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CORRELATION OF ^{137}Cs LEACHABILITY FROM SMALL-SCALE TO LARGE-SCALE WASTE FORMS

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ABSTRACT

A study correlating the leachability of ^{137}Cs from small-scale to large-scale cement forms was performed. The waste forms consisted of (a) 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 (as free water) and (b) boric acid waste (12% solution) incorporated in Portland III cement, with a waste-to-cement ratio of 0.7. ^{137}Cs was added to both waste types prior to solidification. The sample dimensions varied from 1 in. x 1 in. to 22 in. x 22 in. (diameter x height). Leach data extending over a period of 260 days were obtained using a modified IAEA leach test. A method based on semi-infinite plane source diffusion model was applied to interpret 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 was obtained.

INTRODUCTION

Present regulations promulgated by the Nuclear Regulatory Commission (NRC) require that all new commercial power reactor plants licensed have the capability to solidify their waste streams [1]. In addition, the proposed rule 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste," requires either solidification or the use of high integrity containers for the disposal of spent ion exchange resins [2]. Basic concerns in licensing radioactive waste forms and containers are their dimensional stability and the potential for release 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 and extrapolation of long-term performance of waste forms based on short-term laboratory tests.

This paper presents work performed at Brookhaven National Laboratory on the correlation of ^{137}Cs leachability from small-scale (laboratory) samples to large-scale waste forms. The waste forms evaluated in this study are typical of those expected to be generated at nuclear power plants, e.g. organic ion exchange resins and boric acid waste streams solidified in Portland cements [3,4].

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BACKGROUND

Several theoretical and empirical methods based on mass transport and diffusion theory have been developed to predict the leachability of radioisotopes from waste composites [3-12]. A method was recommended in 1970 by the International Atomic Energy Agency (IAEA) for leaching samples and for the analysis and interpretation of leach data [13]. 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 the diffusion rate. The mass transport rate equation describing this diffusion mode can be written as [5,14]:

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

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 diffusion coefficient of the substance for the particular composite matrix,

The underlying assumptions dictated by Equation (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 surface concentration of the isotope in the waste form is zero. Furthermore, the relationship in Equation (1) implies that initially for t = 0, the fraction leached (f) is also zero. However, experimental leach data deviate from this prediction for small values of t, and a more general relationship is suggested [5,15]:

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

where the term (α) represents non-diffusive contributions from the surface of the waste form [7,16]. Furthermore, a linear relationship of the term (α) with the S/V ratio of the waste form was shown to exist [16], indicating that it is indeed surface controlled.

PRESENT WORK

Experiments were conducted to determine if the ^{137}Cs leach data from simulated small-scale laboratory samples could be used in predicting the leaching behavior of larger samples. This report presents the experimental data obtained from 1 x 1 (diameter x height, in inches), 2 x 2, 2 x 4, 3 x 3, 6 x 6, 6 x 12, 12 x 12, and 22 x 22 forms incorporating organic ion exchange resins or boric acid waste solidified in Portland cements. A method is presented to correlate the ^{137}Cs leach data from small-scale samples to large-scale samples.

EXPERIMENTAL

Waste Type and Solidification

The waste form nominal dimensions (diameter x height, in inches) selected for this study were 1 x 1, 2 x 2, 2 x 4, 3 x 3, 6 x 6, 6 x 12, 12 x 12 and

22 x 22 for the organic ion exchange resins waste stream, and 1 x 1, 2 x 2, 2 x 4, 3 x 3 and 6 x 6 for the boric acid waste stream. Cesium-137 was the radioactive tracer used in both formulations because it is one of the major long-lived radioactive components of reactor waste and one of the least reactive with Portland cement.

Details of the methods used to prepare simulated spent ion exchange resin and boric acid wastes are given elsewhere [3,4]. The simulated spent resin waste consists of 33 weight percent IRN-77, Na^+, form cation exchange resin, loaded with ^{137}Cs, and 67 weight percent deionized water added as free water. Simulated boric acid waste consists of a stock solution containing 12 weight percent boric acid adjusted to pH ~12 by the addition of sodium hydroxide. A known amount of ^{137}Cs was added to the solution prior to solidification.

