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PROPERTIES OF RADIOACTIVE WASTES AND WASTE CONTAINERS

QUARTERLY PROGRESS REPORT APRIL-JUNE 1982

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PROPERTIES OF RADIOACTIVE WASTES AND WASTE CONTAINERS

QUARTERLY PROGRESS REPORT April-June 1982

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SUMMARY

Data presented in the previous quarterly progress report indicated that sand is not inert relative to the radiotracers under investigation (¹³⁷Cs, ⁸⁵Sr, and ⁶⁰Co), and was replaced with high density polyethylene beads (HDPE beads). In this report, comparative characterization data on sand and HDPE beads are presented. Water percolation rate, particle size distribution, packing characteristics, total porosity, hydraulic conductivity, and water permeability were determined for the two media. In addition, initial leach data on waste forms emplaced in HDPE beads are reported.

The following observations were made:

- Water percolation rates were slower for the first pass of water through sand-filled systems and faster for subsequent passes (approx. twice as fast). Approximately 52% of the water was retained in the sand after the first pass (1100 mL retained out of 2100 mL introduced into the system).
- For HPDE-filled systems, flow rates were faster for the first pass of water and slightly slower for subsequent passes. Approximately 3% of the water was retained in the HDPE after the first pass (60 mL retained out of 2100 mL introduced into the system).
- Particle size distribution measurements indicate that 90% of the HDPE beads were between 2 to 4 mm in diameter and 98% of the sand grains were between 0.6 and 0.8 mm.
- The ratios of bulk densities to particle densities of both materials were comparable (0.62 for sand and 0.65 for HDPE beads), indicating similar interparticle void space. The total porosities of the two media were 38 and 35 for sand and HDPE beads, respectively.
- Hydraulic conductivity and water permeability of sand were comparable to the reported values in the literature. The values for HDPE beads were within the range reported for sand.

Work performed during this quarter on leaching waste forms in an inert medium is presented in this report. Leach data from this study will be compared with data for similar forms without the inert medium. This comparison will provide information on the effect of partial saturation and drying cycles on the waste form leachability. In turn, this information will provide a more realistic predictive data basis for the leach behavior of waste forms in shallow-land burial sites.

PROPERTIES OF RADIOACTIVE WASTES AND WASTE CONTAINERS QUARTERLY PROGRESS REPORT - APRIL-JUNE 1982

1. Introduction

This report summarizes work performed during this quarter on leaching waste forms in an inert environment. The sand medium was replaced with HDPE beads which are inert relative to the leached radiotracers. Water percolation rates, particle size distribution, and packing characteristics of the two media were determined to evaluate the difference between their water retention characteristics. In addition, preliminary leach data from waste forms emplaced in HDPE beads are presented. The effects of different wet/dry cycles on the leachability of 137Cs and 85Sr from resin/cement composites and of 137Cs, 85Sr, and 60Co from unsolidified resins are being investigated.

2. Comparative Evaluation of Sand and HDPE Beads Media

2.1 Water Percolation Characteristics

Water percolation rates were measured for both sand and HDPE beads leaching columns. The dimensions of the columns were described in the previous quarterly progress report (Morcos and Dayal, 1982). Measurements were performed in triplicate. The volume of water introduced into each system was 2100 mL. Rates were slower for the first pass of water through the sand system, and faster for subsequent passes. For the HDPE beads system, however, the rates were faster for the first pass of water and slightly slower for subsequent passes (Figures 1 and 2). During the first pass, the sand system retained approximately 1100 mL of water after 3 hours of draining under gravity. whereas the HDPE system retained approximately 60 mL after 4.5 min of draining. These effects can be attributed to the different wetting properties of the media and the differences in their total exposed surface areas. When suction was applied to the sand system at the end of each drain period, the volume of residual water was approximately 80 mL. Suction was not applied at the end of each leaching period to the HDPE system, but they were allowed to drain under gravity for 3-5 min.

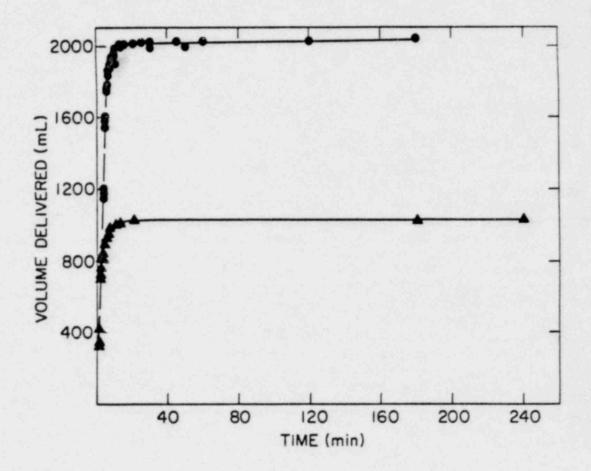


Figure 1

Delivered volume vs time for a sand-filled leaching system consisting of a 2-in.-diam x 4-in.-high waste form surrounded by a 2-in.-thick layer of sand (5700 g of sand). The lower curve (**(**) represents the average of three determinations for the first elution. The upper curve (**()** represents the average data of three determinations for five subsequent elutions. The upper limit of the ordinate represents volume input into column (2100 mL).

