LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES

3 4456 0446624 5

ORNL-4962

Cy 88

Development of Cementitious Grouts for the Incorporation of Radioactive Wastes. Part I: Leach Studies

J. G. Moore
H. W. Godbee
A. H. Kibbey
D. S. Joy

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
CIRCULATION SECTION
4500N ROOM 175
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON.
If you wish someone else to see this report, send in name with report and the library will arrange a loan.
ORNL-118 (6-97)

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION ○ FOR THE U.S. ATOMIC ENERGY COMMISSION

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy $5.45; Microfiche $2.25

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

ORNL-4962

UC-70 - Waste Management

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

DEVELOPMENT OF CEMENTITIOUS GROUTS FOR THE INCORPORATION OF RADIOACTIVE WASTES. PART I: LEACH STUDIES

J. G. Moore
H. W. Godbee
A. H. Kibbey
D. S. Joy

APRIL 1975

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES
3 4456 0446624 5

CONTENTS

PAGE

Abstract 1

1. Introduction 2

2. Experimental Methods and Materials 4

2.1 Test Materials 4
2.2 Leach Test Method 7
2.3 Analytical Methods 10

3. Losses of Radioactivity from Uncured Grout 15

4. Results and Discussion 16

4.1 Replication of Results 17
4.2 Effect of Leachant Renewal Frequency 25
4.3 Effect of Air Curing Time 31
4.4 Comparison of Isotopes 37
4.5 Tests with Added Alkaline-Earth Ions 37

5. Comparison of Results with Theoretical Expressions 41

6. Applications and Projections 49

7. Evaluation of Clay Minerals Other Than Grundite 60

8. Conclusions and Recommendations 62

9. Acknowledgments 66

10. References 66

11. Nomenclature 69

Appendix A: Compilation of Data on Leaching of Cementitious Grouts 73

Appendix B: Solution of Mass Transport Equations for the Case of Diffusion with Instantaneous Reaction in a Semi-Infinite Medium 105

Appendix C: Solution of Mass Transport Equations for the Case of Diffusion with Instantaneous Reaction in a Finite Cylinder 109

DEVELOPMENT OF CEMENTITIOUS GROUTS FOR THE INCORPORATION OF RADIOACTIVE WASTES. PART I: LEACH STUDIES

J. G. Moore, H. W. Godbee
A. H. Kibbey, D. S. Joy

ABSTRACT

The grouts used in the Hydrofracture Facility at Oak Ridge National Laboratory (ORNL) are made by blending ORNL intermediate-level radioactive waste solution with a dry mixture of cementitious materials and selected clays. Laboratory leach studies were made with simulated grouts containing ^{85}Sr, ^{137}Cs, ^{239}Pu, or ^{244}Cm in order to determine how effectively the grouts would retain radionuclides in the unlikely event that they were to come in contact with water. In general, the method of leach testing proposed by the International Atomic Energy Agency (IAEA) was followed. The fraction of each isotope leached varied with the square root of time if the leachant was replaced more frequently than once per day. Less frequent replacement of the leachant depressed or inhibited the fraction leached as a function of time. For the grouts made with Grundite (a clay to retain cesium), the leach rates for the isotopes used in this study were found to be in the order: Cs > Sr > Cm > Pu. Overall results indicate that the grouts can give leach rates comparable to those obtained for wastes incorporated in borosilicate glasses.

Curing a grout specimen up to 28 days in humid air before leaching decreased the amount leached from the specimen. Longer curing periods (up to 365 days) were run with cesium and had no significant effect. The leach rate for cesium was decreased by the addition of Grundite, pottery clay, or Conasauga shale. These additives, as listed, are in the order of increasing effectiveness with respect to cesium retention. The leach rate for strontium was lowered by adding small quantities of stable strontium or strontium and calcium to the waste, and by allowing the resulting precipitate to age before being blended with the dry ingredients to prepare a grout.

Theoretical relationships that consider diffusion and instantaneous reaction (an equilibrium or time-independent relationship between mobile and immobile forms of a species) give good agreement with the data for the 28-day-cured grouts when the leachant was replaced twice per day. Using the derived modified effective diffusivities, D_e's, comparisons were made of the amounts leached from a 55-gal drum of grout

assuming two different forms for the drum - a semi-infinite medium and a finite cylinder. These calculations show that the drum approximates a semi-infinite medium for a few years if the D_e is $\sim 1 \times 10^{-9}$ cm^2/sec, for about 30 years if the D_e is $\sim 3 \times 10^{-11}$ cm^2/sec, and for more than 3000 years if the D_e is $\sim 6 \times 10^{-16}$ cm^2/sec. They also show that the projected amounts leached from a waste product which is assumed to approximate a semi-infinite medium are conservative (on the side of safety) since they are equal to or greater than the amounts predicted by a solution based on the actual geometry of the finite product.

1. INTRODUCTION

This report presents the initial results from studies made to determine the leachability of various radionuclides from cementitious grouts similar to those used in the Hydrofracture Facility at Oak Ridge National Laboratory (ORNL). This facility has been used for the disposal of intermediate-level waste (ILW) for about eight years.

Hydrofracturing is a procedure whereby the radionuclides present in ILW are fixed in a cement matrix deep within a Conasauga shale bed formation located within the area.[1] The ILW is composed of a mixture of all the liquid wastes, other than process waste, that are produced in hot-cell, pilot-plant, and reactor operations, including relatively small volumes of organic reagents and solvents. The solution, after concentration by evaporation, may contain up to 2 Ci of beta-gamma activity per gallon. The major radionuclide is generally ^{137}Cs while ^{90}Sr is present at a level of only 0.1 Ci/gal or less. The major alpha activity is ^{244}Cm with a maximum concentration of about 1 mCi/gal.[2] The waste is formed into a grout by blending it with a mixture of cementitious materials and selected clays. The grout is immediately injected 800 to 1000 ft underground into cracks produced in the shale formations by the pressure of the injected grout. Here the mixture solidifies, fixing the radionuclides. A safety analysis based on the development studies and actual waste injection experience concluded that the procedure is safe, presenting no significant hazard to the public or environment.[3] Since 1966, more than one million gallons of waste solution have been injected without serious difficulty.

A proposal has been made to build a new facility that would have the capability of handling sludges and wastes with higher specific activities than described above. This proposal led to the resumption of a development program to resolve questions concerning the safety and environmental impact of the facility. One of these questions concerned how effectively the grouts would retain the radionuclides in the unlikely event they came in contact with water. Consequently, a series of grout leach studies was undertaken to obtain such data.

Incorporation of radioactive waste in cement has been widely studied[4] and routinely used at nuclear research and production sites[5-7] for more than two decades and at power reactor plants for more than a decade.[8-10] Cement has many favorable characteristics which make it an attractive candidate for a waste binding agent. It is widely available, noncombustible, strong, radiation resistant, and has good chemical and thermal stability at the temperatures reached with low- and intermediate-level wastes. Cement products, which are made by low-temperature processes using established technology, are relatively nonleachable if large quantities of soluble solids are absent. To determine how effectively such products retain the incorporated radioactive wastes, the amount of radioactivity that is leached from the solids by water or other liquids is usually measured as a function of time. Attempts are then made to relate these results to existing environmental situations.[11,12]

The leachability of radionuclides from cement has been studied using a wide variety of methods.[13,14] To permit intercomparison of various products from different processes or different materials, the IAEA has proposed a standard method[15] of measuring and reporting the leaching, by water, of the radioactive components in solid waste products. Insofar as possible, this method was followed in the leach tests reported here. Models based on transport phenomena were investigated to relate the experimental results to disposal situations.

2. EXPERIMENTAL METHODS AND MATERIALS

2.1 Test Materials

The grout used in the hydrofracture procedure is made by combining the desired volume of waste solution with a predetermined weight of dry solids. The dry solids consist of a mixture of Type I Portland cement,[16-19] fly ash, Attapulgite-150 clay, Grundite clay, and a sugar. The recipe for the dry mixture is shown in Table 1. The fly ash, a pozzuolana, is a partial substitute for cement which can improve the retention of strontium. Attapulgite-150 is a drilling clay which suspends the cement, thus reducing phase separation. Grundite is an illite clay which has a high capacity for cesium. The sugar (delta gluconolactone or CFR-1) is a retarder used to prevent the grout from setting or stiffening too rapidly. Simulated waste solutions (Table 2) were used to prepare the grouts used in these leach studies. The compositions are based on chemical analyses of actual waste solutions in ORNL storage tanks W-7 and W-9/10. The simulated waste contained no radioactive material other than the tracer used to follow the isotope of interest during a leach test. Either ^{85}Sr, ^{137}Cs, ^{239}Pu, or ^{244}Cm was added to the waste immediately before preparation of a grout. Prior to the addition of tracer, a sufficient quantity of tributyl phosphate (TBP) was added to the waste to give a final concentration of 400 ppm. The TBP, in conjunction with the CFR-1, results in a relatively free release of air from the grout during stirring and thus prevents foaming.

The simulated waste containing appropriate amounts of TBP and CFR-1 (CFR-1 is added to the dry mix at the Hydrofracture Facility) was placed in a Waring Blender. After addition of the desired tracer, the blender was turned on for about 5 sec at the lowest speed setting. Samples of the resulting solution were taken and were used as the feed in all subsequent activity determinations. Next, the blended dry mix was added to the waste over a period of 15 sec while stirring; then the resulting slurry was stirred for an additional 15 sec at the same setting. At this setting, the blender speed under no-load conditions was 3100 to 3500 rpm; however, some reduction in speed was observed under the load conditions of grout preparation. These variations in mixing speed can affect the

Table 1. Composition of Dry Mix Used in Cementitious Grout Preparations

Constituent	Recipe[a] (parts by wt)	Composition[b] (wt %)
Cement[c]	2.5	38.5
Fly ash[d]	2.5	38.5
Attapulgite-150[e]	1.0	15.4
Grundite[f]	0.5	7.7

[a]CFR-1 or delta gluconolactone, a chemical that retards the setting of cement, is added to this formulation to the extent of 0.003 part by weight.

[b]CFR-1, added as a chemical retarder to setting, amounts to about 0.05 wt %.

[c]Portland Cement Type I (see refs. 16-19 and similar works on cement and concrete products for a description of chemical and physical properties).

[d]Fly ash (a pozzuolanic material) obtained from the coal-fired Kingston Steam Plant, Kingston, Tenn.

[e]Attapulgite-150 is the trade name of a clay product from the polygorskite group of clay minerals (with the general formula $5MgO \cdot 8SiO_2 \cdot 9H_2O$).

[f]Grundite is the trade name of a clay product from the illite group of clay minerals [with the general formula $(OH)_4K_x(Al_4 \cdot Fe_4 \cdot Mg_4 \cdot Mg_6)(Si_{8-x} \cdot Al_x)O_{20}$] from Grundy County, Ill.

Table 2. Composition and Properties of Simulated Waste Solutions Used in Cement Grout Preparations

	Concentration (g-mole/liter)	
	W-7[a]	W-9/10[b]
Constituent		
Na^+	1.78	0.843
NH_4^+	0.003	0.05
Al^{3+}	0.0074	0.0016
NO_3^-	0.835	0.3
OH^-	0.18	0.08
Cl^-	0.093	0.05
CO_3^{2-}	0.255	0.136
SO_4^{2-}	0.094	0.098
Property		
pH	11.52	11.37
Density, g/ml	1.085	1.042

[a]Each liter of solution was prepared with 68.85 g of $NaNO_3$, 27.0 g of Na_2CO_3, 13.35 g of Na_2SO_4, 7.2 g of NaOH, 5.44 g of NaCl, 2.78 g of $Al(NO_3)_3 \cdot 9H_2O$, and 0.24 g of NH_4NO_3.

[b]Each liter of solution was prepared with 25.08 g of $NaNO_3$, 14.42 g of Na_2CO_3, 13.92 g of Na_2SO_4, 3.2 g of NaOH, 2.68 g of NH_4Cl, and 0.6 g of $Al(NO_3)_3 \cdot 9H_2O$.

physical properties of the final grout and can account for some of the differences in leach rates that were observed in duplicate batches of grout (see Sect. 4.1).

A series of preliminary trials was made to determine the solids/liquid ratio to be used in the preparation of leach specimens. Prior to initiation of these leach studies, about 7 lb of dry mix per gallon of waste had been used at the Hydrofracture Facility. However, trials with the laboratory equipment covering dry mix/waste ratios from 5.5 to 7 lb/gal showed that 6 lb/gal produced a wet mixture that handled easily and hardened into acceptable solids for leach tests. At 6.5 to 7 lb/gal, the wet grout mixture was very thick and made the preparation of small specimens without bubbles and other surface flaws difficult. Thus, all but one of the leach specimens in these studies were prepared at a dry mix/waste ratio of 6 lb/gal; specimen 94 was prepared at 7 lb/gal. The compositions of these products, as prepared (i.e., before the loss of any evaporable water), are given in Table 3. The high water/cement ratios (~ 3/1 to 3.5/1) and water/fly ash ratios (~ 1.5/1 to 1.8/1) of the grouts in Table 3 are worth noting. Compared with ordinary cement products and many cement products incorporating radioactive waste, the grouts contain a small percentage of cement. For example, in many instances, cement products incorporating radioactive waste have water/cement ratios of 1 or less. The products made from W-7 at a ratio of 6 lb/gal have a density of 1.40 g/cm^3 and represent 1.31 volumes of product per volume of waste incorporated. The products prepared from W-7 at 7 lb/gal have a density of 1.46 g/cm^3 and represent 1.34 volumes of product per volume of waste incorporated. The products made from W-9/10 at 6 lb/gal have a density of 1.37 g/cm^3 and represent 1.31 volumes of product per volume of waste incorporated. Generally, the grouts have a compressive strength of about 200 to 300 lb/in.^{2}.

2.2 Leach Test Method

In general, the method of leach testing proposed by the IAEA[15] was followed. The leach specimens were prepared by pouring the grout into a 5-cm-ID, 5-cm-high polyethylene container (Fig. 1) immediately after mixing. The container protected all except the upper surface of the grout

Table 3. Composition (in wt %) of Grout Products as Prepared

Component	Weight of Dry Mix[a] Added to Waste[b]		
	6 lb/gal W-7[c]	7 lb/gal W-7[d]	6 lb/gal W-9/10[e]
Cement	15.33	16.77	15.71
Fly ash	15.33	16.77	15.71
Attapulgite-150	6.13	6.71	6.28
Grundite	3.07	3.35	3.14
CFR-1	0.02	0.02	0.02
TBP	0.03	0.03	0.03
NaOH	0.40	0.37	0.18
$Al(NO_3)_3$	0.09	0.08	0.02
NH_4Cl			0.15
NH_4NO_3	0.01	0.01	
$NaNO_3$	3.81	3.58	1.42
NaCl	0.30	0.28	
Na_2SO_4	0.74	0.69	0.79
Na_2CO_3	1.12	1.05	0.82
H_2O	53.62	50.28	55.74

[a]Composition of dry mix is given in Table 1.

[b]Compositions of waste solutions are given in Table 2.

[c]Products have a density of 1.40 g/cm^3 and represent 1.31 volumes per volume of waste incorporated.

[d]Products have a density of 1.46 g/cm^3 and represent 1.34 volumes per volume of waste incorporated.

[e]Products have a density of 1.37 g/cm^3 and represent 1.31 volumes per volume of waste incorporated.

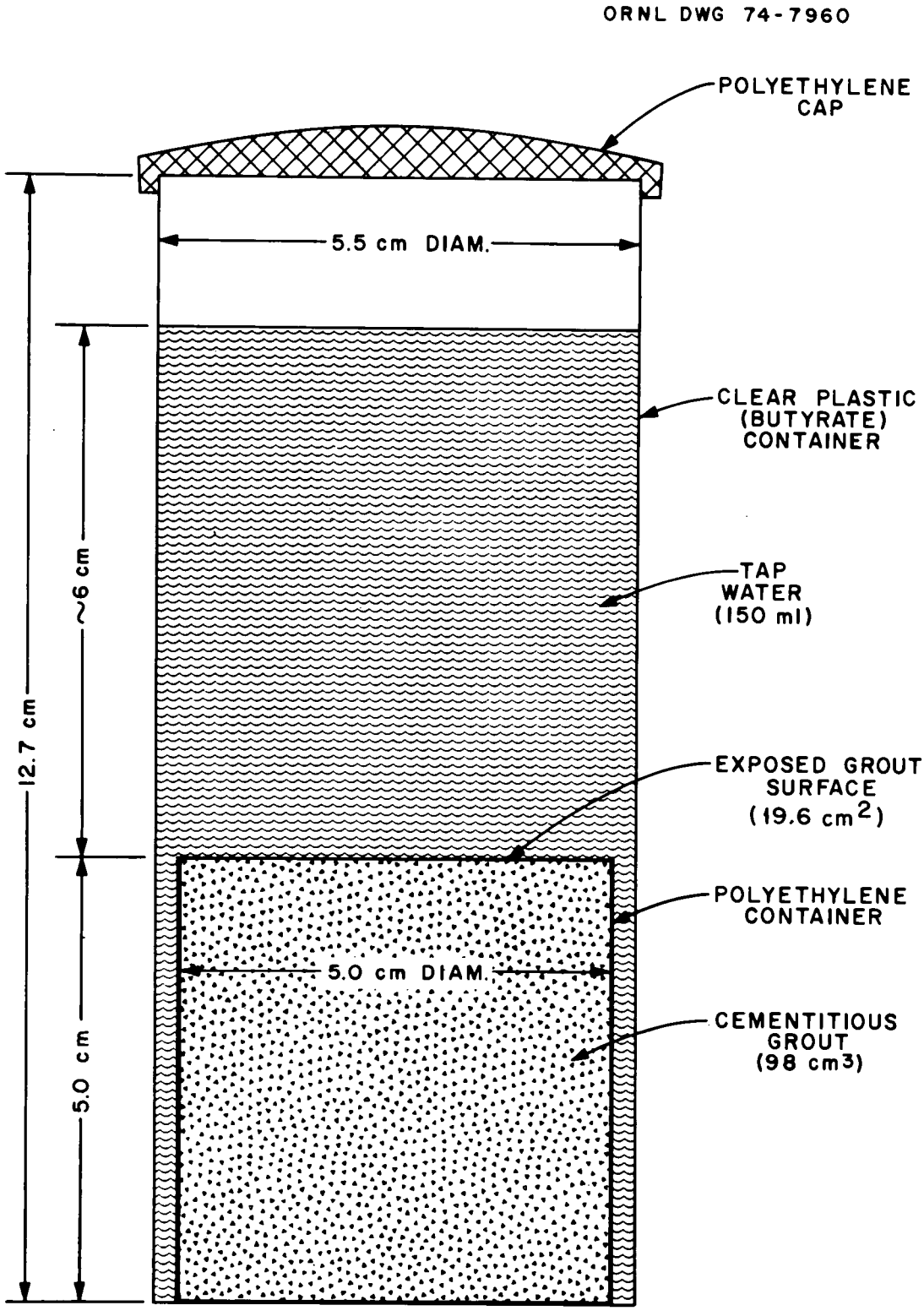

Fig. 1. Schematic of Leach Test Setup.

specimens. Hence, the specimens had a volume of 98 cm^3 and an exposed surface area of 19.6 cm^2. In most cases, the specimens were cured for a predetermined number of days in a humid atmosphere (referred to as "air curing" in this report). This curing was accomplished by placing the grout-filled container in a shallow tray of water. Both the specimen and the tray of water were sealed inside a plastic bag. Visual examination of the cured specimens showed no evidence of a gap between the walls of the plastic container and the grout. After curing, the specimen, in its container, was placed inside a clear plastic (butyrate) cylinder having an inside diameter of 5.5 cm and a height of 12.7 cm. Next, 150 ml of tap water was added to the clear plastic cylinder. This volume is sufficient to cover the exposed grout surface with approximately 6 cm of water. Finally, the outer container was covered with a plastic cap (Fig. 1). The leachate was replaced periodically with 150 ml of fresh tap water. The twenty-nine leach tests, which were carried out at room temperature, are briefly described in Table 4.

