.4)	1.45 ± 0.11	0.000 ± 0.01	1.24 ± 0.08	0.184 (23.9)	1.34 ± 0.09
158	0.863 (14.9)	2.31 ± 0.17	0.000 ± 0.01	1.24 ± 0.08	0.438 (21.0)	1.78 ± 0.13
229	0.000 ± 0.03	2.31 ± 0.17	0.000 ± 0.01	1.24 ± 0.09	0.000 ± 0.04	1.78 ± 0.13

	Composite #4		Composite #5	
Time Days	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100	Incremental Fraction Released x 100[a,b]	Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.608 (29.2)	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.02
3	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.02
4	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.03
5	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.03
6	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.03
7	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.04
8	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.04
9	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.04
10	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.04
15	0.000 ± 0.01	0.608 ± 0.18	0.000 ± 0.01	0.000 ± 0.04
20	0.124 (16.7)	0.731 ± 0.18	0.134 (16.2)	0.134 ± 0.05
29	0.206 (13.0)	0.937 ± 0.18	0.144 (15.7)	0.277 ± 0.05
43	0.299 (11.1)	1.24 ± 0.19	0.103 (18.0)	0.380 ± 0.06
71	0.000 ± 0.01	1.24 ± 0.19	0.421 (9.1)	0.802 ± 0.07
100	0.440 (5.7)	1.68 ± 0.20	0.292 (19.0)	1.09 ± 0.09
158	0.985 (14.5)	2.66 ± 0.25	0.000 ± 0.01	1.09 ± 0.09
229	0.000 ± 0.04	2.66 ± 0.25	0.000 ± 0.04	1.09 ± 0.10

[a] Number in ( ) = 1σ percent counting uncertainty.
[b] For incremental fraction releases equal to zero, the error is based on the minimum detectable limit.

Table A.12

^{85}Sr Incremental and Cumulative Fractions Released From
Boric Acid/Portland III Cement Composites (12% Boric Acid Solution and w/c Ratio of 0.7)

Time Days	Composite #1 Incremental Fraction Released x 100[a,b]	Composite #1 Cumulative Fraction Released x 100	Composite #2 Incremental Fraction Released x 100[a,b]	Composite #2 Cumulative Fraction Released x 100	Composite #3 Incremental Fraction Released x 100[a,b]	Composite #3 Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
3	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
4	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
5	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
6	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
7	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
8	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
9	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.04
10	0.134 (16.2)	0.134 ± 0.04	0.064 (21.4)	0.064 ± 0.04	0.000 ± 0.01	0.000 ± 0.04
15	0.124 (16.7)	0.258 ± 0.05	0.000 ± 0.01	0.064 ± 0.04	0.142 (15.7)	0.142 ± 0.04
20	0.289 (11.1)	0.546 ± 0.06	0.164 (13.3)	0.227 ± 0.05	0.162 (14.4)	0.304 ± 0.05
29	0.206 (13.0)	0.752 ± 0.06	0.227 (11.6)	0.455 ± 0.05	0.253 (11.6)	0.556 ± 0.06
43	0.299 (11.1)	1.05 ± 0.07	0.218 (11.7)	0.673 ± 0.06	0.364 (9.74)	0.921 ± 0.07
71	0.412 (9.27)	1.46 ± 0.08	0.391 (8.86)	1.06 ± 0.07	0.455 (8.69)	1.38 ± 0.08
100	0.000 ± 0.01	1.46 ± 0.08	0.213 (20.9)	1.28 ± 0.08	0.435 (15.8)	1.81 ± 0.10
158	0.891 (15.9)	2.28 ± 0.15	0.000 ± 0.01	1.28 ± 0.08	0.000 ± 0.01	1.81 ± 0.11
229	0.000 ± 0.01	2.28 ± 0.16	0.000 ± 0.04	1.28 ± 0.09	0.000 ± 0.06	1.81 ± 0.11

Time Days	Composite #4 Incremental Fraction Released x 100	Composite #4 Cumulative Fraction Released x 100	Composite #5 Incremental Fraction Released x 100	Composite #5 Cumulative Fraction Released x 100
1	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01	0.000 ± 0.01
2	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.01
3	0.000 ± 0.01	0.000 ± 0.02	0.000 ± 0.01	0.000 ± 0.02
4	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.02
5	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.02
6	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.02
7	0.000 ± 0.01	0.000 ± 0.03	0.000 ± 0.01	0.000 ± 0.03
8	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.03
9	0.000 ± 0.01	0.000 ± 0.04	0.000 ± 0.01	0.000 ± 0.03
10	0.000 ± 0.01	0.000 ± 0.04	0.132 (14.4)	0.132 ± 0.04
15	0.136 (15.7)	0.136 ± 0.05	0.140 (14.1)	0.272 ± 0.04
20	0.300 (10.3)	0.436 ± 0.05	0.296 (9.74)	0.568 ± 0.05
29	0.291 (10.7)	0.726 ± 0.06	0.370 (8.69)	0.938 ± 0.06
43	0.262 (11.1)	0.988 ± 0.07	0.362 (8.88)	1.30 ± 0.07
71	0.087 (18.9)	1.08 ± 0.07	0.519 (5.75)	1.82 ± 0.07
100	0.000 ± 0.01	1.08 ± 0.07	0.291 (17.2)	2.11 ± 0.09
158	1.17 (12.7)	2.25 ± 0.17	0.522 (17.7)	2.63 ± 0.13
229	0.000 ± 0.04	2.25 ± 0.17	0.000 ± 0.03	2.63 ± 0.13

[a] Number in ( ) = 1σ percent counting uncertainty.

[b] For incremental fraction releases equal to zero, the error is based on the minimum detectable limit.

APPENDIX B

LEACH DATA FOR SECTION 4 OF THIS REPORT

Table 8.1

^{137}Cs Incremental and Cumulative Fractions Released From 1 x 1 Organic Cation Exchange Resin/Portland I Cement Composites

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	10.7 (1.1)	10.7 ± 0.1	4.00 (1.8)	4.0 ± 0.1	2.75 (2.1)	2.75 ± 0.06
1	27.8 (0.7)	38.5 ± 0.2	25.1 (0.7)	29.1 ± 0.2	11.9 (1.0)	14.7 ± 0.1
2	10.1 (1.1)	48.6 ± 0.2	11.9 (1.0)	41.0 ± 0.2	6.18 (1.4)	20.8 ± 0.2
3	6.19 (1.4)	54.8 ± 0.3	7.58 (1.3)	48.6 ± 0.2	4.43 (1.7)	25.3 ± 0.2
4	4.60 (1.7)	59.4 ± 0.3	5.71 (1.5)	54.3 ± 0.3	3.44 (1.9)	28.7 ± 0.2
5	3.53 (1.9)	63.0 ± 0.3	4.48 (1.7)	58.8 ± 0.3	2.69 (2.2)	31.4 ± 0.2
6	2.76 (2.1)	65.7 ± 0.3	3.69 (1.9)	62.5 ± 0.3	2.31 (2.3)	33.7 ± 0.2
7	2.30 (2.4)	68.0 ± 0.3	3.14 (2.0)	65.6 ± 0.3	1.87 (2.6)	35.6 ± 0.2
8	1.76 (2.7)	69.8 ± 0.3	2.58 (2.2)	68.2 ± 0.3	1.60 (2.9)	37.2 ± 0.2
9	1.66 (2.9)	71.4 ± 0.3	2.13 (2.5)	70.3 ± 0.3	1.53 (2.9)	38.7 ± 0.2
12	3.23 (2.0)	74.7 ± 0.3	4.60 (1.7)	74.9 ± 0.3	3.50 (1.9)	42.2 ± 0.2
13	0.957 (3.9)	75.6 ± 0.3	1.35 (3.2)	76.3 ± 0.3	1.10 (3.7)	43.3 ± 0.2
14	0.966 (3.9)	76.6 ± 0.3	1.28 (3.3)	77.6 ± 0.3	1.23 (3.4)	44.5 ± 0.2
15	0.780 (4.4)	77.4 ± 0.3	1.07 (3.6)	87.6 ± 0.3	1.05 (3.7)	45.6 ± 0.2
16	0.764 (4.5)	78.1 ± 0.3	1.04 (3.8)	79.7 ± 0.3	1.09 (3.6)	46.7 ± 0.2
19	1.76 (2.8)	79.7 ± 0.3	2.04 (2.6)	81.7 ± 0.3	2.32 (2.3)	49.0 ± 0.3
20	0.622 (3.6)	80.5 ± 0.3	0.697 (4.6)	82.4 ± 0.3	0.889 (3.9)	49.9 ± 0.3
21	0.596 (3.7)	81.1 ± 0.3	0.705 (4.7)	83.1 ± 0.3	0.836 (4.2)	50.7 ± 0.3
22	0.515 (4.1)	81.6 ± 0.3	0.587 (3.7)	83.7 ± 0.3	0.721 (4.6)	12.4 ± 0.3
23	0.474 (4.4)	82.1 ± 0.3	0.576 (3.8)	84.3 ± 0.3	0.781 (4.4)	52.2 ± 0.3
26	1.23 (3.4)	83.3 ± 0.3	1.44 (3.0)	85.7 ± 0.3	1.92 (2.6)	54.1 ± 0.3

Table B.1, Continued

^{137}Cs Incremental and Cumulative Fractions Released from 1 x 1 Organic Cation Exchange Resin/Portland I Cement Composites

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
27	0.457 (6.0)	83.8 ± 0.3	0.543 (3.9)	86.3 ± 0.3	0.733 (4.5)	54.9 ± 0.3
28	0.401 (5.0)	84.2 ± 0.3	0.485 (4.5)	86.7 ± 0.3	0.641 (3.5)	55.5 ± 0.3
29	0.437 (4.6)	84.6 ± 0.3	0.424 (4.7)	82.7 ± 0.3	0.661 (3.5)	56.2 ± 0.3
30	0.379 (4.9)	85.0 ± 0.3	0.360 (5.2)	87.5 ± 0.3	0.635 (3.5)	56.8 ± 0.3
33	1.05 (2.8)	86.0 ± 0.3	1.04 (1.6)	88.6 ± 0.3	1.66 (1.3)	58.5 ± 0.3
34	0.355 (5.9)	86.4 ± 0.3	0.337 (3.6)	88.9 ± 0.3	0.586 (2.5)	59.1 ± 0.3
35	0.335 (6.3)	86.7 ± 0.3	0.303 (3.6)	89.2 ± 0.3	0.530 (2.5)	59.6 ± 0.3
36	0.284 (3.9)	87.0 ± 0.3	0.286 (3.8)	89.5 ± 0.3	0.484 (2.8)	60.1 ± 0.3
37	0.229 (4.8)	87.2 ± 0.3	0.260 (4.2)	89.8 ± 0.3	0.467 (2.9)	60.5 ± 0.3
40	0.621 (2.4)	87.9 ± 0.3	0.644 (2.3)	90.4 ± 0.3	1.28 (1.1)	61.8 ± 0.3
41	0.278 (4.0)	88.1 ± 0.3	0.241 (4.6)	90.6 ± 0.3	0.521 (2.1)	62.4 ± 0.3
42	0.448 (2.4)	94.3 ± 0.3	0.438 (2.5)	96.8 ± 0.3	0.670 (2.0)	68.6 ± 0.3
49	1.30 (0.93)	95.6 ± 0.3	1.15 (1.0)	97.9 ± 0.3	2.53 (0.71)	71.1 ± 0.3
56	1.11 (1.5)	96.7 ± 0.3	0.889 (1.6)	98.8 ± 0.3	2.35 (1.0)	73.5 ± 0.3
63	1.02 (1.5)	97.7 ± 0.3	0.768 (1.7)	99.6 ± 0.3	2.36 (1.0)	75.8 ± 0.3
70	1.10 (1.5)	98.8 ± 0.3	0.883 (1.6)	100.4 ± 0.3	2.26 (1.0)	78.1 ± 0.3
77	0.876 (1.6)	99.7 ± 0.3	0.513 (2.1)	101.0 ± 0.3	1.83 (1.1)	79.9 ± 0.3
84	0.797 (1.8)	100.5 ± 0.3	0.759 (1.7)	101.7 ± 0.3	1.78 (1.2)	81.7 ± 0.3
112	1.62 (2.7)	102.1 ± 0.3	0.650 (4.3)	102.4 ± 0.3	4.74 (1.6)	86.4 ± 0.3
140	1.34 (1.0)	103.4 ± 0.3	0.51 (1.4)	102.9 ± 0.3	4.33 (5.3)	90.8 ± 0.3
169	0.76 (2.8)	104.2 ± 0.3	0.36 (4.0)	103.2 ± 0.3	3.22 (13.8)	94.0 ± 0.3

[a] Number in ( ) = 1σ percent counting uncertainty.

Table 8.2

^{137}Cs Incremental and Cumulative Fractions Released From
2 x 2 Organic Cation Exchange Resin/Portland I Cement Composites

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	2.32 (1.4)	2.32 ± 0.03	1.54 (1.7)	1.54 ± 0.03	1.16 (2.0)	1.16 ± 0.02
1	7.71 (0.8)	10.0 ± 0.1	5.70 (0.9)	7.24 ± 0.06	5.24 (0.9)	6.4 ± 0.05
2	3.31 (1.2)	13.4 ± 0.1	2.73 (0.4)	9.97 ± 0.06	3.13 (1.2)	9.5 ± 0.07
3	2.26 (1.4)	15.6 ± 0.1	2.04 (1.5)	12.0 ± 0.1	2.21 (1.4)	11.7 ± 0.1
4	7.76 (1.6)	17.4 ± 0.1	1.67 (1.7)	13.7 ± 0.1	1.83 (1.6)	13.6 ± 0.1
5	1.45 (1.8)	18.8 ± 0.1	1.40 (1.8)	15.1 ± 0.1	1.51 (1.7)	15.1 ± 0.1
6	1.23 (2.0)	20.1 ± 0.1	1.20 (2.0)	16.3 ± 0.1	1.30 (1.9)	16.4 ± 0.1
7	1.90 (2.1)	21.1 ± 0.1	1.09 (2.1)	17.4 ± 0.1	1.15 (2.0)	17.5 ± 0.1
8	0.917 (2.3)	22.1 ± 0.1	0.896 (2.3)	18.3 ± 0.1	1.[illegible]5 (2.3)	1.95 ± 0.1
9	0.858 (2.3)	22.9 ± 0.1	0.872 (2.3)	19.1 ± 0.1	8.85 (1.5)	21.6 ± 0.1
12	1.89 (1.6)	24.8 ± 0.1	1.94 (1.5)	21.0 ± 0.1	2.11 (1.5)	21.6 ± 0.1
13	0.729 (2.5)	25.5 ± 0.1	0.656 (2.7)	21.7 ± 0.1	0.718 (2.6)	22.3 ± 0.1
14	0.709 (2.5)	26.2 ± 0.1	0.665 (2.6)	22.4 ± 0.1	0.760 (2.5)	23.1 ± 0.1
15	0.597 (2.8)	26.8 ± 0.1	0.3[illegible]1 (2.7)	23.0 ± 0.1	0.659 (2.6)	23.7 ± 0.1
16	0.572 (2.8)	27.4 ± 0.1	0.572 (2.8)	23.6 ± 0.1	0.662 (2.6)	24.4 ± 0.1
19	0.135 (1.8)	28.8 ± 0.1	1.35 (1.8)	25.0 ± 0.1	1.50 (1.8)	25.9 ± 0.1
20	0.549 (2.9)	29.3 ± 0.1	0.526 (3.0)	25.5 ± 0.1	0.562 (2.9)	26.4 ± 0.1
21	0.513 (3.0)	29.8 ± 0.1	0.501 (2.9)	26.0 ± 0.1	0.559 (2.9)	27.0 ± 0.1
22	0.471 (3.1)	30.3 ± 0.1	0.486 (3.1)	26.4 ± 0.1	0.527 (3.0)	27.5 ± 0.1
23	0.501 (3.0)	30.8 ± 0.1	0.494 (3.1)	27.0 ± 0.1	0.497 (3.1)	28.0 ± 0.1
26	1.20 (2.0)	32.0 ± 0.1	1.20 (1.9)	28.2 ± 0.1	1.39 (1.8)	29.4 ± 0.1

Table B.2, Continued

^{137}Cs Incremental and Cumulative Fractions Released From
2 x 2 Organic Cation Exchange Resin/Portland I Cement Composites

Time Days	Composite #1 Incremental Fraction Released x 100	Composite #1 Cumulative Fraction Released x 100	Composite #2 Incremental Fraction Released x 100	Composite #2 Cumulative Fraction Released x 100	Composite #3 Incremental Fraction Released x 100	Composite #3 Cumulative Fraction Released x 100
27	0.480 (3.1)	32.5 ± 0.1	0.443 (3.2)	28.6 ± 0.1	0.529 (3.0)	29.9 ± 0.1
28	0.416 (3.3)	32.9 ± 0.1	0.432 (3.3)	29.0 ± 0.1	0.519 (3.0)	30.5 ± 0.1
29	0.425 (3.2)	33.3 ± 0.1	0.444 (3.2)	29.5 ± 0.1	0.514 (3.0)	31.0 ± 0.1
30	0.418 (3.3)	33.7 ± 0.1	0.402 (3.4)	29.9 ± 0.1	0.475 (3.1)	31.4 ± 0.1
33	1.12 (0.9)	34.8 ± 0.1	1.16 (0.9)	31.0 ± 0.1	1.34 (0.8)	32.8 ± 0.1
34	0.484 (1.4)	35.3 ± 0.1	0.485 (1.4)	31.5 ± 0.1	0.503 (1.4)	33.3 ± 0.1
35	0.428 (1.5)	35.8 ± 0.1	0.429 (1.5)	32.0 ± 0.1	0.325 (2.0)	33.6 ± 0.1
36	0.381 (1.5)	36.1 ± 0.1	0.411 (1.5)	32.4 ± 0.1	0.317 (2.0)	33.9 ± 0.1
37	0.390 (1.5)	36.5 ± 0.1	0.391 (1.5)	32.8 ± 0.1	0.460 (1.4)	34.4 ± 0.1
40	0.943 (1.0)	37.5 ± 0.1	0.919 (1.0)	33.7 ± 0.1	1.09 (0.9)	36.0 ± 0.1
41	0.436 (1.5)	37.9 ± 0.1	0.440 (1.5)	34.1 ± 0.1	0.479 (1.3)	36.0 ± 0.1
42	0.374 (1.6)	38.3 ± 0.1	0.489 (1.4)	34.6 ± 0.1	0.446 (1.4)	36.4 ± 0.1
49	0.336 (1.6)	38.6 ± 0.1	0.343 (1.6)	34.9 ± 0.1	0.410 (1.5)	36.8 ± 0.1
56	1.65 (0.75)	40.3 ± 0.1	1.61 (0.80)	36.7 ± 0.1	1.89 (0.71)	38.7 ± 0.1
63	1.31 (0.85)	41.6 ± 0.1	1.58 (0.76)	38.1 ± 0.1	1.79 (0.72)	40.5 ± 0.1
70	1.71 (0.73)	43.3 ± 0.1	1.56 (0.77)	39.7 ± 0.1	1.81 (0.71)	42.3 ± 0.1
77	1.31 (0.85)	44.6 ± 0.1	1.37 (0.80)	41.1 ± 0.1	1.49 (0.77)	43.8 ± 0.1
84	1.32 (0.84)	45.9 ± 0.1	1.37 (0.80)	42.5 ± 0.1	1.55 (0.77)	45.3 ± 0.1
112	3.58 (1.1)	49.5 ± 0.1	3.86 (1.1)	46.3 ± 0.1	4.33 (1.0)	49.7 ± 0.1
140	3.50 (0.37)	53.0 ± 0.1	3.75 (0.36)	50.1 ± 0.1	4.14 (0.33)	53.8 ± 0.1
169	2.92 (0.88)	55.9 ± 0.1	3.00 (0.88)	53.1 ± 0.1	3.26 (0.85)	57.1 ± 0.1

[a] Number in ( ) = 1σ percent counting uncertainty.

Table B.3

^{137}Cs Incremental and Cumulative Fractions Released From 2 x 4 Organic Cation Exchange Resin/Portland I Cement Composites

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	1.14 (2.5)[a]	1.14 ± 0.03	1.06 (2.7)	1.06 ± 0.03	0.999 (2.7)	1.00 ± 0.03
1	3.81 (1.4)	4.95 ± 0.06	4.15 (1.3)	5.21 ± 0.06	4.13 (1.3)	5.13 ± 0.06
2	2.40 (1.8)	7.35 ± 0.07	2.55 (1.7)	7.76 ± 0.08	2.46 (1.7)	7.59 ± 0.08
3	0.950 (4.0)	8.30 ± 0.08	2.02 (1.9)	9.78 ± 0.09	2.01 (1.9)	9.60 ± 0.08
4	1.60 (2.1)	9.90 ± 0.09	1.62 (2.1)	11.4 ± 0.1	1.59 (2.1)	11.2 ± 0.1
5	1.32 (2.4)	11.2 ± 0.1	1.43 (2.3)	12.8 ± 0.1	1.37 (2.3)	12.6 ± 0.1
6	1.16 (1.3)	12.4 ± 0.1	1.20 (2.5)	14.0 ± 0.1	1.21 (2.5)	13.8 ± 0.1
7	1.08 (2.6)	13.5 ± 0.1	1.06 (2.7)	15.1 ± 0.1	1.09 (2.6)	14.9 ± 0.1
8	0.988 (2.8)	14.4 ± 0.1	0.934 (2.9)	16.0 ± 0.1	0.901 (2.9)	15.8 ± 0.1
9	0.982 (2.7)	15.4 ± 0.1	0.905 (2.9)	16.9 ± 0.1	0.854 (3.0)	16.6 ± 0.1
12	2.00 (1.9)	17.4 ± 0.1	2.00 (1.9)	18.9 ± 0.1	1.99 (1.9)	18.6 ± 0.1
13	0.725 (3.2)	18.1 ± 0.1	0.765 (3.1)	19.7 ± 0.1	0.760 (3.1)	19.4 ± 0.1
14	0.771 (3.1)	18.9 ± 0.1	0.762 (3.1)	20.5 ± 0.1	0.749 (3.2)	20.1 ± 0.1
15	0.706 (3.3)	19.6 ± 0.1	0.690 (3.3)	21.2 ± 0.1	0.710 (3.2)	20.8 ± 0.1
16	0.695 (3.3)	20.3 ± 0.1	0.607 (3.4)	21.8 ± 0.1	0.656 (3.4)	21.5 ± 0.1
19	1.59 (2.1)	21.9 ± 0.1	1.58 (2.2)	23.3 ± 0.1	1.57 (2.2)	23.0 ± 0.1
20	0.626 (3.4)	22.5 ± 0.1	0.610 (3.5)	24.0 ± 0.1	0.570 (3.6)	23.6 ± 0.1
21	0.591 (3.5)	23.1 ± 0.1	0.594 (3.5)	24.5 ± 0.1	0.598 (3.5)	24.2 ± 0.1
22	0.581 (3.6)	23.7 ± 0.1	0.580 (3.6)	25.1 ± 0.1	0.566 (3.7)	24.8 ± 0.1
23	0.061 (3.6)	24.3 ± 0.1	0.538 (3.7)	25.7 ± 0.1	0.567 (3.7)	25.4 ± 0.1
26	1.45 (2.2)	25.8 ± 0.1	1.32 (2.4)	27.0 ± 0.1	1.23 (2.5)	26.6 ± 0.1

Table B.3, Continued

^{137}Cs Incremental and Cumulative Fractions Released from
2 x 4 Organic Cation Exchange Resin/Portland I Cement Composites

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
27	0.585 (3.5)	26.3 ± 0.1	0.553 (3.6)	27.5 ± 0.1	0.531 (3.8)	27.1 ± 0.1
28	0.568 (3.6)	26.9 ± 0.1	0.517 (3.9)	28.1 ± 0.1	0.510 (2.8)	27.8 ± 0.1
29	0.533 (3.8)	27.5 ± 0.1	0.562 (2.6)	28.6 ± 0.1	0.536 (3.7)	28.2 ± 0.1
30	0.516 (3.7)	28.0 ± 0.1	0.533 (2.6)	29.1 ± 0.1	0.474 (3.0)	28.6 ± 0.1
33	1.40 (1.0)	29.4 ± 0.1	1.35 (1.0)	30.5 ± 0.1	1.33 (1.1)	30.0 ± 0.1
34	0.561 (1.6)	29.9 ± 0.1	0.555 (1.6)	31.1 ± 0.1	0.559 (1.6)	30.5 ± 0.1
35	0.535 (1.7)	30.5 ± 0.1	0.518 (1.7)	31.6 ± 0.1	0.531 (1.7)	31.0 ± 0.1
36	0.530 (1.7)	31.0 ± 0.1	0.512 (1.7)	32.1 ± 0.1	0.513 (1.7)	31.6 ± 0.1
37	0.527 (1.7)	31.5 ± 0.1	0.457 (1.8)	32.5 ± 0.1	0.473 (1.7)	32.0 ± 0.1
40	1.22 (1.1)	32.7 ± 0.1	1.07 (1.2)	33.6 ± 0.1	1.10 (1.1)	33.1 ± 0.1
41	0.556 (1.6)	33.3 ± 0.1	0.525 (1.7)	34.1 ± 0.1	0.519 (1.7)	33.7 ± 0.1
42	0.518 (1.7)	33.8 ± 0.2	0.479 (1.7)	34.6 ± 0.2	0.470 (1.7)	34.1 ± 0.2
49	2.10 (1.9)	35.9 ± 0.2	2.00 (1.9)	36.6 ± 0.2	0.396 (1.9)	34.5 ± 0.2
56	1.99 (0.86)	37.9 ± 0.2	1.87 (0.87)	38.5 ± 0.2	1.90 (0.90)	36.4 ± 0.2
63	1.85 (8.8)	39.7 ± 0.2	1.75 (0.93)	40.2 ± 0.2	1.78 (0.91)	38.2 ± 0.2
70	1.86 (8.8)	41.6 ± 0.2	1.78 (0.91)	42.0 ± 0.2	1.83 (0.89)	40.1 ± 0.2
77	1.60 (0.97)	43.2 ± 0.2	1.52 (0.98)	43.5 ± 0.2	1.57 (0.99)	41.6 ± 0.2
84	1.59 (9.7)	44.8 ± 0.2	1.49 (0.99)	45.0 ± 0.2	1.57 (0.99)	43.2 ± 0.2
112	4.32 (1.3)	49.1 ± 0.2	4.04 (1.4)	49.1 ± 0.2	4.31 (1.3)	47.5 ± 0.2
140	3.87 (0.44)	53.0 ± 0.2	3.84 (0.44)	52.9 ± 0.2	4.10 (0.43)	51.6 ± 0.2
169	3.16 (1.1)	56.1 ± 0.2	3.18 (1.1)	56.1 ± 0.2	3.26 (1.1	54.9 ± 0.2

[a]Number in ( ) = 1_σ percent counting uncertainty.