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. This formulation was chosen because preliminary test samples maintained their physical integrity during a prolonged immersion period (4-5 weeks) and because it provided good workability of the mixture during the mixing stage. Procedures used for preparing small and large samples were described in earlier reports [3,4]. A summary of the dimensions and compositions of the IRN-77 resin/cement waste forms is given in Table I.

Table I
Dimensions and compositions of IRN-77 resin waste forms

	Composite				Components (g)			^{137}Cs Added to Composite (μCi)
Sample Size[a] (in. x in.)	Diameter (in.)	Height (in.)	V/S (cm)	Weight (g)	Cement	Waste		
					Portland I	IRN-77[b]	H_2O	
1 x 1	0.93	0.94	0.396	20	12.5	2.5	5.0	1
	0.93	0.94	0.396	c	12.5	2.5	5.0	1
	0.93	0.94	0.396	c	12.5	2.5	5.0	1
2 x 2	1.83	1.89	0.784	150	93.8	18.8	37.5	10
	1.83	1.89	0.784	150	93.8	18.8	37.5	10
	1.83	1.89	0.784	150	93.8	18.8	37.5	10
2 x 4	1.85	3.62	0.936	290	181.3	36.3	72.5	10
	1.85	3.62	0.936	290	181.3	36.3	72.5	10
	1.85	3.62	0.936	290	181.3	36.3	72.5	10
3 x 3	3.00	3.34	1.32	734	460	92.0	184	10
	3.00	3.34	1.32	735	460	92.0	184	10
	3.00	3.34	1.32	735	460	92.0	184	10
6 x 6	6.06	5.79	2.53	c	3,250	650	1,300	500
	6.06	5.73	2.52	c	3,250	650	1,300	500
	6.06	5.71	2.51	c	3,250	650	1,300	500
6 x 12	6.00	11.6	3.03	9,620	6,139	1,228	2,456	1,000
	6.00	12.5	3.07	9,250	6,139	1,228	2,456	1,000
	6.00	11.1	3.00	9,430	6,139	1,228	2,456	1,000
12 x 12	12.5	11.5	5.14	40,000	24,900	4,990	9,980	10,000
	12.3	11.5	5.09	40,100	24,900	4,990	9,980	10,000
	12.3	11.5	5.09	39,800	24,900	4,990	9,980	10,000
22 x 22	21.5	21.5	9.10	228,340	143,700	28,740	51,480	20,000

[a] Each sample size prepared in triplicate, except 22 x 22.
[b] Rohm and Haas Amberlite organic cation exchange resin.
[c] Hot weighed.

The simulated boric acid waste solution was heated to 77°C prior to solidification in Portland III cement. The ratio of waste-to-cement was 0.7 and the nominal dimensions (in inches) of the solidified samples were 1 x 1, 2 x 2, 2 x 4, 3 x 3, and 6 x 6. The dimensions and compositions of simulated boric acid waste forms are summarized in Table II. Further details of the solidification procedure were given elsewhere [3,4].