2.3 Analytical Methods

Samples of all feed and leachate solutions containing ^{85}Sr or ^{137}Cs were analyzed with a Nuclear-Chicago Automatic Gamma Well System, Model 8725. The samples were counted by utilizing the narrow differential mode in which the response from a small section of the isotope's characteristic energy spectrum is measured. The section is determined by the base setting of the instrument. Each time analyses were made, the base setting was adjusted for the maximum output obtained when counting an appropriate dilution of one of the feed solutions used in the grout preparations. Each leachate sample and its corresponding feed sample were then counted at this base setting. The fraction of the element in question that was removed from the grout by the leachant was calculated from these counts by using Eq. (1):

$$\frac{a_n}{A_o} = \frac{(c_\ell)(V_\ell)}{(c_f)(V_f)} \quad , \qquad (1)$$

Table 4. Description of Cementitious Grout Specimens Prepared[a] for Leaching Studies[b]

Grout No.	Type of Simulated Waste[c]	Solid[a]/Waste[c] Ratio (lb/gal)	Radioactivity in Specimen Isotope	Radioactivity in Specimen µCi/ml	Curing Time Before Leaching[d] (days)	Nominal Frequency of Leachant Renewal[e]	Length of Run (days)	Remarks
13A	W-7	6	^{85}Sr	0.435	0	f	196	Duplicate of 13D
13B	W-7	6	^{85}Sr	0.435	7	f	189	Duplicate of 13E
13C	W-7	6	^{85}Sr	0.435	28	f	168	Duplicate of 13F
13D	W-7	6	^{85}Sr	0.435	0	f	196	Duplicate of 13A
13E	W-7	6	^{85}Sr	0.435	7	f	189	Duplicate of 13B
13F	W-7	6	^{85}Sr	0.435	28	f	168	Duplicate of 13C
70	W-7	6	^{85}Sr	5.08	7	f	254	Stable Sr and Ca added[g]
87	W-7	6	^{85}Sr	9.0	112	f	203[h]	Stable Sr added[i]
92	W-7	6	^{85}Sr	9.0	168	f	121[h]	Stable Sr added[i]
16A	W-7	6	^{137}Cs	0.276	0	f	449[h]	Duplicate of 16D
16B	W-7	6	^{137}Cs	0.276	7	f	273	Duplicate of 16E
16C	W-7	6	^{137}Cs	0.276	28	f	421[h]	Duplicate of 16F
16D	W-7	6	^{137}Cs	0.276	0	f	280	Duplicate of 16A
16E	W-7	6	^{137}Cs	0.276	7	f	442[h]	Duplicate of 16B
16F	W-7	6	^{137}Cs	0.276	28	f	252	Duplicate of 16C
86	W-7	6	^{137}Cs	1.36	84	f	245[h]	
91	W-7	6	^{137}Cs	1.36	168	f	161[h]	
93A	W-7	6	^{137}Cs	2.76	28	1 per day	112[h]	
93C	W-7	6	^{137}Cs	2.76	28	2 per day	113[h]	
94A	W-7	7	^{137}Cs	2.76	28	1 per day	112[h]	
94C	W-7	7	^{137}Cs	2.76	28	2 per day	113[h]	
3A	W-9/10	6	^{137}Cs	0.554	28	1 per day	70[h]	Duplicate of 3C
3C	W-9/10	6	^{137}Cs	0.554	28	1 per day	70[h]	Duplicate of 3A
35A	W-7	6	^{239}Pu	0.10	0	f	118[h]	
35B	W-7	6	^{239}Pu	0.10	7	f	111[h]	
35C	W-7	6	^{239}Pu	0.10	28	f	90[h]	
36A	W-7	6	^{244}Cm	0.10	0	f	118[h]	
36B	W-7	6	^{244}Cm	0.10	7	f	111[h]	
36C	W-7	6	^{244}Cm	0.10	28	f	90	

Table 4 (continued)

---

[a]All specimens were prepared with dry mix of the composition given in Table 1.

[b]All studies were made with tap water as the leachant.

[c]Compositions of simulated wastes are given in Table 2.

[d]Length of time the specimen was cured in air before leaching commenced.

[e]For exact renewal times, see tables in Appendix A.

[f]For the most part, in accordance with the IAEA-recommended procedure as given in ref. 15.

[g]Specimens were prepared by addition of 1 g of Sr^{2+} and 0.25 g of Ca^{2+} (as $SrCl_2$ and $CaCl_2$) per liter of waste.

[h]These runs are still in progress.

[i]Specimens were prepared by addition of 0.05 g of Sr^{2+} (as $SrCl_2$) per liter of waste.

where

a_n = amount of the species in the leachate,

A_o = amount of the species in the grout specimen at the beginning of the leach test or time t = 0,

c_ℓ = net ^{85}Sr (or ^{137}Cs) counts min^{-1} ml^{-1} in the leachate at time t,

c_f = net ^{85}Sr (or ^{137}Cs) counts min^{-1} ml^{-1} in the feed sample at time t,

V_ℓ = volume of the leachate, ml,

V_f = volume of waste solution used to prepare the grout specimen, ml.

That the right side of Eq. (1) is equal to the incremental fraction of the element leached may be demonstrated in the following discussion. Since the number of counts obtained for each isotope is directly proportional to the number of radioactive atoms,

$$\frac{c_\ell}{c_f} = \frac{h_\ell}{h_f} \quad , \tag{2}$$

where

h_ℓ = number of ^{85}Sr (or ^{137}Cs) atoms per ml of leachate at time t,

h_f = number of ^{85}Sr (or ^{137}Cs) atoms per ml of feed sample at time t.

With the assumption that stable strontium (or cesium) from the waste and from the dry mix are leached at the same rate and that no separation of isotopes has occurred,

$$\frac{s_\ell + h_\ell}{h_\ell} = \frac{s_f + h_f}{h_f} \quad , \tag{3}$$

where

s_ℓ = number of stable strontium (or cesium) atoms per ml of leachate,

s_f = number of stable strontium (or cesium) atoms per ml of feed sample.

Substitution of Eq. (3) into Eq. (2) yields:

$$\frac{c_\ell}{c_f} = \frac{s_\ell + h_\ell}{s_f + h_f} \quad . \tag{4}$$

In these studies with strontium (or cesium), the number of stable atoms of the isotope is much larger than the number of radioactive atoms; that is, s>>h. The stable strontium/^{85}Sr atom ratio ranged from about 5 x 10^7/1 to 1 x 10^9/1, and the stable cesium/^{137}Cs atom ratio from about 9 x 10^3/1 to 9 x 10^4/1. Thus, for these isotopes, Eq. (4) is equivalent to

$$\frac{c_\ell}{c_f} = \frac{s_\ell}{s_f} \quad . \tag{5}$$

Multiplication of both sides of Eq. (5) by $(V_\ell)/(V_f)$ gives:

$$\frac{(c_\ell)(V_\ell)}{(c_f)(V_f)} = \frac{(s_\ell)(V_\ell)}{(s_f)(V_f)} = \frac{a_n}{A_o} \quad . \tag{6}$$

The amount of each actinide in the sample was obtained by determining ^{239}Pu and ^{244}Cm via an extraction-scintillation technique. A 100-ml sample of the leachate in question was contacted for 5 min with 15 ml of an extractive scintillator, which was a toluene solution of 0.5 M di(2-ethylhexyl)phosphoric acid containing 200 g of naphthalene and 4 g of 2-(4'-biphenyl)-6-phenylbenzoxazole per liter. The two phases were allowed to separate for 30 min. Then, a portion of the extractive scintillator was counted in a Packard Tri-Carb Liquid Scintillation Spectrometer. Preliminary experiments with known concentrations showed that, without further treatment, plutonium was essentially completely extracted from the alkaline leachates (pH ~ 11.5 to 13). To obtain similar results with curium, it was necessary to adjust the leachates to a pH of about 3 with hydrochloric acid prior to contact with the organic extractant. Appropriate dilutions of the feed samples were analyzed similarly. Since the half-life of ^{239}Pu (or ^{244}Cm) is long compared with the length of a leach test, the incremental fraction leached was calculated using Eq. (1), where c_ℓ and c_f are net ^{239}Pu (or ^{244}Cm) counts min^{-1} ml^{-1} in the leachate and feed sample respectively.

3. LOSSES OF RADIOACTIVITY FROM UNCURED GROUT

Brief consideration was given to possible short-term losses of radioactivity from the grouts. Although the grouts in this study were designed to minimize the amount of liquid that separates out during setting (phase separation), it was considered necessary to obtain data concerning the amount of cesium and strontium that might be lost to phase separation should it occur. In addition, information was needed (for an impact statement) to assess the amount of radioactivity that might be lost (in accident situations) if freshly prepared uncured grout should be contacted immediately by relatively large quantities of rapidly moving water.

Even though the grout specimens used in the static leach tests had been prepared with 6 or 7 lb of dry mix per gallon of waste, the grouts used in the phase-separation tests were prepared using a solid-waste ratio of only 4.5. This lower ratio was necessary in order to obtain a sufficient volume of separated liquid for analysis. These phase-separation-study grouts were traced with either ^{85}Sr or ^{137}Cs. Immediately after mixing, aliquots of each grout were placed in a 100-ml graduated cylinder and allowed to stand for 2 hr. The volume and the radioactive content of the supernatant liquid were then measured. In the test with strontium, the separated liquid amounted to about 3% of the total grout volume and contained about 0.02% of the total strontium in the system. In the test with cesium, the separated liquid amounted to about 2% of the total grout volume and contained about 0.1% of the total cesium in the system. Such phase separation can lead to an especially active, contaminated, or non-representative surface, and the aqueous-phase strontium and cesium concentrations could be indicative of the amounts of these elements that would be leached or readily washed off over a relatively short period of time.

To estimate the loss of radioactivity in the unlikely event that a freshly prepared grout were exposed to a large body of rapidly moving water, uncured grouts traced with either ^{85}Sr, ^{137}Cs, ^{239}Pu, or ^{244}Cm were mixed with ten volumes of tap water by rotating at 30 rpm for 1, 3, and 5 hr. The mixtures were then allowed to settle for 1 hr prior to sampling the supernatant liquid. After the initial sampling, the mixture was allowed

to settle further for a total of 24 hr before a second sample was taken. In both the initial and second samples, approximately 0.7% of the strontium and 7% of the cesium had transferred to the aqueous phase, which indicates that neither mixing nor settling time had an effect. This was not true for plutonium and curium. In the case of these actinides, the amount of radioactivity in the aqueous phase decreased with both mixing time and settling time. After 1-hr mixing and 1-hr settling periods, about 2% of either actinide was in the supernatant liquid. However, after a 5-hr mixing period and a 1-hr settling interval, the total actinide activity in the supernatant liquid had decreased to 1% for plutonium and 0.3% for curium. Under all three mixing conditions, the actinide activity in the supernatant liquid after the 24-hr settling time decreased to approximately 0.02% of the total present for plutonium and 0.05% for curium.

The studies on phase separation described in this section are presented largely to illustrate a possible mechanism for producing a contaminated surface. It should be mentioned again that the grouts made in actual practice and those used in the static leach tests do not have a solids/waste ratio as low as that of the grouts used in the phase separation studies. The studies on loss of activity from freshly prepared grout are included mainly to illustrate the relative solubilities and low intrinsic solubilities of these elements (see Sects. 4.4, 5, and 6).

4. RESULTS AND DISCUSSION

Twenty-nine tests were made to evaluate the leaching of cementitious grouts by tap water at room temperature. These include 9 individual tests with strontium (^{85}Sr tracer), 14 with cesium (^{137}Cs tracer), 3 with plutonium (^{239}Pu), and 3 with curium (^{244}Cm). The tests with plutonium and curium, as well as several of those with cesium, are still in progress. The curing time in humid air prior to leaching of the specimens ranged from about 10 min (called 0-day cure) to 168 days. During leaching, the time interval between replacement of the leachant with fresh tap water varied from one-fourth day to 136 days. The strontium data are given in

Tables A-1 through A-9 (Appendix A), the cesium data in Tables A-10 through A-23, the plutonium data in Tables A-24 through A-26, and the curium data in Tables A-27 through A-29. The results in these tables are presented as incremental fraction leached (a_n/A_o) during each leachant renewal period (t_n) and as cumulative fraction leached ($\Sigma a_n/A_o$) vs cumulative leaching time (Σt_n). The results are presented in this section as plots of the product of the cumulative fraction leached and the volume-to-surface ratio [$(\Sigma a_n/A_o)(V/S)$] vs cumulative leaching time, where V is the volume of the specimen in cm^3 and S is the surface area exposed to leachant in cm^2. The ordinate of each plot is converted to fraction leached by multiplying by the exposed surface/volume ratio (S/V), which is 0.2 cm^{-1} for the specimens in this study. These data form the basis for the following discussion.

4.1 Replication of Results

The results shown in Fig. 2 for the leaching of cesium from 28-day-cured specimens 3A and 3C represent duplicate tests in which the specimens were made from the same batch of grout. The cumulative fraction leached from 3C was about 1.3 times greater than that from 3A after 70 days of leaching. The results presented in Figs. 3-5 represent duplicate tests for the leaching of cesium in which the specimens were made from different batches of grout. Specimens 16A, 16B, and 16C were from one batch, while 16D, 16E, and 16F were the corresponding specimens from another batch. After 240 days of leaching, the cumulative fraction of cesium leached was about 1.05 times greater for 16D than for 16A (0-day-cured specimens shown in Fig. 3), about 1.1 times greater for 16E than for 16B (7-day-cured specimens shown in Fig. 4), and about 1.3 times greater for 16F than for 16C (28-day-cured specimens shown in Fig. 5). Similarly, the data shown in Figs. 6-8 for the leaching of strontium represent duplicate tests in which the specimens were from different batches of grout. Specimens 13A, 13B, and 13C were from one batch of grout, while the corresponding specimens 13D, 13E, and 13F were from another. After 160 days of leaching, the cumulative fraction of strontium leached was about 1.4 times greater from 13A than from 13D (0-day-cured specimens shown in Fig. 6),

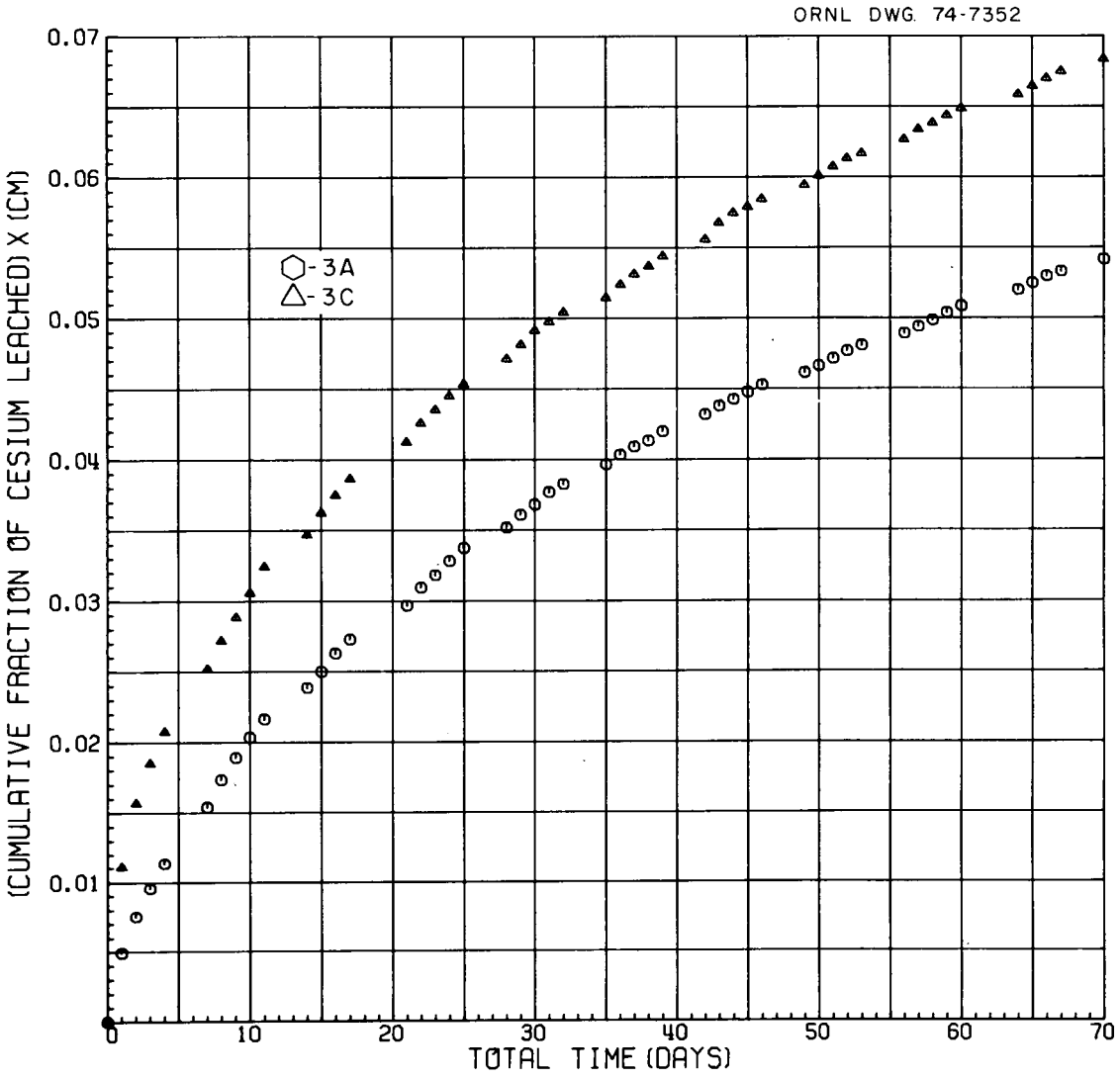

Fig. 2. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts No. 3A and 3C. (The specimens were from the same batch of cementitious grout, were cured 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-22 and A-23 of Appendix A. These specimens were prepared with W-9/10 waste instead of W-7, the compositions of which are given in Table 2.)