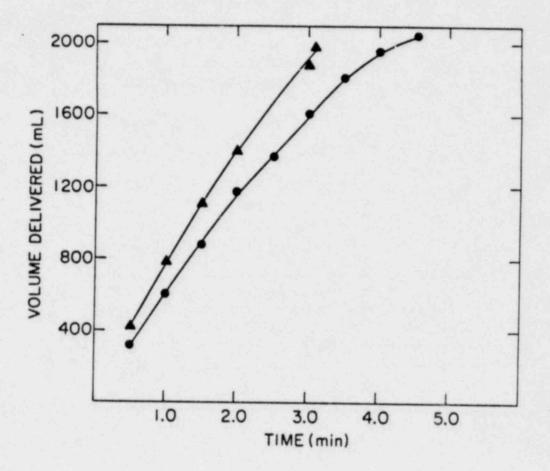


Figure 2 Delivered volume vs time for a HDPE bead-filled leaching system consisting of a 2-in.-diam x 4-in.-high waste form surrounded by a 2-in.-thick layer of HDPE beads (2200 g of HDPE beads). The upper curve (A) represents the average data of three determinations for the first elution. The lower curve (•) represents the average data of three determinations for the subsequent (second) elution. The upper limit of the ordinate represents volume input into column (2100 mL).

2.2 Particle Size Distributions of Filler Media

Particle size distributions of sand and of HDPE beads were determined using standard sieves ranging from mesh numbers 5 to 35. Results are summarized in Table 1.

Table 1

Particle Size Distributions of Sand and of HDPE Beads

Particle S (mm)	ize	Weight Distributio HDPE ^a	on in Percent Sand ^b
>4.8		1.3	
4.8-4.0		7	
4.0-3.4		20	
3.4-2.0		70	
2.0-1.2		1.4	
<1.2		0.3	0.02
1.0-0.8			1.5
0.8-0.6			98.0
0.6-0.5			0.22
<0.5			0.32

bOne determination using 1 kg of sand.

Figure 3 shows a comparison between the sizes of HDPE beads and sand grains. The HDPE beads exhibit a platy habit.

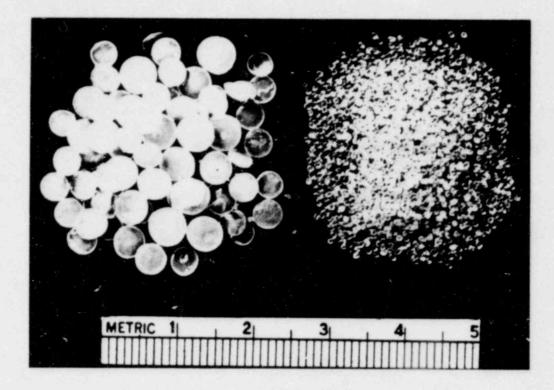


Figure 3 Visual comparison of HDPE beads (left) and sand (right) used in the experiment. Note the platy habit of the HDPE beads.

2.3 Packing Characteristics and Total Porosity of Sand and HDPF Beads

The ratio (D_b/D_p) of the bulk density (D_b) of the filler medium to its particle density (D_p) was used to compare the volume of the interstitial void space in a sand column to that in a HDPE beads column. The bulk density is defined as the density of the packed material; particle density is the density of the solid material. The closer the value of D_b/D_p is to one, the smaller is the interstitial space. Table 2 summarizes the bulk and particle densities for the two filler media and their D_b/D_p .

Table 2

Bulk and Particle Densities of Packed Sand and HDPE Beads

Filler Material	$D_b (g/cm^3)a$	L_p (g/cm ³)	D _b /D _p
Sand	1.66+2%	2.65	0.62
HDPE Beads	0.63+3%	0.96	0.65

^aAverage of three determinations.

The bulk densities were calculated as follows: Mass of the filler material required to fill a known volume (920 cc) divided by the mass of the material used to fill the volume. The method of filling the known volume was similar to that used in filling the leaching columns.