Table B.4

^{137}Cs Incremental and Cumulative Fractions Released From
3 x 3 Organic Cation Exchange Resin/Portland I Cement Composites

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	1.30 (3.1)[a]	1.30 ± 0.04	0.914 (3.7)	0.91 ± 0.03	0.623 (4.5)	0.62 ± 0.03
1	2.77 (2.1)	4.07 ± 0.07	2.40 (2.3)	3.31 ± 0.07	2.65 (2.2)	3.27 ± 0.06
2	1.30 (3.1)	5.37 ± 0.08	1.23 (3.2)	4.54 ± 0.08	1.35 (3.1)	4.62 ± 0.08
3	0.997 (3.5)	6.37 ± 0.09	0.901 (3.8)	5.45 ± 0.08	1.01 (3.5)	5.63 ± 0.08
4	0.878 (2.7)	7.25 ± 0.09	0.780 (2.9)	6.23 ± 0.09	0.905 (5.3)	6.54 ± 0.10
5	0.812 (2.8)	8.06 ± 0.10	0.685 (3.1)	6.91 ± 0.09	0.800 (5.8)	7.34 ± 0.11
6	0.717 (3.0)	8.78 ± 0.10	0.665 (3.0)	7.57 ± 0.09	0.761 (5.8)	8.10 ± 0.12
7	0.658 (3.0)	9.44 ± 0.10	0.584 (3.2)	8.16 ± 0.09	0.709 (6.1)	8.81 ± 0.12
8	0.669 (3.0)	10.1 ± 0.1	0.582 (3.2)	8.74 ± 0.10	0.623 (6.5)	9.43 ± 0.13
9	0.629 (3.2)	10.7 ± 0.1	0.531 (3.6)	9.27 ± 0.10	0.670 (6.2)	10.1 ± 0.1
12	1.17 (3.2)	11.9 ± 0.1	1.10 (3.4)	10.4 ± 0.1	1.27 (3.2)	11.4 ± 0.1
13	0.543 (3.5)	12.4 ± 0.1	0.482 (5.7)	10.9 ± 0.1	0.563 (6.7)	11.9 ± 0.1
14	0.523 (3.4)	13.0 ± 0.1	0.452 (3.6)	11.3 ± 0.1	0.589 (6.6)	12.5 ± 0.2
15	0.493 (3.6)	13.5 ± 0.1	0.451 (3.6)	11.8 ± 0.1	0.526 (7.0)	13.0 ± 0.2
16	0.467 (3.8)	13.9 ± 0.1	0.444 (3.7)	12.3 ± 0.1	0.478 (7.4)	13.5 ± 0.2
19	0.896 (2.7)	14.8 ± 0.1	0.812 (2.8)	13.0 ± 0.1	1.17 (4.6)	14.7 ± 0.2
20	0.456 (2.5)	15.3 ± 0.1	0.423 (2.4)	13.4 ± 0.1	0.443 (12.0)	15.1 ± 0.2
21	0.470 (2.4)	15.7 ± 0.1	0.413 (2.4)	13.8 ± 0.1	0.443 (12.0)	15.6 ± 0.2
22	0.427 (2.4)	16.2 ± 0.1	0.406 (2.5)	14.3 ± 0.1	0.439 (12.1)	16.0 ± 0.2
23	0.425 (2.4)	16.6 ± 0.1	0.419 (2.4)	14.7 ± 0.1	0.457 (11.9)	16.5 ± 0.2
26	0.848 (1.8)	17.4 ± 0.1	0.769 (1.7)	15.4 ± 0.1	0.967 (8.1)	17.4 ± 0.2

Table B.4, Continued

^{137}Cs Incremental and Cumulative Fractions Released from
3 x 3 Organic Cation Exchange Resin/Portland I Cement Composites

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
27	0.422 (2.4)	17.9 ± 0.1	0.390 (2.6)	15.8 ± 0.1	0.428 (12.1)	17.9 ± 0.2
28	0.387 (2.6)	18.3 ± 0.1	0.390 (2.6)	16.2 ± 0.1	0.401 (12.6)	18.3 ± 0.2
29	0.424 (2.4)	18.7 ± 0.1	0.395 (2.6)	16.6 ± 0.1	0.434 (12.2)	18.7 ± 0.2
30	0.405 (2.5)	19.0 ± 0.1	0.375 (2.7)	17.0 ± 0.1	0.423 (12.2)	19.1 ± 0.2
33	0.639 (2.2)	19.7 ± 0.1	0.704 (2.0)	17.7 ± 0.1	0.791 (1.8)	19.9 ± 0.2
34	0.404 (2.2)	20.1 ± 0.1	0.413 (2.4)	18.1 ± 0.1	0.424 (2.4)	20.3 ± 0.2
35	0.409 (2.5)	20.5 ± 0.1	0.380 (2.7)	18.5 ± 0.1	0.395 (2.6)	20.7 ± 0.2
36	0.382 (2.6)	20.9 ± 0.1	0.379 (2.7)	18.9 ± 0.1	0.414 (2.4)	21.1 ± 0.2
37	0.396 (2.6)	21.3 ± 0.1	0.381 (2.7)	19.2 ± 0.1	0.377 (2.7)	21.5 ± 0.2
40	0.645 (2.2)	21.0 ± 0.1	0.722 (1.9)	20.0 ± 0.1	0.756 (1.8)	22.3 ± 0.2
41	0.410 (2.5)	22.4 ± 0.1	0.452 (2.2)	20.4 ± 0.1	0.451 (2.2)	22.7 ± 0.2
42	0.399 (2.5)	22.8 ± 0.1	0.374 (2.7)	20.8 ± 0.1	0.401 (2.5)	23.1 ± 0.2
49	0.232 (3.3)	23.0 ± 0.1	0.192 (3.9)	21.0 ± 0.1	0.236 (3.2)	23.4 ± 0.2
56	1.14 (1.4)	24.1 ± 0.1	1.08 (1.5)	22.1 ± 0.1	1.14 (1.4)	24.5 ± 0.2
63	1.09 (1.5)	25.2 ± 0.1	1.06 (1.6)	23.1 ± 0.1	1.12 (1.5)	25.6 ± 0.2
70	1.14 (1.4)	26.4 ± 0.1	1.12 (1.5)	24.2 ± 0.1	1.18 (1.5)	26.8 ± 0.2
77	1.01 (1.6)	27.4 ± 0.1	0.928 (1.6)	25.2 ± 0.1	0.976 (1.6)	27.8 ± 0.2
84	0.977 (1.6)	28.3 ± 0.1	0.948 (1.6)	26.1 ± 0.1	0.972 (1.6)	28.8 ± 0.2
112	2.69 (2.2)	31.0 ± 0.1	2.40 (2.3)	28.5 ± 0.1	2.64 (2.2)	31.4 ± 0.3
140.0	2.70 (0.71)	33.7 ± 0.1	2.53 (0.70)	31.1 ± 0.1	2.70 (0.70)	34.1 ± 0.3
169.0	2.26 (1.7)	36.0 ± 0.2	2.14 (1.7)	33.2 ± 0.1	2.26 (1.7)	36.4 ± 0.3

[a] Number in ( ) = 1 σ percent counting uncertainty.

Table B.5

^{137}Cs Incremental and Cumulative Fractions Released From 6 x 6 Organic Cation Exchange Resin/Portland I Cement Composites

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	0.580 (0.9)[a]	0.58 ± 0.01	0.489 (1.0)	0.49 ± 0.01	0.469 (1.0)	0.47 ± 0.01
1	2.71 (0.4)	3.29 ± 0.01	1.95 (0.5)	2.44 ± 0.01	1.55 (0.6)	2.02 ± 0.01
2	1.72 (5.2)	5.01 ± 0.02	1.19 (0.6)	3.63 ± 0.01	0.877 (0.7)	2.90 ± 0.01
3	1.35 (0.6)	6.36 ± 0.02	0.944 (0.7)	4.57 ± 0.01	0.708 (0.8)	3.60 ± 0.01
4	2.92 (0.4)	9.28 ± 0.02	1.94 (0.5)	6.51 ± 0.02	1.50 (0.6)	5.10 ± 0.02
7	0.818 (1.1)	10.1 ± 0.1	0.533 (1.3)	7.05 ± 0.02	0.431 (1.5)	5.54 ± 0.02
8	0.665 (1.2)	10.8 ± 0.1	0.452 (1.4)	7.50 ± 0.02	0.360 (1.6)	5.90 ± 0.02
9	0.729 (1.1)	11.5 ± 0.1	0.470 (1.4)	7.97 ± 0.02	0.407 (1.5)	6.30 ± 0.02
10	0.634 (1.2)	12.1 ± 0.1	0.410 (1.5)	8.38 ± 0.02	0.335 (1.7)	6.64 ± 0.02
13	1.63 (0.3)	13.9 ± 0.1	1.09 (0.4)	9.47 ± 0.02	0.945 (0.5)	7.58 ± 0.02
14	0.561 (0.6)	14.3 ± 0.1	0.369 (0.7)	9.84 ± 0.02	0.316 (0.8)	7.90 ± 0.02
15	0.514 (0.6)	14.8 ± 0.1	0.333 (0.7)	10.2 ± 0.1	0.286 (0.8)	8.18 ± 0.02
16	0.482 (0.6)	15.3 ± 0.1	0.322 (0.8)	10.5 ± 0.1	0.262 (0.8)	8.45 ± 0.02
17	0.432 (0.7)	15.8 ± 0.1	0.286 (0.8)	10.8 ± 0.1	0.251 (0.9)	8.70 ± 0.02
20	1.07 (0.4)	16.8 ± 0.1	0.741 (0.5)	11.5 ± 0.1	0.647 (0.5)	9.34 ± 0.02
21	0.381 (0.7)	17.2 ± 0.1	0.277 (0.8)	11.8 ± 0.1	0.243 (0.9)	9.58 ± 0.02
22	0.359 (0.74)	17.6 ± 0.1	0.263 (0.87)	12.1 ± 0.1	0.232 (0.90)	9.82 ± 0.1
29	1.85 (0.72)	19.4 ± 0.1	1.31 (0.85)	13.4 ± 0.1	1.20 (0.89)	11.0 ± 0.1
36	1.65 (0.34)	21.1 ± 0.1	1.19 (0.40)	14.6 ± 0.1	1.05 (0.43)	12.1 ± 0.1
43	1.47 (0.36)	22.5 ± 0.1	1.09 (0.42)	15.7 ± 0.1	0.963 (0.44)	13.0 ± 0.1
50	1.49 (0.36)	24.0 ± 0.1	1.11 (0.41)	16.8 ± 0.1	0.930 (0.45)	14.0 ± 0.1
57	1.31 (0.38)	25.3 ± 0.1	0.963 (0.44)	17.7 ± 0.1	0.792 (0.49)	14.8 ± 0.1
64	1.28 (0.39)	26.6 ± 0.1	0.952 (0.45)	18.7 ± 0.1	0.800 (0.49)	15.6 ± 0.1
92	3.40 (0.53)	30.0 ± 0.1	2.64 (0.60)	21.3 ± 0.1	2.32 (0.64)	17.9 ± 0.1
120	3.05 (0.18)	33.1 ± 0.04	2.44 (0.20)	23.8 ± 0.03	2.23 (0.21)	20.1 ± 0.03
149	2.57 (0.43)	35.6 ± 0.04	2.15 (0.47)	25.9 ± 0.03	1.88 (0.50)	22.0 ± 0.03

[a] Number in ( ) = 1 σ percent counting uncertainty.

Table B.6

^{137}Cs Incremental and Cumulative Fractions Released From 6 x 12 Organic Cation Exchange Resin/Portland I Cement Composites

Time Days	Composite #1		Composite #2		Composite #3	
	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	0.305 (1.7)[a]	0.305 ± 0.005	0.286 (1.8)	0.286 ± 0.005	0.279 (1.77)	0.279 ± 0.005
1	1.76 (0.71)	2.07 ± 0.01	1.96 (0.67)	2.25 ± 0.01	1.69 (0.72)	1.96 ± 0.01
2	1.18 (0.86)	3.24 ± 0.02	1.33 (0.82)	3.58 ± 0.02	1.07 (0.90)	3.04 ± 0.02
3	0.950 (0.97)	4.19 ± 0.02	1.05 (0.92)	4.63 ± 0.02	0.862 (1.0)	3.90 ± 0.02
4	0.779 (1.1)	4.97 ± 0.02	0.861 (1.0)	5.49 ± 0.02	0.704 (1.1)	4.60 ± 0.02
8	2.31 (0.62)	7.28 ± 0.03	2.43 (0.60)	7.92 ± 0.03	1.92 (0.68)	6.52 ± 0.02
9	0.517 (1.3)	7.79 ± 0.03	0.511 (1.3)	8.43 ± 0.03	0.387 (1.5)	6.91 ± 0.02
10	0.478 (1.4)	8.27 ± 0.03	0.464 (1.4)	8.90 ± 0.03	0.370 (1.5)	7.28 ± 0.03
11	0.362 (1.6)	8.63 ± 0.03	0.356 (1.6)	9.25 ± 0.03	0.290 (1.7)	7.57 ± 0.03
14	0.895 (1.0)	9.53 ± 0.03	0.917 (0.98)	10.2 ± 0.1	0.758 (1.1)	8.32 ± 0.03
15	0.285 (1.8)	9.81 ± 0.03	0.296 (1.7)	10.5 ± 0.1	0.252 (1.9)	8.58 ± 0.03
16	0.268 (1.8)	10.1 ± 0.1	0.273 (1.8)	10.7 ± 0.1	0.235 (2.0)	8.81 ± 0.03
17	0.281 (1.8)	10.4 ± 0.1	0.292 (1.7)	11.0 ± 0.1	0.239 (1.9)	9.05 ± 0.03
18	0.267 (1.8)	10.6 ± 0.1	0.286 (1.8)	11.3 ± 0.1	0.241 (1.9)	9.29 ± 0.03
21	0.759 (1.1)	11.4 ± 0.1	0.800 (1.0)	12.1 ± 0.1	0.670 (1.1)	10.0 ± 0.1
22	0.247 (1.9)	11.6 ± 0.1	0.265 (1.8)	12.4 ± 0.1	0.241 (1.9)	10.2 ± 0.1
23	0.245 (1.9)	11.9 ± 0.1	0.259 (1.8)	12.6 ± 0.1	0.238 (1.9)	10.4 ± 0.1
24	0.220 (2.0)	12.1 ± 0.1	0.248 (1.9)	12.9 ± 0.1	0.226 (2.0)	10.7 ± 0.1
25	0.202 (2.1)	12.3 ± 0.1	0.235 (1.9)	13.1 ± 0.1	0.215 (2.0)	10.9 ± 0.1
28	0.461 (1.4)	12.8 ± 0.1	0.585 (1.2)	13.7 ± 0.1	0.559 (1.2)	11.4 ± 0.1
29	0.145 (2.4)	12.9 ± 0.1	0.195 (2.1)	13.9 ± 0.1	0.192 (2.1)	11.6 ± 0.1

Table B.6, Continued

^{137}Cs Incremental and Cumulative Fractions Released From
6 x 12 Organic Cation Exchange Resin/Portland I Cement Composites

	Composite #1		Composite #2		Composite #3	
Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
30	0.158 (2.3)	13.1 ± 0.1	0.218 (2.0)	14.1 ± 0.1	0.223 (2.0)	11.9 ± 0.1
31	0.159 (2.4)	13.2 ± 0.1	0.215 (2.0)	14.3 ± 0.1	0.217 (2.0)	12.1 ± 0.1
32	0.138 (2.6)	13.4 ± 0.1	0.201 (2.1)	14.5 ± 0.1	0.208 (2.0)	12.3 ± 0.1
35	0.363 (1.6)	13.7 ± 0.1	0.539 (1.3)	15.1 ± 0.1	0.573 (1.2)	12.9 ± 0.1
36	0.142 (2.5)	13.9 ± 0.1	0.201 (2.1)	15.3 ± 0.1	0.227 (1.9)	13.1 ± 0.1
37	0.121 (2.7)	14.0 ± 0.1	0.182 (2.2)	15.5 ± 0.1	0.188 (2.2)	13.3 ± 0.1
38	0.126 (2.7)	14.1 ± 0.1	0.170 (2.3)	15.6 ± 0.1	0.193 (2.1)	13.5 ± 0.1
39	0.132 (2.5)	14.2 ± 0.1	0.173 (2.2)	15.8 ± 0.1	0.196 (2.1)	13.7 ± 0.1
42	0.317 (1.7)	14.6 ± 0.1	0.450 (1.4)	16.3 ± 0.1	0.508 (1.3)	14.2 ± 0.1
43	0.122 (2.7)	14.7 ± 0.1	0.170 (2.3)	16.4 ± 0.1	0.204 (2.1)	14.4 ± 0.1
44	0.115 (2.8)	14.8 ± 0.1	0.165 (2.3)	16.6 ± 0.1	0.192 (2.2)	14.6 ± 0.1
49	0.484 (1.3)	15.3 ± 0.1	0.691 (1.1)	17.3 ± 0.1	0.794 (1.1)	15.4 ± 0.1
50	0.116 (2.7)	15.4 ± 0.1	0.173 (2.2)	17.5 ± 0.1	0.214 (2.0)	15.6 ± 0.1
51	0.119 (2.7)	15.5 ± 0.1	0.162 (2.3)	17.6 ± 0.1	0.195 (2.1)	15.8 ± 0.1
52	0.135 (2.6)	15.7 ± 0.1	0.176 (2.3)	17.8 ± 0.1	0.222 (2.0)	16.0 ± 0.1
53	0.116 (2.8)	15.8 ± 0.1	0.151 (2.4)	17.9 ± 0.1	0.189 (2.2)	16.2 ± 0.1
56	0.281 (1.8)	16.0 ± 0.1	0.387 (1.5)	18.3 ± 0.1	0.495 (1.3)	16.7 ± 0.1
63	0.526 (1.3)	16.6 ± 0.04	0.693 (1.1)	19.0 ± 0.04	0.949 (1.0)	17.6 ± 0.04
70	0.501 (1.3)	17.1 ± 0.04	0.535 (1.3)	19.6 ± 0.04	0.843 (1.0)	18.5 ± 0.04
77	0.490 (1.4)	17.6 ± 0.04	0.484 (1.4)	20.1 ± 0.04	0.793 (1.1)	19.3 ± 0.04
84	0.474 (1.4)	18.1 ± 0.04	0.472 (1.4)	20.5 ± 0.04	0.764 (1.1)	20.0 ± 0.04
91	0.474 (1.4)	18.5 ± 0.04	0.472 (1.4)	21.0 ± 0.04	0.685 (1.1)	20.7 ± 0.07
98	0.546 (1.3)	19.1 ± 0.04	0.539 (1.3)	21.5 ± 0.04	0.613 (1.2)	21.3 ± 0.04
105	0.477 (1.4)	19.6 ± 0.04	0.498 (1.3)	22.0 ± 0.04	0.561 (1.3)	21.9 ± 0.04

[a] Number in ( ) = 1 σ percent counting uncertainty.

Table B.7

^{137}Cs Incremental and Cumulative Fractions Released From 12 x 12 Organic Cation Exchange Resin/Portland I Cement Composite #I

Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	0.117 (1.3)[a]	0.117 ± 0.002
1	0.825 (0.48)	0.943 ± 0.004
2	0.582 (0.58)	1.53 ± 0.005
5	1.89 (0.32)	3.42 ± 0.008
6	0.951 (0.45)	4.37 ± 0.009
7	0.346 (0.75)	4.71 ± 0.01
8	0.285 (0.82)	5.00 ± 0.01
9	0.254 (0.87)	5.25 ± 0.01
12	0.640 (0.55)	5.89 ± 0.01
13	0.226 (0.92)	6.12 ± 0.01
14	0.188 (1.0)	6.31 ± 0.01
15	0.173 (1.1)	6.48 ± 0.01
16	0.300 (0.80)	6.78 ± 0.01
19	0.361 (0.73)	7.14 ± 0.01
20	0.120 (1.3)	7.26 ± 0.01
21	0.124 (1.2)	7.38 ± 0.01
22	0.122 (1.3)	7.51 ± 0.01
23	0.123 (1.3)	7.63 ± 0.01
27	0.421 (0.68)	8.05 ± 0.01
28	0.115 (1.3)	8.16 ± 0.01
29	0.107 (1.3)	8.27 ± 0.01
30	0.109 (1.3)	8.38 ± 0.01
33	0.242 (0.90)	8.62 ± 0.01
34	0.088 (1.5)	8.71 ± 0.01
35	0.088 (1.5)	8.80 ± 0.01
36	0.090 (1.5)	8.89 ± 0.01
37	0.091 (1.5)	8.98 ± 0.01
40	0.220 (0.93)	9.20 ± 0.01
41	0.089 (1.5)	9.29 ± 0.01
42	0.076 (1.6)	9.37 ± 0.01
43	0.081 (1.6)	9.45 ± 0.01
44	0.087 (1.5)	9.53 ± 0.01
47	0.200 (0.98)	9.73 ± 0.01
54	0.428 (0.7)	10.2 ± 0.01
61	0.383 (0.7)	10.6 ± 0.01
68	0.384 (0.7)	10.9 ± 0.01
75	0.371 (0.7)	11.3 ± 0.01
82	0.369 (0.7)	11.7 ± 0.02
89	0.356 (0.7)	12.0 ± 0.02
96	0.405 (0.7)	12.4 ± 0.02
124	1.35 (0.4)	13.8 ± 0.02

[a] Number in ( ) = 1σ percent counting uncertainty.