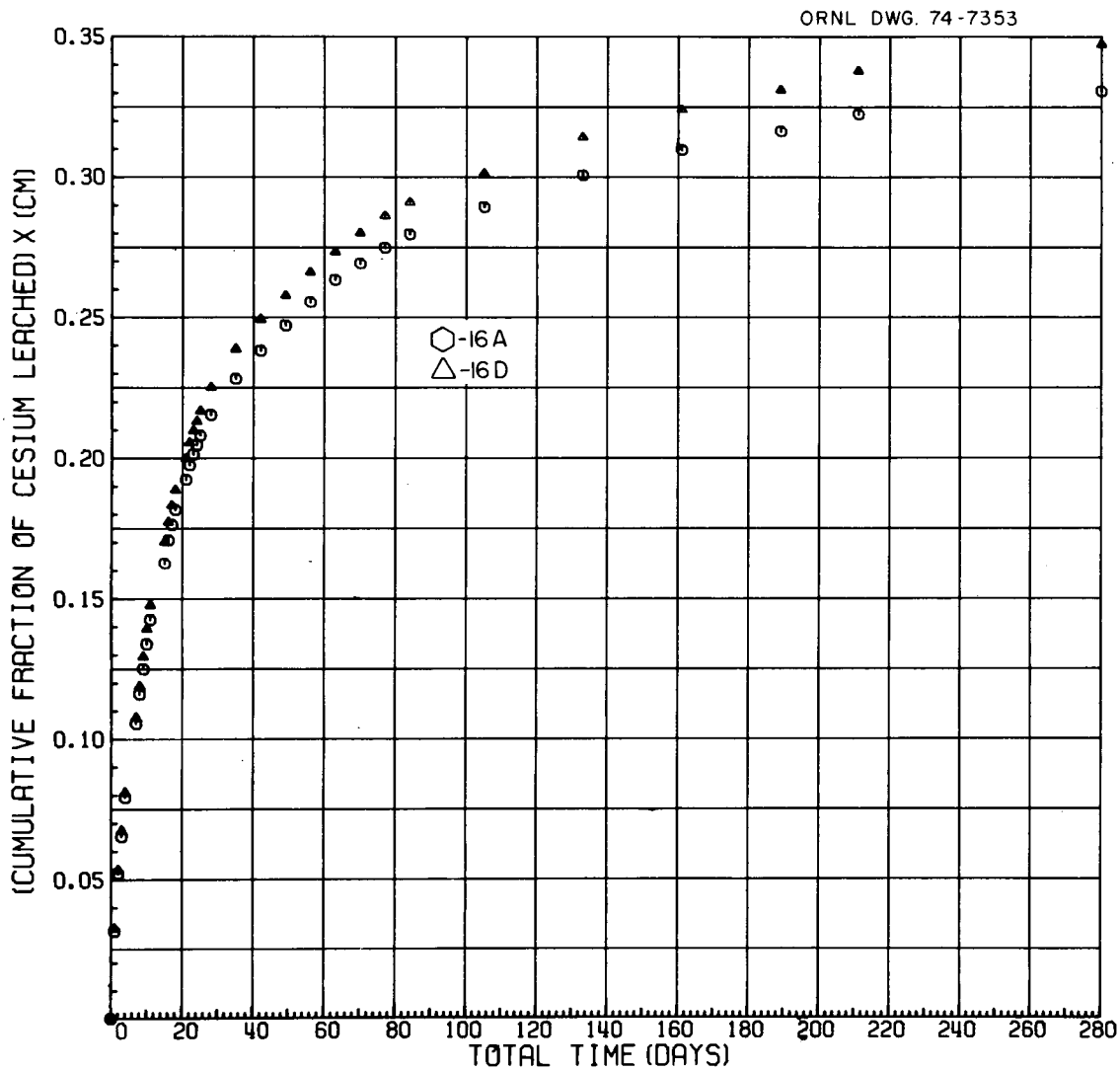

Fig. 3. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts No. 16A and 16D. (The specimens were from different batches of cementitious grout, were cured 0 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-10 and A-13 of Appendix A.)

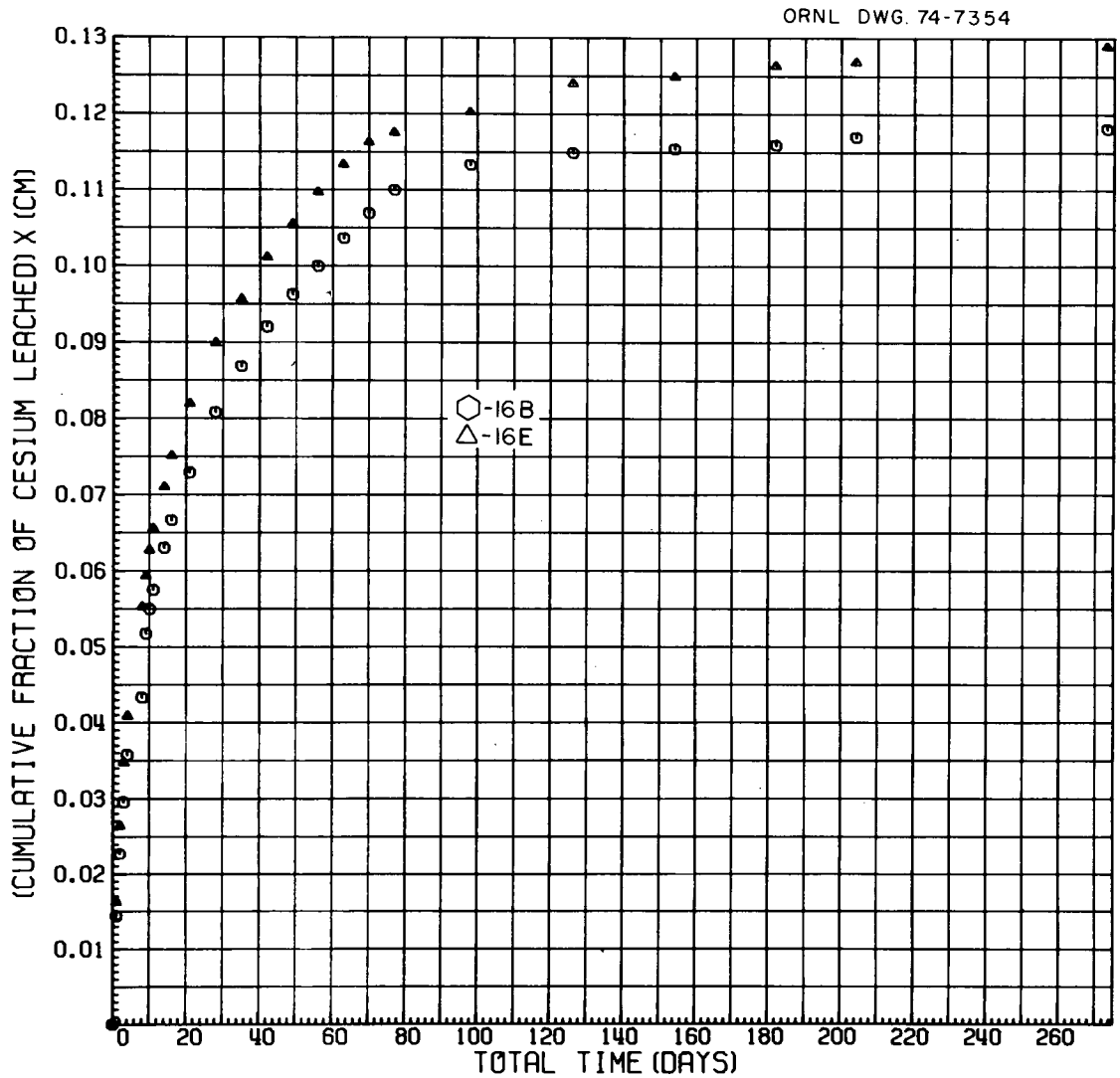

Fig. 4. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts No. 16B and 16E. (The specimens were from different batches of cementitious grout, were cured 7 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-11 and A-14 of Appendix A.)

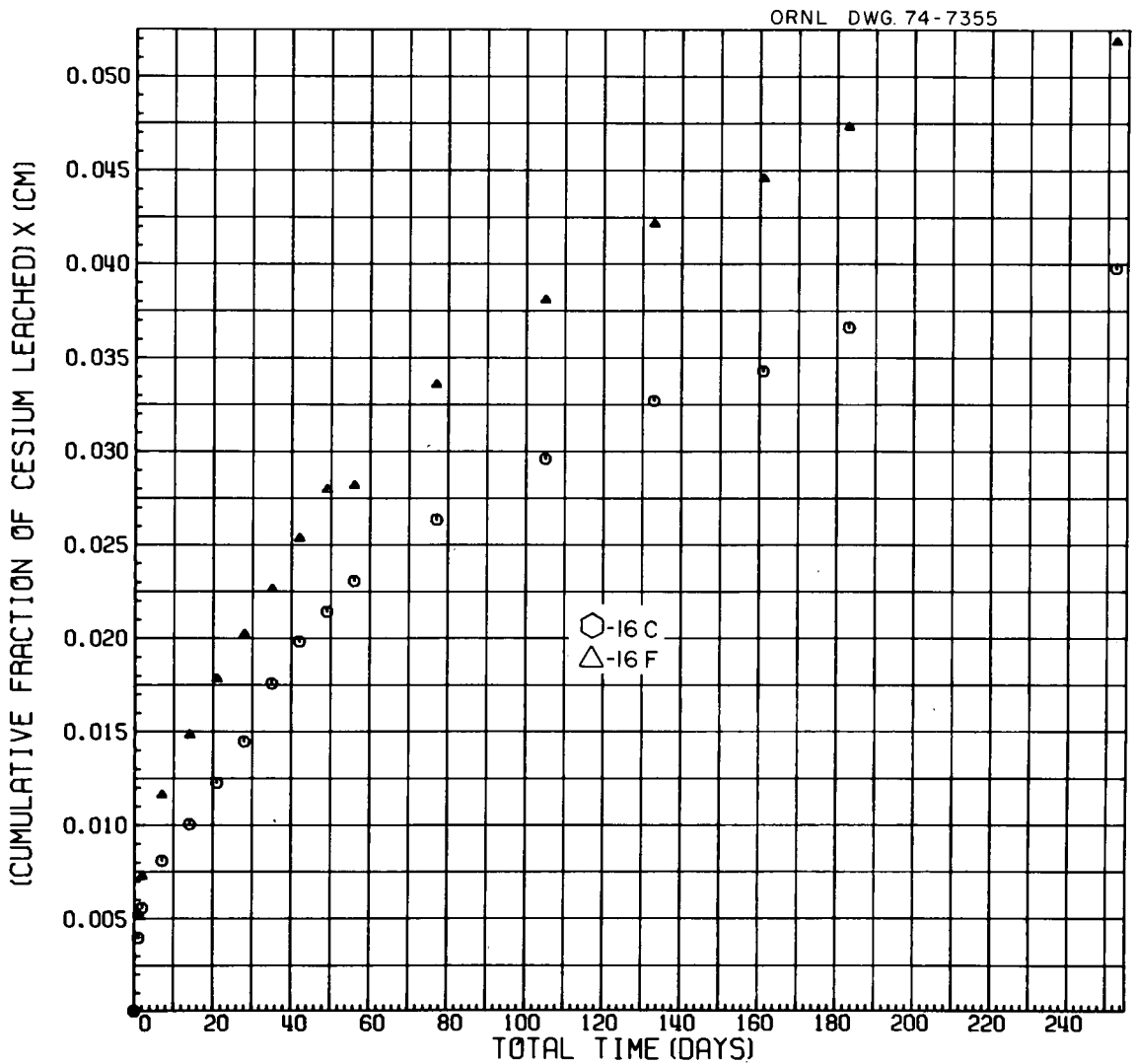

Fig. 5. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts No. 16C and 16F. (The specimens were from different batches of cementitious grout, were cured for 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-12 and A-15 of Appendix A.)

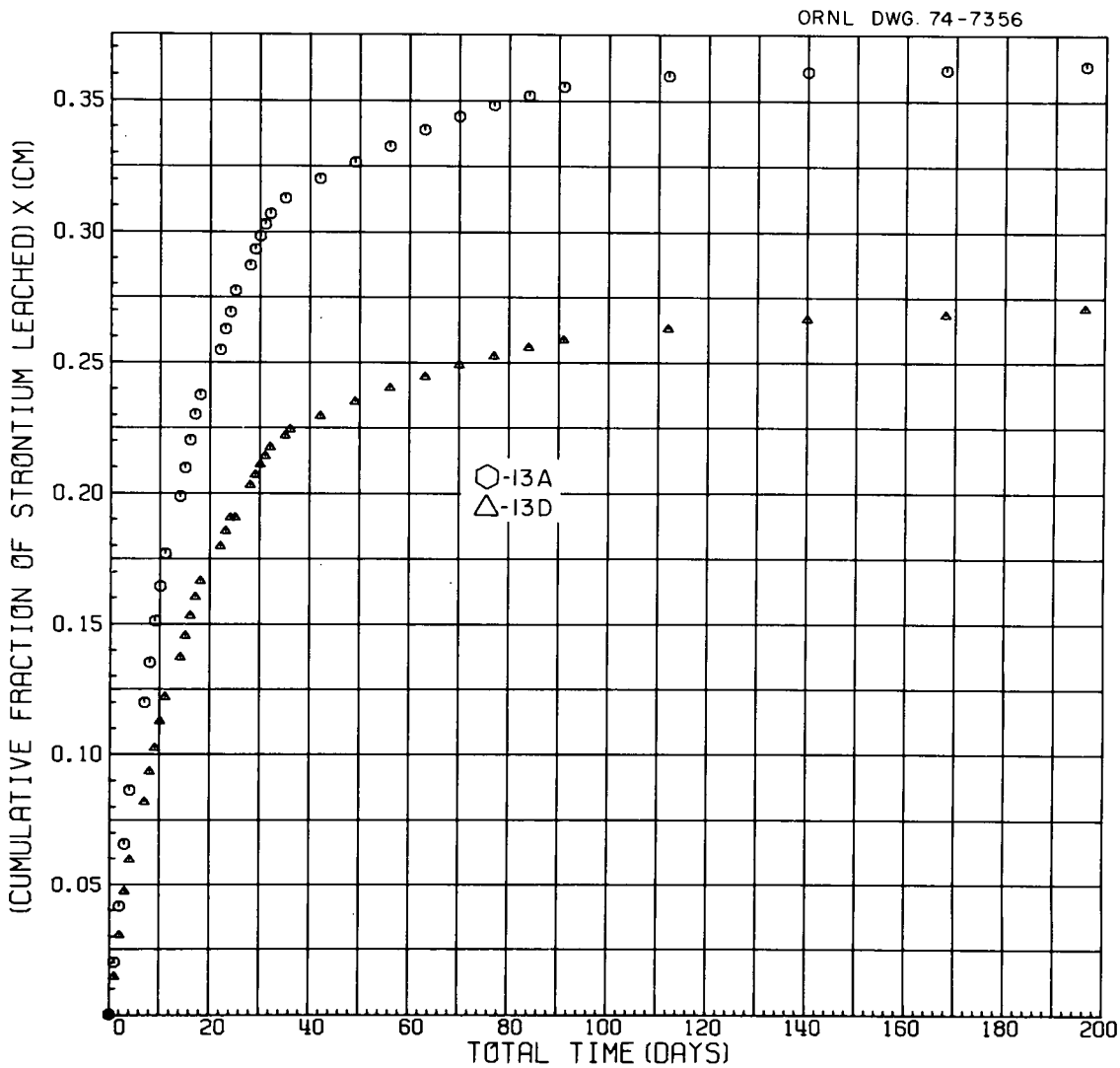

Fig. 6. Product of the Cumulative Fraction of Strontium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 13A and 13D. (The specimens were from different batches of cementitious grout, were cured 0 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-1 and A-4 of Appendix A.)