Table II
Dimensions and compositions of boric acid waste forms

Sample Size[a] (in. x in.)	Composite				Components (g)				^{137}Cs Added to Composite (μCi)
	Diameter (in.)	Height (in.)	V/S (cm)	Weight[b] (g)	Cement	Waste			
					Portland III	H_3BO_3	NaOH	H_2O	
1 x 1	0.97	0.94	0.408	19.8	11.8	0.9	0.9	6.5	1
	0.97	0.93	0.405	19.8	11.8	0.9	0.9	6.5	1
	0.97	0.91	0.402	19.5	11.8	0.9	0.9	6.5	1
2 x 2	1.83	1.89	0.783	149	88.2	6.6	6.7	48.4	20
	1.83	1.89	0.783	150	88.2	6.6	6.7	48.4	20
	1.83	1.89	0.783	150	88.2	6.6	6.7	48.4	20
2 x 4	1.86	3.62	0.939	288	170.6	12.8	13.0	93.6	20
	1.86	3.62	0.939	288	170.6	12.8	13.0	93.6	20
	1.86	3.58	0.937	289	170.6	12.8	13.0	93.6	20
3 x 3	3.06	3.27	1.32	697	412	30.8	31.5	22.6	20
	3.06	3.27	1.32	698	412	30.8	31.5	22.6	20
6 x 6	5.91	6.02	2.52	c	2,941	220	225	1614	1,000
	5.91	5.98	2.51	c	2,941	220	225	1614	1,000
	5.91	6.10	2.53	c	2,941	220	225	1614	1,000

[a] Each sample size prepared in triplicate, except 3 x 3.
[b] Weight of waste form after removal from mold.
[c] Not weighed.

Waste Form Leaching

The composites were leached in deionized water using a modified IAEA leaching procedure described earlier [18-20]. 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 this 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.

RESULTS

The incremental and cumulative fractional releases (CFR) for the organic ion exchange resin/cement and boric acid/cement composites have been reported in an earlier report [3]. Since the solution for the mass transport diffusion model (Eq. 2) infers that CFR is a linear function of $t^{1/2}$, the CFR data were plotted vs $t^{1/2}$. The complete set of plots for the organic ion exchange resin/cement composites and the boric acid/cement composites were presented in a topical report [3]. The average cumulative fractional release curves were normalized for V/S variation in the waste forms. Some of the resin/cement composites physically deteriorated during the course of the experiment. Two of the 1 x 1, one of the 6 x 6 and the larger waste forms (6 x 12, 12 x 12, and 22 x 22) disintegrated at various times during the experiment. Leach data obtained from these forms were considered only while the forms remained intact. Further details are given elsewhere [3,4].

DISCUSSION

Interpretation of Leach Data

A semi-empirical approach based on the semi-infinite plane source diffusion model is used to interpret the leach data. This approach is applied to both the organic ion exchange resin/cement and the boric acid/cement composites. Rewriting Equation (2) yields:

$$f = [S/V \cdot 2(D/\pi)^{1/2}] \cdot t^{1/2} + \alpha \qquad (3)$$

This relationship describes a straight line with slope = $S/V \cdot 2(D/\pi)^{1/2}$ and intercept α when f is plotted vs $t^{1/2}$.

Organic Ion Exchange Resin/Cement Composites. Appropriate linear regions of the ^{137}Cs release curves were selected visually [3,4]. The selection was based on the fact that the dominant diffusional mass transport reaction occurs in the linear regions and that the initial reactions are primarily surface-controlled and not representative of the overall release curve. The average CFR's from the intact forms for each size studied were calculated from the linear regions, and were plotted vs $t^{1/2}$ as shown in Figure 1. The data from the 1 x 1 forms were not considered since they did not fit the diffusional model; the surface-to-volume ratio was the largest for this size (2.53 cm^{-1}). As a result, the surface effects are expected to be most pronounced in this particular size. 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 III, together with the coefficients of determination as defined by:

$$R^2_{(S/V)} = \left(\frac{\alpha\Sigma(\mathrm{CFR})_i + b\Sigma(t^{1/2})_i \times (\mathrm{CFR})_i - 1/n(\Sigma(\mathrm{CFR})_i)^2}{\Sigma(\mathrm{CFR})_i^2 - 1/n(\Sigma(\mathrm{CFR})_i)^2} \right)_{(S/V)} \qquad (4)$$