The total porosity (S_t) is defined as $100(1-D_b/D_p)$. St for sand and HDPE beads were calculated to be 38 and 35 percent, respectively. These S, values indicate that the void volumes in the two media are comparable.

2.4 Hydraulic Conductivity of Sand and HDPE Bead Columns

The hydraulic conductivity and water permeability for sand and HDPE beads were determined according to the method outlined by A. Klute (A. Black, Editor-in-Chief) for saturated soils. The hydraulic conductivities were calculated using the relationship:

(1)

(2)

 $K = (Q/At)(L/\Delta H)$

where

- K = hydraulic conductivity
 - Q = volume of water that passes through the soil sample within time t
 - A = cross sectional area of the soil column
 - L = length of the soil column
 - AH = hydraulic head difference.

The permeability of sand and HDPE bead columns were calculated using the relationship:

$$K' = \frac{\eta}{\rho g} R$$

where

K' = permeability of the material to water, cm²

n = viscosity of water at the experimental temperature, dyne sec/cm² (poises)

- ρ = density of water, g/cm³
- $g = acceleration of gravity, cm/sec^2$
- K = hydraulic conductivity, cm/sec.

The results are summarized in Table 3.

Medium	Hydraulic Conductivity ^a (cm/sec)	Permeability to Water (cm ²)
Sand	6.65 x 10 ⁻²	7.72 x 10 ⁻⁷
HDPE Beads	1.19 x 10 ⁻¹	1.39 x 10 ⁻⁶

Hydraulic Conductivity and Permeability of Sand and HDPE

Reported values (Davis and DeVriest, 1966) of the hydraulic conductivity range from 10^{-3} to 1 cm/sec for clean sand. The corresponding reported water permeability values are 9.9×10^{-9} to 9.9×10^{-6} cm².

The values shown in Table 3 for sand agree with the reported values and those for HDPE beads indicate that the hydraulic conductivity and permeability of HDPE beads are within the range of sand.

3. Leach Data for Waste Form in HDPE Bead-Filled Columns

Waste form preparation and experimental setup for leach studies were outlined in the previous quarterly report. The sand medium was replaced with HDPE beads based on information presented earlier (Morcos and Dayal, 1982). Preliminary leach data for waste forms emplaced in HDPE and extending over a 19-day period are presented below.

3.1 Leaching Regimens for Waste Forms Emplaced in HDPE Beads

Two leaching regimens were followed for resin/cement composites. In the first regimen, a constant one-day dry period was maintained between varying wet (saturation) periods. In the second regimen, a constant saturation period was maintained between varying dry periods. Each regimen was followed on three sets of identical waste forms. Each set was exposed to fixed wet and dry cycles and consisted of triplicate waste forms. Diagrams of the leaching regimens are shown in Figures 4 and 5. Deionized water was used as the leach-ant (2100 mL per leaching period).

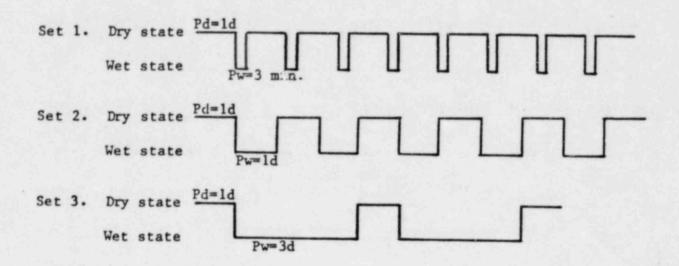
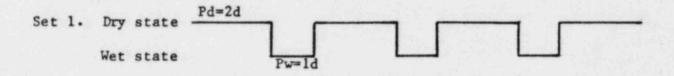


Figure 4 Leaching Regimen 1: A constant one-day dry period is maintained for three different wet periods (Regimen 1); Pd = dry period and Pw = wet period.



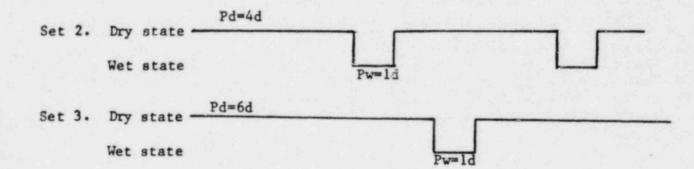


Figure 5 Leaching Regimen 2: A constant one-day wet period is maintained for three different dry periods (Regimen 2); Pd = dry period and Pw = wet period.