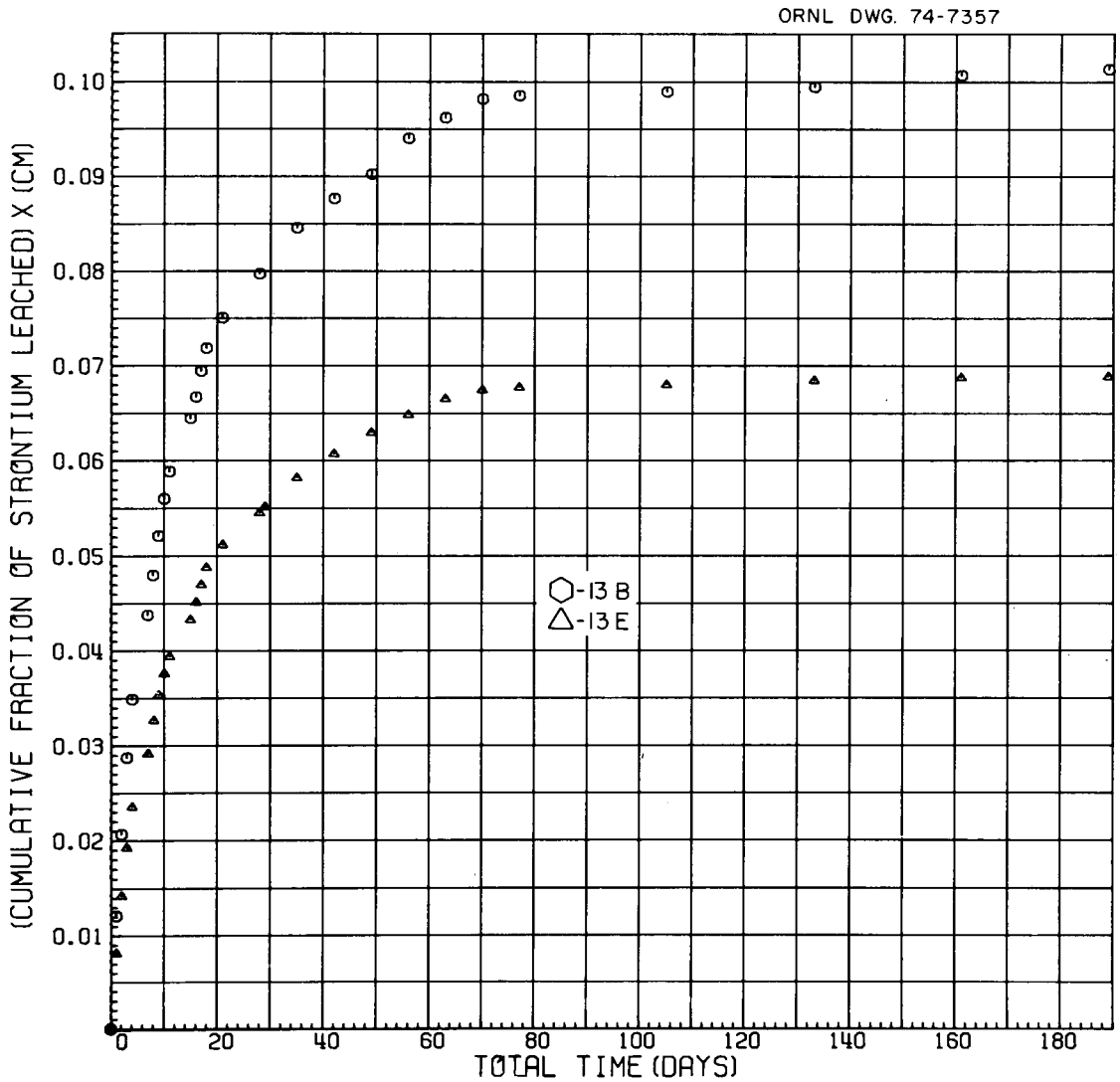

Fig. 7. Product of the Cumulative Fraction of Strontium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 13B and 13E. (The specimens were from different batches of cementitious grout, were cured 7 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-2 and A-5 of Appendix A.)

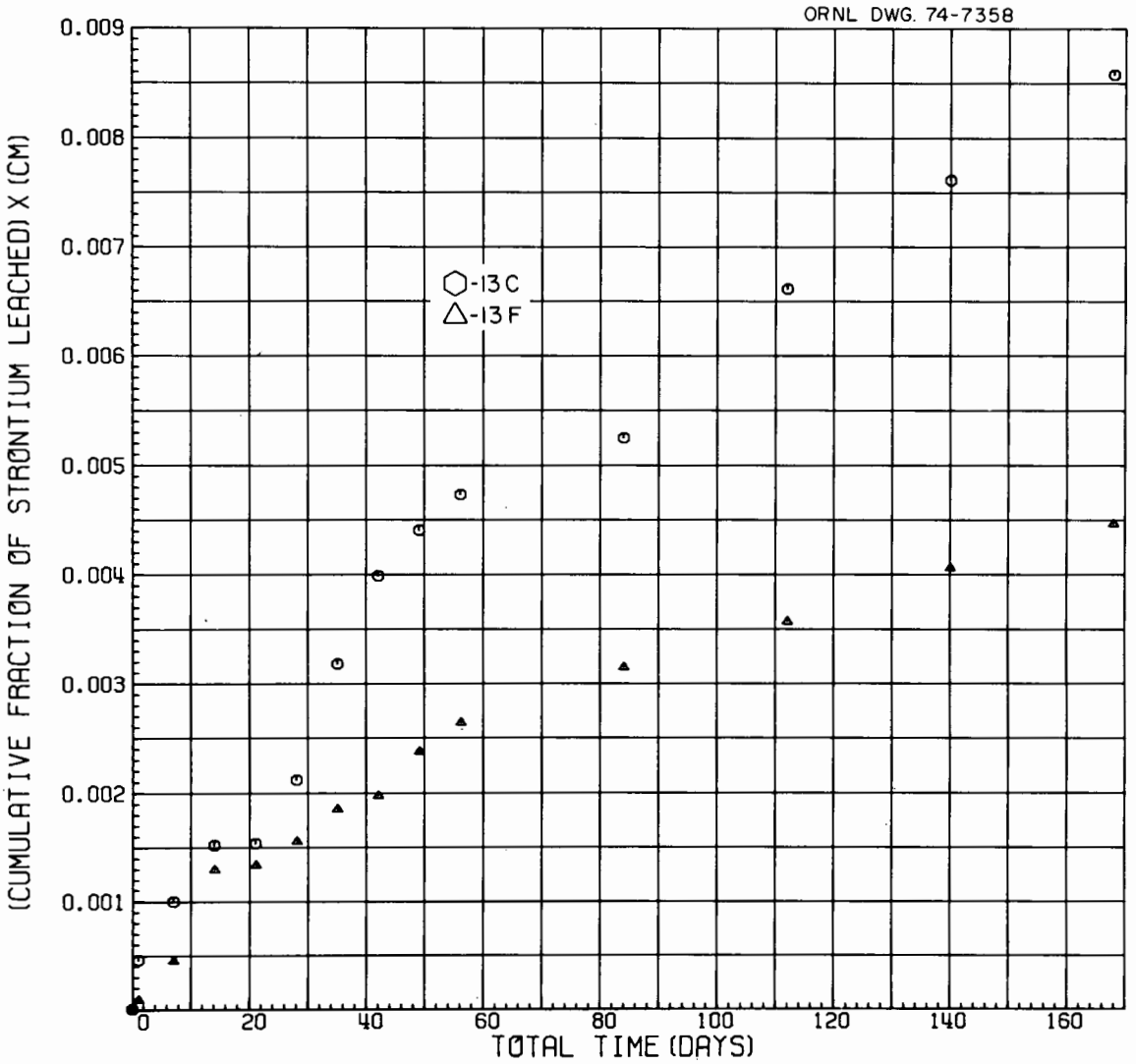

Fig. 8. Product of the Cumulative Fraction of Strontium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 13C and 13F. (The specimens were from different batches of cementitious grout, were cured 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-3 and A-6 of Appendix A.)

about 1.5 times greater from 13B than from 13E (7-day-cured specimens shown in Fig. 7), and about 2 times greater from 13C than from 13F (28-day-cured specimens shown in Fig. 8).

These comparisons indicate that the values for the fraction leached from replicate tests may vary by as much as a factor of 2 (e.g., Fig. 8). This emphasizes the difficulty frequently encountered in duplicating results with complex, heterogeneous materials such as cements, concretes, mortars, and grouts. The physical and chemical properties of such systems often depend on factors such as mixing speed, mixing time, curing time, and curing conditions, which are difficult to control precisely. Curing conditions would include wet (underwater) vs air curing, temperature, and pressure, as well as the humidity and carbon dioxide content of the air.

4.2 Effect of Leachant Renewal Frequency

In the course of the tests with one or two leachant renewals per day, it became apparent that the longer sampling periods represented by weekends (which introduced a 2- to 3-day sampling period) were influencing the leach results. In other words, the leach curve was shifted (to the right) by the longer weekend leaching periods but returned, approximately, to the slope that was observed during the previous week (see Figs. 9 and 10). Figure 9 presents results for the leaching of cesium from specimens 93A and 93C (from one batch of grout); Fig. 10 gives the data for 94A and 94C (from a second batch of grout). Each of these specimens had been cured 28 days prior to leaching. In the case of the A specimens, the leachant was replaced approximately once per day for the first two months and then approximately once per week for approximately three months. The leachant for the C specimens was replaced approximately twice per day for the first two months and then once per day for nearly three months.

Although not out of the range of repeatability, the results for each pair of specimens (93A-93C in Fig. 9 and 94A-94C in Fig. 10) suggest that the cumulative fraction leached (for each product) increases as the leachant renewal frequency increases. To illustrate the effect that sampling frequency has on the fraction leached, the sampling schedules of specimens

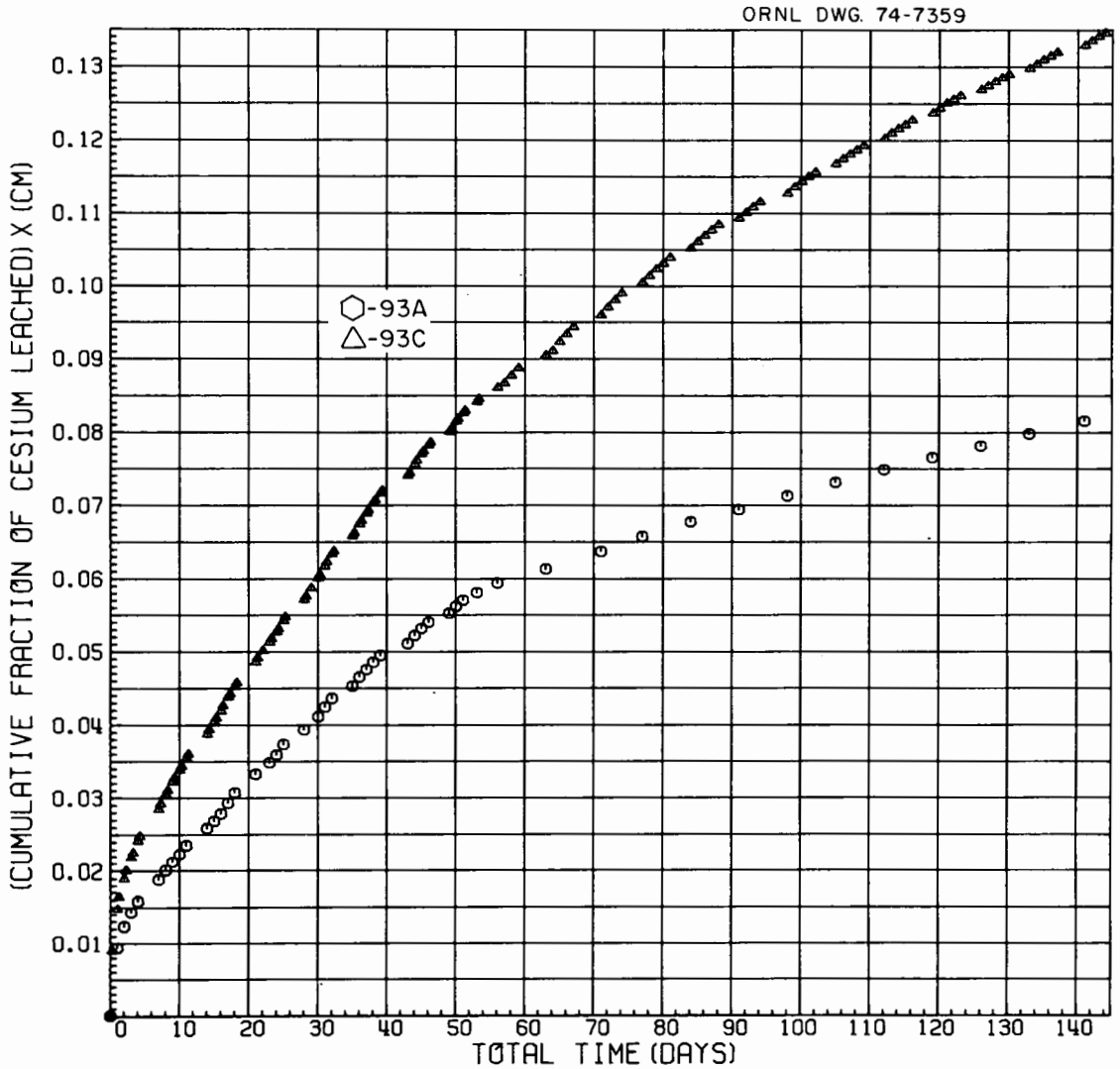

Fig. 9. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 93A and 93C. (The specimens were from the same batch of cementitious grout, were cured 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-18 and A-19 of Appendix A.)

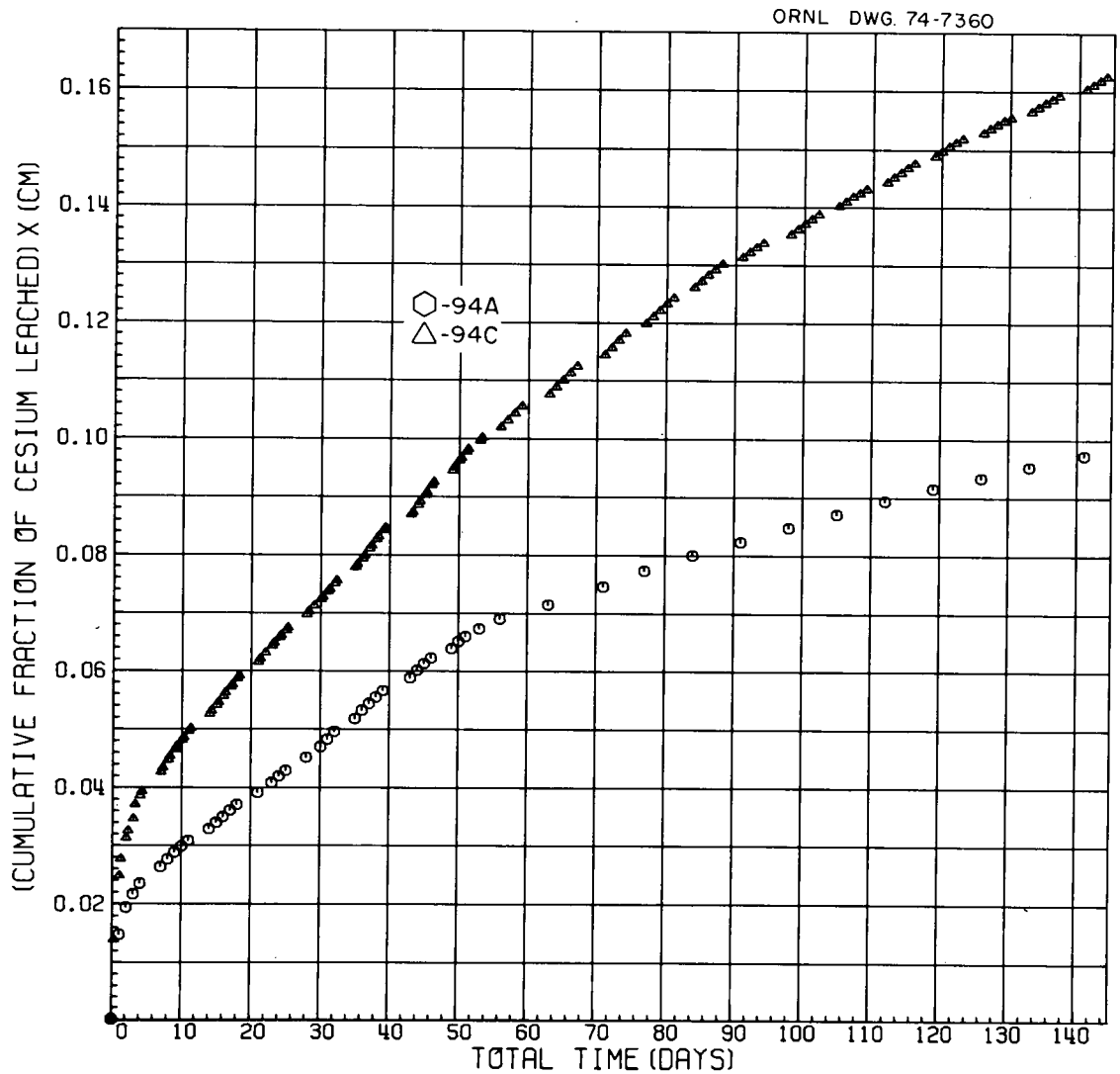

Fig. 10. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 94A and 94C. (The specimens were from the same batch of cementitious grout, were cured 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-20 and A-21 of Appendix A. These specimens contained 7 lb instead of 6 lb of dry mix per gallon of waste. The dry mix formula is given in Table 1.)

86 and 91 were varied several times during the leach tests. In these tests, cesium was the isotope being considered. Specimens 86 and 91 were cured 84 days and 168 days, respectively, prior to leaching. The results, which are plotted in Figs. 11 and 12, show that the slope of the leach curve changes abruptly when the sampling frequency changes. These figures also show that the curve is shifted to the right and its slope decreases when leachants remain in contact with the specimen for relatively long periods of time compared with the nominal sampling interval previously in effect. Consider, for example, the region in Fig. 11 from about the 49th to the 92nd day, when the sampling frequency was nominally once per week, and the region from about the 92nd to the 176th day, when the sampling frequency was once per day (with several slightly longer sampling periods interspersed, which appear as gaps between the data points). The slope of the cumulative-fraction-leached-vs-time curve is markedly different for each of the regions, and a significant gap or shift is caused by the longer interval between the 115th and the 135th days. From about the 176th to the 210th day, the leachant renewal frequency was again once per week; from the 210th to the 245th day, it was twice per week. Similar considerations of Fig. 12 reveal the same type of pattern.

These results suggest that the leach data are being influenced by the concentration of the species (the isotope of concern as well as the matrix material) in the leachant and/or surface concentration effects. Speaking broadly, this implies that the desired experimental conditions (mathematically, boundary conditions) of essentially negligible concentrations of <u>all</u> species in the leachant and of essentially negligible concentration of the isotope of concern at the leachant-solid interface are probably not being achieved. Apparently, with glass, asphalt, and common cement products, the IAEA standard leach test tacitly assumes that these conditions will be satisfied if a daily sampling schedule is maintained until the leach rate has become virtually constant or if the pH of the leachant (initially at a pH of about 7 for the demineralized water in the IAEA test) does not rise above 8 (ref. 15).