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

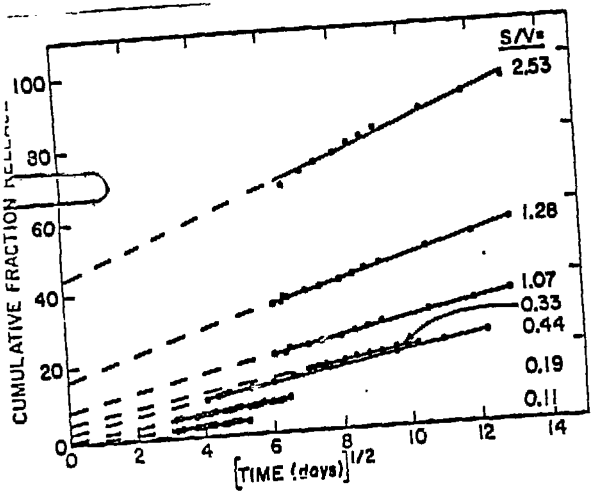

Figure 1. Solid lines represent least squares linear regression fits through average CFR's for resin/cement composites of varying sizes. Dashed lines represent extrapolation of the best fit lines to determine α, the intercept.

Table III
Slopes and intercepts of CFR vs $t^{1/2}$ plots for resin/cement composites

Size (in. x in.)	S/V (cm^{-1})	Slope[a]	Intercept[a] (α)	Coefficient of Determination[b]
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	1.5[illegible]	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 from the general relationship CFR = $\alpha + b(t)^{1/2}$ using data in the linear region as explained in the text.

[b] The coefficient of determination is defined in Equation 4.

Since the slopes of the lines (Table III) are represented by $[S/V \times 2(D/\pi)]^{1/2}$, plotting $[S/V \times 2(D/\pi)]^{1/2}$ versus S/V would yield a line with a slope equal to $2(D/\pi)^{1/2}$, i.e., for $z = (S/V)$, then $d(\text{slope})/dz = 2(D/\pi)$. A plot of the slopes vs S/V is shown in Figure 2, indicating a linear relationship. A least squares linear regression on the points (slopes, S/V) (Table III) yields an expression of the form: slope = a + bx, with $R^2 = 0.90$, a = 0.75, b = 2.07, where $b = 2(D/\pi)^{1/2}$.

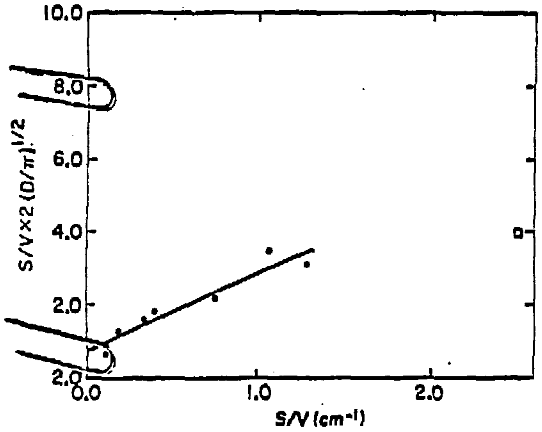

Figure 2. Plot of $[S/V \cdot 2(D/\pi)^{1/2}]$, representing slopes of the lines shown in Figure 1, vs (S/V) for resin/cement composites. The solid line is a least squares linear regression on the points for S/V less than 2.53 (i.e., forms larger than 1 x 1).

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

$$(\text{slope})_{(S/V)} = 2.07 \times \frac{S}{V} + 0.75 \tag{5}$$

A similar fit was performed on the intercepts of the lines and the S/V ratios, as shown in Figure 1, and the S/V ratios, resulting in the relationship:

$$(\alpha)_{S/V} = 1.80 + 12.6 \frac{S}{V} \tag{6}$$

A plot of these intercepts $[(\alpha)_{S/V}]$ versus their corresponding S/V ratios is shown in Figure 3.

Combining Equations (5) and (6) yields:

$$(\text{CFR}) = (2.07 \cdot S/V + 0.75)t^{1/2} + (12.6 \cdot S/V - 1.80) \tag{7}$$

Thus, the cumulative fractional release for a given time t from a sample with a geometric surface-to-volume ratio of S/V can be calculated using Equation (7). The cumulative fractional releases calculated for several S/V values for different time intervals are summarized in Table IV, together with the experimental data. A good agreement is observed between the calculated values and the experimental data measured over the linear regions of CFR vs $t^{1/2}$ plots.