Leach data using regimen 1 are presented in Appendix A (Tables A.1, A.2, and A.3); and those based on regimen 2, set 1 are summarized in Appendix B (Tables B.1 and B.2). Data from regimen 2, sets 2 and 3 will be available the next reporting period. The errors quoted represent only the statistical errors associated with the counting. These data are also graphically shown in Figures 6 through 17 for regimen 1 and Figures 18 and 21 for set 1 of regimen 2. Each pair of figures shown on a page represents leaching data of three replicate samples and the average cumulative fractional release (CFR) of the three replicates. The average CFR curves have been normalized for volume to geometric surface area. The CFR data are plotted vs t1/2 (total experimental time)1/2.

3.1.2 Unsolidified Resins

Leaching of unsolidified resins in HDPE medium was initiated according to regimen 1 (Figure 4). Deionized water and simulated Barnwell trench water were used as leachants. The chemical composition of simulated Barnwell groundwater (Czyscinski and Weiss, 1981) is summarized in Table 4.

Leaching resins in sand with deionized water showed no measurable leached radioactivity above background over a period of 14 days. These experiments have been discontinued. Leach data using simulated Barnwell trench water will be included in the next quarterly report.

Table 4

Cations	Concentration mmoles/L	Anions	Concentration mmoles/L
Na+1	1.34	HCO3-1	1.10
Na ⁺¹ K ⁺¹	0.075	S04=2	0.48
NH4 ⁺¹ Ca ⁺² Mg ⁺² Mn ⁺²	0.26	c1 ²¹	0.38
Ca ⁺²	0.35	NO3-1	0.30
Mg+2	0.060	-	
Mn+2	0.013		

Composition of Synthetic Barnwell Trench Water^a

 $SiO_2 = 0.12 \text{ mmoles/Lb}$

^aThe composition simulates water from trench sump 6D1 at Barnwell (Czyscinski and Weiss, 1981). The pH of the solution is adjusted to 6.0 with hydrochloric acid.

bAdded as sodium metasilicate Na2SiO3.9H2O.

(Continued on Page 18)

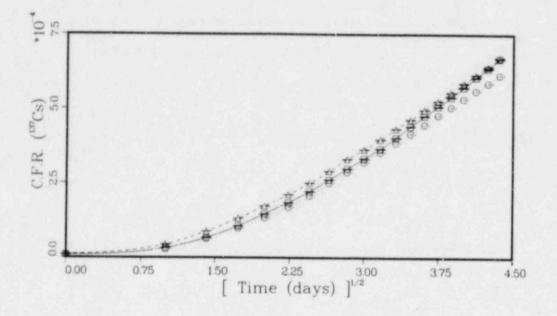


Figure 6 137Cs CFR vs (time)1/2 for resin/cement composites placed in sand medium. The leaching conditions were: 3-5 min saturation and 1 day dry periods (w/c=0.6).

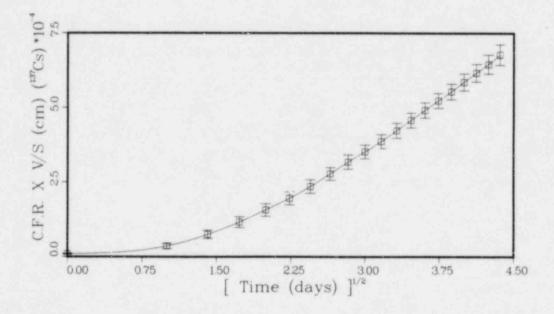


Figure 7 Volume/surface area normalized average CFR of 137Cs vs (time)1/2 ior resin/cement composites placed in sand medium. The leaching conditions were: 3-5 min saturation and 1 day dry periods (w/c=0.6; V/S=1.04 cm).

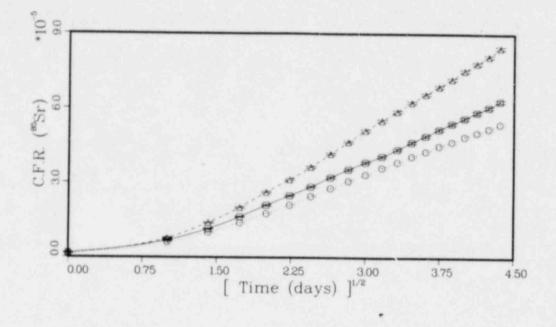


Figure 8 85Sr CFR vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 3-5 min saturation and 1 day dry periods (w/c=0.6).