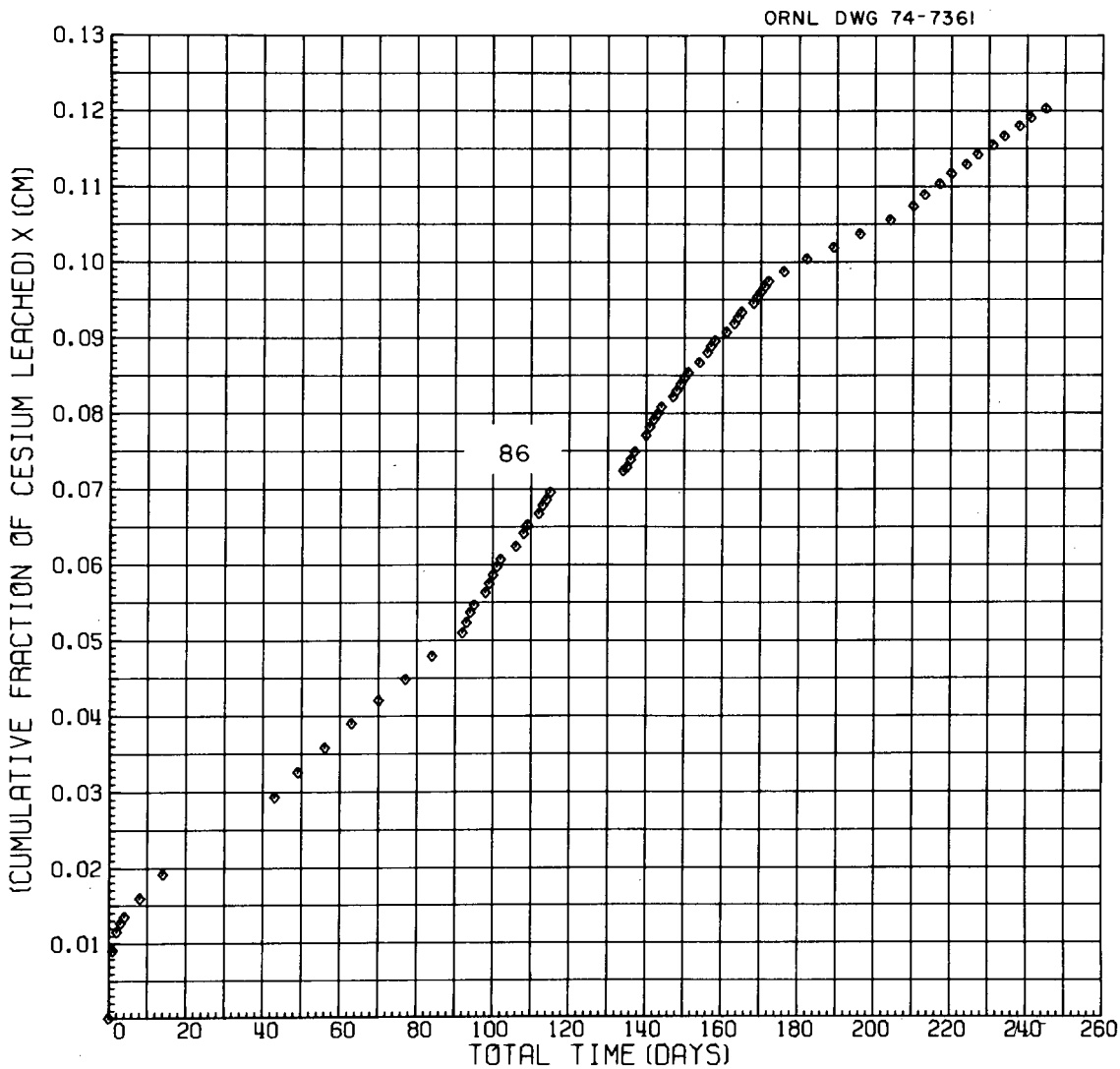

Fig. 11. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grout 86. (The specimen was cured 84 days before leaching commenced and had a volume/exposed surface ratio of 5. The data points are given in Table A-16 of Appendix A.)

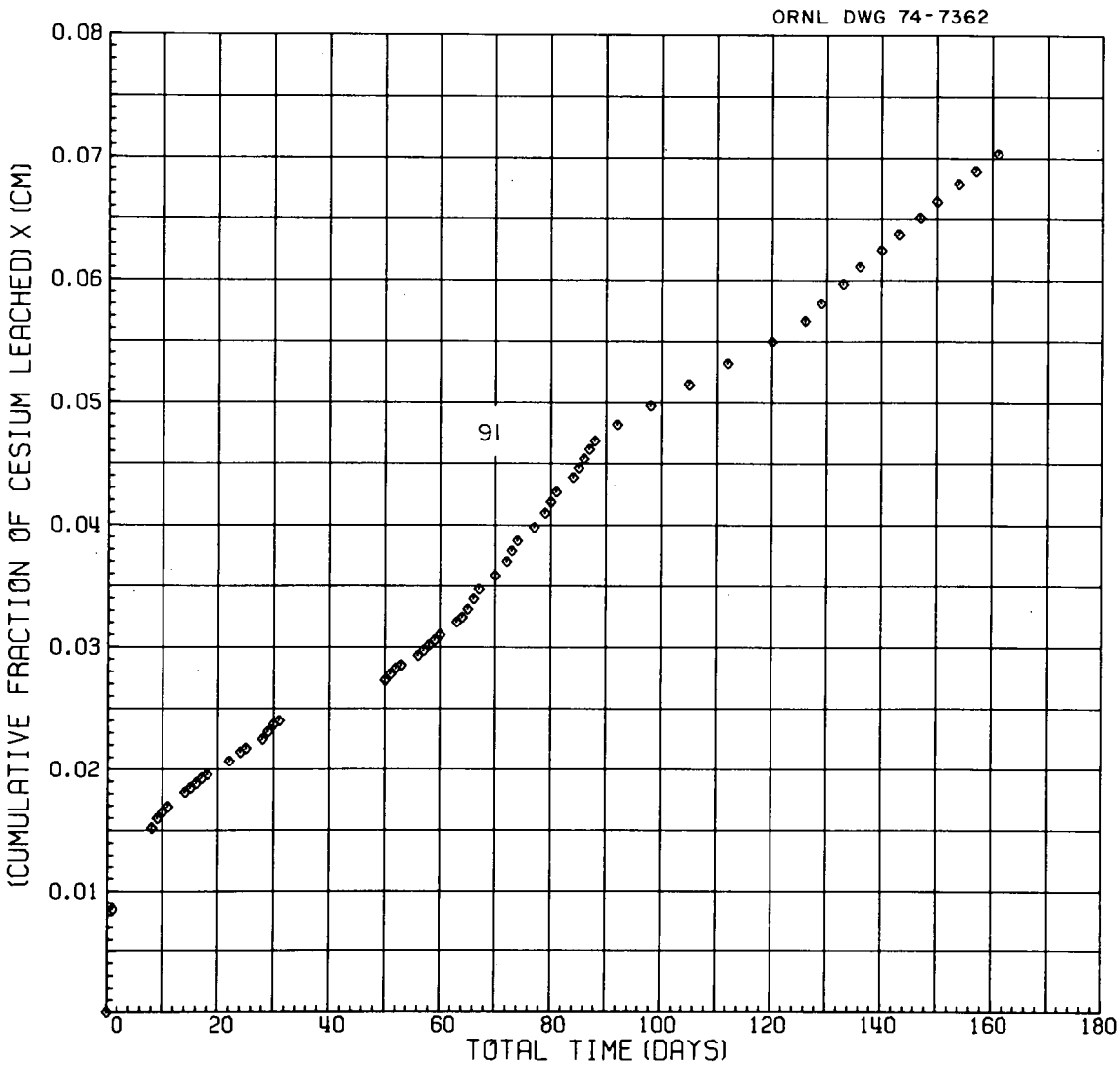

Fig. 12. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grout 91. (The specimen was cured 168 days before leaching commenced and had a volume/exposed surface ratio of 5. The data points are given in Table A-17 of Appendix A.)

4.3 Effect of Air Curing Time

As mentioned previously in Sect. 2.2, curing time refers to the length of time that the specimen was allowed to cure in air before leaching with water was initiated. No attempt is made to analyze the results in terms of total (i.e., air plus wet) curing time since the sets of data are not mutually comparable with respect to isotope, sampling frequency, etc.

Figure 13 shows the results obtained for the leaching of strontium from specimens 13A (0-day cure), 13B (7-day cure), and 13C (28-day cure), all of which were prepared from the same batch of grout. Figure 14 gives the results obtained by leaching cesium from specimens 16A (0-day cure), 16B (7-day cure), and 16C (28-day cure), which are also from one batch of grout. The results presented in Figs. 15 and 16 are for plutonium and curium specimens, respectively, with curing times of 0, 7, and 28 days. The specimens are from the same batch of grout in each case.

Comparison of the leach data for the 0-, 7-, and 28-day-cured specimens of each isotope in Figs. 13-16 leads to the conclusion that longer air curing times, up to 28 days at least, decrease the cumulative fraction leached from a product. This conclusion appears to hold even when allowance is made for the ability to duplicate a test and for the effect of sampling frequency (the leachant was replaced more frequently on the specimens with shorter curing times).

Figure 17 compares the results obtained for the leaching of cesium from specimens 16C and 16F (28-day cure), 86 (84-day cure), and 91 (168-day cure). These results suggest that a curing time of longer than 28 days has no significant effect on the cumulative fraction of cesium leached from a specimen. This conclusion gains further support when allowance is made for the different sampling frequencies (86 and 91 were sampled more frequently than 16C and 16F) and for the ability to duplicate specimens and repeat leach tests. A leach test currently in progress with a grout cured for 365 days in air, in which the sampling frequency is similar to that of 93A (Table A-15 in Appendix A), is giving values that are about 50% less than those of 93A after 70 days of leaching. The 365-day-cured specimen and the 28-day-cured specimen (93A) were

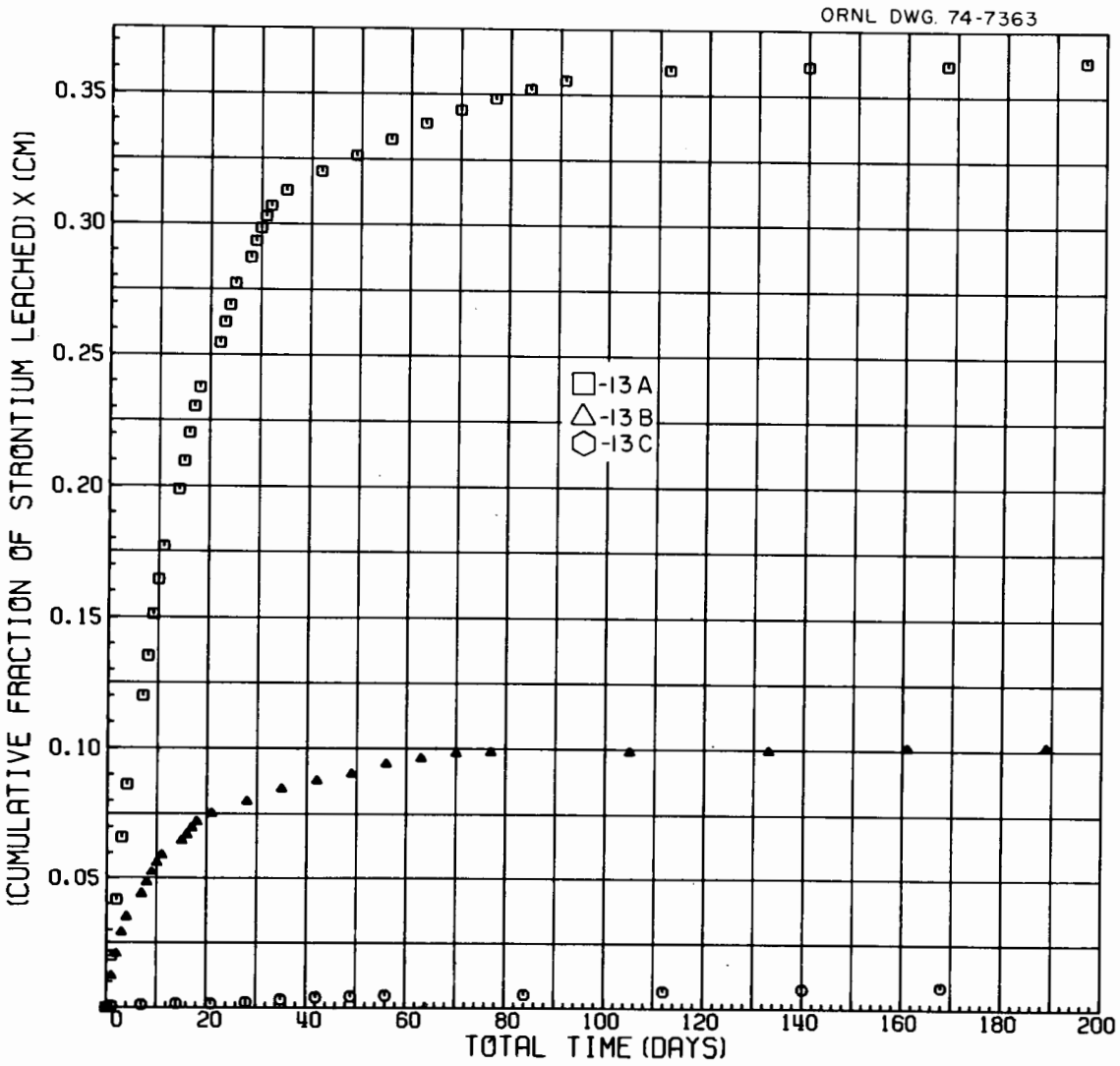

Fig. 13. Product of the Cumulative Fraction of Strontium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 13A, 13B, and 13C. (The specimens were from the same batch of cementitious grout, were cured 0, 7, and 28 days, respectively, before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-1, A-2, and A-3 of Appendix A.)

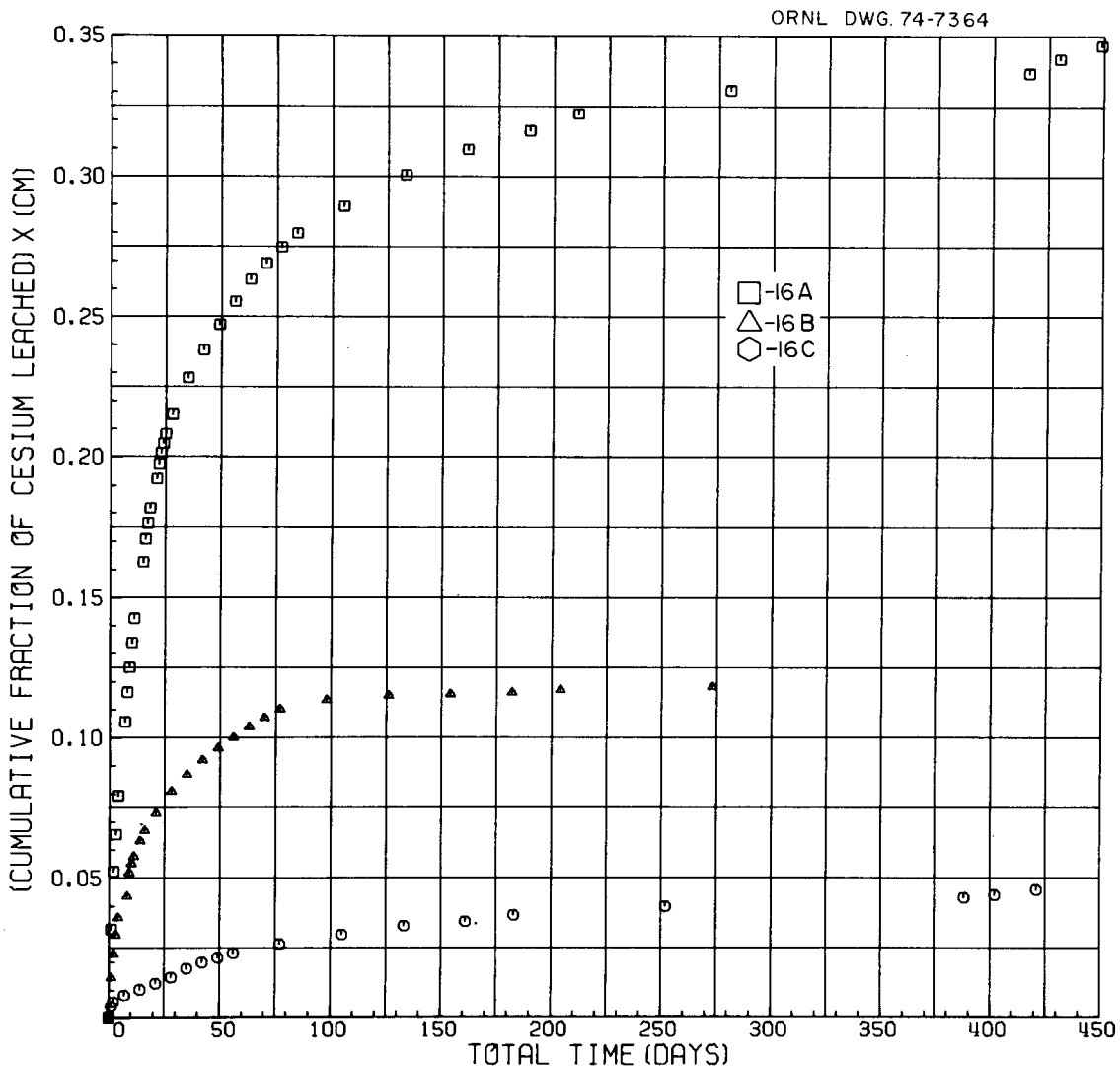

Fig. 14. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 16A, 16B, and 16C. (The specimens were from the same batch of cementitious grout, were cured 0, 7, and 28 days, respectively, before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-10, A-11, and A-12 of Appendix A.)