Table B.7, Continued

^{137}Cs Incremental and Cumulative Fractions Released From 12 x 12 Organic Cation Exchange Resin/Portland I Cement Composites #2 and 3

Time Days	Composite #2 Incremental Fraction Released x 100	Composite #2 Cumulative Fraction Released x 100	Composite #3 Incremental Fraction Released x 100	Composite #3 Cumulative Fraction Released x 100
0.07	0.148 (1.3)[a]	0.148 ± 0.002	0.097 (1.9)	0.097 ± 0.002
1	1.00 (0.5)	1.16 ± 0.005	1.14 (0.5)	1.24 ± 0.006
2	0.654 (0.6)	1.81 ± 0.007	0.791 (0.6)	2.03 ± 0.007
3	0.507 (0.7)	2.32 ± 0.007	0.578 (0.7)	2.61 ± 0.008
4	0.438 (0.7)	2.75 ± 0.008	0.483 (0.7)	3.09 ± 0.009
7	0.964 (0.5)	3.72 ± 0.009	1.02 (0.5)	4.11 ± 0.01
8	0.322 (0.9)	4.04 ± 0.01	0.327 (0.9)	4.43 ± 0.01
9	0.272 (0.9)	4.31 ± 0.01	0.271 (1.0)	4.70 ± 0.01
10	0.219 (1.0)	4.53 ± 0.01	0.227 (1.1)	4.93 ± 0.01
11	0.198 (1.1)	4.73 ± 0.01	0.208 (1.1)	5.14 ± 0.01
14	0.491 (0.7)	5.22 ± 0.01	0.442 (0.8)	5.58 ± 0.01
15	0.183 (1.1)	5.40 ± 0.01	0.189 (1.2)	5.77 ± 0.01
16	0.165 (1.2)	5.57 ± 0.01	0.169 (1.2)	5.94 ± 0.01
17	0.159 (1.2)	5.73 ± 0.01	0.160 (1.3)	6.10 ± 0.01
18	0.151 (1.2)	5.88 ± 0.01	0.186 (1.2)	6.28 ± 0.01
21	0.373 (0.8)	6.25 ± 0.01	0.391 (0.8)	6.68 ± 0.01
22	0.152 (1.2)	6.40 ± 0.01	0.142 (1.4)	6.82 ± 0.01
23	0.134 (1.8)	6.54 ± 0.01	0.131 (1.4)	6.95 ± 0.01
24	0.110 (1.5)	6.65 ± 0.01	0.138 (1.4)	7.09 ± 0.01
25	0.134 (1.3)	6.78 ± 0.01	0.126 (1.4)	7.21 ± 0.01
28	0.362 (0.8)	7.14 ± 0.01	0.324 (0.9)	7.54 ± 0.01
29	0.115 (1.4)	7.26 ± 0.01	0.108 (1.6)	7.64 ± 0.01
30	0.108 (1.5)	7.36 ± 0.01	0.101 (1.6)	7.75 ± 0.01
31	0.108 (1.5)	7.47 ± 0.01	0.120 (1.5)	7.87 ± 0.01
32	0.102 (1.5)	7.57 ± 0.01	0.104 (1.6)	7.97 ± 0.01
35	0.282 (0.9)	7.86 ± 0.01	0.284 (1.0)	8.25 ± 0.01
36	0.093 (1.6)	7.95 ± 0.01	0.097 (1.7)	8.35 ± 0.01
37	0.94 (1.6)	8.04 ± 0.01	0.092 (1.7)	8.44 ± 0.01
38	0.094 (1.6)	8.14 ± 0.01	0.093 (1.7)	8.54 ± 0.02
39	0.098 (1.6)	8.24 ± 0.01	0.093 (1.7)	8.63 ± 0.02
42	0.280 (0.9)	8.52 ± 0.01	0.313 (0.9)	8.94 ± 0.02

[a] Number in ( ) = 1σ percent counting uncertainty.

Table B.8

^{137}Cs Incremental and Cumulative Fractions Released From 22 x 22 Organic Cation Exchange Resin/Portland I Cement

Time Days	Incremental Fraction Released x 100	Cumulative Fraction Released x 100
0.07	0.05 (2.8)[a]	0.05 ± 0.001
1	0.44 (0.9)	0.48 ± 0.004
2	0.31 (1.1)	0.80 ± 0.01
3	0.26 (1.2)	1.06 ± 0.01
4	0.22 (1.3)	1.28 ± 0.01
7	0.49 (0.9)	1.77 ± 0.01
8	0.16 (1.5)	1.93 ± 0.01
9	0.14 (1.7)	2.07 ± 0.01
10	0.11 (1.9)	2.18 ± 0.01
11	0.11 (1.9)	2.23 ± 0.01
14	0.31 (1.1)	2.60 ± 0.01
15	0.10 (2.0)	2.70 ± 0.01
16	0.08 (2.2)	2.78 ± 0.01
17	0.08 (2.2)	2.86 ± 0.01
18	0.08 (2.2)	2.94 ± 0.01
21	0.20 (1.4)	3.13 ± 0.01
22	0.07 (2.4)	3.20 ± 0.01
23	0.06 (2.5)	3.27 ± 0.01
24	0.07 (2.4)	3.33 ± 0.01
25	0.07 (2.4)	3.40 ± 0.01
28	0.18 (1.5)	3.58 ± 0.01

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[a] Number in ( ) = 1σ percent counting uncertainty.

APPENDIX C

ANALYSIS OF IRRADIATED ION EXCHANGE MATERIALS

ANALYSIS OF IRRADIATED ION EXCHANGE MATERIALS

by

R. C. McFarland
Neely Nuclear Research Center
School of Nuclear Engineering
Georgia Institute of Technology
Atlanta, Georgia 30332

FINAL RESEARCH REPORT
Project A60-611
Contract S26325-S
Brookhaven National Laboratory
Associated Universities, Inc.
Upton, NY 11973

TABLE OF CONTENTS

1. STATEMENT OF THE PROBLEM

Solid adsorbents and ion exchange materials, used in the decontamination of high level liquid radioactive waste, will receive large radiation doses from the radioactive material which they remove from the waste stream. Radiation damage to the adsorbent material is responsible for the observed effects of loss of exchange capacity and chemical decomposition of the base material. Since the decontamination process is relatively rapid, the loss of exchange capacity due to radiation damage is a minor consideration in the actual decontamination of liquid rad waste. On the other hand, radiolytic decomposition of the base material and loss of exchange capacity can be major concerns when the highly radioactive adsorbent materials are buried for long periods of time in sealed containers. The possibility that radiation decomposition of adsorbent materials could produce gases in sufficient quantity to cause over pressurization of burial containers led to this investigation.

This has been a two phase project. Phase one funded by Chem-Nuclear Systems, Incorporated had as its objectives to determine the pressure buildup and gas composition as a function of gamma dose in burial canisters of the type being considered for use at Three Mile Island. Funded by Brookhaven National Laboratory, the second phase was to determine the physical and chemical properties of the radiolytic products and to determine the effects of these products on the stainless steel containers. The second phase was not begun until one year after the samples were irradiated.

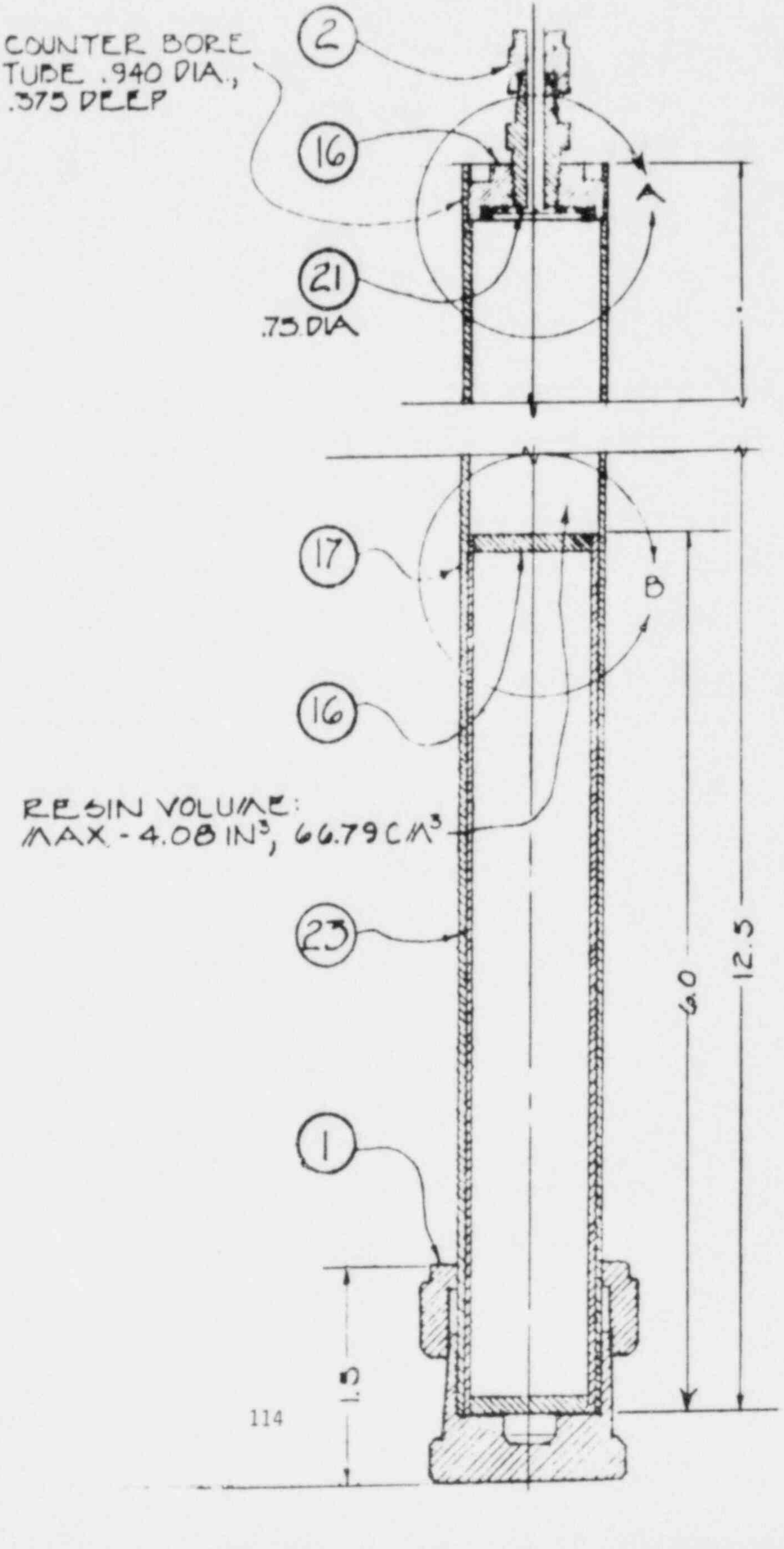
The initial part of the first phase was to measure the pressure versus gamma radiation dose, in separate simulated burial containers holding organic cation resin, organic anion resin, and activated charcoal. In order to design this experiment it was first necessary to estimate the radiation dose to an adsorbent material loaded with TMI high level radioactive waste and sealed in a burial container. The calculation of the estimated radiation dose is given in Appendix B. Once the magnitude of the total dose was determined, the gamma irradiator design was finalized. The details of the Co-60 irradiator and irradiation capsules are given in section 2. Finally, the simulated burial containers were irradiated in the Co-60 gamma irradiator at 5×10^6 rad/hr. field until the total accumulated dose reached 5×10^9 rads or the capsule reached the pressure limit of 200 psig. Pressure versus gamma dose curves are given in seciton 3.1. The remainder of the first phase was to determine H_2 and hydrocarbon content inside the resin irradiation capsules at several different dose levels. The gas composition was determined using gas chromatography and the methods and results are discussed in Section 4.3.

Phase two of this project commenced with opening of the irradiated resin containers from phase one and the determination of the physical properties of the irradiated material. The physical properties of the irradiated resin materials are given in section 3. Chemical analyses were performed on the liquid, solid and gas phases taken from the capsules and the results are reported in section 4. The corrosion effects were evaluated and the results reported in appendix A.

2. IRRADIATION CAPSULE CONSTRUCTION AND IRRADIATOR DESIGN

In order to meet the time schedule of this project, it was decided that the pressure test and gas chromatograph test capsules should be constructed from commercially available fittings to the maximum extent possible. The construction material chosen was stainless steel in order to match the proposed burial containers as closely as possible. While stainless steel might influence the chemical reactions and be undesirable from a purely scientific point of view, it was a good compromise considering radiation resistance, pressure limitations, chemical reactivity, and the desire to simulate the actual burial containers. In addition, it was required that no organic materials, other than those being tested, be used in any of the units. At the high dose levels involved, organic O rings and bellows could break down and contaminate the gases or release the gaseous products. All stainless steel valves and pressure gauges were used in the construction of these containers.

Figure 1 shows the pressure test capsules. Specifications for the fittings and tubing are given in figure 5. The pressure test capsules required an internal stainless steel spacer plug (item 23) to raise the material into the radiation field and to provide the correct material to void ratio. From data supplied by Chem-Nuclear Systems it was estimated that the burial containers would have a resin to toal volume ratio of 0.875. The pressure test capsule shown in figure 1 has a resin to total volume ratio of 0.866. In order to get this ratio as close to the actual ratio as possible, the pressure gauges had to have a small internal volume and had to be connected to the capsule with a minimum length of 1/8" stainless steel tubing. To keep the tubing connections short, the gauges were located on a rack on the back wall of the hot cell about 5 feet from the capsules. The gauges were outside the most intense radiation



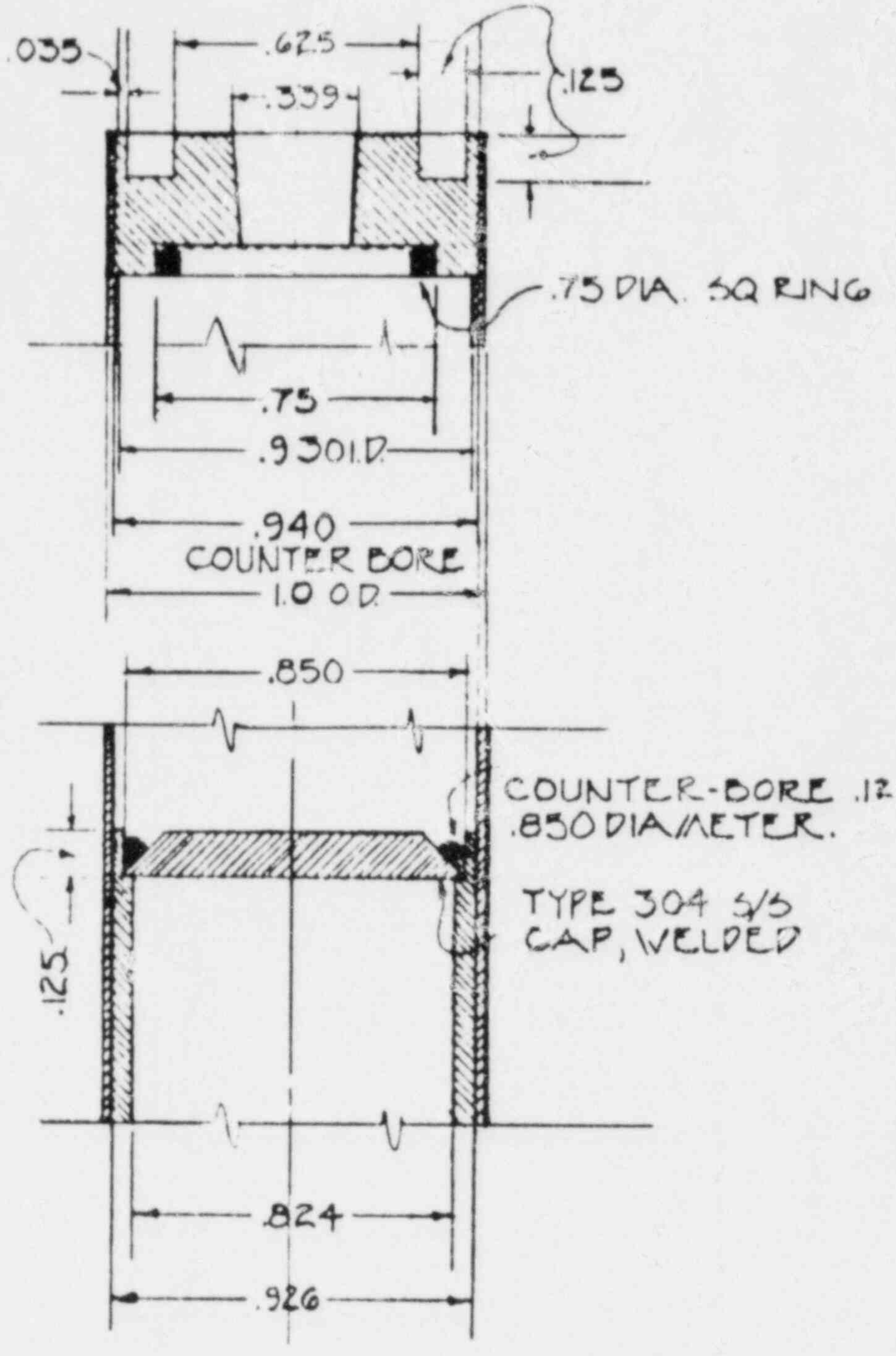
SECTION THRU RESIN PRESSURE CAPSULE

FIGURE 1

FIGURE 2

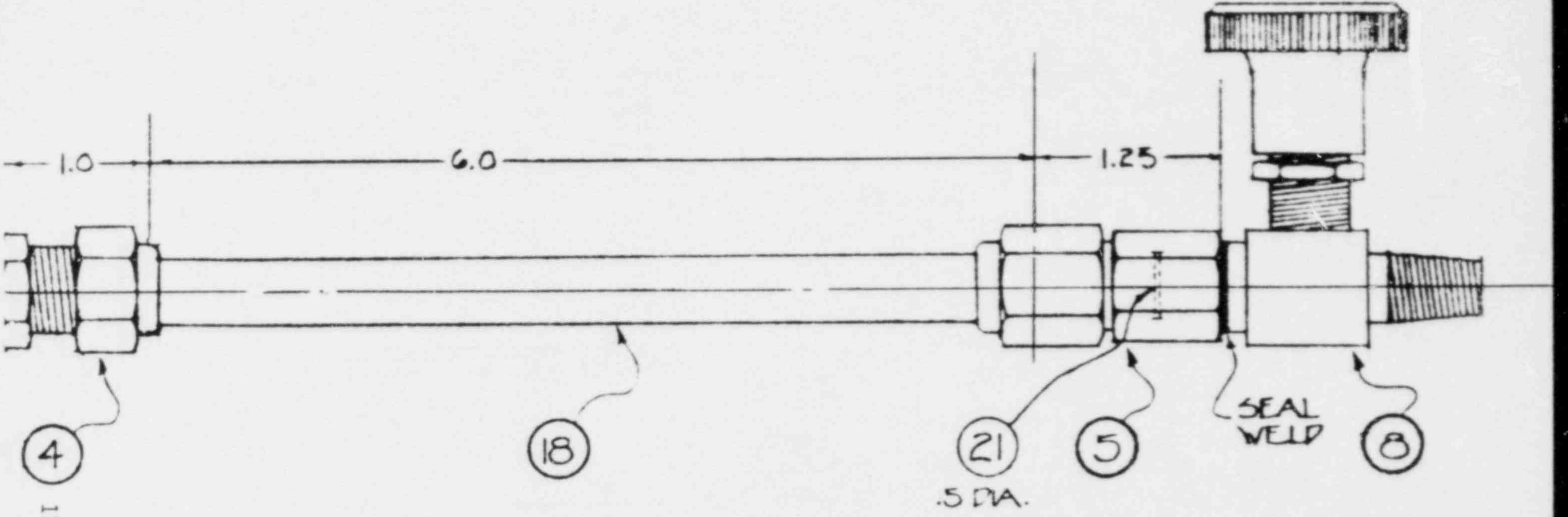
DETAIL-A

SCALE: x2



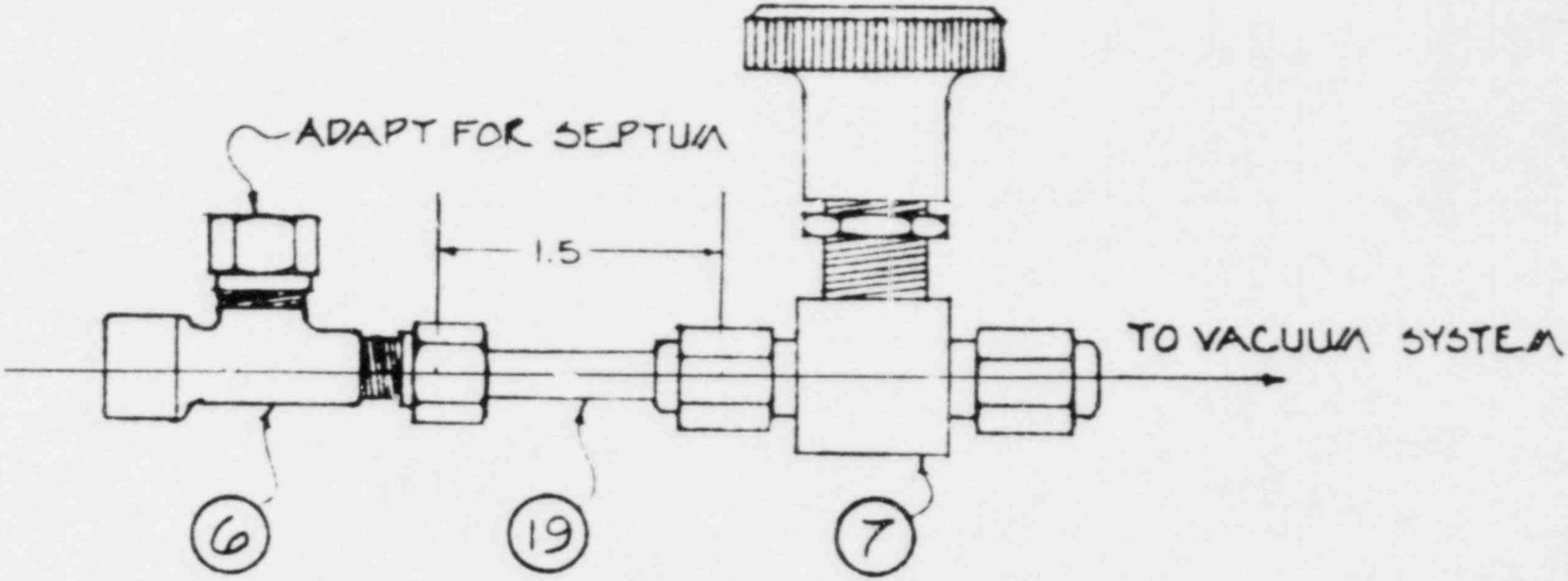
DETAIL-B

SCALE: x2



GAS CHROMATOGRAPH CAPSULE

FIGURE 4

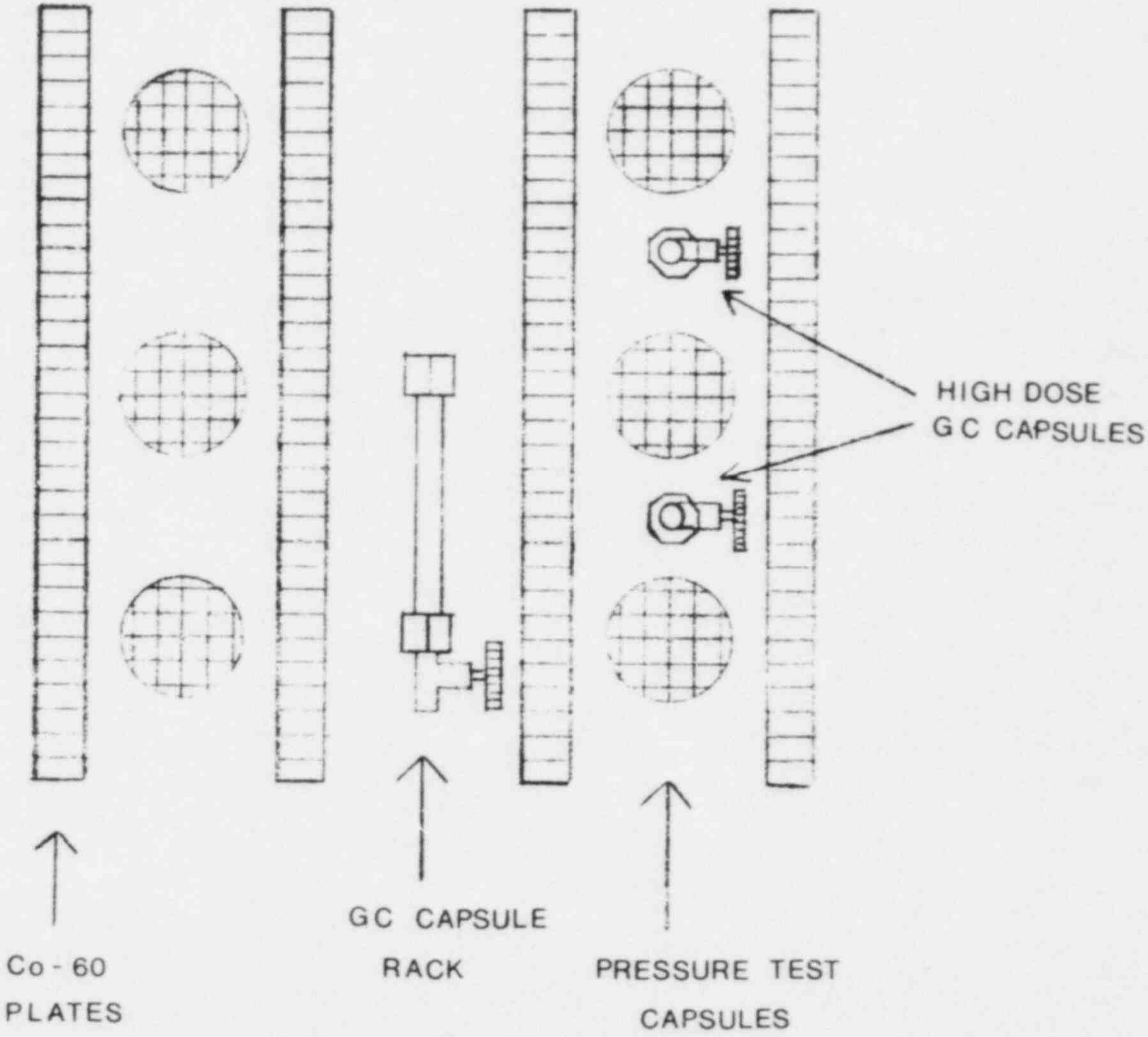


GAS CHROMATOGRAPH
SAMPLE ASSEMBLY

FIGURE 5

ITEM	QUAN.	CAT PAGE	DESCRIPTION	SWAGELOK CODE
1	8 ea.	20	CAP-1"	SS-1610-C
2	8	7	MALE CONNECTOR 1/8-1/4	SS-200-1-2
3	8	10	FEMALE CONNECTOR 1/8-1/4	SS-200-7-4
4	10	20	CAP-1/2"	SS-810-C
5	10	10	FEMALE CONNECTOR 1/2T-1/4P	SS-810-7-4
6	10	12	FEMALE RUN TEE 1/4"	SS-400-3-4TFT
7	9	NUPRO 7	H SERIES BELOWS VALVE	SS-4H
8	9	NUPRO 7	H SERIES BELOWS VALVE	SS-4H2
16	4 FT.		1"D. ROUND S/S ROD	
17	9 FT.		1" O.D. S/S TUBE, 0.035 WALL SMLS	
18	6 FT.		1/2" O.D. S/S TUBE, 0.035 WALL SMLS	
19	2 FT.		1/4" O.D. S/S TUBE, 0.035 WALL SMLS	
20	40 FT.		1/8" O.D. S/S TUBE, 0.032 WALL SMLS	
21	6 ea.		2" SQUARE, 10 MICRON SCREEN S/S	
22	7 ea.		2 1/2" D. PRESS. GAGE 0-200 PSI, S/S	
23	4 FT.		3/4" S/S PIPE, SCH. 40, TYP. 304	

FIGURE 6
Co-60 IRRADIATOR



field but the glass faces still had to be removed because of radiation darkening. The gauges were Ashcroft 0-200 psi gauges which were calibrated using a dead weight gauge tester. At the inside top of the pressure test capsule there was a stainless steel screen (10 micron openings) to prevent particles from entering the tubing leading to the gauge. Figure 2 shows the details of the top of the pressure capsule and the spacer plug.