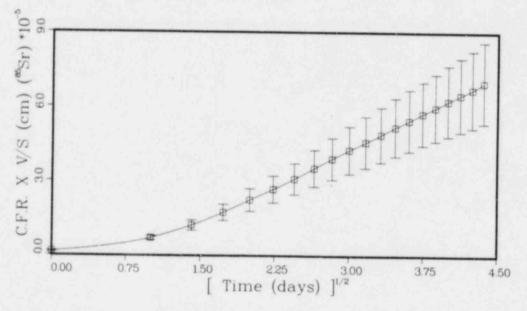


Figure 9 Volume/surface area normalized average CFR of ⁸⁵Sr vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 3-5 min saturation and 1 day dry periods (w/c=0.6; V/S=1.04 cm).

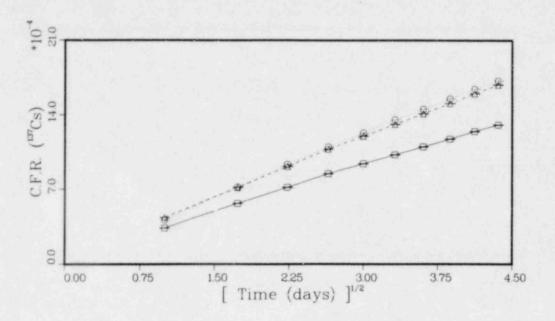


Figure 10 137Cs CFR vs (time)1/2 for resin/cement composites placed in sard medium. The leaching conditions were: 1 day saturation and 1 day dry periods (w/c=0.6).

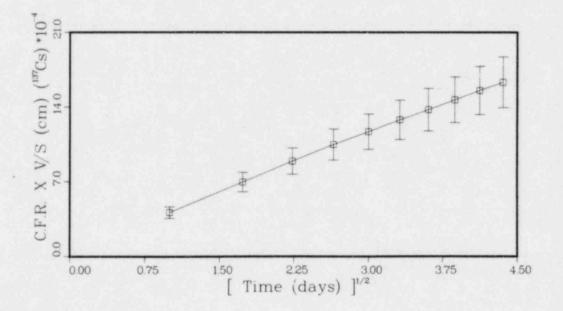


Figure 11 Volume/surface area normalized average CFR of 137Cs vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 1 day dry periods (w/c=0.6; V/S=1.04 cm).

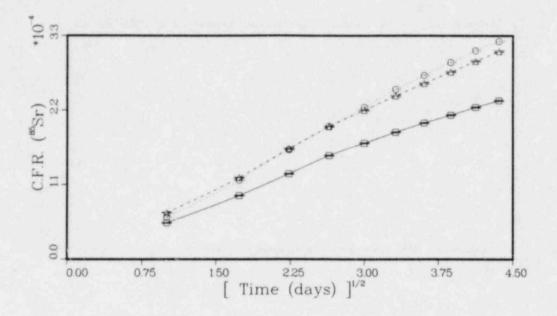


Figure 12 ⁸⁵Sr CFR vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 1 day dry periods (w/c=0.6).

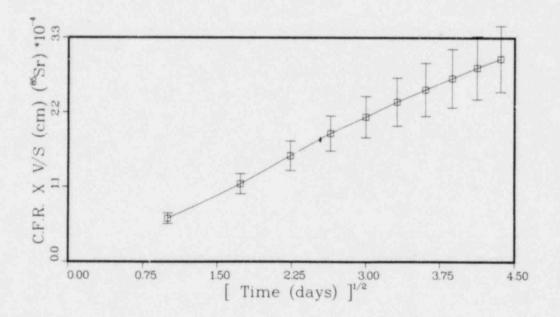


Figure 13 Volume/surface area normalized average CFR of ⁸⁵Sr vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 1 day dry periods (w/c=0.6; V/S=1.04 cm).

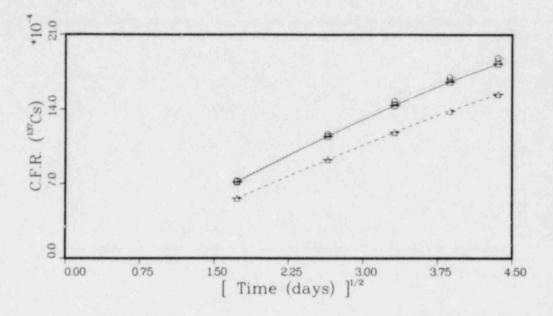


Figure 14 137Cs vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 3 days saturation and 1 day dry periods (w/c=0.6).