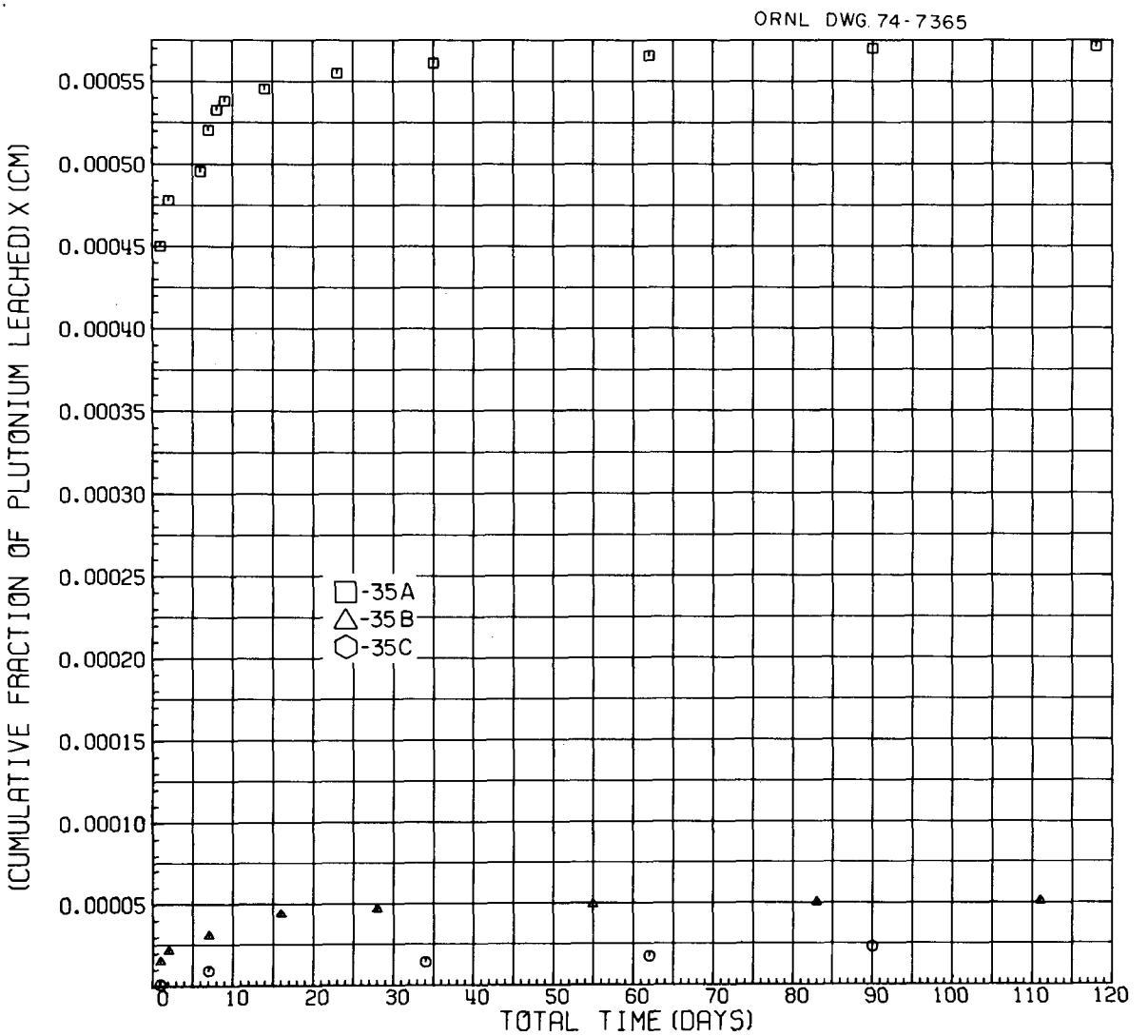

Fig. 15. Product of the Cumulative Fraction of Plutonium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 35A, 35B, and 35C. (The specimens were from the same batch of cementitious grout, were cured 0, 7, and 28 days, respectively, before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-24, A-25, and A-26 of Appendix A.)

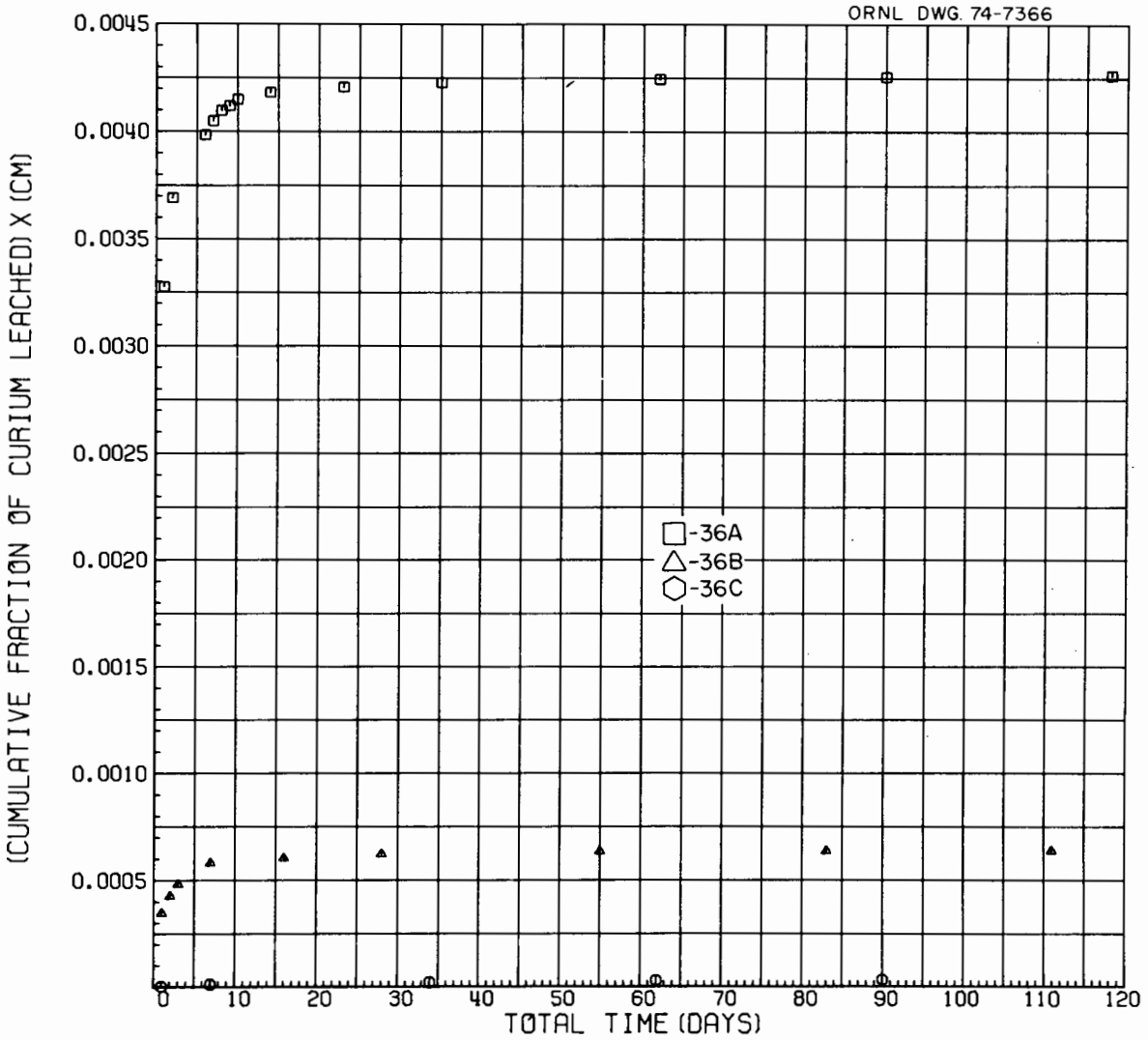

Fig. 16. Product of the Cumulative Fraction of Curium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 36A, 36B, and 36C. (The specimens were from the same batch of cementitious grout, were cured 0, 7, and 28 days, respectively, before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-27, A-28, and A-29 of Appendix A.)

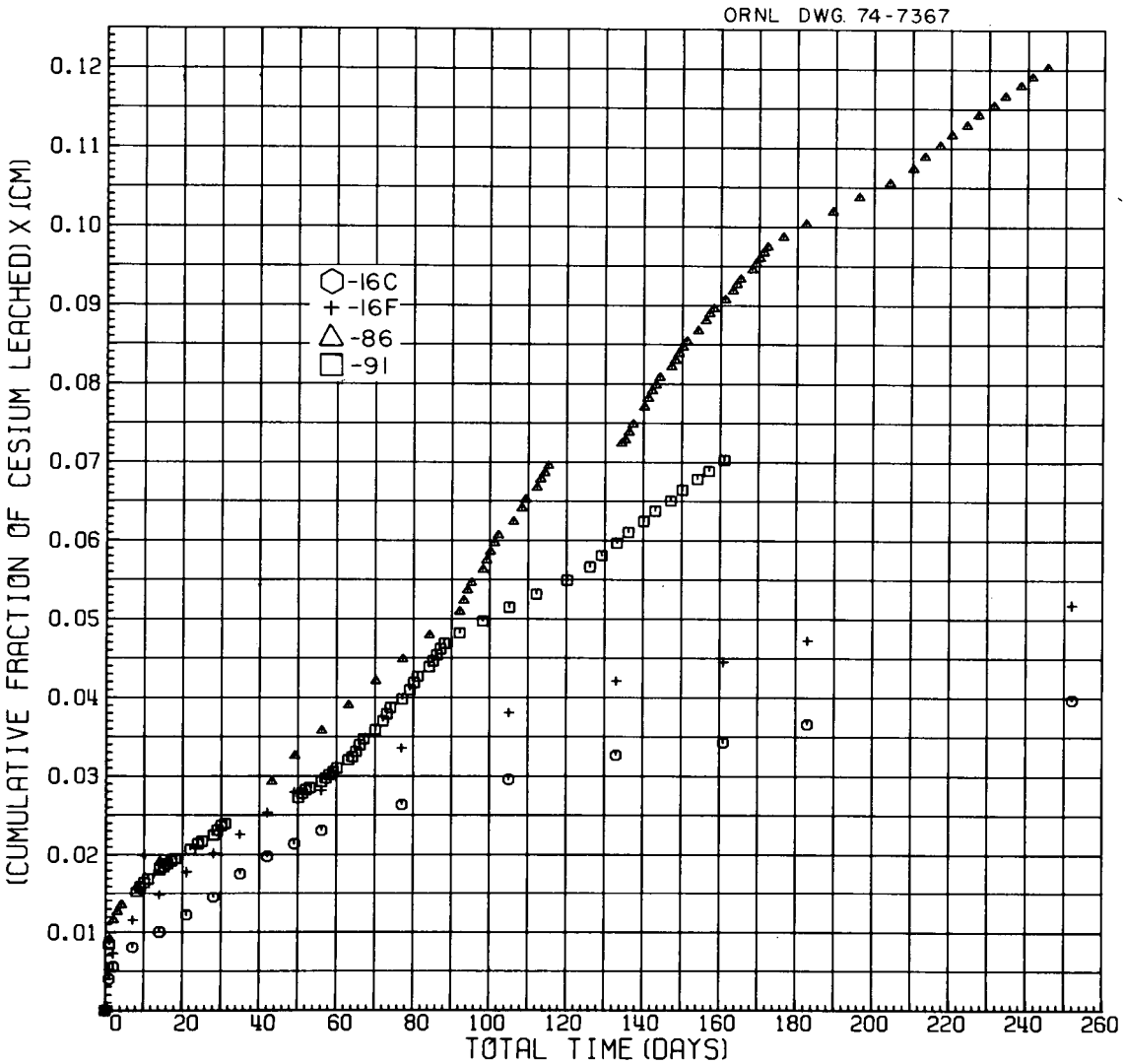

Fig. 17. Product of the Cumulative Fraction of Cesium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 16C, 16F, 86, and 91. (Specimens 86 and 91 were from the same batch of cementitious grout and were cured 84 and 168 days, respectively, before leaching commenced. Specimens 16C and 16F were from different batches of cement grout and were cured 28 days before leaching commenced. All specimens had a volume/exposed surface ratio of 5. The data points are given in Tables A-12, A-15, A-16, and A-17 of Appendix A.)

prepared from different batches of grout (6 lb of dry mix per gal of W-7 waste). However, this difference does not appear to have real significance when the ability to prepare duplicate specimens and to repeat a leach test is considered.

4.4 Comparison of Isotopes

Figure 18 compares the cumulative fraction of cesium leached from two 28-day-cured specimens with the fraction of strontium leached from two 28-day-cured specimens. After about 160 days, the fraction of cesium leached was about 6 times greater than the fraction of strontium leached. The cumulative fraction of plutonium leached from a 28-day-cured specimen is compared with the cumulative fraction of curium leached from a 28-day-cured specimen in Fig. 19. At the end of 90 days, the fraction of curium leached is about 1-1/3 times greater than the fraction of plutonium leached. A cross comparison of Figs. 18 and 19 shows that, at the end of 90 days, the cumulative fraction of cesium leached is about 1400 times greater than the fraction of plutonium leached. Similar comparisons of the 0- and 7-day-cured specimens for these isotopes (Figs. 13-16) lead to the same observation, namely, that the cumulative fractions leached are in the following order: Cs > Sr > Cm > Pu. Qualitatively, the known solubilities of these isotopes in basic solutions would support this order of releases in cases where intrinsic solubility is a major factor influencing the fraction leached. Also, the empirical rules for ionic selectivity on an exchange material would predict the following order of sorption based on valence: $Cs^{+} < Sr^{2+} < Cm^{3+} < Pu^{4+}$.

4.5 Tests with Added Alkaline-Earth Ions

Figure 20 presents the results of the leaching of strontium from 7-day-cured specimens 13B, 13E, and 70. Specimen 70 was prepared using W-7 waste (Table 2), to which 1 g of Sr^{2+} and 0.25 g of Ca^{2+} (as the chlorides) were added per liter. The precipitate was allowed to age 30 days before being incorporated into a grout. The three specimens were from different batches of grout made with 6 lb of dry mix per gallon of W-7 waste. A comparison of the results shown in Fig. 20 leads to the

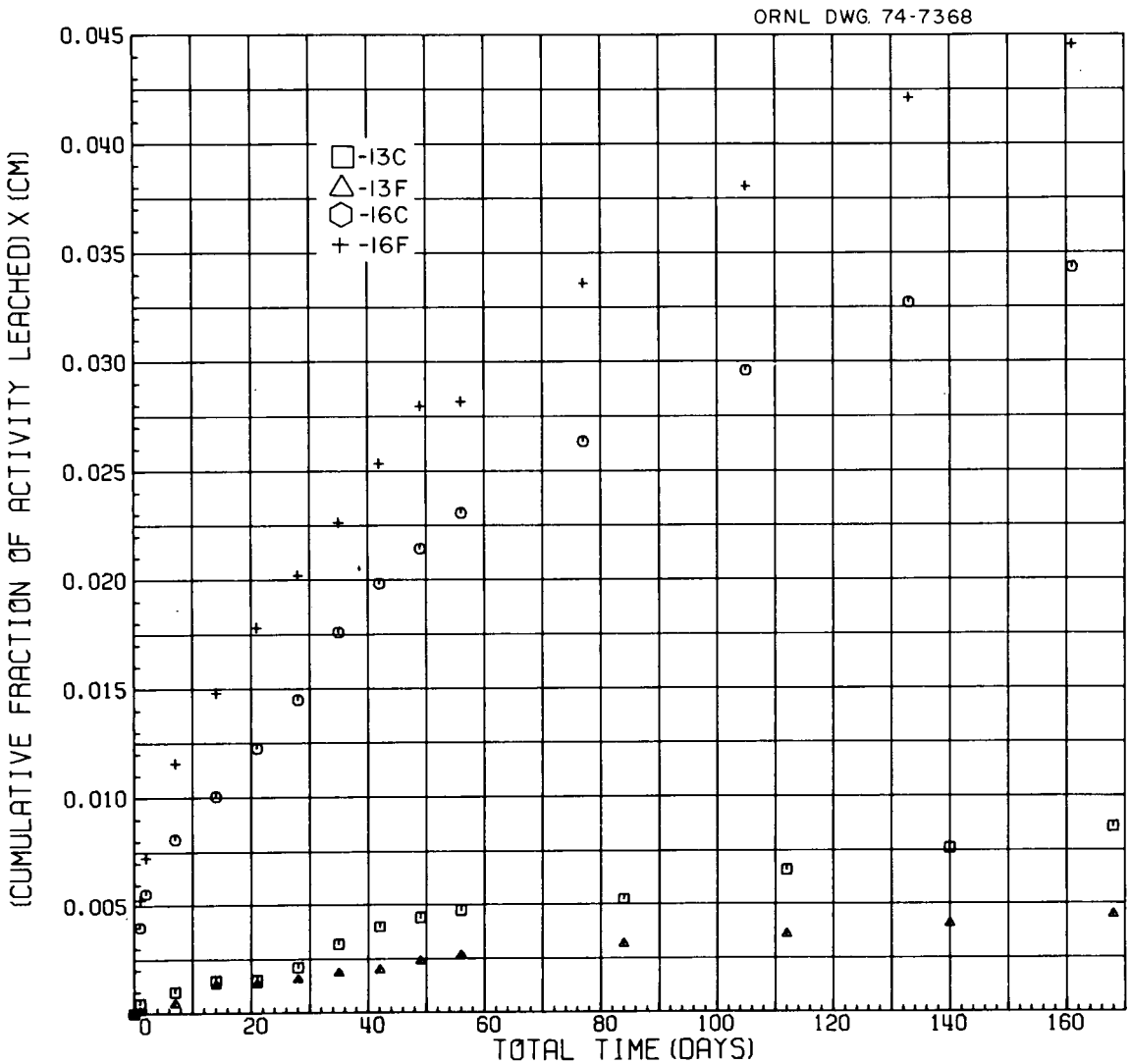

Fig. 18. Product of the Cumulative Fraction of Activity Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 13C, 13F, 16C, and 16F. (Specimens 13C and 13F contained ^{85}Sr; specimens 16C and 16F contained ^{137}Cs. All specimens were from different batches of cementitious grout, were cured 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-3, A-6, A-12, and A-15 of Appendix A.)