To determine the gas composition versus dose, the gas chromatograph capsules shown in figure 3 were constructed. As with the pressure test capsules, these were also all stainless steel construction. The resin was weighed into these capsules which had an internal volume of 18.9 cc. The void volume was calculated assuming that the wet resin had a density of 1.1 gm/cc. After irradiation, the samples were removed for analysis using the gas chromatograph sample assembly shown in figure 4. The sample assemble was screwed onto the gas chomatograph capsule and evacuated with the capsule valve closed. After evacuation the sample assembly valve was closed and the capsule valve was opened. Samples could then be taken from the septum side arm for analysis by gas chromatography.

To simulate the decontamination of TMI liquid radwaste, samples of cation resin (DOW HCR-S), anion resin (DOW SBR-OH) and activated charcoal supplied by Chem-Nuclear Systems, were converted to the sodium and borate forms and loaded into the irradiation capsules described above. Sodium borate solution was passed through the resin samples until the pH of the effluent from the column was identical to the pH of the original solution. In the case of the activated charcoal, no simple indication of exhaustion could be found. To pretreat the activated charcoal, an amount of sodium borate solution, equal to the amount needed to convert the anion resin to the borate form, was passed through the charcoal sample. After pretreatment with sodium borate solution free liquid was removed by pulling air through the column for about 2 minutes. The adsorbent material was then transferred to the pressure test and GC irradiation capsules using a tap fill procedure. Adsorbent loading in the irradiation capsules matched the anticipated loading of the proposed TMI clean-up canisters to within 10%.

The Co-60 irradiator shown in figure 6 was assembled to irradiate the test capsules in the Georgia Tech hot cell. The Co-60 was in the form of four plates each containing 6.25 K Ci of Co-60 arranged as shown in figure 6. The outside spaces held the pressure test capsules and the long term gas chromatograph capsules. Thermocouples were attached to the pressure capsules, the long term GC capsules and several other places on the irradiator. Compressed air was piped into the hot cell and used for cooling the capsules. Before starting irradiation, dose rates were measured at several positions inside the irradiation assemble. The dose rates were measured using Harshaw TL-800 lithium borate thermoluminescent dosimeters. These thermoluminiscent dosimeters were calibrated against a Farmer dosimeter, model 2502/3, which had been calibrated using NBS Co-60 at M. D. Anderson Hospital, Houston, Texas. In the outside space the dose rates were: at the top 3" above center, 4.46 X 10^6 rads/hr.; center, 5.87 X 10^6 rads/hr. and 3" above bottom, 4.86 X 10^6 rads/hr. This gives an average of 5.0 X 10^6 rads/hr. The center space for the G.C. capsules had dose rates of 4.3 X 10^6 rads/hr. at the top; 5.4 X 10^6 rads/hr. at the center, and 4.60 rads/hr. at the bottom. The average dose rate in the GC sample was 4.8 X 10^6 rads/hr.

3.1 CAPSULE PRESSURE AS A FUNCTION OF GAMMA DOSE

The filled capsules of cation resin (DOW HCR-S), anion resin (DOW SBR-OH) and activated charcoal supplied by Chem-Nuclear Systems were placed in the Co-60 irradiator as shown in figure 6 and irradiated at 5 X 10^6 rads/hr., in the Georgia Tech hot cell. The capsule pressure was monitored visually using a monocular sighting through the hot cell window in order not to disturb the irradiation. The capsule temperature was determined using thermocouples with the reader located outside the hot cell. The temperature stabilized within a few hours of the beginning of the irradiation and remained between 30 C and 45 C throughout the test. The pressure versus gamma ray dose curves for the anion resin, cation resin, and activated charcoal are presented in figures 7, 8, and 9, respectively. The pressure tests on the resin samples were terminated when the pressure reached the limit of the pressure gauge which was 200 psig. Duplicate pressure test capsules containing each of the adsorbent materials were prepared and irradiated. One of the anion pressure test capsules developed an interval leak in the spacer plug during the irradiation and the results from that capsule were not reported. The close agreement between the duplicate cation resin tests indicate the reproducibility of the tests.

3.2 PHYSICAL PROPERTIES OF IRRADIATED MATERIAL

Eight GC capsules from the first phase of this project remained undisturbed after one year and were available for phase two of this project. These capsules were opened and the physical properties of the material inside were noted. Unlike the pressure test capsules discussed later these capsules were not expected to be under pressure due to the fact that they had been contained only by a punctured rubber septum for this long period of time. Actually two of the capsules were still under pressure, cation 5 X 10^9 rads and anion

FIGURE 7
ANION RESIN

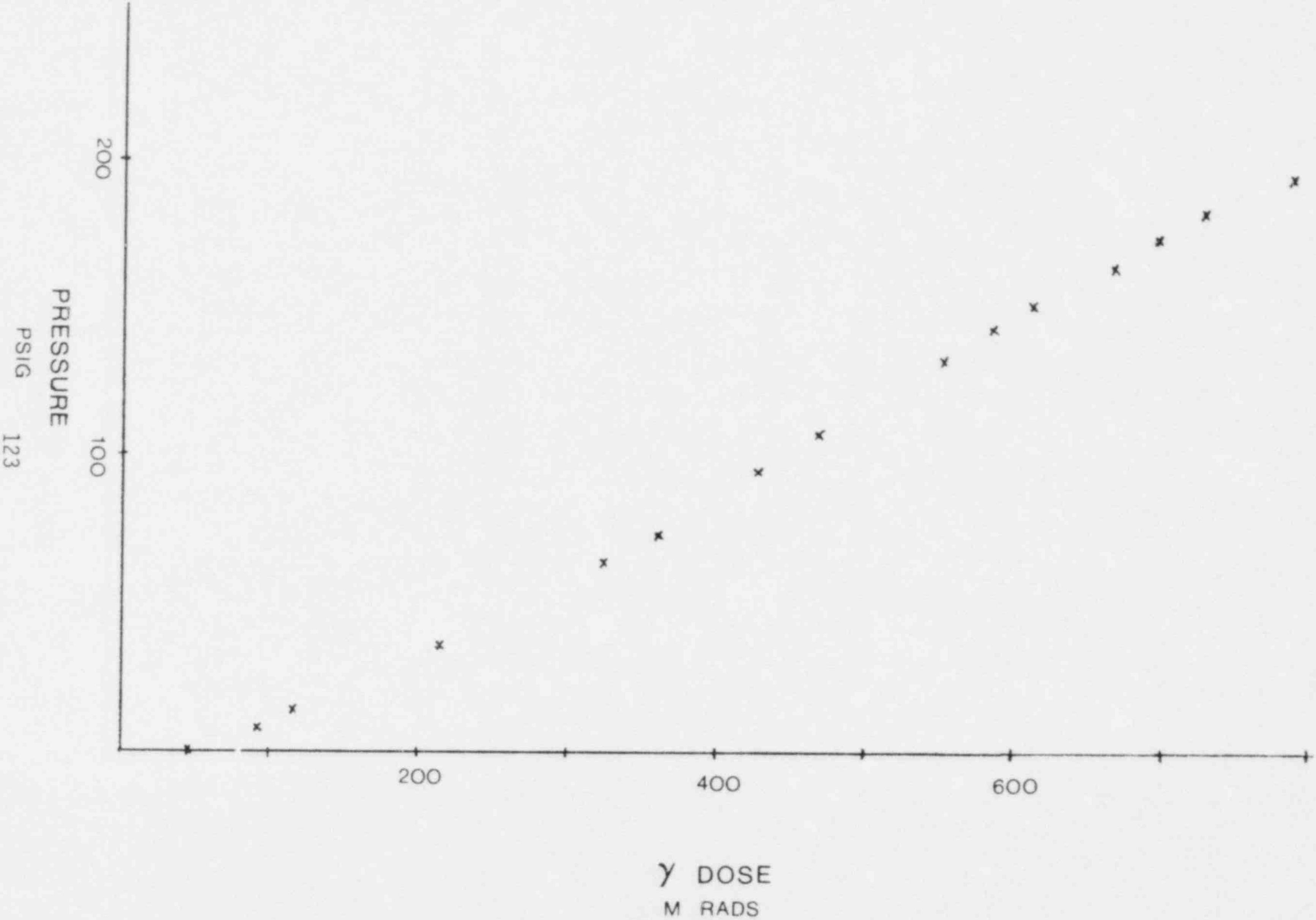


FIGURE 8
CATION RESIN

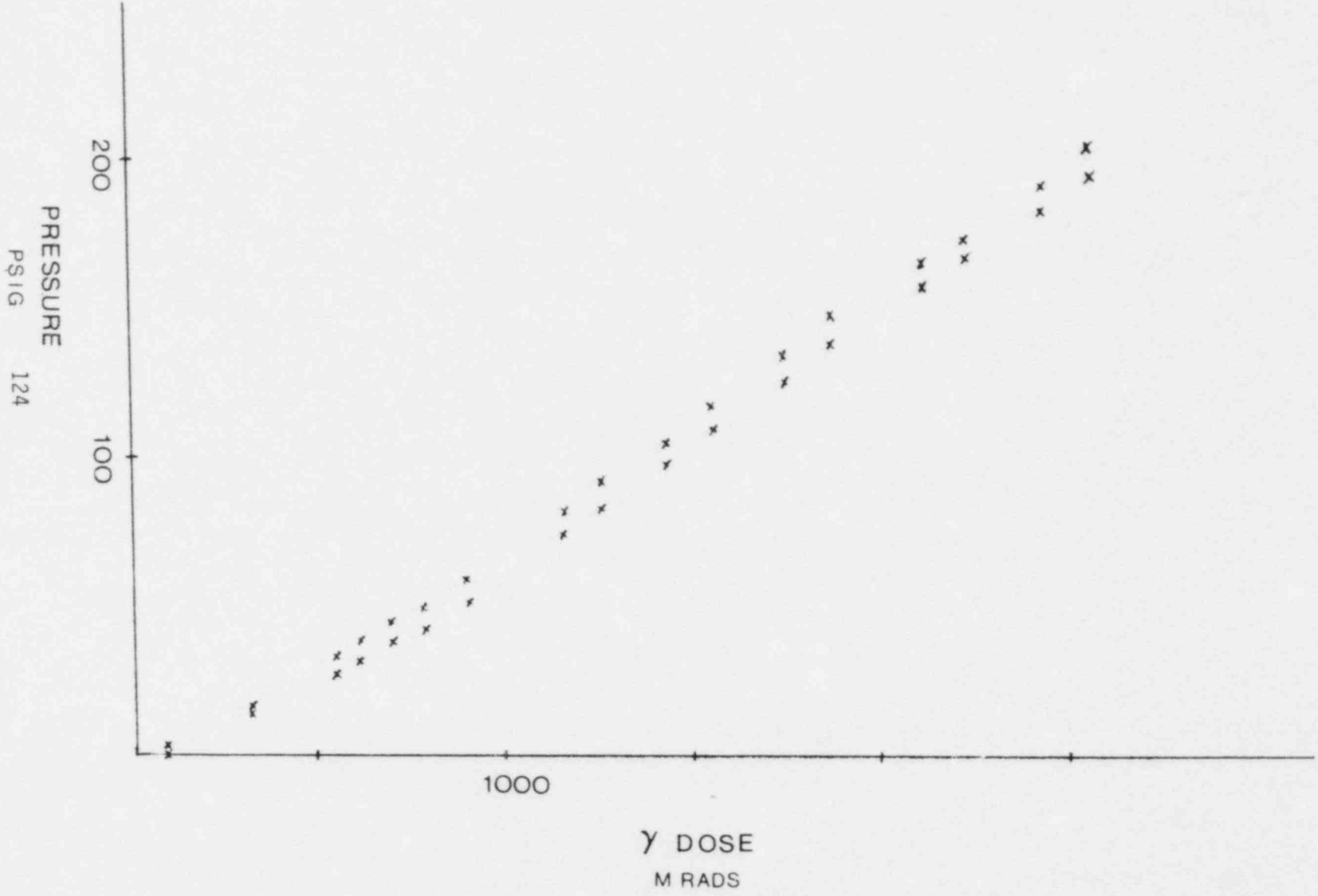
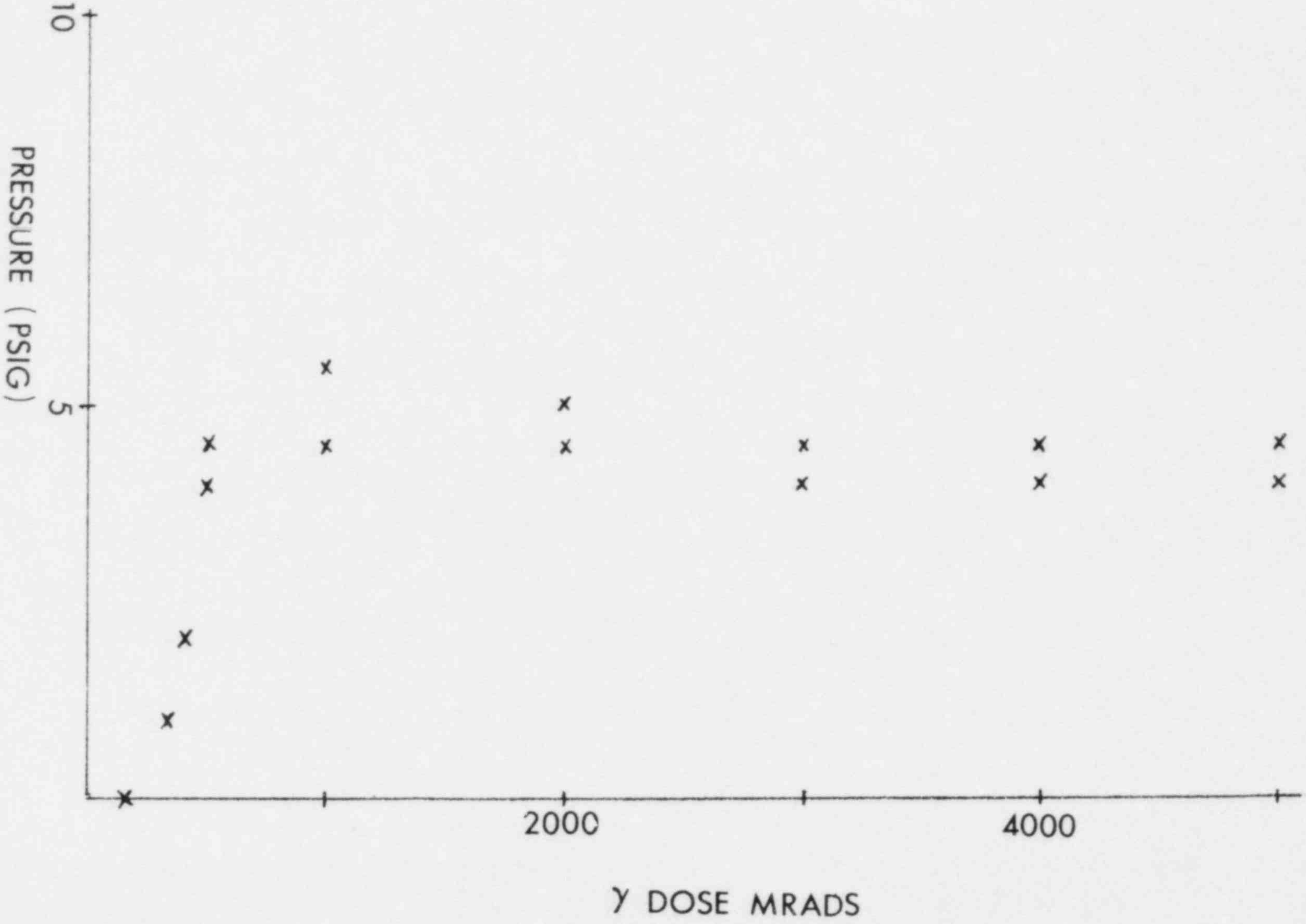


FIGURE 9
CHARCOAL



10^9 rads. After removal, samples of the solid material were soaked in water and the properties of the remaining solids were recorded. This water soak test was intended to simulate the opening of TMI resin canisters and the use of water to transfer the resin to additional radioactive waste treatment facilities.

Anion resin irradiated to 10^8 rads has very little liquid present and a strong smell of amines. When viewed under a microscope at 100X some large fractured pieces were seen and some of the spherical beads appear to be shedding layers of material. Most of the beads were transparent and slightly brown. Among the remaining spherical beads no drastic changes from unirradiated resin in the size distribution or density were apparent. The water-soaked beads showed no changes.

Anion resin irradiated to 10^9 rads results in a flowing slurry of liquid and resin. Fractured pieces of beads were visible under a microscope and some of the beads had rough scaling surfaces. No changes in the size distribution or density of the remaining spherical beads were apparent in either the slurry or the water soaked sample. The liquid was watery and smelled more of ammonia than amines.

Cation resin irradiated to 10^8 rads showed very few changes. The beads were slightly darker than unirradiated beads. No additional liquid was found in these samples and no noticeable odor was detected. No fractured pieces of resin beads could be detected. When the irradiated beads were soaked in water no changes were noticeable. In the dry state the beads showed a tendency to cling together or to cling to any surface close by.

At 3×10^8 to 5×10^8 rads the cation resin beads darken considerable but are not completely black. After water is added a floculent red-brown precipitate becomes apparent. From microscopic examinations it appears that this fine precipitate forms from larger pieces of material broken down by the added water. This precipitate is very fine and remains in suspension for several hours after the mixture is shaken. Attempts to filter this precipitate after shaking were unsuccessful, since the fine material clogged the filter. When the material had settled down for several hours it could be filtered through a 0.2 micron filter with more very fine brown material remaining on the filter. Again the remaining spherical beads have the same approximate size distribution and density as the unirradiated resin and water soaking showed no changes in the beads themselves.

When the exposure is increased to 10^9 rads and 5×10^9 rads, cation resin shows no large additional changes in physical appearance. The red-brown, fine precipitate is present. No additional liquid is found. More fractured pieces of beads can be seen. The beads are all dark black and tend to cling together or to any surface close at hand. The only effect of water soaking appears to be the production of the floculent precipitate.

Four pressure test capsules remaining from the first phase of the project were opened. Commercially available devices for sampling aerosol cans were tested for application to the sampling of these pressure capsules. The pressure capsules described in section 2 were not originally designed to be sampled since phase two was a much later addition to this project. It was found that the stainless steel pressure capsules could not be penetrated in a manner that would allow retention of the gas using any conceivable type of puncturing device. Measurement of the gas volume and sampling of the radiolytic gases was accomplished using a one liter gas sampling bag sealed around the connection between the 1/8" tubing and the gauge.

The air was removed from the bag by an aspirator pump through a hypodermic needle inserted into the septum port on the sampling bag. After the bag was evacuated a known quantity of Xe-133 in 0.10 ml of air was injected into the bag to measure the expanded volume of radiolytic gas by isotope dilution. Following the addition of Xe-133 the connection between the 1/8" tubing and the gauge was broken and the radiolytic gases were allowed to escape into the gas sampling bag. The flexible sampling bag was sufficiently large to allow the radiolytic gases to expand until the pressure inside the bag was equal to the atmospheric pressure outside. The gases were mixed with the Xe-133 by kneading the bag for several minutes. A 0.8 ml sample was removed from the sampling bag and counted for Xe-133 in a Ge(Li) gamma spectrometer system. The volume of radiolytic gas in the sampling bag was calculated by comparing the concentration of Xe-133 in the 0.8 ml sample to the original amount injected.

The gas volume calculated for anion resin 7.9 X 10^8 rads was 680 cc at 20°C and 410 cc for cation resin 2.6 X 10^9 rads. From these gas volume measurements it is obvious that the effect of neglecting interstitial void volume between beads and the gas solubility in the liquid-resin mixture introduced a serious error in phase one gas composition measurements on these samples with high resin loading. In the pressure test capsules the bed volume was 67 cc with an estimated 10 cc volume in the 1/8" line and the pressure gauge. At 200 psi, which is the pressure at which these capsules were removed from the radiation field, the gas in the anion capsules should occupy 50 cc and in the cation capsules about 30 cc. Results from Gas analyses on samples from the pressure test capsules were corrected to the measured volumes.

4.1 CHEMICAL PROPERTIES OF THE LIQUID PHASE

Immediately upon opening the irradiation capsules the pH of the liquid inside was measured. Since the amount of liquid in most cases was small and inseparable from the resin phase, samples of the resin liquid mixture were placed on wide range pH paper and the pH was estimated. Additional samples were placed on the appropriate narrow range pH paper for closer measurement. Anion resin liquid material was in the basic pH region between 7.5 and 9.0. The pH decreased with increasing gamma ray dose from 8.5 - 9.0 at 10^8 rads to the 8.5 to 7.5 range at 10^9 rads. Cation resin showed a general increase in pH with gamma dose. These pH measurements are given in table 1.

In order to perform chemical analyses on the liquid phase it was necessary to add to a known amount of water a weighed portion of the resin-liquid mixture to separate the phases. This water soaking was especially necessary in the low dose samples where the liquid phase was very small and held inside the resin.