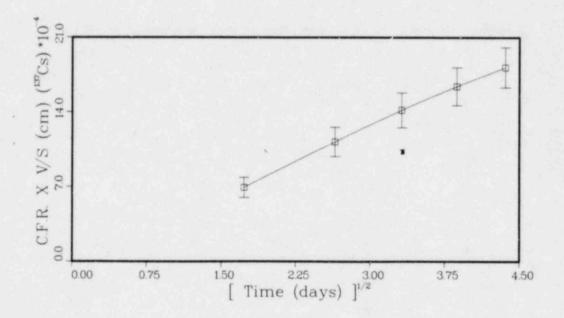


Figure 15 Volume/surface area normalized average CFR of 137Cs vs $(time)^{1/2}$ for resin/cement composites placed in sand medium. The leaching conditions were 3 days saturation and 1 day dry periods (w/c=0.6; V/S=1.04 cm).

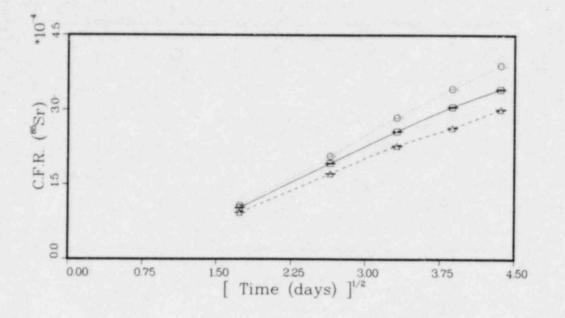


Figure 16 ⁸⁵Sr vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 3 days saturation and 1 day dry periods (w/c=0.6).

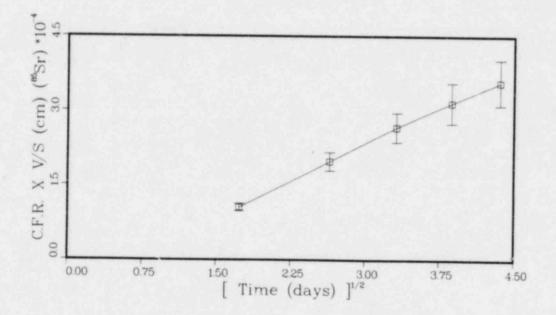


Figure 17 Volume/surface area normalized average CFR of ⁸⁵Sr vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were 3 days saturation and 1 day dry periods (w/c=0.6; V/S=1.04 cm).

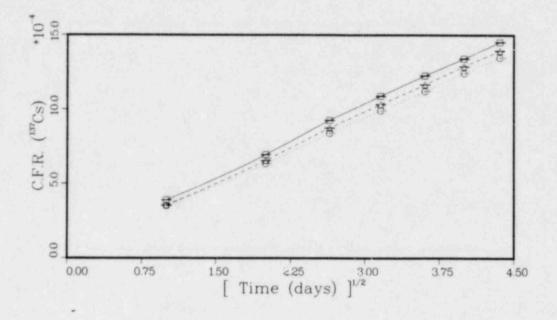


Figure 18 ¹³⁷Cs CFR vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 2 days dry periods (w/c=0.6).

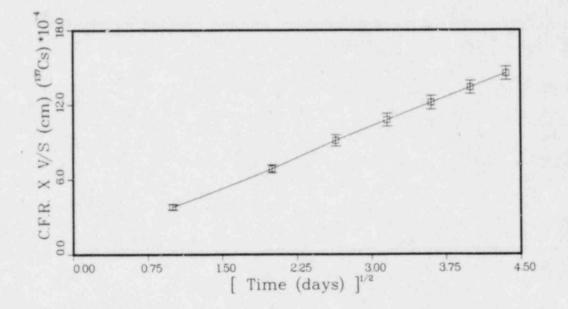


Figure 19 Volume/surface area normalized average CFR of 137Cs vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 2 days dry periods (w/c=0.6; V/S=1.04 cm).

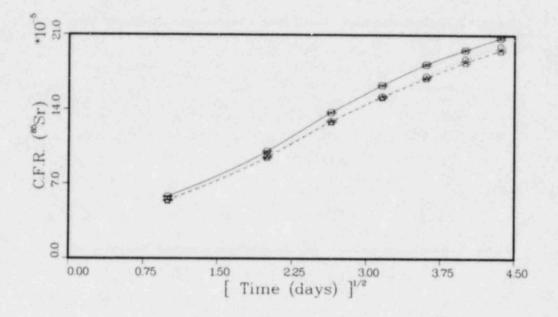


Figure 20 ⁸⁵Sr CFR vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 2 days dry periods (w/c=0.6).