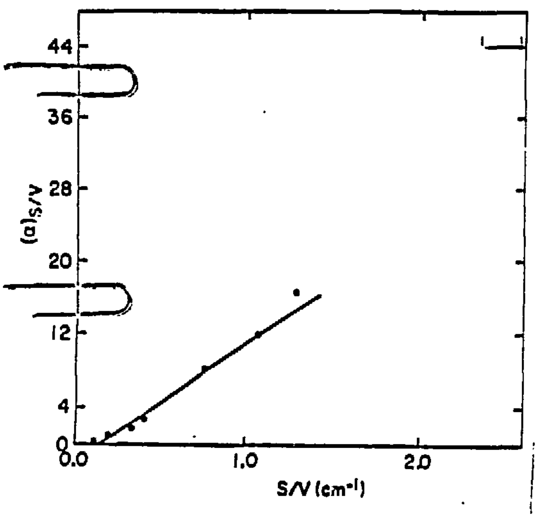

Figure 3. Plot of the intercepts $[(\alpha)_{S/V}]$ of the lines shown in Figure 1 vs S/V for resin/cement composites. The solid line is a least-squares linear regression on the points for S/V less than 2.53 (i.e., forms larger than 1 x 1).

Table IV
Experimental and calculated CFR (%) for IRN-77/cement composites

Dimension	S/V (cm^{-1})	Mode	Time (Days) 9	15	21	30	42	56	88	112	126	169	240	260
2 x 2	1.28	Expt. Calc.		24.5 27.5	27.6 29.9	31.7 32.9	36.3 36.4	38.6 39.8	*	48.5 50.3	*	55.4 58.6	*	62.7 66.1
2 x 4	1.07	Expt. Calc.		20.5 23.2	23.9 25.3	28.6 27.9	34.2 30.9	37.6 33.9	*	48.6 43.1	*	55.7 50.3	*	63.3 59.5
3 x 3	0.76	Expt. Calc.		12.8 16.8	15.0 18.4	18.4 20.5	22.2 22.9	23.6 25.2	*	30.3 32.4	*	35.2 38.0	*	40.8 45.3
6 x 6	0.40	Expt. Calc.	7.2 7.9	9.2 9.3	10.7 10.5	12.2 11.9	14.4 13.5	16.3 15.1	*	19.6 19.9	*	24.0 23.8	29.1 27.7	*
6 x 12	0.33	Expt. Calc.	7.7 6.7	9.6 7.9	11.2 8.9	13.0 10.2	15.0 11.7	17.0 13.1	*	*	23.3 20.9	*	*	*
12 x 12	0.19	Expt. Calc.	5.3 4.0	6.5 5.0	7.4 5.8	8.4 6.9	9.4 8.0	*	*	*	*	*	*	*
22 x [illegible]	0.11	Expt. Calc.	2.1 2.5	2.7 3.4	3.1 4.7	3.7 4.9	4.5 5.9	*	8.3 8.8	*	*	*	*	*

*Experimental data not available for these time periods.

Boric Acid/Cement Composites. The cumulative fractional releases from the boric/acid cement composites are shown as a function of $t^{1/2}$ in Figure 4. Data from the 2 x 4, 3 x 3, and 6 x 6 samples, after 9 days of leaching, represent the main diffusion-controlled mass transport reaction. This set of data for the post 9-day linear regions is used in calculating CFR's for the boric acid waste/cement composites. The 1 x 1 and 2 x 2 leach data were not considered here for the same reasons given for the 1 x 1 resin/cement composites.