Samples of the water extract from the irradiated cation resin material were analyzed for total organic carbon, total organic sulfur, sulfate, organic acids, aldehydes, alcohols, and peroxides. Water extracts from the irradiated anion resins were analyzed for total organic carbon, total nitrogen, ammonia, amines, organic acids, aldehydes, alcohols, and peroxides. Organic carbon, nitrogen, and sulfur analyses were performed by Atlantic Microlab, Incorporated. Carbon was determined by a modified Pregl procedure, nitrogen by a modified Dumas procedure, and sulfur by Schoniger flask combustion followed by titration. Sulfate, ammonia, and amines were determined by ion exchange chromatography using conductivity detection. The results of these analyses are reported in table 2 and 3 as concentration of analyte in the original

TABLE 1. pH OF LIQUID PHASE

Resin Type	Dose (rads)	pH	Comments
Anion	10^8	8.5-9.0	
Anion	10^9	8.0-8.5	
Anion	10^9	7.5-8.0	Capsule pressurized
Cation	10^8	2.5-3.0	
Cation	3×10^8	4.0-4.5	
Cation	5×10^8	2.5-3.0	
Cation	10^9	4.0-4.5	
Cation	5×10^9	4.0	Capsule pressurized

irradiated liquid-resin mixture. The water extracts were analyzed for organic acids by gas chromotography and liquid chromotography. The gas chromotographic analyses were performed using the method described by Ottenstein and Bartley(1,2). No simple organic acids of carbon number C_2 to C_{10} were detected by this method. This was confirmed by ion exchange chromotography using both conductivity and UV detection. Several anionic species were detected using the two detectors but the retention times did not match any of the literature values. Attempts to extract these species from the effluent of the liquid chromatograph were unsuccessful. It appears that most of the ionic species present are multifunctional species and are very sensitive to the solution conditions. This is evidenced by the fact that liquid chromatography runs using different buffers (borate or carbonate) gave results which did not vary in a predictable manner. Variations in the elution pattern were observed with the column condition and the length of time the sample was exposed to air.

Gas chromatographic analyses for alcohols, aldehydes, and ketones gave results similar to the organic acid determinations. No simple compounds could be detected by gas chromatography but polarographic analyses indicated the presence of carbonyl groups either as aldehydes or ketones. Using the polarographic method of Heyrovsky (3) a possible indication of the presence of a small amount of peroxide was obtained. The polarographic peak found in the samples was small and shifted slightly negative from that in the peroxide standards. The effect of other functional groups may account for the shift. Infra red analyses proved to be of little use in verifying any of these results due to the difficulty in removing the compounds from the water extracts.

The results of the chemical analyses on the water extracts from anion resins show some distinct differences from those reviewed by Gangwer, Gold-

TABLE 2. CHEMICAL ANALYSIS OF WATER EXTRACTS FROM IRRADIATED ANION RESINS

Dose (rads)	C(%)	N(%)	Ammonia (%)	Monomethyl Amine (%)	Dimethyl Amine (%)	Trimethyl Amine (%)
10^8	4.1	0.72	0.002	0.015	0.41	2.1
10^9	6.5	1.4	0.027	0.19	2.6	0.79
10^9	4.8	1.3	0.044	0.26	2.2	0.81

TABLE 3. CHEMICAL ANALYSIS OF WATER EXTRACTS FROM IRRADIATED CATION RESINS

Dose (rads)	C(%)	Organic Sulfur (%)	Sulfate (%)
10^8	2.2	0	0.59
3×10^8	2.8	0	2.9
5×10^8	2.9	0	2.5
10^9	3.6	0	4.1
5×10^9	4.1	0	5.3

stein, and Pillay(4). Organic carbon analyses indicate significant weight loss from the resin due to carbon compounds other than amines. The most striking difference appears to be the amine yields. At 10^8 rads the primary product is trimethylamine as in the previous studies. When the dose is increased to 10^9 rads the trimethylamine yield decreases drastically and more dimethyl and monomethyl amines are produced. It is not known what effect the borate ion has in this system, which may account for the differences with previous work. Calculation of the total nitrogen found as ammonia and the various amines gives a number slightly smaller than the total nitrogen value determined by the Dumas method. This small difference may indicate the presence of some other forms of nitrogen but it is very close to the experimental error in the five determinations.

4.2 CHEMICAL PROPERTIES OF IRRADIATED SOLID MATERIAL

Samples of the solid material from the anion and cation capsules were submitted to Atlantic Microlab for carbon, hydrogen, nitrogen, and sulfur analysis. Carbon and hydrogen were determined by the modified Pregl method, nitrogen by the modified Dumas procedure, and sulfur by Schoniger flash combustion and titration. The samples were drained of any liquid and dried at 40°C under vacuum. Since the resin was not washed some of the liquid products probably remained inside the beads. The results are summarized in Table 4 and expressed as percent of wet weight.

The analyses on the cation resin solids indicates that the solid material remains of essentially the same elemental constitution up to 5×10^9 rads. The carbon to hydrogen and carbon to sulfur ratios change by less than 10%. On the other hand, the anion resin solids show changes with increasing dose. The carbon to hydrogen ratio increases by 25% between unirradiated resin and

TABLE 4. ELEMENTAL ANALYSIS OF ION EXCHANGE RESINS

Resin	Dose	C (%)	H (%)	N (%)	S (%)
Cation	0	43.55	4.78	0.0	9.98
Cation	10^8	40.37	4.24	-	12.60
Cation	3×10^8	40.30	4.49	-	11.08
Cation	5×10^8	41.10	4.35	-	12.52
Cation	10^9	38.42	4.66		11.97
Cation	5×10^9	42.25	4.72	-	10.36
Anion	0	47.09	7.54	3.18	0.0
Anion	10^8	48.79	7.49	3.10	-
Anion	10^9	54.02	7.30	2.69	-
Anion	10^9	60.43	7.49	2.50	-

resin irradiated to 10^9 rads. The carbon to nitrogen ratio increases by 60% over the same dose interval. Loss of aliphatic amine groups probably accounts for these changes. Unfortunately, these solids were so hydroscopic that infra-red analyses showed only the presence of water.

4.3 CHEMICAL PROPERTIES OF THE GAS PHASE

Samples of the radiolytic gases were withdrawn from the gas sampling bags used to trap the radiolytic gases as described in section 3.2 Gas samples were drawn through hypodermic tubing into evacuated 25 cc glass vials fitted with rubber septa. Hydrocarbon gases were determined by gas chromatography on a ¼"X 6' stainless steel column packed with n-octane / porasil c using a flame ionization detector. Hydrogen was determined by gas chromatography on a 1/8" X 3' stainless steel column packed with Porapak R using a catalytic detector(5). Thermal conductivity gas chromatography was used to determine CO_2, CO, N_2, and O_2 on a 1/8" X 6' Carbosieve S column. Sulfur gases were estimated by injecting an absorbant solution of Na_2CO_3, $NaHCO_3$, and H_2O_2 into the vial, converting the sulfur gases to SO_4^{-2}, and determining the sulfate by the method described in section 4.1. The results of these analyses on radiolytic gases from the pressure test capsules are given in table 5. The following figures show the results of the pressure test capsule analyses as well as measurements made using the GC test capsules in phase one of this project after correction for the gas volume effects discussed in section 3.2.

The results shown in table 5 for the cation resin radiolytic gases are in rough agreement with earlier results summarized by Gangwer, Goldstein, and Pillay(4) for H_2, CO_2 and CO. The cation resin used in this study was in the

TABLE 5 ANALYSIS OF GASES FROM PRESSURE TEST CAPSULES

Resin	Dose (rads)	Total gas (cc/gm resin)	H_2 (cc/gm)	CO_2 (cc/gm)	CO (cc/gm)	CH_4 (cc/gm)	O_2 (cc/ gm)	N_2 (cc/ gm)	Sulfur gases (μgS/gm)
Anion	7.9×10^8	12.6	6.8	1.1	1.3	0.81	0.56	0.68	-
Cation	2.5×10^9	6.8	2.8	0.83	1.0	0.12	0.71	-	0.09

FIGURE 10

H_2 PRODUCTION

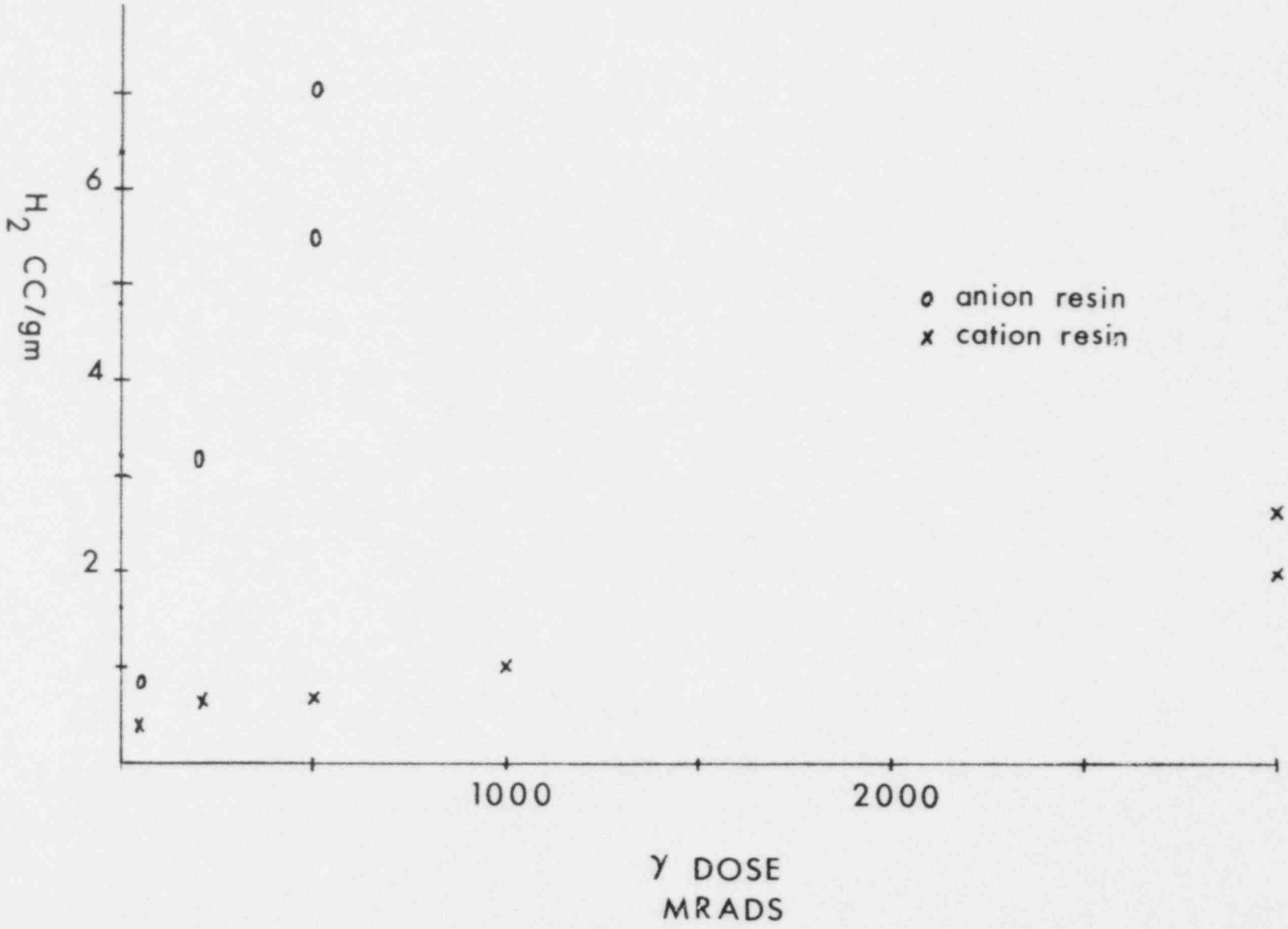


FIGURE 11

METHANE PRODUCTION

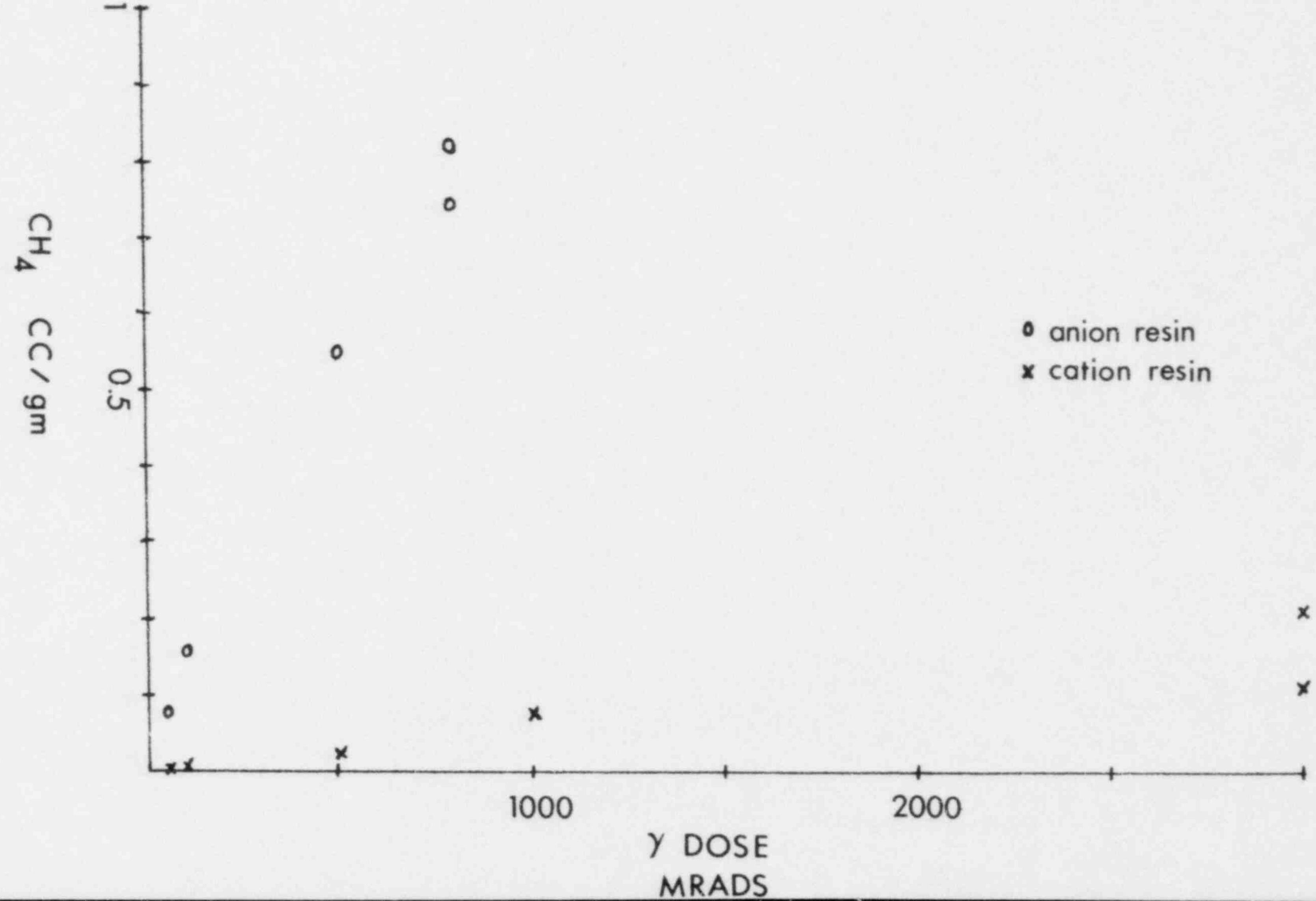


FIGURE 12
ETHANE PRODUCTION

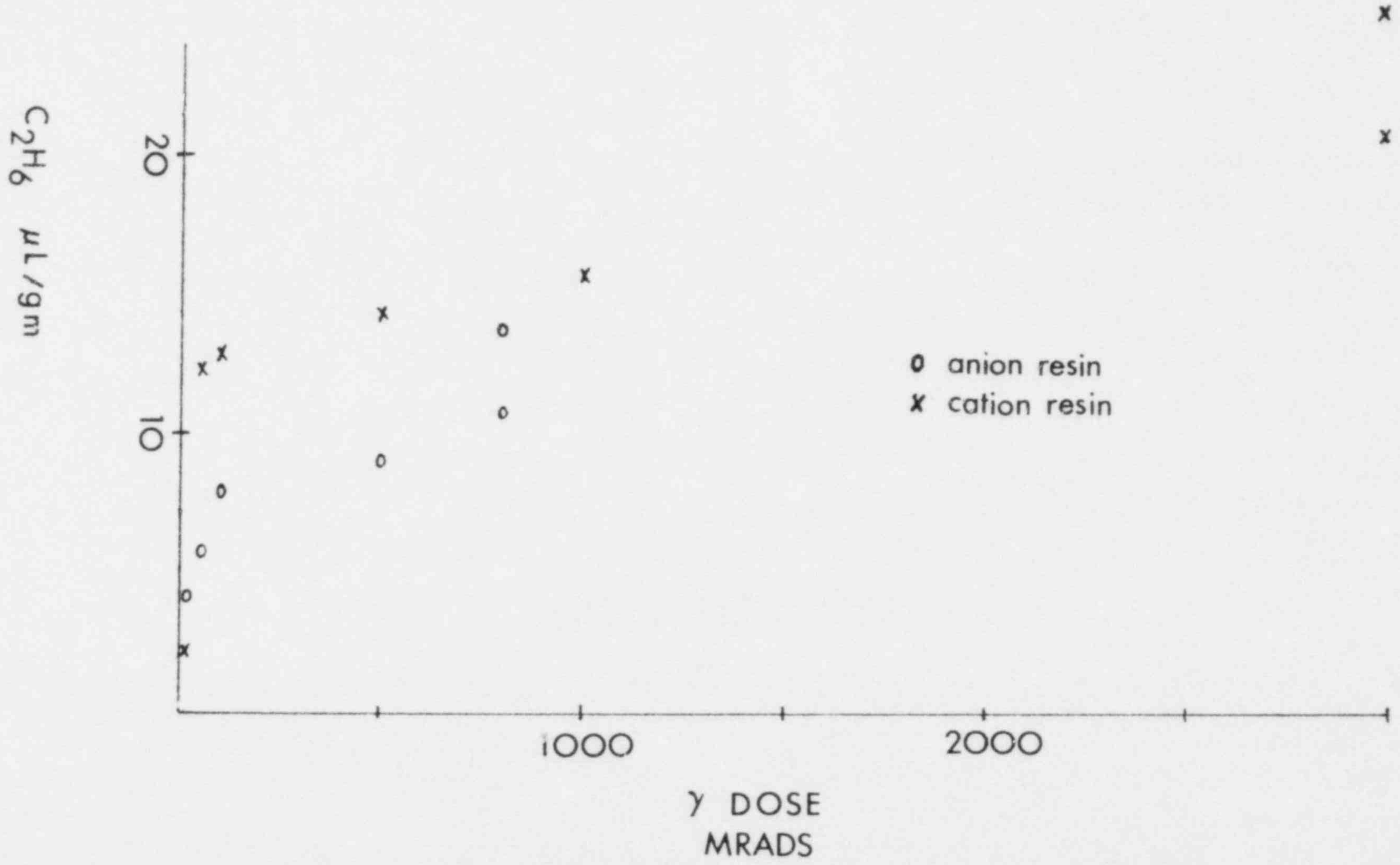


FIGURE 13

PROPANE PRODUCTION

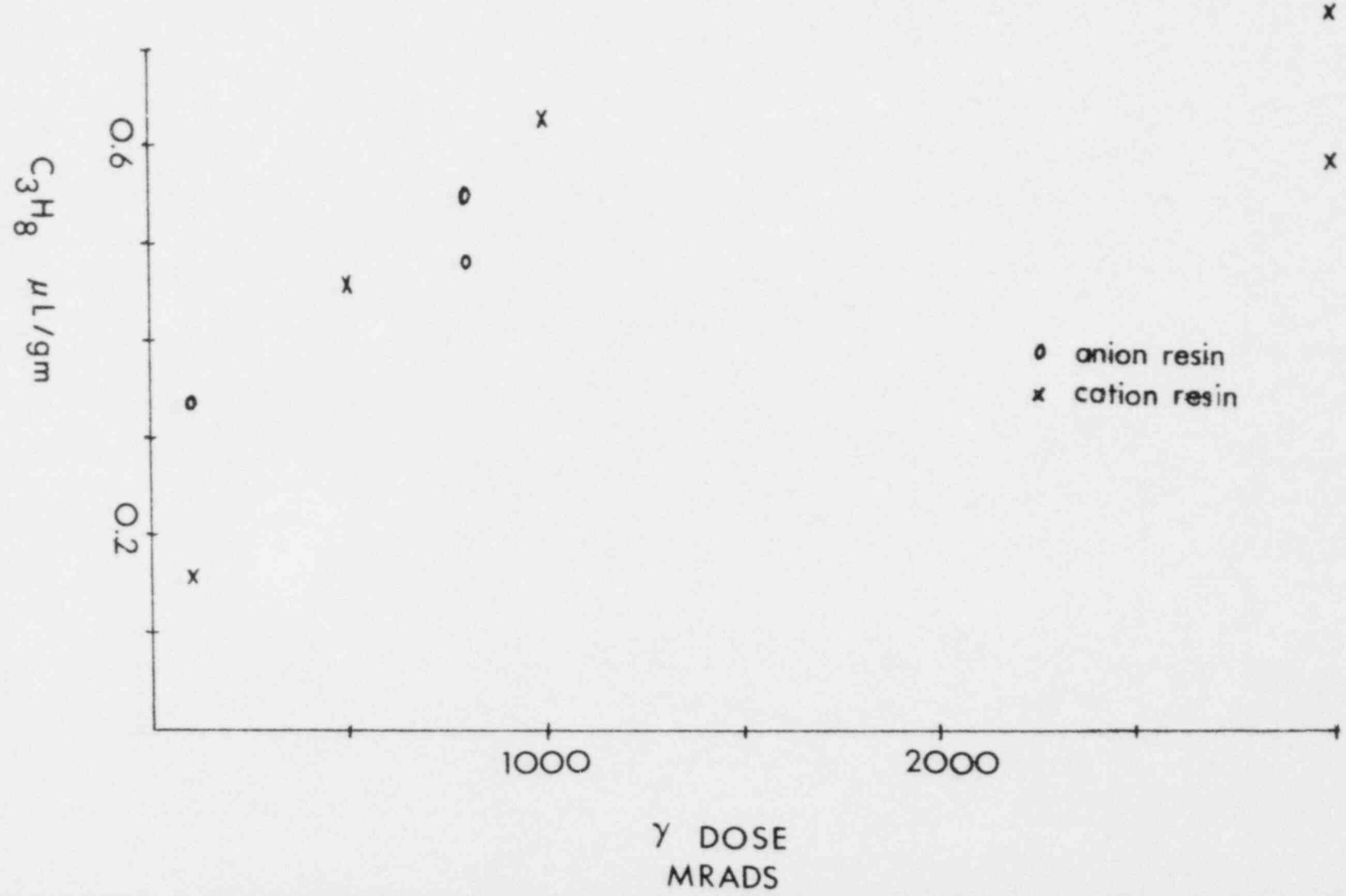
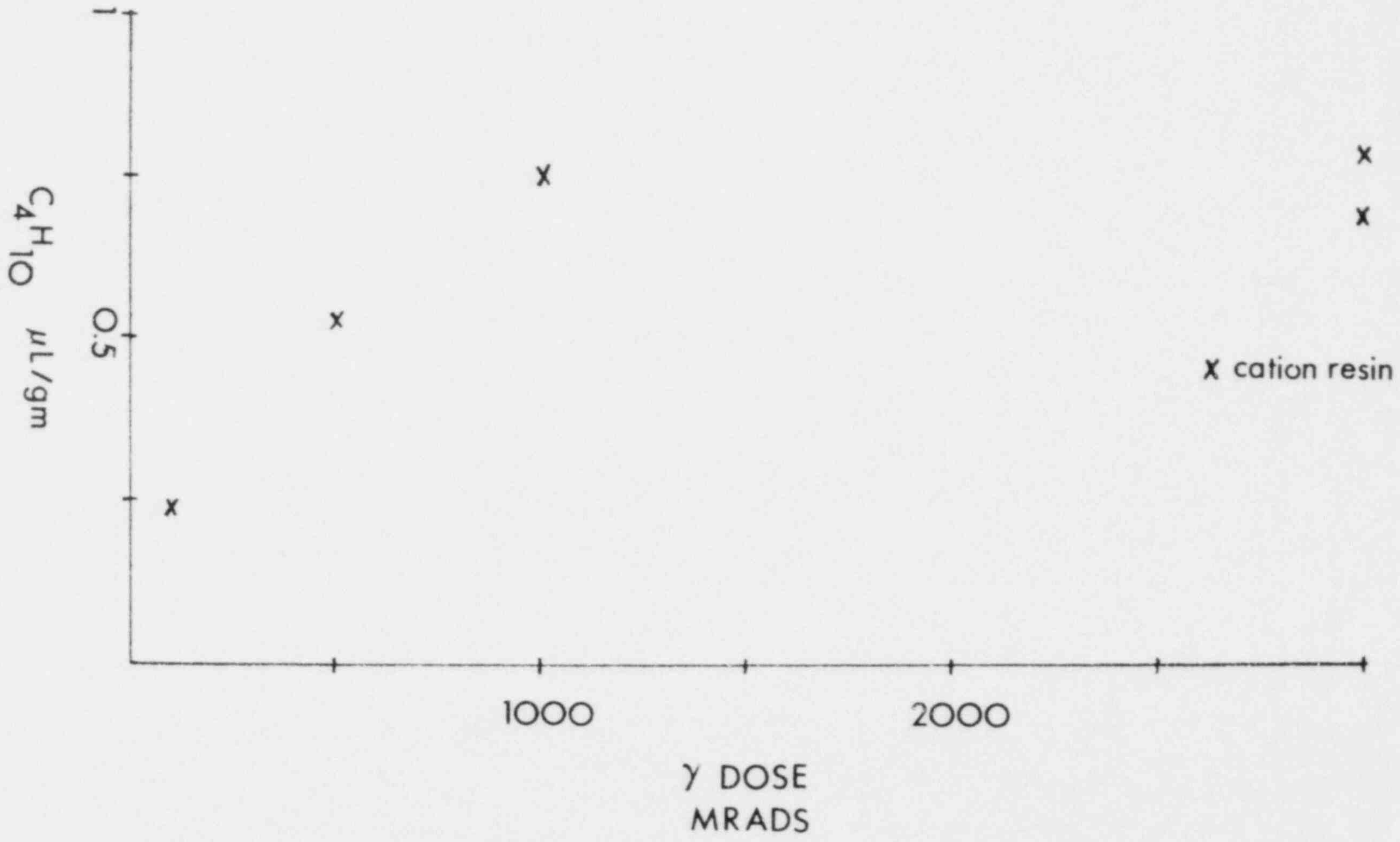


FIGURE 14
BUTANE PRODUCTION



sodium form with approximately 30% water content. The effects of the stainless steel surfaces and the contact with the radiolytic gases under high pressure are not known. The pressure test capsules were allowed to sit under pressure at room temperature for about one year between phase one and phase two of this project which probably had some effect on the more reactive gases. Previous analyses on anion resin radiolytic gases are very sketchy and comparison is further complicated by the fact that the resin used in this study was in the borate form. This study was initially undertaken as an engineering test to determine the pressure buildup in a few very specific situations, as a result, the later more basic scientific phase of this work has had to cope with capsules and equipment which were not ideal from the standpoint of basic research design.