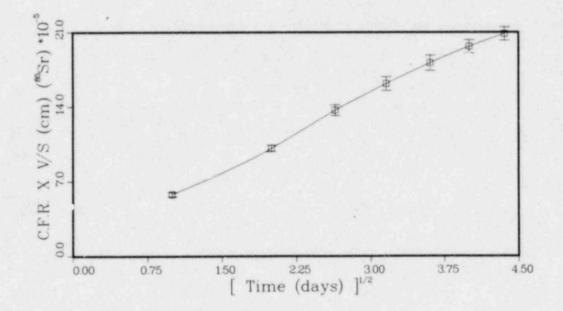


Figure 21 Volume/surface area normalized average CFR of ⁸⁵Sr vs (time)^{1/2} for resin/cement composites placed in sand medium. The leaching conditions were: 1 day saturation and 2 days dry periods (w/c=0.6; V/S=1.04 cm).

4. Current Progress

We are now reducing the leach data presented above to compare it with leach data for similar waste forms without the inert medium. This comparison will provide information on the effect of partial saturation and drying cycles on the waste form leachability. In comparison to earlier experiments in which complete saturation conditions were maintained continuously, we believe that short-term drying periods in the present experiments might allow diffusive migration of the radionuclides towards the surface of the waste form, thus resulting in an episodic release during the subsequent saturation period.

5. References

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APPENDIX A

Leach data for resin/cement composites leached in HDPE beads with a constant dry period (1 d) and 3 different saturation periods (3-5 min, 1 d and 3 d).

Tables A.1, A.2 and A.3 present ^{137}Cs incremental and cumulative fractional release data normalized for V/S.

Tables A.4, A.5 and A.6 present 85 Sr incremental and cumulative fractional release data normalized for V/S.

137Cs Average Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (3-5 min Saturation Periods Followed by 1 d Dry Periods)

TIME (DAYS)	AV. FRACT. REL. X V/S (CM)	AV. CUM. FRAC. REL. X V/S (CM)	ERROR
	.9641E-05	.9641E-05	.1421E-05
1.000 2.000	.2615E-04 .3894E-04	.3579E-04 .7473E-04	.8668E-05 .1391E-04
3.000	.4095E-04	.1157E-03	.1794E-04
4.000	.4077E-04	.1565E-03	.2092E-04
5.000	.3914E-04	.1956E-03	.2192E-04
6.000	.3958E-04	.2352E-03	.2269E-04
7.000	.4254E-04	.2777E-03	.2108E-04
8.000	.3974E-04	.3175E-03	.2355E-04
9.000	.3447E-04	.3519E-03	.2298E-04
10.00	.3449E-04	.3864E-03	.2315E-04
11.00	.3640E-04	.4228E-03	.2494E-04
12.00	.3455E-04	.4574E-03	.2479E-04
13.00	.3380E-04	.4912E-03	.2550E-04
14.00	.3209E-04	.5233E-03	.2477E-04
15.00	.3041E-04	.5537E-03	.2622E-04
16.00	.3219E-04	.5859E-03	.2825E-04
17.00	.3070E-04	.6166E-03	.2963E-04
18.00	.2901E-04	.6456E-03	.3183E-04
19.00	.3188E-04	.6775E-03	.3439E-04

137Cs Average Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (1 d Saturation Periods Followed by 1 d Dry Periods)

TIME (DAYS)	AV. FRACT. REL. X V/S (CM)	AV. CUM. FRAC. REL. X V/S (CM)	ERROR
1.000	.4119E-03	.4119E-03	.5363E-04
3.000	.2833E-03	.6952E-03	.9057E-04
5.000	.1954E-03	.8906E-03	.1231E-03
7.000	.1560E-03	.1047E-02	.1451E-03
9.000	.1199E-03	.1167E-02	.1651E-03
11.00	.1113E-03	.1278E-02	.1845E-03
13.00	.9567E-04	.1373E-02	.1993E-03
15.00	.9225E-04	.1466E-02	.2141E-03
17.00	.8825E-04	.1554E-02	.2265E-03
19.00	.7712E-04	.1631E-02	.2382E-03

Table A.3

137Cs Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (3 d Saturation Periods Followed by 1 d Dry Periods)

TIME	AV. FRACT.	AV. CUM.	ERROR
(DAYS)	REL. X V/S	FRAC. REL. X V/S (CM)	
		× •/3 (CH/	
3.000	.6914E-03	.6914E-03	.9369E-04
7.000	.4299E-03	.1121E-02	.1376E-03
11.00	.2953E-03	.1417E-02	.1630E-03
15.00	.2233E-03	.1640E-02	.1774E-03
19.00	.1788E-03	.1819E-02	.1879E-03

⁸⁵Sr Average Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (3-5 min Saturation Periods Followed by 1 d Dry Periods)