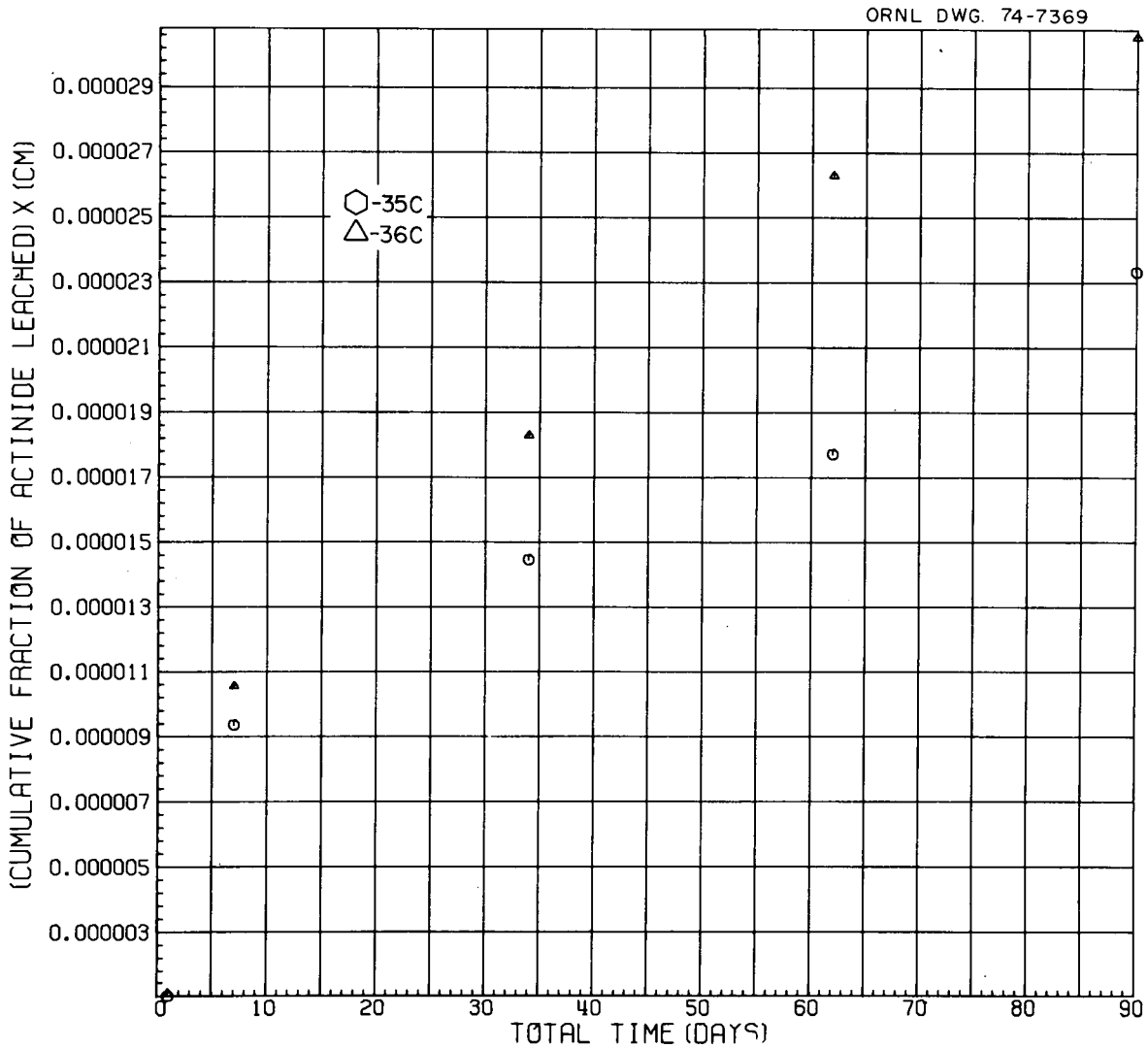

Fig. 19. Product of the Cumulative Fraction of Actinide Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 35C and 36C. (Specimen 35C contained ^{239}Pu; specimen 36C contained ^{244}Cm. The specimens were from different batches of cementitious grout, were cured 28 days before leaching commenced, and had a volume/exposed surface ratio of 5. The data points are given in Tables A-26 and A-29 of Appendix A.)

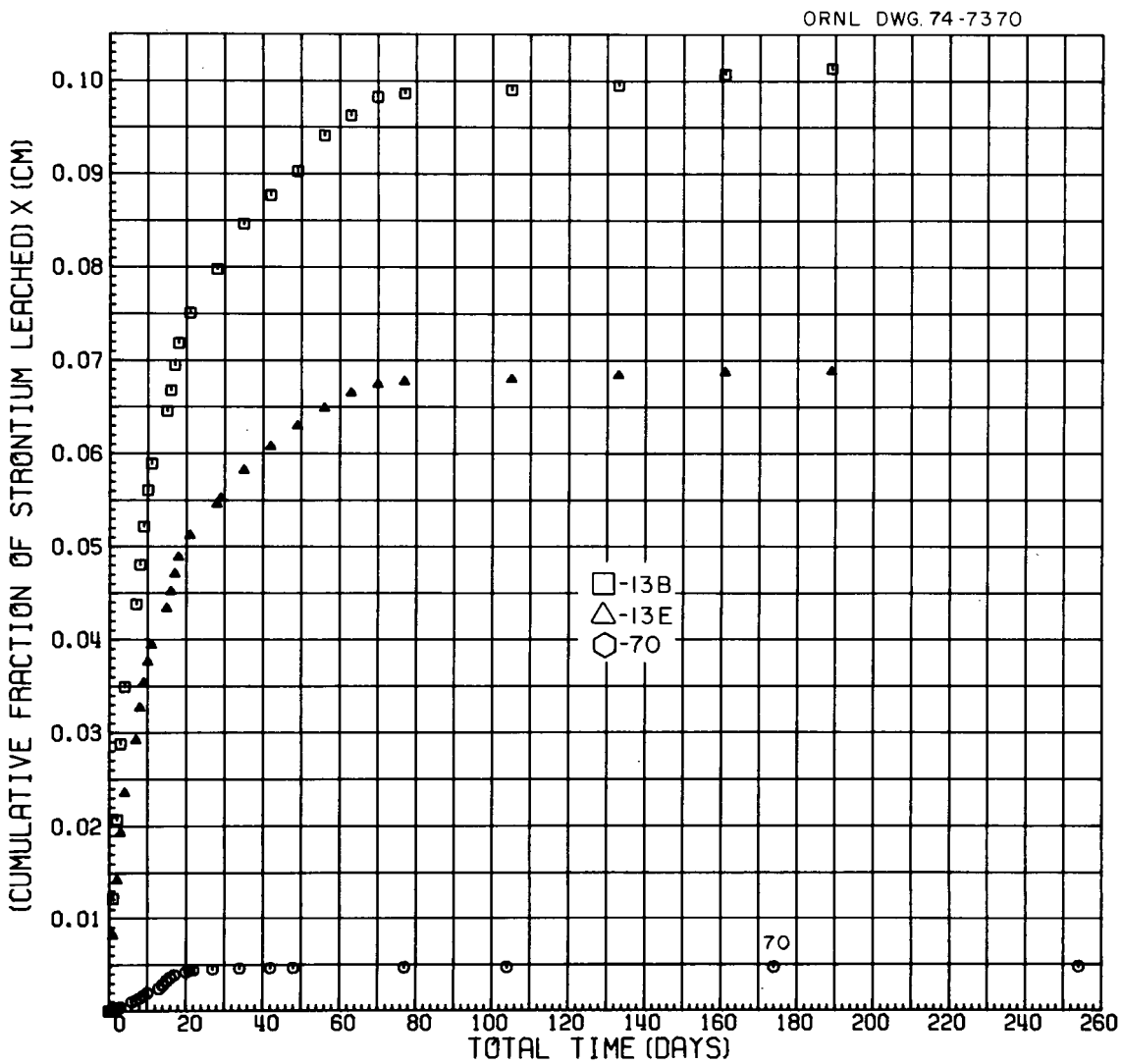

Fig. 20. Product of the Cumulative Fraction of Strontium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_0)(V/S)]$ Plotted Against Time for Cementitious Grouts 13B, 13E, and 70. (The specimens were from different batches of cementitious grout. Specimen 70 was prepared with waste to which 1 g of Sr^{2+} and 0.25 g of Ca^{2+} were added per liter. All specimens were cured 7 days and had a volume/exposed surface ratio of 5. The data points are given in Tables A-2, A-5, and A-7 of Appendix A.)

conclusion that the cumulative fraction of strontium leached from specimen 70 (with the added Sr^{2+} and Ca^{2+}) is a factor of ~ 20 lower than the fraction leached from specimens 13B and 13E (no Sr^{2+} or Ca^{2+} added). However, it should be kept in mind that specimen 70 (106 mg of Sr^{2+}) contains more strontium than 13B and 13E (30 mg of Sr^{2+} present as impurities in the ingredients, cf. Tables A-7 with Tables A-2 and A-5 in Appendix A). Thus, the total amount of strontium leached from specimen 70 is only a factor of about 6 lower than the amount leached from 13B and 13E. On the other hand, this decrease in the amounts leached is sufficiently large (out of the range of replication of results) to suggest that the effort to take advantage of the low solubility product constant of difficultly soluble strontium compounds and the dilution of radioactive strontium with stable strontium is worthwhile. Additional tests that compare the amount, as well as the fraction, leached as a function of the amount of alkaline earth added to the waste are needed to evaluate the solubility product constant and the isotopic dilution effect.

Four specimens were prepared with W-7 waste to which 0.05 g of Sr^{2+} was added per liter and then allowed to cure for 28, 56, 112, and 168 days respectively. After leaching commenced, the 28- and 56-day cured specimens began, inexplicably, to pit and spall and were discarded. The results for the 112- and 168-day cured specimens are given in Fig. 21. Although these results are not directly comparable to any other data presented in this study, they are given in the interest of completeness and also to show the lowest cumulative-fraction-leached-vs-time results obtained with strontium.

5. COMPARISON OF RESULTS WITH THEORETICAL EXPRESSIONS

As pointed out in Sect. 4.2, the desired boundary condition of essentially negligible concentration of the isotope of concern at the leachate-solid interface was probably not realized in these tests with cementitious grouts. Apparently, the reason for this was too infrequent replacement of leachant. Often, the theoretical expressions applied (e.g., ref. 12) to such systems assume that this boundary condition is either met or closely

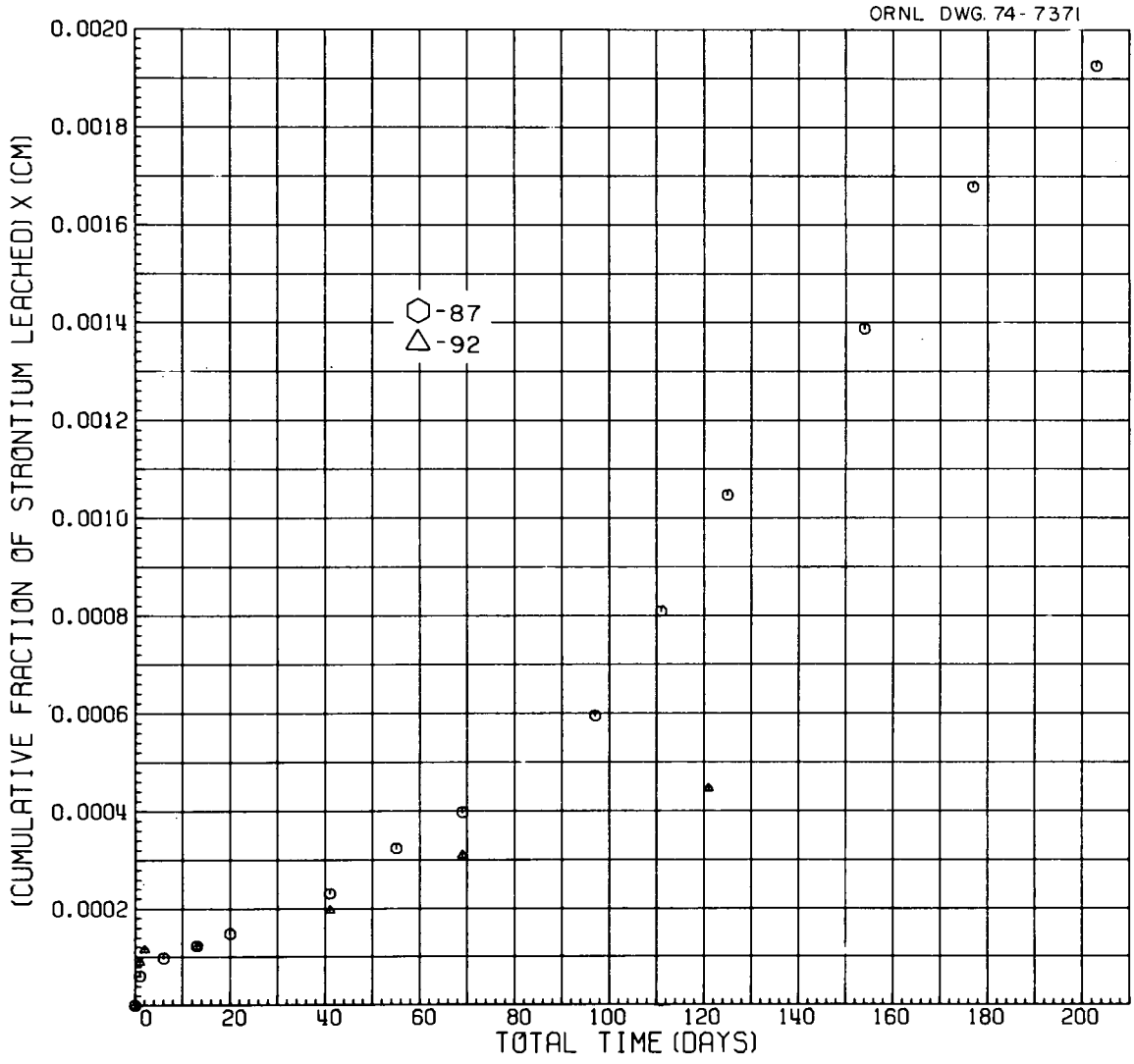

Fig. 21. Product of the Cumulative Fraction of Strontium Leached and the Volume/Surface Ratio $[(\Sigma a_n/A_o)(V/S)]$ Plotted Against Time for Cementitious Grouts 87 and 92. (The specimens were from the same batch of cementitious grout, and were prepared with waste to which 0.05 g of Sr^{2+} was added per liter. Specimen 87 was cured 112 days and specimen 92 was cured 168 days before leaching commenced. Each had a volume/exposed surface ratio of 5. The data points are given in Tables A-8 and A-9 of Appendix A.)

approximated. To illustrate how sampling frequency may be influencing the parameters obtained in empirical and theoretical modeling studies, the data for the leaching of cesium from each of four 28-day-cured specimens (93A, 93C, 94A, and 94C) have been fitted to a theoretical expression which takes into account diffusion and instantaneous reaction (i.e., an equilibrium or time-independent relationship between two forms of a species).

If the mechanisms by which less-mobile forms of a species are converted to more-mobile (diffusible) forms are very rapid compared with diffusional processes, local equilibrium can be assumed to exist between the two forms of the species. Further, if the concentration of less-mobile forms is assumed to be directly proportional to that of the more-mobile forms, the relationship between them is given by:

$$F = KC, \tag{7}$$

where K is the proportionality or equilibrium constant (dimensionless), F is the concentration of less-mobile forms of the species (amount/cm^3), and C is the concentration of more-mobile forms of the species (amount/cm^3).

Parametric studies like those made by Bell[11] show that a specimen such as recommended by the IAEA test procedure[15] will approximate a semi-infinite medium for very long periods of time with most of the products considered to be acceptable waste forms. Therefore, let us consider a semi-infinite medium with a uniform initial concentration of more- and less-mobile forms of a species and with a relationship between the forms as given in Eq. (7). If the surface concentration of more-mobile forms is maintained at zero for time greater than zero, the solution of mass transport equations (Appendix B) for the case of diffusion with reaction as in Eq. (7) yields

$$\left(\frac{\Sigma a_n}{A_o}\right)\left(\frac{V}{S}\right) = 2\left(\frac{D_e}{\pi}\right)^{1/2} t^{1/2} \tag{8}$$

for the cumulative fraction of the species that has left the medium in time t. In Eq. (8), D_e is a modified effective diffusivity (cm^2/sec)

which is defined in Appendix B as $\mathcal{D}_e/(1 + K)$, t is time (sec), V is the volume of the specimen (cm^3), and S is the surface area exposed to leachant (cm^2). Equation (8) is the same as that obtained for the corresponding case of diffusion only, except that a modified effective diffusivity (which includes the equilibrium constant) is used.

The data obtained for specimens 93A and 94A (Tables A-18 and A-20 in Appendix A) during the first 56 days of leaching were fitted to Eq. (8) to give D_e's of 5.4×10^{-10} cm^2/sec and 7.4×10^{-10} cm^2/sec respectively. Comparisons of the experimental values with the cumulative fraction leached as predicted by Eq. (8) using these D_e's are shown in Figs. 22 and 23. As may be seen, the fits are good when we consider that Eq. (8) is a one-constant expression. The sampling frequency during the 56-day period was once per day; after 56 days, it was approximately once per week until the end of the test (at 141 days).

All the data for specimens 93A and 94A were fitted to Eq. (8) to give D_e's of 5.0×10^{-10} cm^2/sec and 6.9×10^{-10} cm^2/sec respectively. The predicted curves and the data points are shown in Figs. 24 and 25. The less frequent sampling after 56 days appeared to depress the amount leached to some extent, as shown by the distinct decrease in the slope of the leach curves after 56 days. This decrease in slope is reflected in D_e as an apparent decrease, that is, 5.4×10^{-10} vs 5.0×10^{-10} cm^2/sec for 93A (cf. Fig. 22 with Fig. 24) and 7.4×10^{-10} vs 6.9×10^{-10} cm^2/sec for 94A (cf. Fig. 23 with Fig. 25). Of course, this result could be explained by other factors such as a time-dependent D_e or depletion mechanism (curing of the grout) that tends to convert more-mobile forms of the species to less-mobile forms with time.