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ANALYSIS OF IRRADIATED ION EXCHANGE MATERIALS: CORROSION EFFECTS

by

M. Marek and J. G. Rinker
Metallurgy Program, School of Chemical Engineering

GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia 30332

FINAL RESEARCH REPORT
Sub-project

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Associated Universities, Inc.
Upton, N.Y. 11973

OBJECTIVE

To evaluate the corrosion effects on stainless steel containers which were exposed to irradiated ion-exchange resins.

MATERIALS

The containers were made of AISI type 304 stainless steel tubing stock. The nominal composition of type 304 steel is: 18-20% Cr, 8-10.5% Ni, max 0.08% C, max 2% Mn, max 1% Si, max 0.045% P, max 0.030% S, balance Fe. The steel used in the program was not analyzed for actual composition.

The containers were made available for metallurgical examination after the resin had been removed. The samples were identified by letter codes; an unexposed Control was also included in the examination. The sample codes, nominal sizes, radiation doses, and final pH data for the environment are shown in Table 1.

Only the internal surfaces of the tubes were exposed to the resin and the products of its decomposition. The length of the containers which had been exposed was about 6.5 in. for samples A, C, and D, and about 4 in. for samples P - X and Control.

PROGRAM OF TESTS

The following tests were conducted on each container:

1. Examination of the interior surfaces by low-power microscopy.
2. Examination of the interior surfaces by scanning electron microscopy (SEM).
3. Metallographic examination of both radial and axial sections.
4. Mechanical tensile testing to determine yield strength, ultimate tensile strength, and elongation at fracture.

A limited, energy-dispersive X-ray analysis was performed on some of the samples to determine the composition of deposits on the surface exposed to the resin.

PREPARATION OF SPECIMENS

The as-received samples were cleaned ultrasonically in distilled water. A short section was cut from each tube and further cut into small pieces suitable for metallographic and SEM examination. The remaining exposed part of each container was prepared for mechanical testing. Samples A, C, and D were machined to produce tensile strip specimens of the shape and dimensions shown in Fig. 1. Samples F - X and Control were tested in the tubular form according to ASTM E-8. Figure 2 shows the dimensions of the specimens and the brass plugs used for the reinforcement of the ends.

TESTING PROCEDURES

The low-power optical microscopy and metallographic examinations were performed using the standard techniques and facilities of the Metallurgical Laboratories of the School of Chemical Engineering. The SEM examination was performed in the Georgia Tech Engineering Experiment Station using the Cambridge Model 150 Scanning Electron Microscope. The tensile tests were done in the Fracture and Fatigue Laboratory using the MTS Model 810 testing system, a 1 in. gauge length extensometer, and an X-Y recorder for the recording of the load-elongation curves. The crosshead speed was 0.02 mm/sec.

RESULTS

It was found that the samples could be divided into two groups according to the observed corrosion effects. Group I consisted of samples A, S, V, and X; samples of this group had been exposed to the anionic resin and showed little, if any, evidence of corrosion attack. Group II consisted of samples C, D, P, Q, T, U, and W, which had been exposed to the cationic resin and showed various degrees of deterioration by corrosion. The results of the tests will be presented separately for the two groups.

Group I. The low-power optical examination of the exposed surfaces failed to reveal any difference between the unexposed control and samples S, V, and X. No control was available for comparsion with sample A; the examination of sample A did not reveal any surface features which would indicate a corrosion attack. The metallographic examination also showed no difference between the structure of the control and the structure of samples S, V, and X. Figure 3a shows a micrograph representative of the structure of the control and Figure 3b shows the structure of sample V; examination of samples S and X showed same structures.

The SEM examination showed that the surfaces of the exposed samples were cleaner than the surface of the control (Figure 4). No evidence of corrosion attack was observed on sample S (Figure 5) and sample A (Figure 6). Samples V and X showed a slight etching of the grain boundaries (Figures 7 and 8).

The results of the mechanical tests (Table 2) show that all the differences in properties in this group were within the limits of experimental error.

Group II. The appearance of the exposed surfaces of the samples in this group, as revealed by both low-power and SEM microscopy, indicated that substantial corrosion attack had taken place. The attack was usually localized in circular areas which may have been the areas of contact between beads of the resin and the metal. The attacked areas were etched and pitted; on the more severely corroded samples the etching and pitting resulted in relatively large (up to about 0.8 mm in diameter), shallow depressions In some cases smaller but deep pits were observed. Following is a brief description of the surface features observed on the individual samples:

Sample C. Large areas of the surface were covered with crusty deposits (Figure 9a); where the deposits peeled off, the metal surface was etched. Numerous small, shallow pits (Figure 9b) and occasional deep pits were observed as well as a few large and deep depressions. Relatively clean areas showed evidence of grain boundary attack (Figure 10).

Sample D. Many relatively small pits, which were deeper than those on Sample C, were observed (Figures 11 and 12). Large areas were covered with a dark deposit. Some very deep pits were found, as well as some very large, but shallow, areas of attack.

Sample P. Large areas of the surface were covered with a deposit. Where the deposit had peeled off. the surface was pitted and etched (Figure 13). The examination of cross sections (Figure 14) showed that the attack resulted in shallow depressions, about 50 to 70 μm deep.

Sample Q. Appearance was similar to that of Sample P, showing extensive pitting under the deposits (Figure 15). Depressions caused by the attack were more localized but deeper than on Sample P.

Sample T. Relatively clean surface that showed etched circular areas, some of which contained deeper pits in the center (Figure 16).

Sample U. Examination revealed a relatively small number of sites of attack in the form of circular, deep pits. Most of the depressions were covered with heavy deposits (Figure 17a). Heavy crystallographic etching was observed at the rims of the pits (Figure 17b).

Sample W. Large, circular areas were corroded into relatively deep depressions. Heavy deposits were found in the pits (Figure 18). Some of the area outside the pits was covered with a crust. Clean surface areas showed evidence of grain boundary attack. Examination of cross sections showed shallow depressions up to 200 μm deep (Figure 19).

A semi-quantitative, energy-dispersive X-ray analysis of the deposits in the pits showed high percentages of sulfur, nickel, and chromium, indicating that the deposits were primarily sulfates of these two alloying elements.

In spite of the relatively severe attack on some of the samples, the results of the mechanical tests (Table 2) did not show a degradation of strength. The only significant degradation of properties was observed for samples C and U which showed a decreased elongation at fracture.

DISCUSSION

The results of the metallographic and surface examinations of the containers show that the anionic resins caused little if any attack on the Type 304 stainless steel. The very slight etching of grain boundaries on samples V and X, which received the highest radiation doses, was not considered significant. The pressurized container, A, did not show any more attack than the other samples. There was no effect of the exposure on the mechanical properties.

The lack of deterioration by the anionic resin and its products of decomposition seems to be mainly due to the pH of the environment, which was in the neutral to slightly alkaline range. These conditions are favorable for the resistance of stainless steel to the corrosion. The lack of corrosion is also evidence of the absence of aggressive species in the environment. The cationic resin and its products of decomposition caused serious deterioration of the stainless steel. The deterioration was generally higher for higher radiation doses, sample T being least attacked and sample W most attacked. There were, however, no sharp differences between samples which received slightly different radiation doses. The deterioration seemed to be related mainly to the acidic nature of the environment, which ranged from pH 2.5 to pH 4.5. At this level of acidity, the Type 304 stainless steel is less corrosion resistant than in neutral environments. The presence of sulfates in the electrolyte indicates that the environment was essentially a sulfuric acid solution. A detailed chemistry of the environment was not available.

The localized nature of the attack indicates that at the areas of contact between the resin and the container, and/or under the crusty deposits of the products of decomposition, formation of localized corrosion cells resulted in a severe local attack. The attack was probably intensified by local chemistry changes, which may have included further acidification and oxygen concentration differences. The mechanism of the attack may have included other aggressive species; since the detailed chemistry of the environment was not available, these factors could not be thoroughly evaluated.

Although the depth of the attack was as high as 20% of the wall thickness in the deepest pits, the mechanical strength, as determined by the tensile tests, was not affected. This can be explained by the localized nature of the attack; even the deepest pits represented only a small fraction of the total cross section. The only noticeable effect on mechanical properties occurred when a pit was located in a critical area, such as at the edge of the specimen, and the resulting localization of deformation led to lower values of elongation at fracture.

In spite of the lack of effect on mechanical strength the observed corrosion attack represents a serious degradation of the stainless steel containers. The form of the attack indicates that, at longer exposures, corrosion would probably result in perforation of the container walls.

CONCLUSIONS

1. The irradiated anionic resin and its products of decomposition did not significantly attack the Type 304 stainless steel containers.
2. The irradiated cationic resin and its products of decomposition caused significant localized corrosion of the Type 304 stainless steel containers. The attack was in the form of etching under surface deposits, and pitting. The pits were of different sizes, including some large and deep depressions.
3. Although the corrosion attack did not cause loss of strength of the test cylinders, the localized nature of the attack might cause performation of the walls at longer exposures.

Table 1

Sample Codes, Nominal Sizes, and Exposure Data

Code	Nominal O.D. (in.)	Nominal wall thickness (in.)	Resin type	Dose (rads)	pH[2]
A[1]	1.0	0.037	anion	7.9×10^8	
C[1]	1.0	0.037	cation	2.5×10^9	
D[1]	1.0	0.037	cation	2.5×10^9	
P	0.5	0.034	cation	1×10^9	4.0-4.5
Q	0.5	0.034	cation	3×10^8	4.0-4.5
S	0.5	0.034	anion	1×10^8	8.5-9
T	0.5	0.034	cation	1×10^8	2.5-3
U	0.5	0.034	cation	5×10^8	2.5-3
V	0.5	0.034	anion	1×10^9	7.5-8
W	0.5	0.034	cation	5×10^9	4
X	0.5	0.034	anion	1×10^9	8-8.5
Control	0.5	0.034	none	none	

---

1) Pressurized containers

2) pH of the environment in the containers at the end of the exposure, as determined in the main project.

Table 2

Results of the Tensile Tests of the Containers

Sample Code	Yield Strength (MPa/psi) MPa(psi)	Ultimate Tensile Strength (MPa/psi) MPa (psi)	Elongation at Fracture %
A	250 (36,200)	ND	68
	250 (36,300)		68
C	243 (35,200)	ND	41
	228 (33,000)		58
D	247 (35,800)	ND	55
	249 (36,100)		60
P	310 (44,950)	642 (93,180)	61
Q	356 (51,600)	658 (95,400)	59
S	331 (46,060)	650 (94,290)	63
T	312 (45,750)	646 (93,750)	72
U	333 (48,280)	639 (92,640)	47
V	325 (47,170)	646 (93,750)	63
W	ND	642 (93,180)	61
X	312 (45,220)	650 (94,290)	63
Control	314 (45,480)	650 (94,290)	64

Notes: Samples A, C, and D were made of a tubing stock different from the one used for samples P - X and Control.

ND: Not Determined

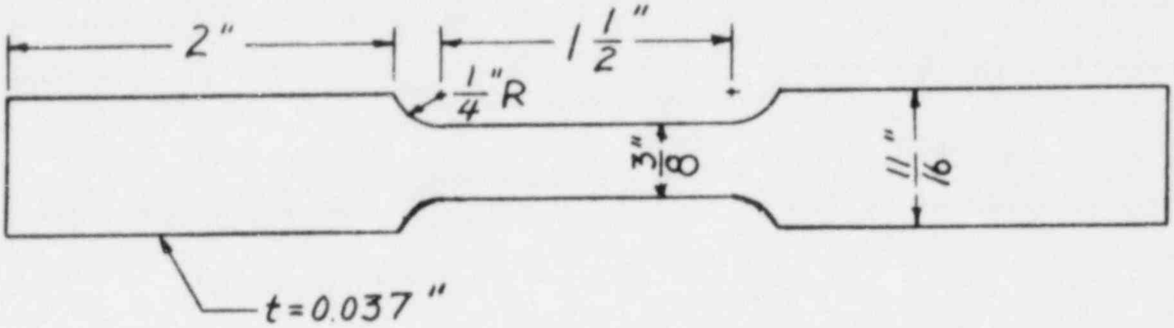

Figure 1. Tensile Specimen Used in Tests of Samples A, C, and D.

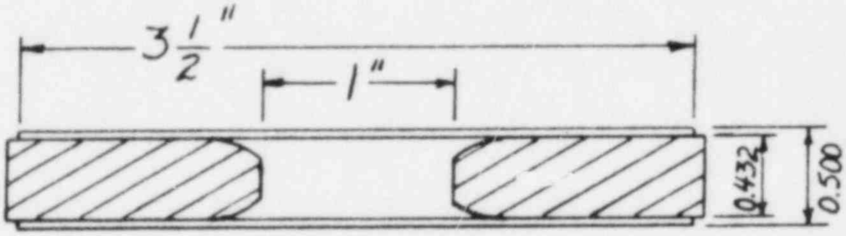

Figure 2. Tubular Tensile Specimen Used in Tests of Samples P to X and Control.

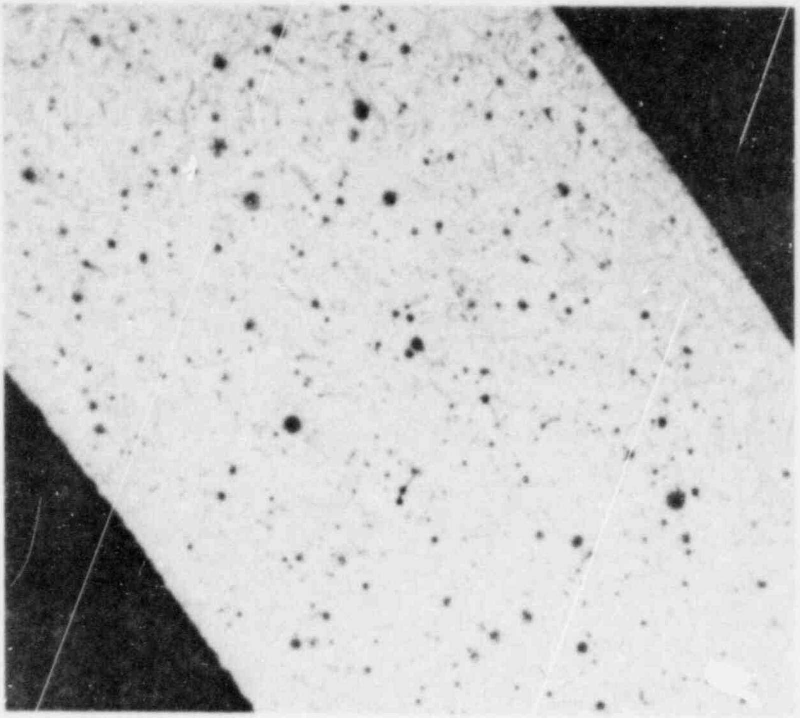

Figure 3a. Etched Radial Metallographic Section of Control (100X).

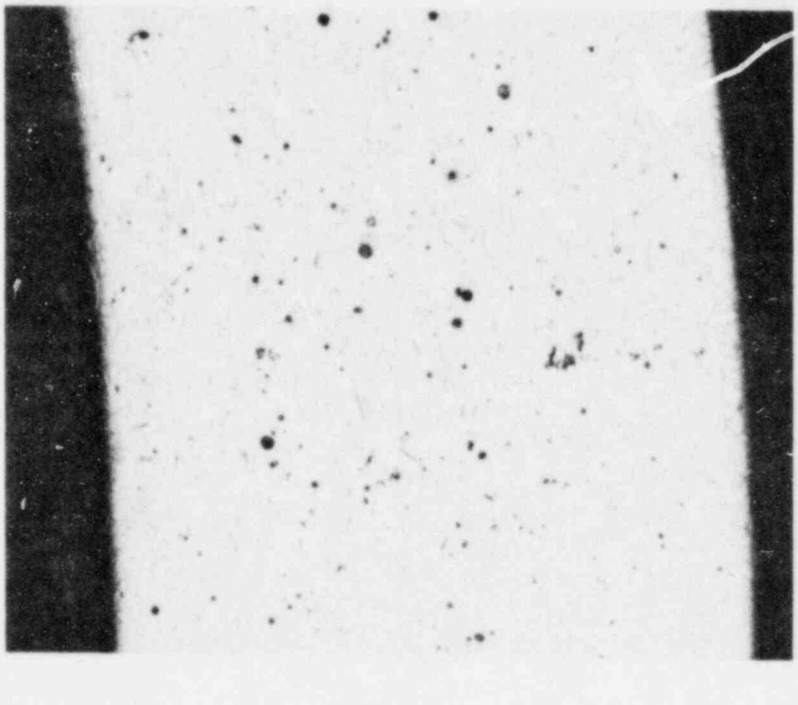

Figure 3b. Etched Radial Metallographic Section of Sample V (100X).

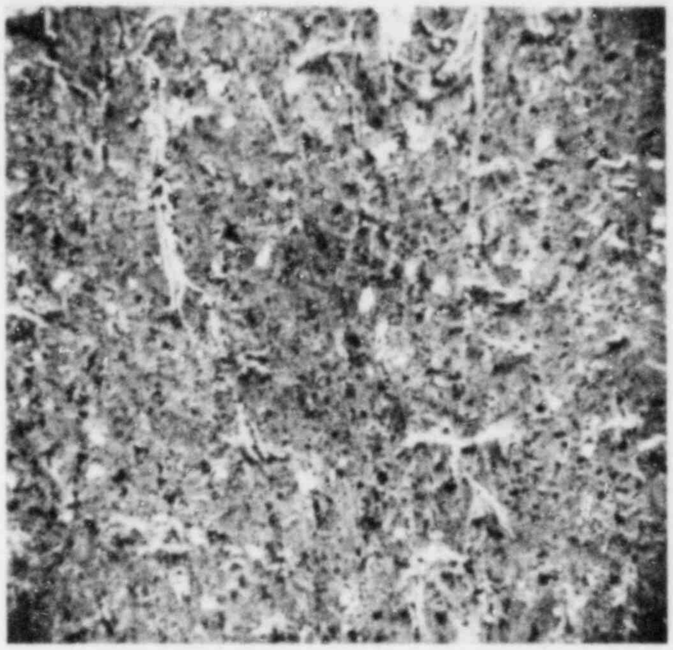

Figure 4a. SEM Micrograph of the Surface of Control (530X).

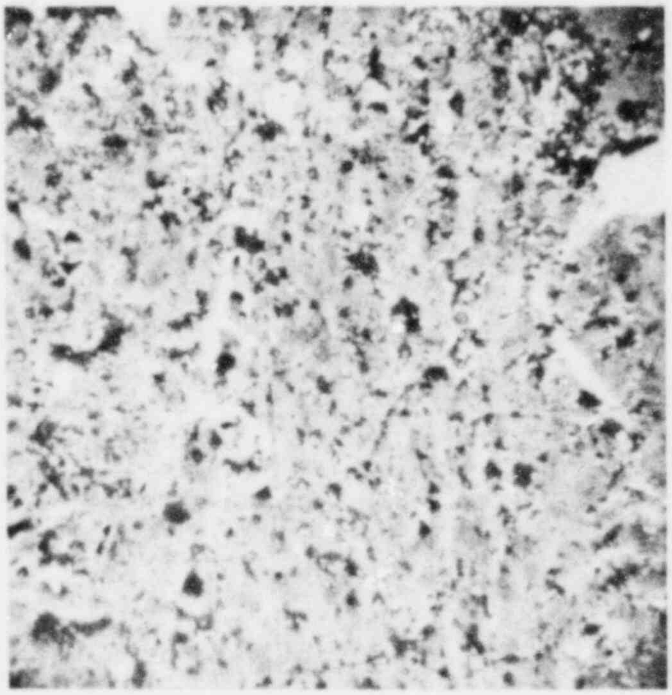

Figure 4b. SEM Micrograph of the Surface of Control (1,500X).

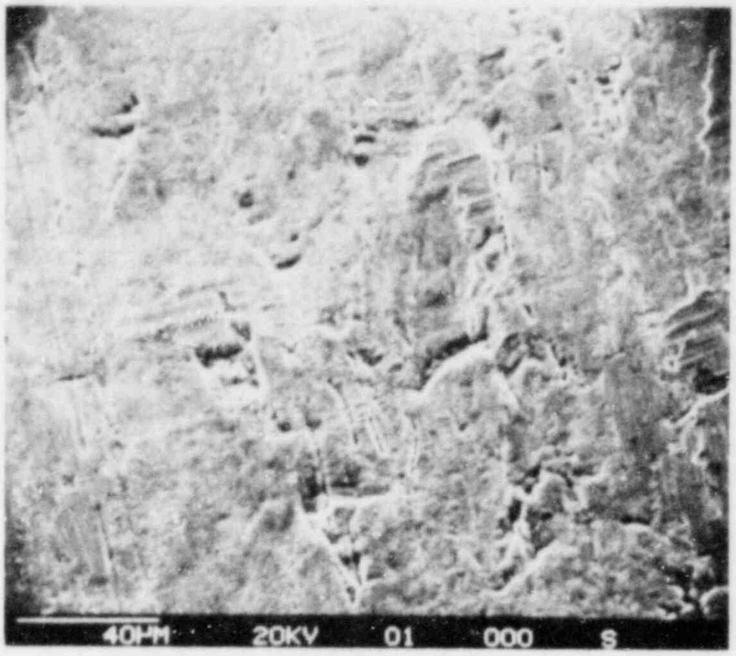

Figure 5a. SEM Micrograph of the Surface of Sample S (500X).

Figure 5b. SEM Micrograph of the Surface of Sample S (1,500X).