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TIME (DAYS)	AV. FRACT. REL. X V/S (CM)	AV. CUM. FRAC. REL. X V/S (CM)	ERROR
•	.1562E-05	.1562E-05	.2485E-06
1.000	.5489E-05	.7051E-05	.8561E-06
2.000	.5123E-05	.1217E-04	.2058E-05
3.000 -	.5137E-05	.1731E-04	.3226E-05
4.000	.5033E-05	.2234E-04	.4513E-05
5.000	.4341E-05	.2668E-04	.5371E-05
6.000	.4197E-05	.3088E-04	.6256E-05
7.000	.4299E-05	.3518E-04	.7347E-05
8.000	.3846E-05	.3903E-04	.8397E-05
9.000	.3504E-05	.4253E-04	.9540E-05
10.00	.3164E-05	.4569E-04	.1034E-04
11.00	.3043E-05	.4874E-04	.1106E-04
12.00	.3037E-05	.5178E-04	.1173E-04
13.00	.2712E-05	.5449E-04	.1240E-04
14.00	.2769E-05	.5726E-04	.1280E-04
15.00	.2451E-05	.5971E-04	.1347E-04
16.00	.2651E-05	.6236E-04	.1425E-04
17.00	.2444E-05	.6480E-04	.1488E-04
18.00	.2245E-05	.6705E-04	.1554E-04
19.00	.2739E-05	.6979E-04	.1635E-04

⁸⁵Sr Average Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (1 d Saturation Periods Followed by 1 d Dry Periods)

TIME (DAYS)	AV. FRACT. REL. X V/S (CM)	AV. CUM. FRAC. REL. X V/S (CM)	ERROR
1.000	.6380E-04	.6380E-04	.7893E-05
3.000	·5066E-04	.1145E-03	.1483E-04
5.000	+4143E-04	+1559E-03	.2180E-04
7.000	+3290E-04	.1888E-03	.2575E-04
9.000	.2442E-04	.2132E-03	.3039E-04
11.00	.2203E-04	.2352E-03	.3541E-04
13.00	.1816E-04	.2534E-03	.3907E-04
15.00	.1660E-04	.2700E-03	.4309E-04
17.00	.1566E-04	.2857E-03	.4639E-04
19.00	.1326E-04	.2989E-03	.4870E-04

Table A.6

⁸⁵Sr Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (3 d Saturation Periods Followed by 1 d Dry Periods)

TIME	AV. FRACT.	AV. CUM.	ERROR
(DAYS)	REL. X V/S (CM)	FRAC. REL. X V/S (CM)	
3.000	.1053E-03	.1053E-03	.7393E-05
7.000	.9244E-04	.1977E-03	.1831E-04
11.00	.6863E-04	.2664E-03	.2949E-04
15.00	+4925E-04	.3156E-03	.4067E-04
19.00	.4150E-04	.3571E-03	.4578E-04

APPENDIX B

Leach data for resin/cement composites leached in HDPE beads with a constant wet period (1 d) and one dry period (2 d).

Tables B.1 presents 137Cs incremental and cumulative fractional release data normalized for V/S.

Table B.2 presents 85 Sr incremental and cumulative fractional release data normalized for V/S.

Table B.1

137Cs Average Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (1 d Saturation Periods Followed by 2 d Dry Periods)

TIME (DAYS)	AV. FRACT. REL. X V/S (CM)	AV. CUM. FRAC. REL. X V/S (CM)	ERROR
1.000	.3798E-03	.3798E-03	.2171E-04
4.000	.3065E-03	.6863E-03	.3172E-04
7.000	.2267E-03	.9130E-03	.4503E-04
10.00	.1650E-03	.1078E-02	.5102E-04
13.00	.1385E-03	.1217E-02	.5500E-04
16.00	.1239E-03	.1340E-02	.5201E-04
19.00	.1117E-03	.1452E-02	.5395E-04

Table B.2

⁸⁵Sr Average Incremental and Cumulative Fractional Releases Normalized for V/S From Resin/Cement Composites Leached in HDPE Medium (1 d Saturation Periods Followed by 2 d Dry Periods)

TIME (DAYS)	AV. FRACT. REL. X V/S (CM)	AV. CUM. FRAC. REL. X V/S (CM)	ERROR
1.000 4.000 7.000 10.00 13.00 16.00 19.00	.5793E-04 .4316E-04 .3576E-04 .2487E-04 .1947E-04 .1541E-04 .1192E-04	.5793E-04 .1011E-03 .1368E-03 .1617E-03 .1812E-03 .1966E-03 .2085E-03	.2012E-05 .3075E-05 .5040E-05 .6447E-05 .7216E-05 .6190E-05 .6199E-05