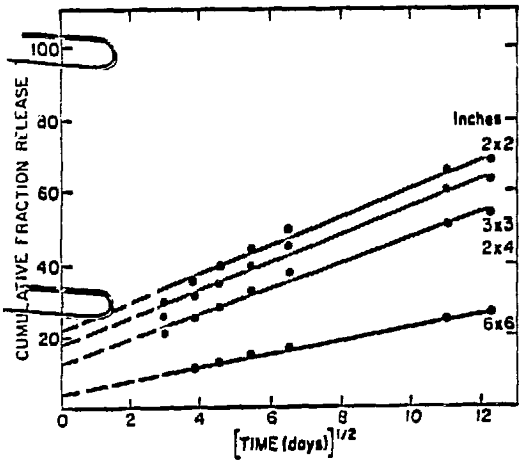

Figure 4. Solid lines represent least squares linear regression fits through average CFR's for boric acid/cement composites of varying sizes. Dashed lines represent extrapolation of the best fit lines to determine α, the intercept.

The slopes, intercepts, and coefficients of determination based on the leach data for the 2 x 4, 3 x 3, and 6 x 6 samples are summarized in Table V. Fitting the slopes and intercepts versus S/V yields:

$$(\text{Slope})_{(S/V)} = 3.15 \cdot S/V + 0.70 \tag{8}$$

and

$$(\alpha)_{S/V} = 18.69 \cdot S/V - 2.63 \tag{9}$$

Combining Equations (8) and (9) yields:

$$\text{CFR} = (3.15 \cdot S/V + 0.70)t^{1/2} + (18.69\ S/V - 2.63) \tag{10}$$

Using Equation (10), the CFR for several V/S ratios of boric acid/cement composites were calculated for different time intervals. These values are summarized in Table VI, together with their corresponding experimental values. A reasonably good agreement between the calculated and experimental data is observed.

Table V
Slopes and intercepts of CFR vs $t^{1/2}$ plots for boric acid/cement composites

Size (in. x in.)	S/V (cm^{-1})	Slope[a]	Intercept[a]	Coefficient of Determination[b]
2 x 2	1.28	4.05	20.2	0.98
2 x 4	1.07	3.89	16.7	0.99
3 x 3	0.76	3.41	12.7	0.99
6 x 6	0.40	1.79	4.2	1.00

[a] Slopes and intercepts are obtained from the general relationship $\mathrm{CFR} = \alpha + b(t)^{1/2}$, using leach data beyond 9 days.

[b] The coefficient of determination is defined in Equation 4.

Table VI
Experimental and calculated CFR (%) for boric acid/cement composites

Dimension (in. x in.)	S/V (cm^{-1})	Mode	Time (Days) 9	15	21	30	42	122	150
2 x 2	1.28	Expt.	29.6	35.2	39.1	43.9	49.7	65.1	68.1
		Calc.	35.5	39.6	43.0	37.2	52.0	73.6	79.3
2 x 4	1.07	Expt.	26.1	31.2	34.7	39.5	44.7	59.8	63.0
		Calc.	29.6	33.1	36.0	39.7	43.8	62.3	67.2
3 x 3	0.76	Expt.	20.9	25.3	28.5	32.5	37.3	50.4	53.2
		Calc.	20.9	23.6	25.8	28.6	31.6	45.8	49.5
6 x 6	0.40	Expt.	9.3	11.1	12.4	14.2	16.4	24.1	25.9
		Calc.	10.7	12.4	13.8	15.6	17.6	26.5	28.9

CONCLUSIONS

The observed ^{137}Cs leach data for resin/cement waste forms and boric acid/cement waste forms can be represented by a diffusional mass transport relationship. The initial release of ^{137}Cs was primarily surface-controlled, the effect being most pronounced in the smallest waste form studied. The semi-empirical relationship was used to estimate the cumulative fractional release from forms varying in size from 2 x 2 to 22 x 22 for a given leaching period. For both types of waste forms a reasonably good agreement between the experimental and predicted cumulative fractional release is obtained, indicating that ^{137}Cs release data from small-scale laboratory samples can be used to predict leachability of large-scale samples.

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