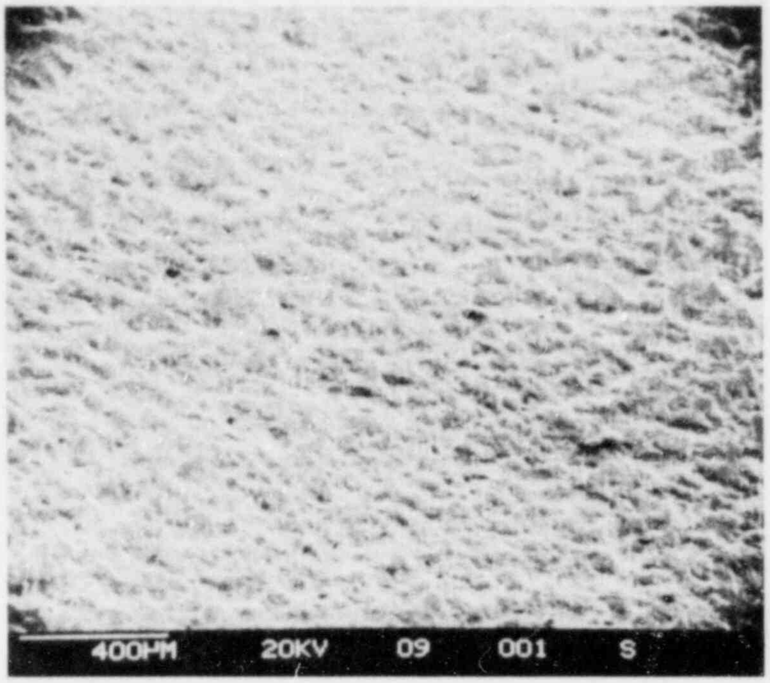

Figure 6a. SEM Micrograph of the Surface of Sample A (50X).

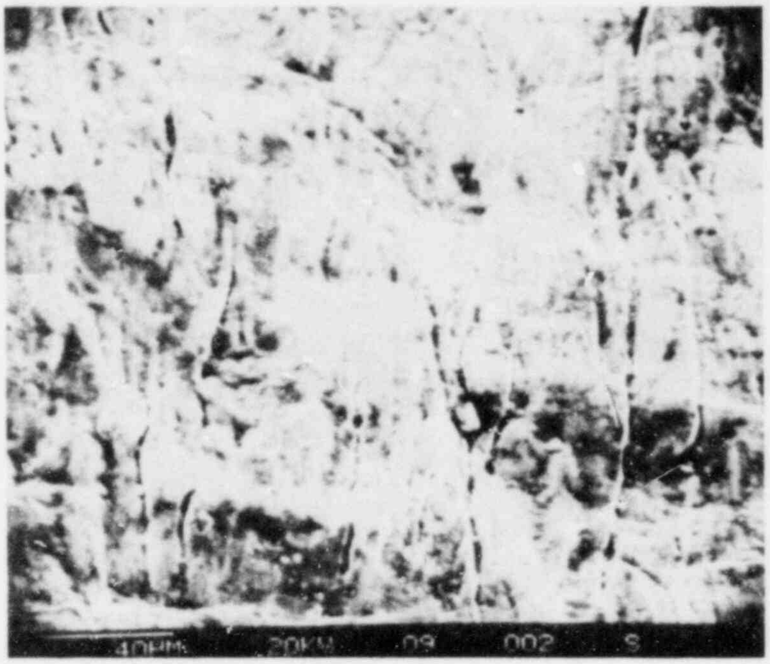

Figure 6b. SEM Micrograph of the Surface of Sample A (500X).

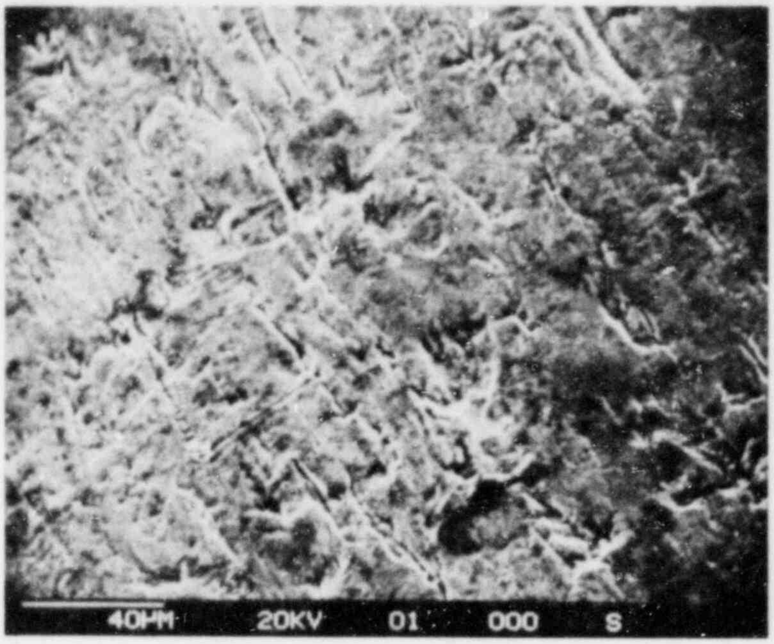

Figure 7a. SEM Micrograph of the Surface of Sample V (500X).

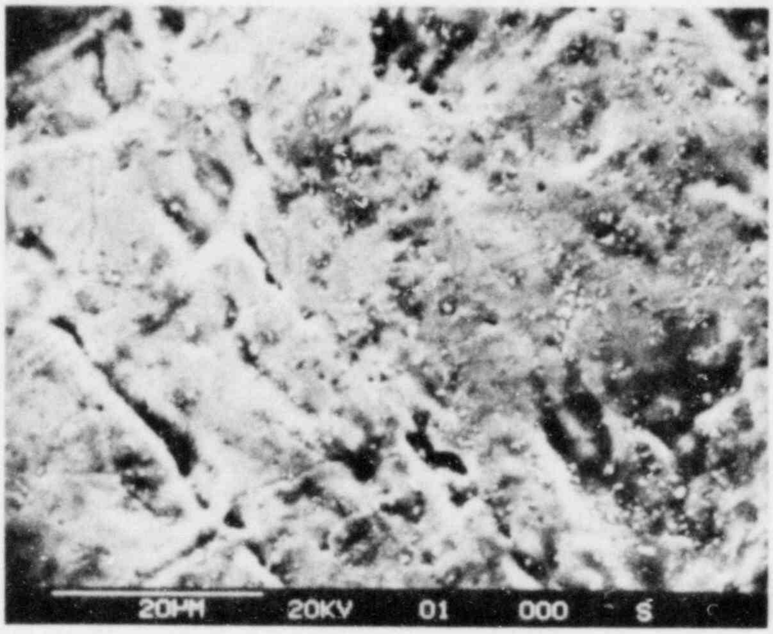

Figure 7b. SEM Micrograph of the Surface of Sample V (1,500X).

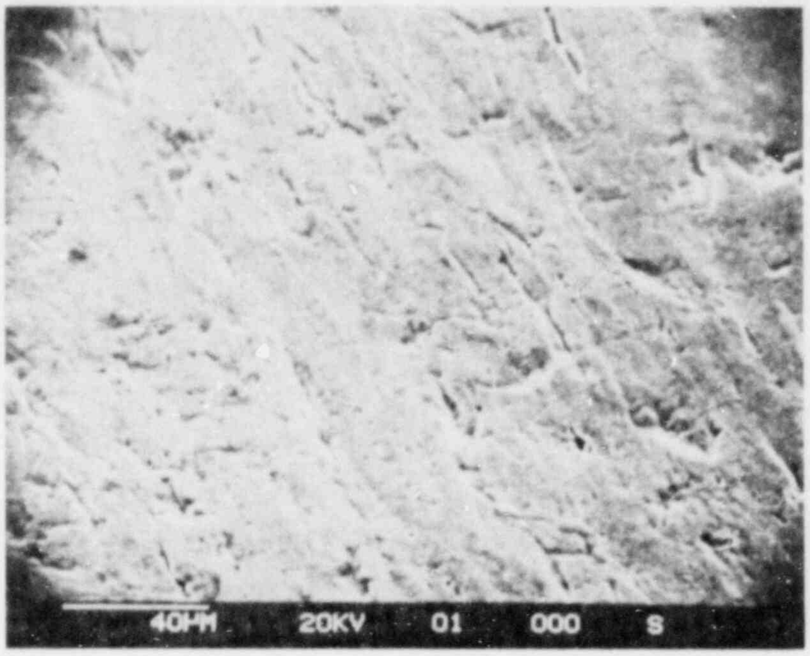

Figure 8a. SEM Micorgraph of the Surface of Sample X (500X).

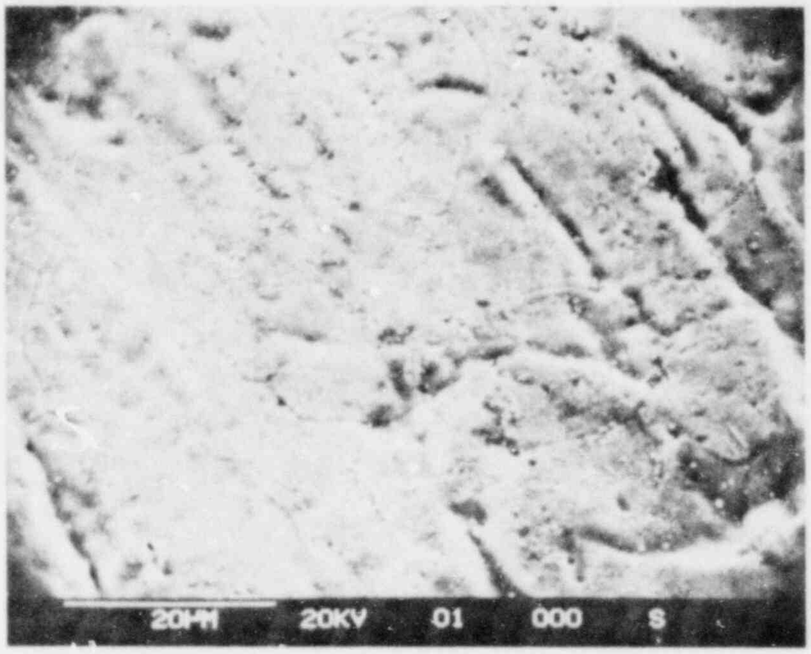

Figure 8b. SEM Micrograph of the Surface of Sample X (1,500X).

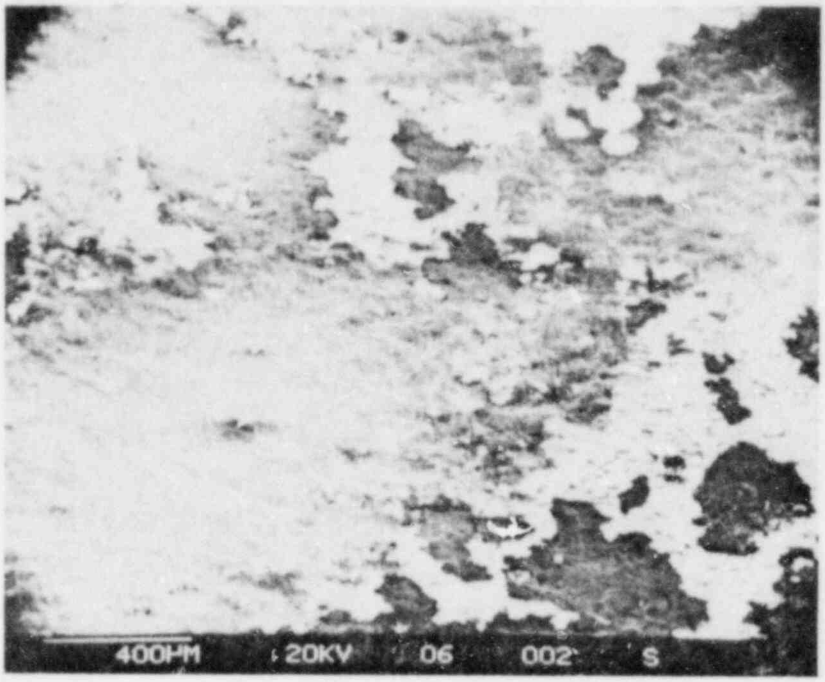

Figure 9a. SEM Micrograph of the Surface of Sample C (50X).

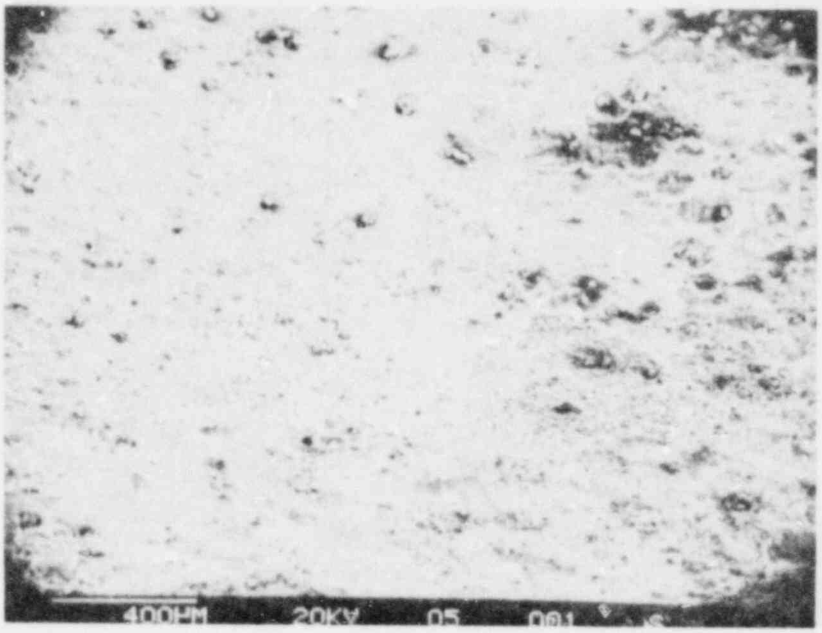

Figure 9b. SEM Micrograph of the Surface of Sample C (50X).

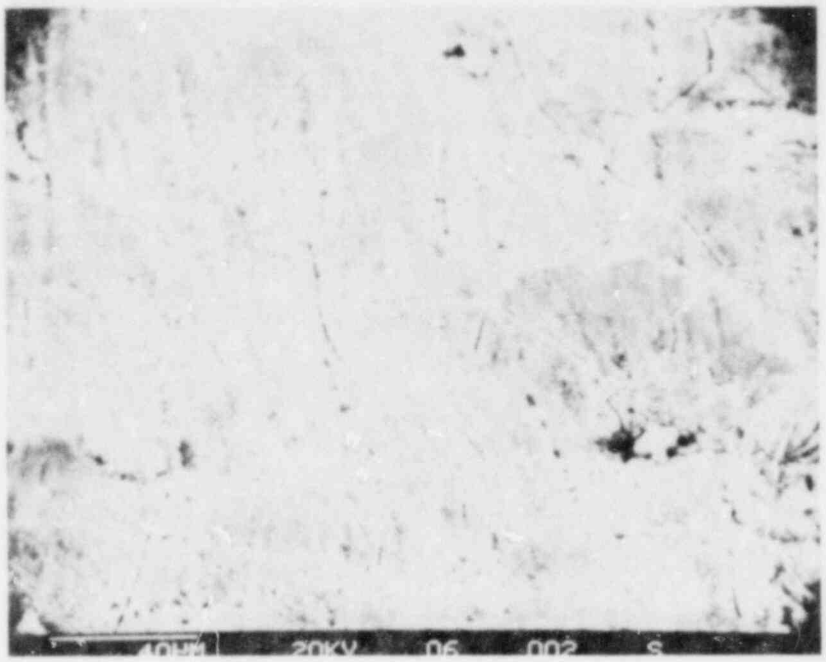

Figure 1 . SEM Micrograph of the Surface of Sample C (500X).

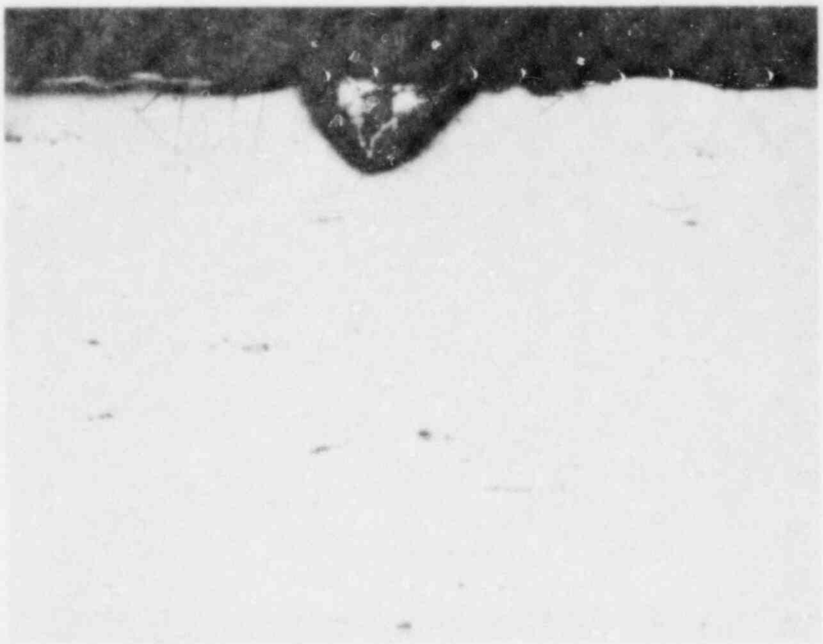

Figure 11. Etched Axial Metallographic Section of Sample D (250X).

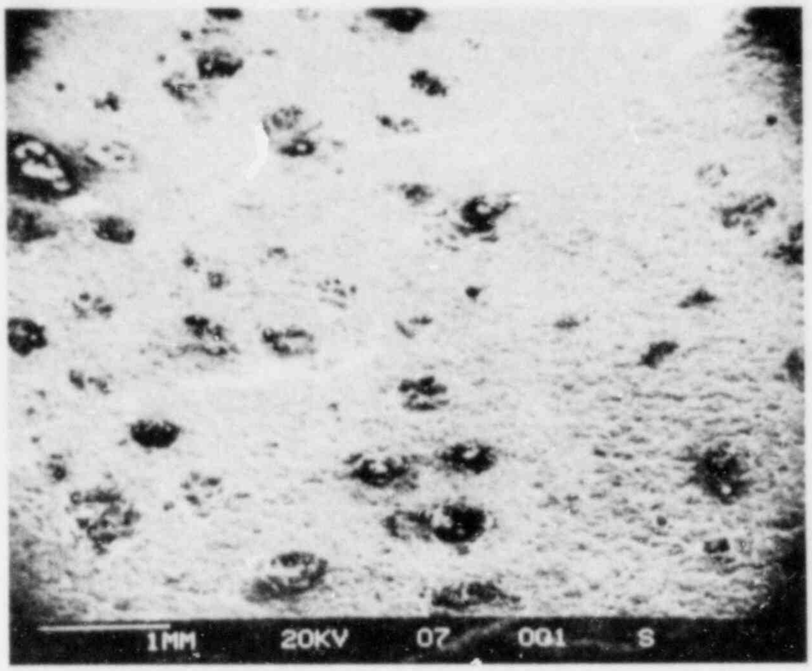

Figure 12a. SEM Micrograph of the Surface of Sample D (20X).

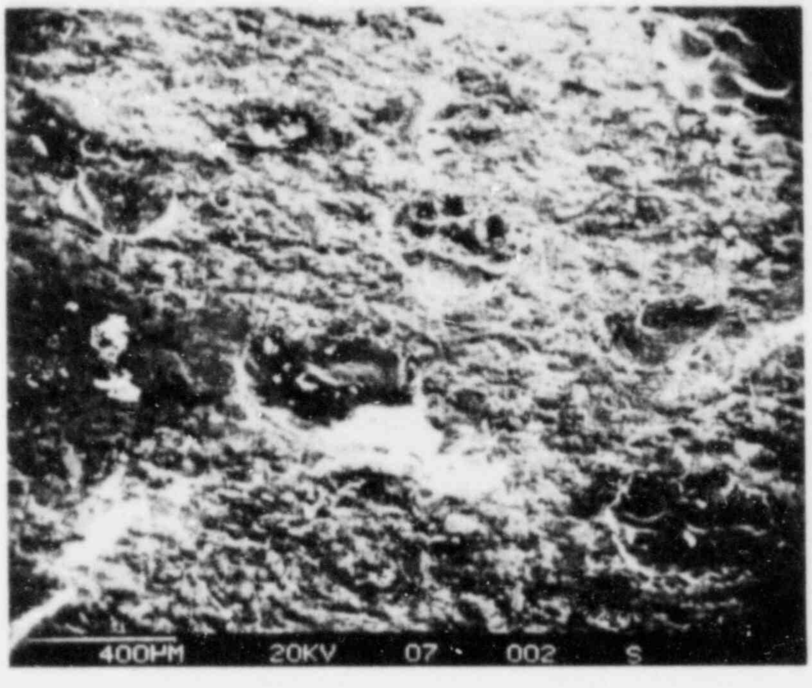

Figure 12b. SEM Micrograph of the Surface of Sample D (50X).

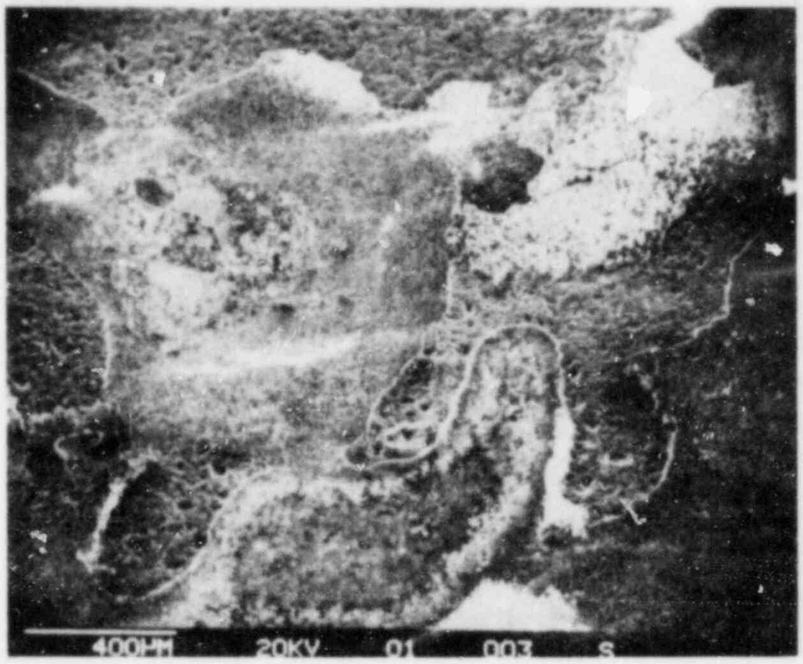

Figure 13. SEM Micrograph of the Surface of Sample P (50X).

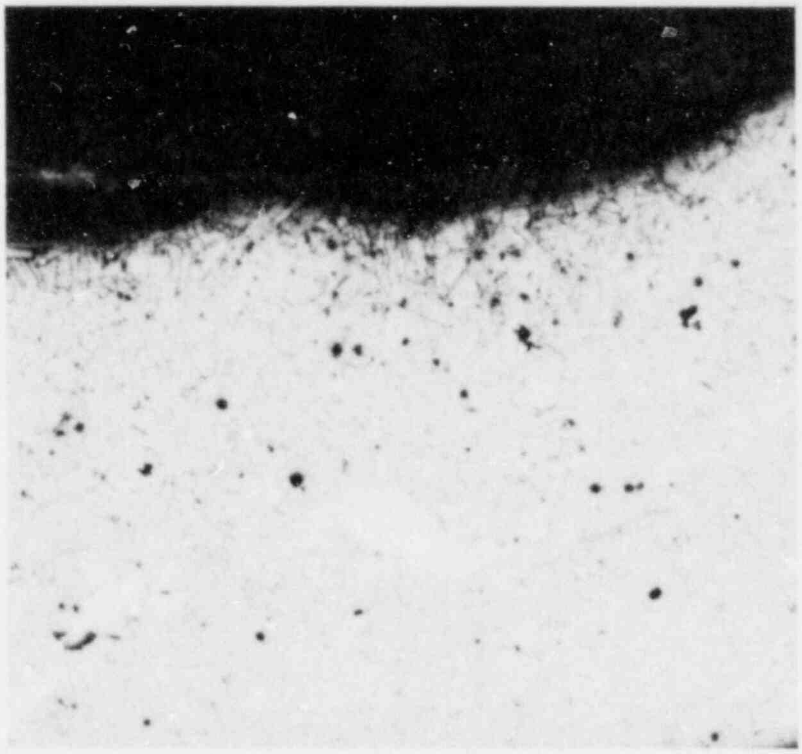

Figure 14. Etched Radial Metallographic Section of Sample P (100X).

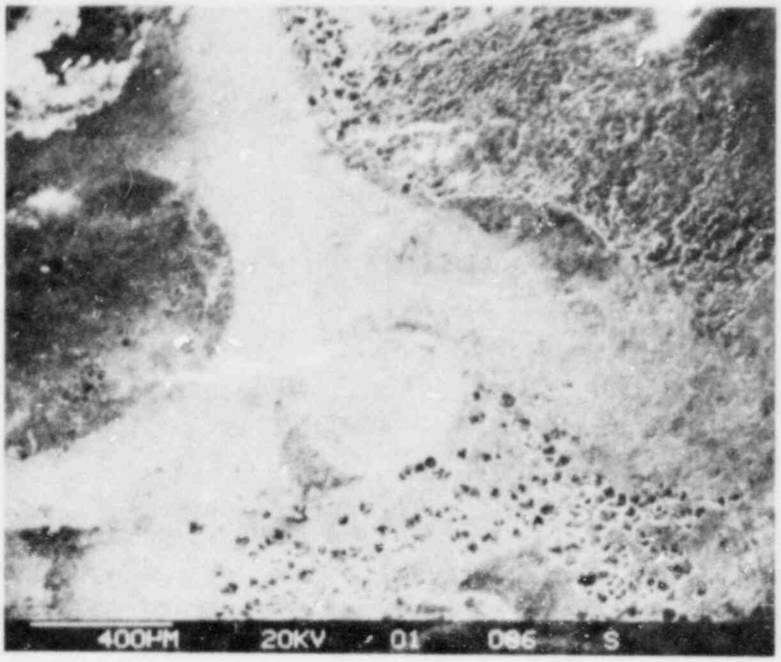

Figure 15a. SEM Micrograph of the Surface of Sample Q (50X).

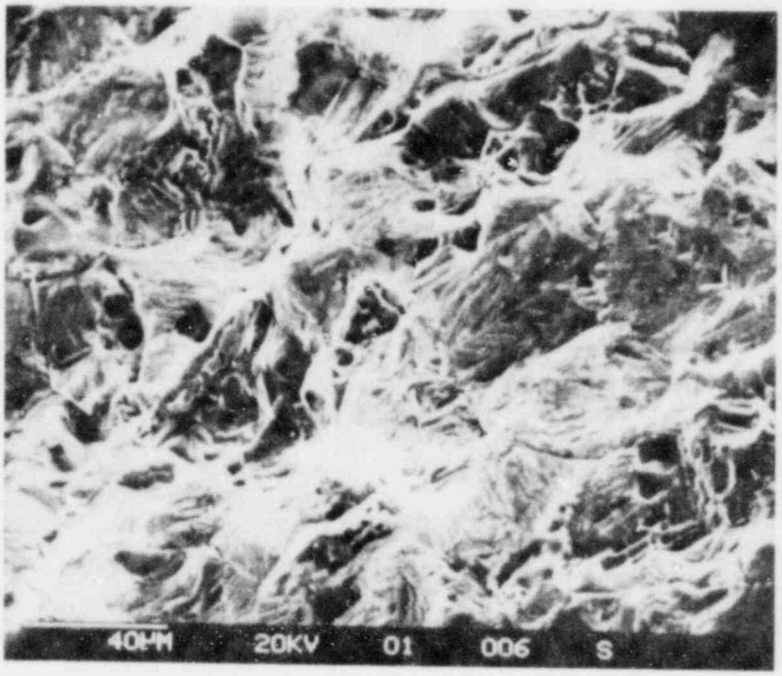

Figure 15b. SEM Micrograph of the Surface of Sample Q (500X).

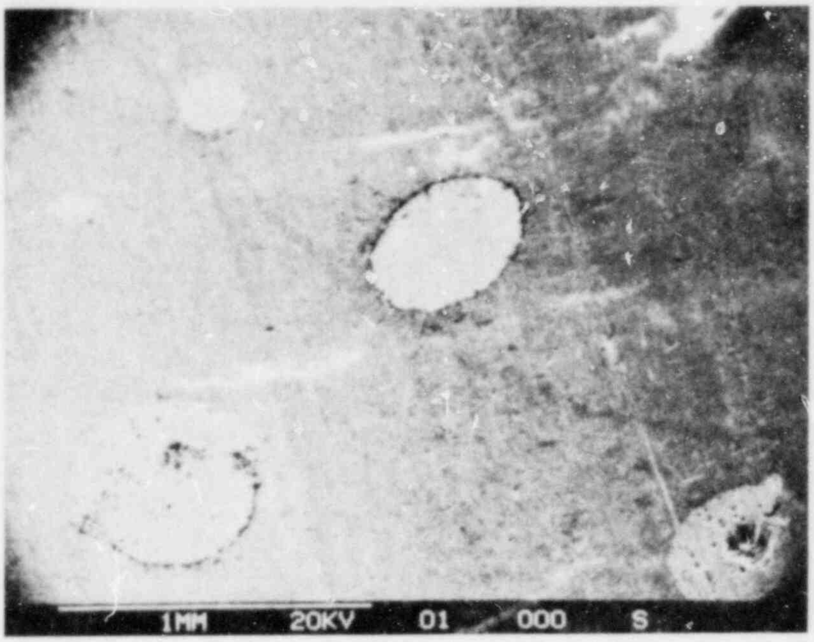

Figure 16a. SEM Micrograph of the Surface of Sampel T (42X).

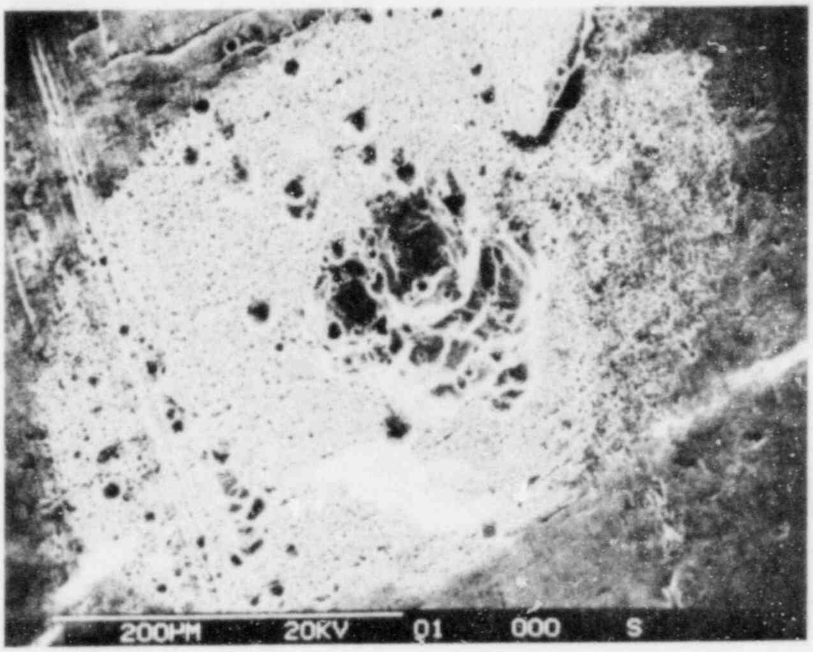

Figure 16b. SEM Micrograph of the Surface of Sample T (235X).

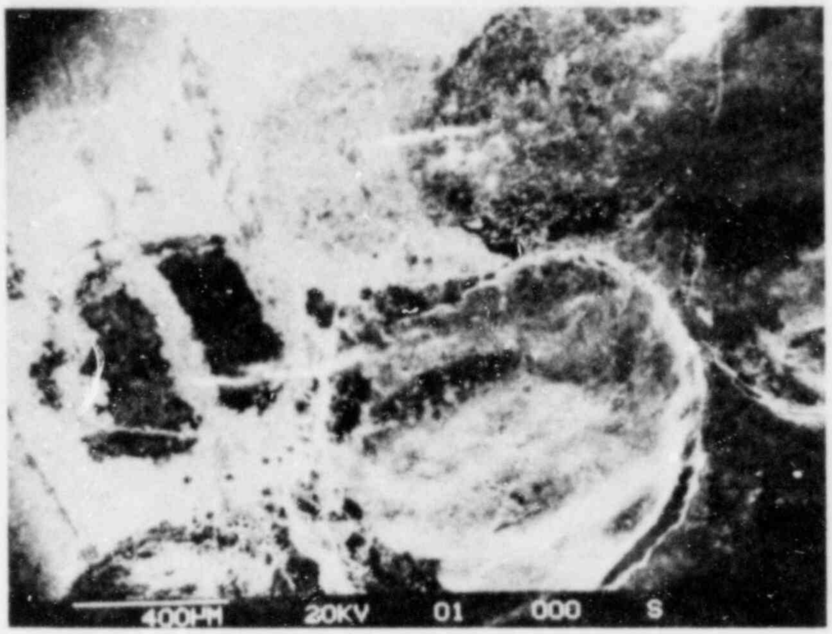

Figure 17a. SEM Micrograph of the Surface of Sample U (50X).

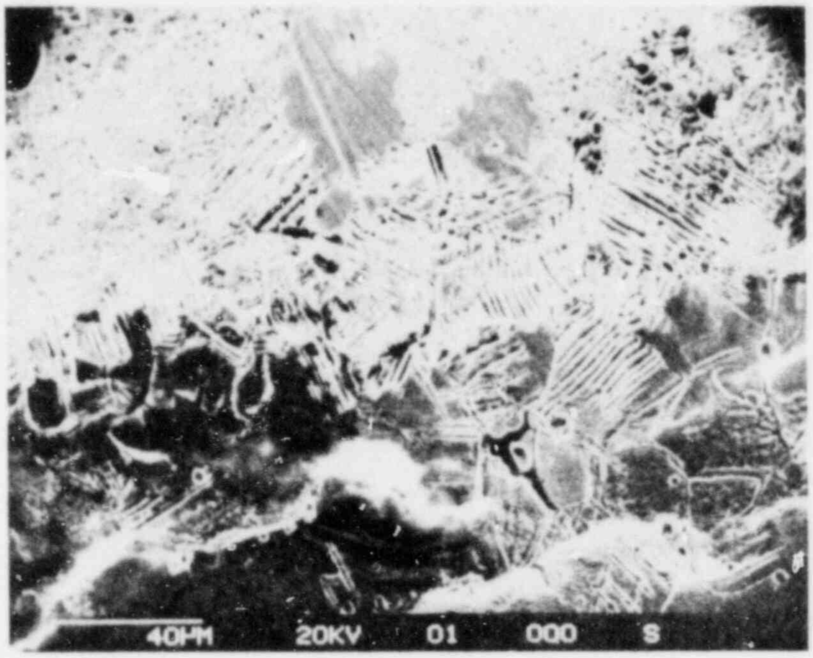

Figure 17b. SEM Mcirograph of the Surface of Sample U (500X).

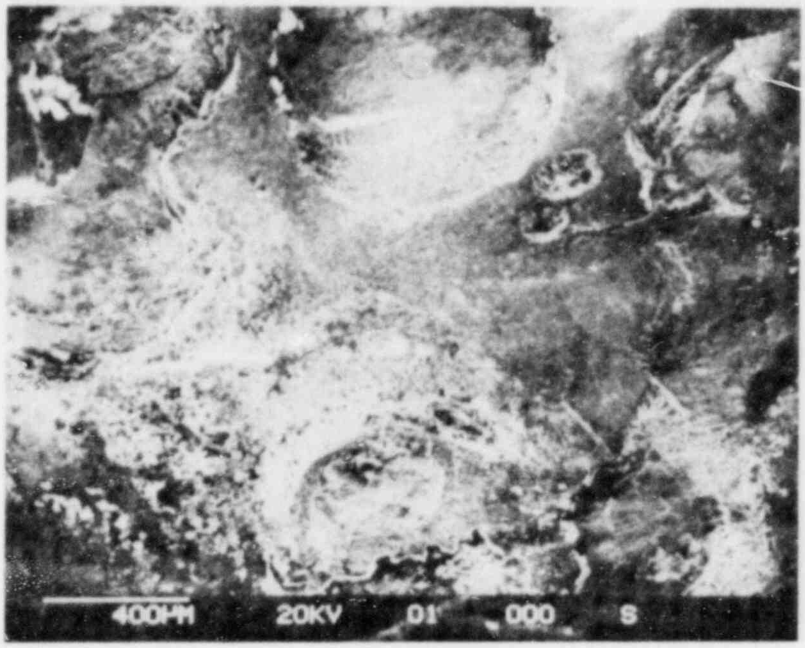

Figure 18a. SEM Micrograph of the Surface of Sample W (50X).

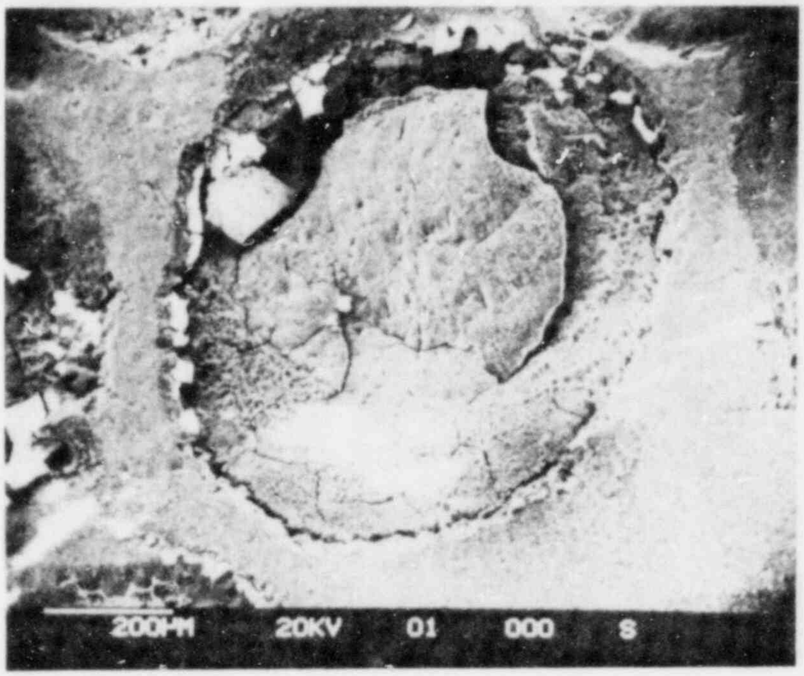

Figure 18b. SEM Micrograph of the Surface of Sample W (85X).

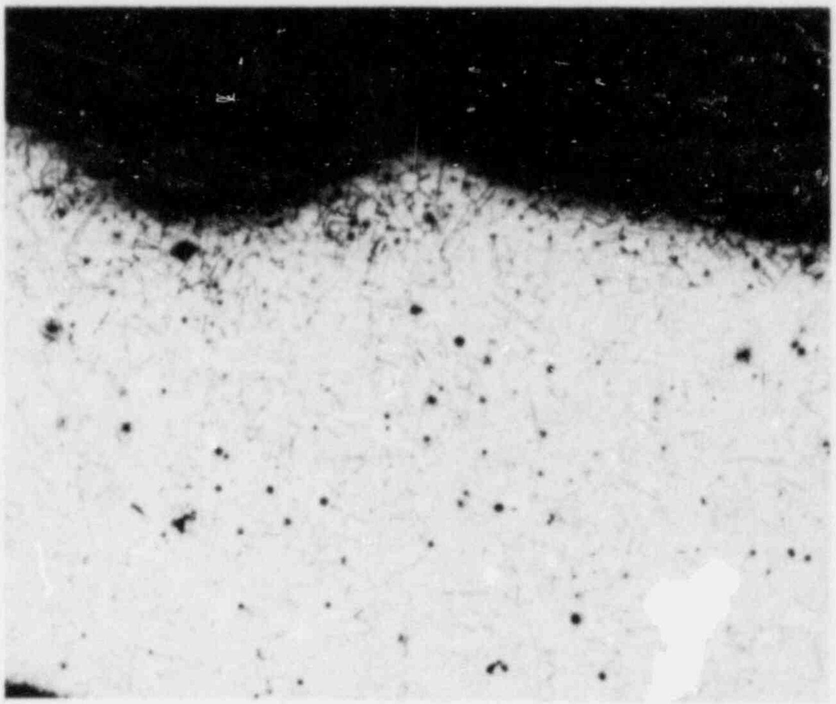

Figure 19a. Etched Radial Metallographic Section of Sample W (100X).

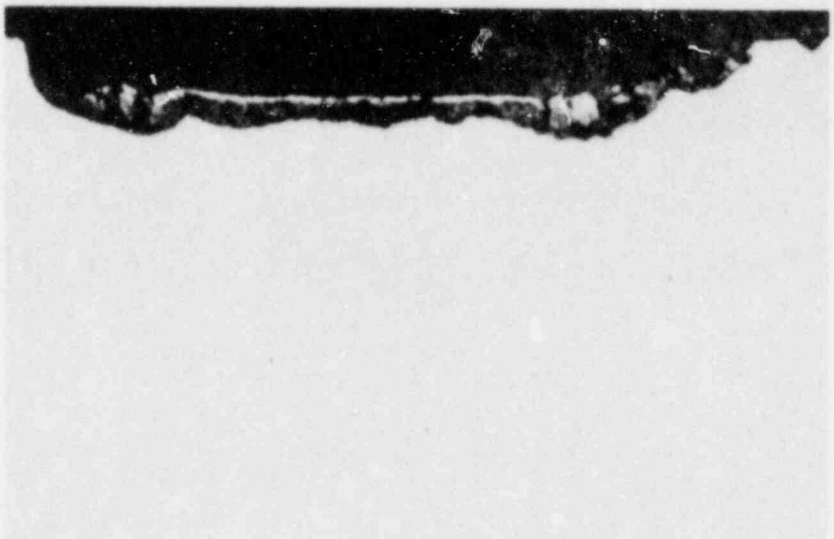

Figure 19b. Unetched Radial Metallographic Section of Sample W (100X).

APPENDIX B - CALCULATION OF DOSE TO BURIED ADSORBENT MATERIAL

Container dimensions - h=136 cm, r=30.5 cm

Resin Load/Container - $10 ft^3 = 2.84 \times 10^5 cm^3$

Assumed Activity Loading (per container)

420 Ci	Cs-137
50 Ci	Cs-134
70 Ci	Sr-90
120 Ci	Ce-144

Beta Dose

Dose Rate =

$$\frac{(Q\ Ci)\ (3.7.10^{10} dps/Ci)\ (\overline{E}\ MeV)\ (1.6 \times 10^{-6} ergs/MeV)\ (3600\ Sec/hr)}{(W gms)\ (100\ ergs/gm/rad)}$$

Assume:

Resin Density = 1gm/cc, or 2.84×10^5 gms Resin per Container

Dose Rate From Cs-137 β

β_1 1.18 MeV (6.0%)

β_2 0.514 MeV (93.5%)

0.557 MeV Average Maximum Energy (Weighted Average)
0.186 MeV Average Beta Energy (1/3 X 0.557)

$$DR = \frac{(420 Ci)\ (3.7 \times 10^{10} dps)\ (0.186\ MeV)\ (1.6 \times 10^{-6} ergs/MeV)\ (3600 sec/hr)}{(2.84 \times 10^5\ gms)\ (100\ ergs/gm/rad)}$$

= 586 Rads/hr

$\lambda = 0.0231\ yr^{-1}$ or $6.326 \times 10^{-5} day^{-1}$

$$\text{Ten Year Dose } D_{10} = \frac{(586\text{Rads/Hr})\ (24\text{hr/day})\ (1-e^{-\lambda 3652.5})}{6.326 \text{ X } 10^{-5}\text{day}^{-1}} = \underline{4.50 \text{ X } 10^{7} \text{ Rads}}$$

$$D\infty = \frac{(586 \text{ Rads/Hr})\ (24\text{hrs/day})}{6.326 \text{ X } 10^{-5}} = \underline{2.22 \text{ X } 10^{8} \text{ Rads}}$$

Dose Rate From Cs-134β

β_1 0.662 MeV (71%)

β_2 0.089 MeV (28%)

0.495 MeV Average Maximum Energy (Weighted Average)
0.165 MeV Average Beta Energy (1/3 X 0.495)

DR = (50) (0.165) (7.50) = 62Rads/hr

$\lambda = 9.257 \text{ X } 10^{-4}\text{day}^{-1}$

$D_{10} = (62)\ (24)\ (1-e^{-\lambda 3652.5}) = \underline{1.55 \text{ X } 10^{6}\text{Rads}}$

$$D\infty = \frac{(62)\ (24)}{9.257 \text{ X } 10^{-4}} = \underline{1.6 \text{ X } 10^{6}\text{Rads}}$$

Dose Rate From Sr-90β

β_1 0.546 MeV (100%)

β_2 2.27 MeV (100%)

2.82 MeV Maximum Beta Energy

0.94 MeV Average Beta Energy

DR = (70) (.94) (7.50) = 494 Rads/hr

$D_{10} = (494)\ (24)\ (1-e^{-\lambda 3652.5}) = \underline{3.834 \text{ X } 10^{7} \text{ Rads}}$

$$D_\infty = \frac{(494)\ (24)}{6.778 \times 10^{-5}} = \underline{1.75 \times 10^{8} \text{Rads}}$$

Dose Rate From Ce-144β

β_1 0.33 MeV (75%)

β_2 0.16 MeV (25%)

β 2.996 MeV (98%)

Daughter

3.284 MeV Maximum Average Energy

1.10 MeV Average Beta Energy

$DR = (120Ci)\ (1.10)\ (7.50) = \underline{990 \text{ Rads/hr}}$

$\lambda = 2.44 \times 10^{-3} \text{day}^{-1}$

$D_{10} = (990)\ (24)\ (1-e^{-\lambda 3652.5}) = \underline{9.73 \times 10^{6} \text{Rads}}$

$$D_\infty = \frac{(990)\ (24)}{2.44 \times 10^{-3}} = \underline{9.74 \times 10^{6} \text{Rads}}$$

Total Beta Dose

$D_{10} = 4.60 \times 10^{7} + 1.55 \times 10^{6} + 3.83 \times 10^{7} + 9.73 \times 10^{6} = \underline{9.56 \times 10^{7} \text{Rads}}$

$D_\infty = 2.22 \times 10^{8} + 1.6 \times 10^{6} + 1.75 \times 10^{8} + 9.74 \times 10^{6} = \underline{4.08 \times 10^{8} \text{Rads}}$

Gamma Dose

Dose Rate = $C\ \Gamma\ g$ Rads/hr, Where

$C = m \ Ci/Cm^3$

Γ = gamma ray constant

g = average geometry factor

Cylinder $r = 30.5$ cm, $h = 136$ cm; $g \sim 150$ assumes tissue equivalent.

Dose Rate From Cs-137γ

$\Gamma = 3.3$ Rads Cm^2/hr - mCi

$$DR = \frac{(3.3)\ (4.2 \times 10^5 \text{ mCi})\ (150)}{2.84 \times 10^5 \text{Cm}^3} = 832 \text{ Rads/hr}$$

$D_{10} = 5.74 \times 10^7$ Rads

$D_\infty = 2.80 \times 10^8$ Rads

Dose Rate From Cs-134γ

$\Gamma = 8.7$

$$DR = \frac{(5 \times 10^4 \text{ mCi})\ (817)\ (150)}{2.84 \times 10^5 \text{Cm}^3} = 230 \text{ Rads/hr}$$

$D_{10} = 5.75 \times 10^6$ Rads

$D_\infty = 5.95 \times 10^6$ Rads

Dose Rate From Sr-90 - Gamma Dose Is Zero

Dose Rate From Ce-144γ

$\Gamma = 0.4$

$$DR = \frac{(1.2 \times 10^5 \text{ mCi})\ (0.4)\ (150)}{2.84 \times 10^5 \text{Cm}^3} = 25.4 \text{ Rads/hr}$$

$D_{10} = 2.49 \times 10^5$ Rads

$D_\infty = 2.52 \times 10^5$ Rads

Total Gamma Dose

$D_{10} = 5.74 \times 10^{7} + 5.75 \times 10^{6} + 0 + 2.49 \times 10^{5} = \underline{6.34 \times 10^{7} \text{Rads}}$

$D_{\infty} = 2.80 \times 10^{8} + 5.59 \times 10^{6} + 0 + 2.52 \times 10^{5} = \underline{2.86 \times 10^{8} \text{Rads}}$

Total Dose - Beta and Gamma

$D_{10} = D_{10}\beta_{\text{total}} + D_{10}\gamma_{\text{total}}$

$= 9.56 \times 10^{7} + 6.34 \times 10^{7} = \underline{1.59 \times 10^{8} \text{Rads}}$

$D_{\infty} = D_{\infty}\beta_{\text{total}} + D_{\infty}\gamma_{\text{total}}$

$= 4.08 \times 10^{8} + 2.86 \times 10^{8} = \underline{6.94 \times 10^{8} \text{Rads}}$

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