The data for specimens sampled more frequently than 93A and 94A were fitted to Eq. (8) in an effort to determine whether the apparent decrease in D_e with decreasing sampling frequency is caused mainly by a failure to meet boundary conditions or whether it is the result of trying to fit data to a single-constant equation when it should be fitted to a several-constants (several-mechanisms) equation. The data for 93C and 94C (Tables A-19 and A-21 in Appendix A) represent tests in which leachant was replaced

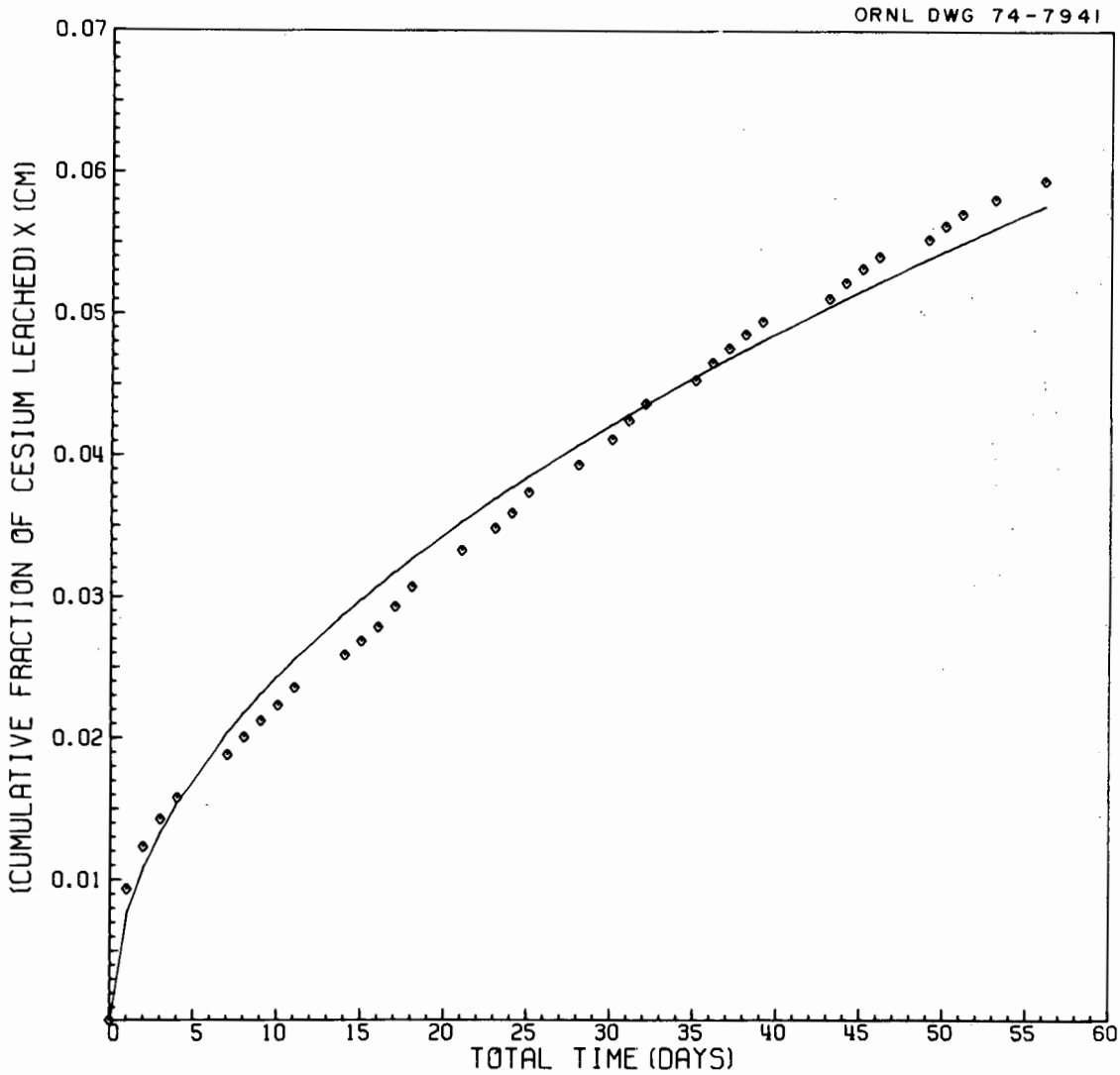

Fig. 22. Comparison of the Cumulative Fraction of Cesium Leached from Cementitious Grout 93A (up to 56 days) to That Predicted by an Equation Based on Transport Phenomena. [Theoretical curve calculated using Eq. (8) with $D_e = 5.4 \times 10^{-10}$ cm²/sec.]

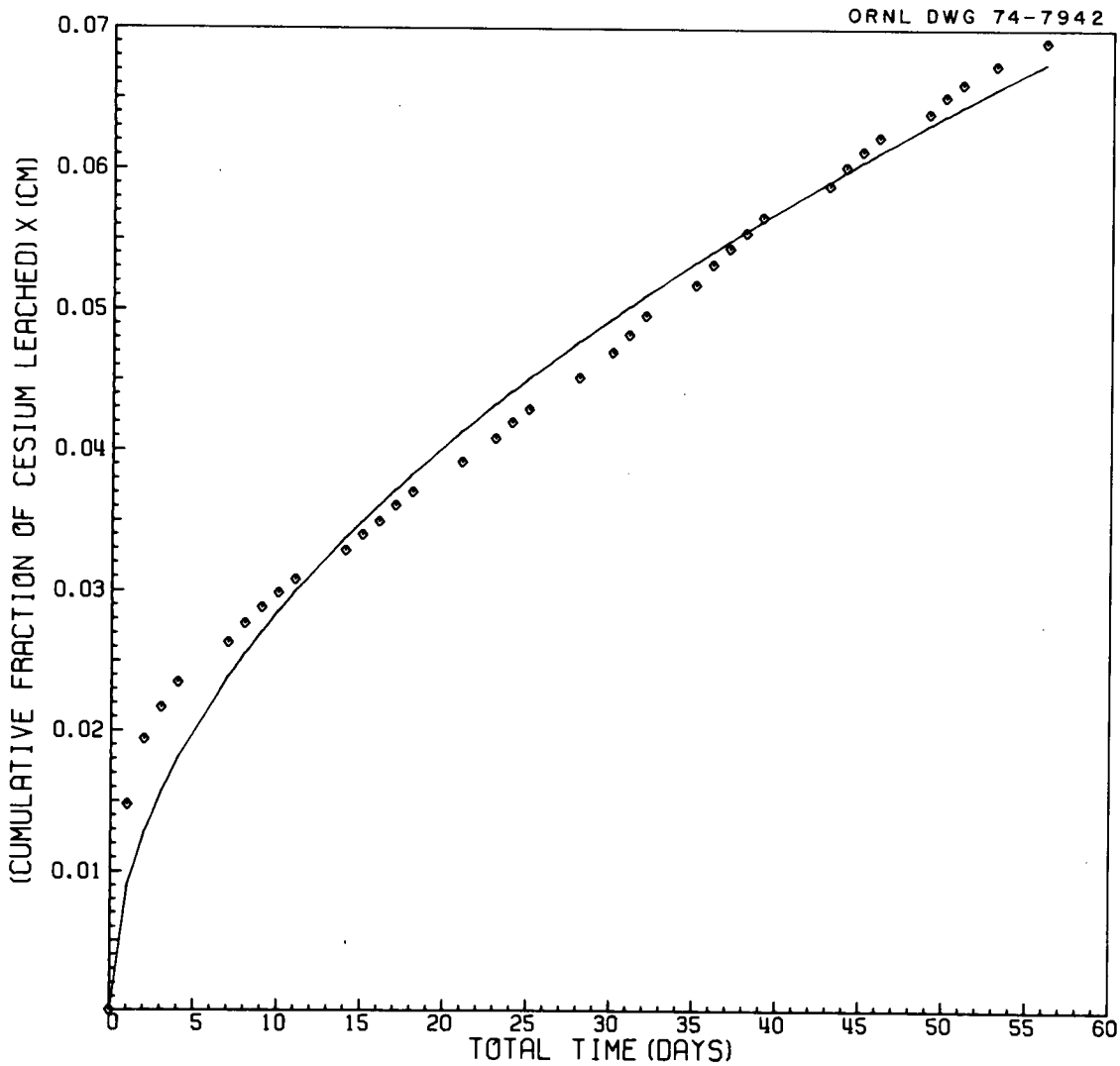

Fig. 23. Comparison of the Cumulative Fraction of Cesium Leached from Cementitious Grout 94A (up to 56 days) to That Predicted by an Equation Based on Transport Phenomena. [Theoretical curve calculated using Eq. (8) with $D_e = 7.4 \times 10^{-10}$ cm^2/sec.]

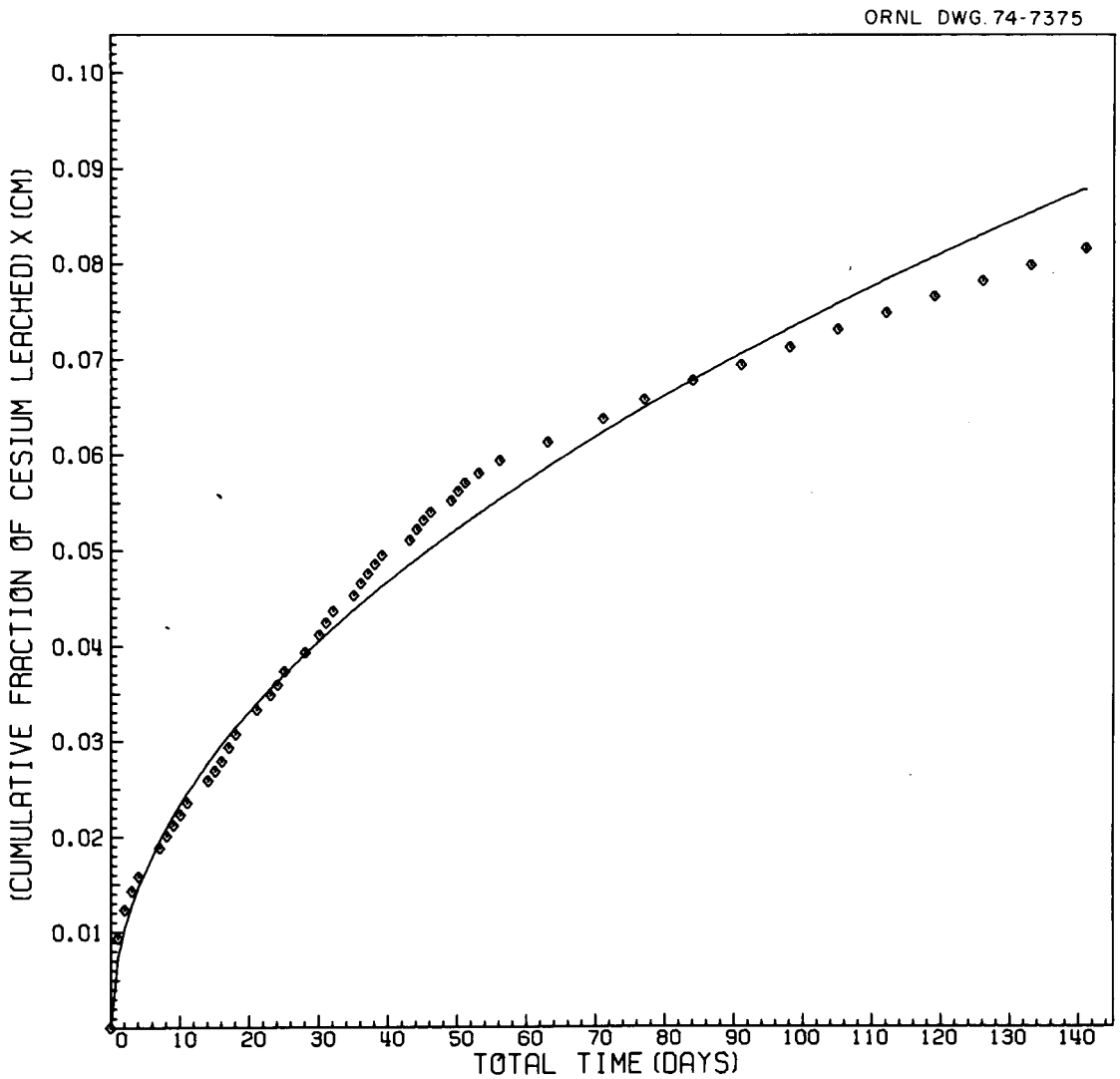

Fig. 24. Comparison of the Cumulative Fraction of Cesium Leached from Cementitious Grout 93A to That Predicted by an Equation Based on Transport Phenomena. [Theoretical curve calculated using Eq. (8) with $D_e = 5.0 \times 10^{-10}$ cm^2/sec.]

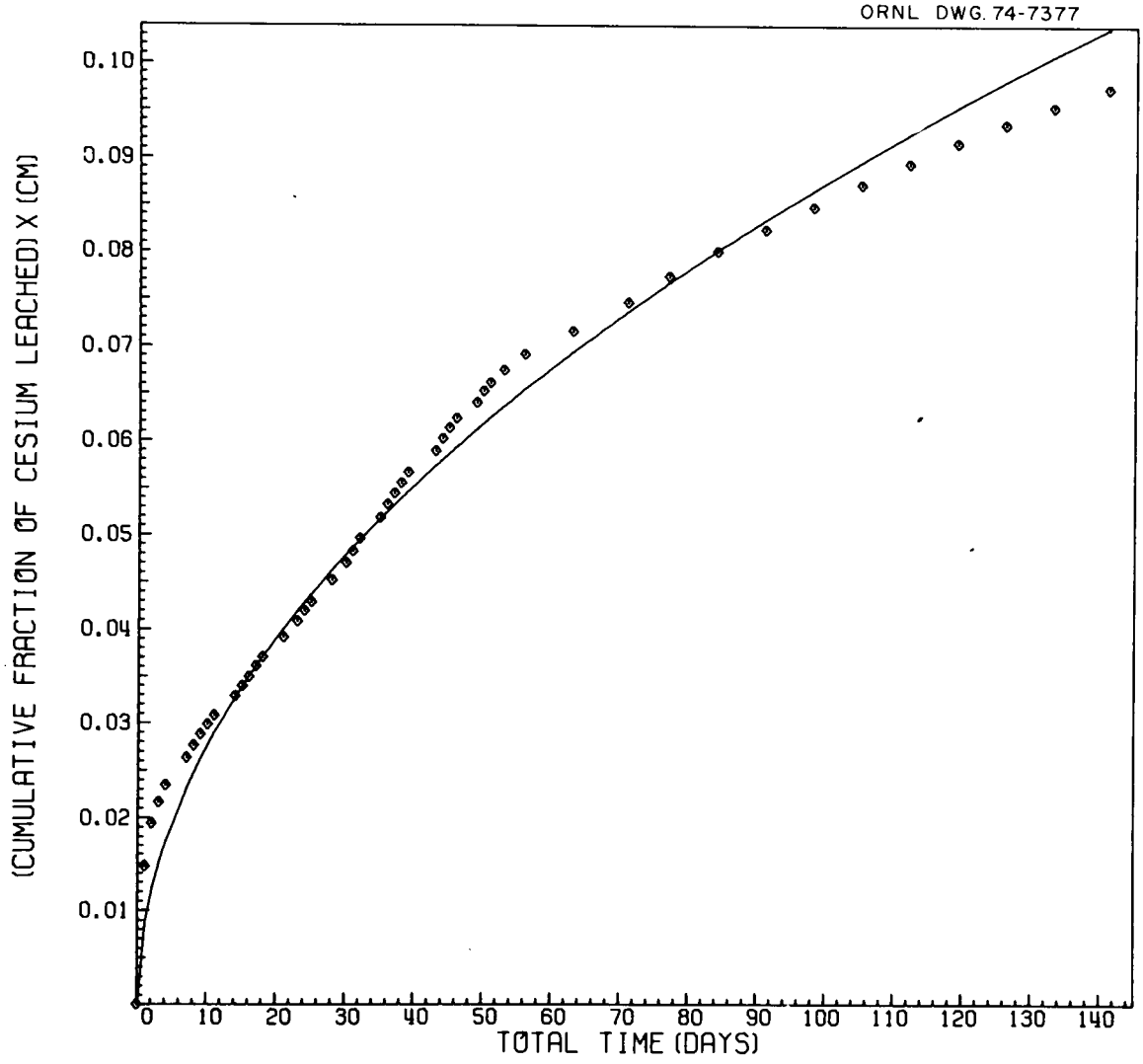

Fig. 25. Comparison of the Cumulative Fraction of Cesium Leached from Cementitious Grout 94A to That Predicted by an Equation Based on Transport Phenomena. [Theoretical curve calculated using Eq. (8) with $D_e = 6.9 \times 10^{-10}$ cm^2/sec.]

approximately twice per day during the first two months and then once per day during the next several months. The D_e's obtained for 93C and 94C are 1.2×10^{-9} cm^2/sec and 1.7×10^{-9} cm^2/sec, respectively. The data points and predicted curves using these D_e's are compared in Figs. 26 and 27, respectively. Since specimens 93A and 93C are from the same batch of grout and the data were collected over the same interval of time, sampling frequency is the only factor (other than repeatability) which can explain the difference between apparent D_e's of 5.0×10^{-10} cm^2/sec for 93A and 1.2×10^{-9} cm^2/sec for 93C (cf. Fig. 24 with Fig. 26). The same statement holds for apparent D_e's of 6.9×10^{-10} cm^2/sec for 94A and 1.7×10^{-9} cm^2/sec for 94C (cf. Fig. 25 with Fig. 27).

As the sampling frequency is increased, the boundary condition of Eq. (8) should be more nearly approached. Tests with more frequent changing of the leachant are needed to determine the minimum number of changes per unit time that are required to yield results which are independent of leachant renewal frequency. Inspection of Figs. 22-27 shows that the goodness-of-fit to Eq. (8) improves as the sampling frequency increases. Equation (8) appears to provide a reasonably good explanation of the leach behavior of specimens 93C and 94C. Tests that meet prescribed boundary conditions and that are carried out for longer periods of time are needed to determine whether other mechanisms whose effects are small at short times (very small rate constants) are important in the long-term storage of such products.

6. APPLICATIONS AND PROJECTIONS

In the foregoing considerations, the isotope being removed from the waste product has been considered to be a stable one. Solutions of the mass transport equations that account for radioactive decay of an isotope escaping from a product are readily obtained from solutions for stable isotopes, as shown in refs. 11 and 12. For example, it is important to know the amount of a radioactive isotope that has escaped from a waste product where decay (but not production) of the isotope in the source (product) and in the environment (leachate) has been taken into account.

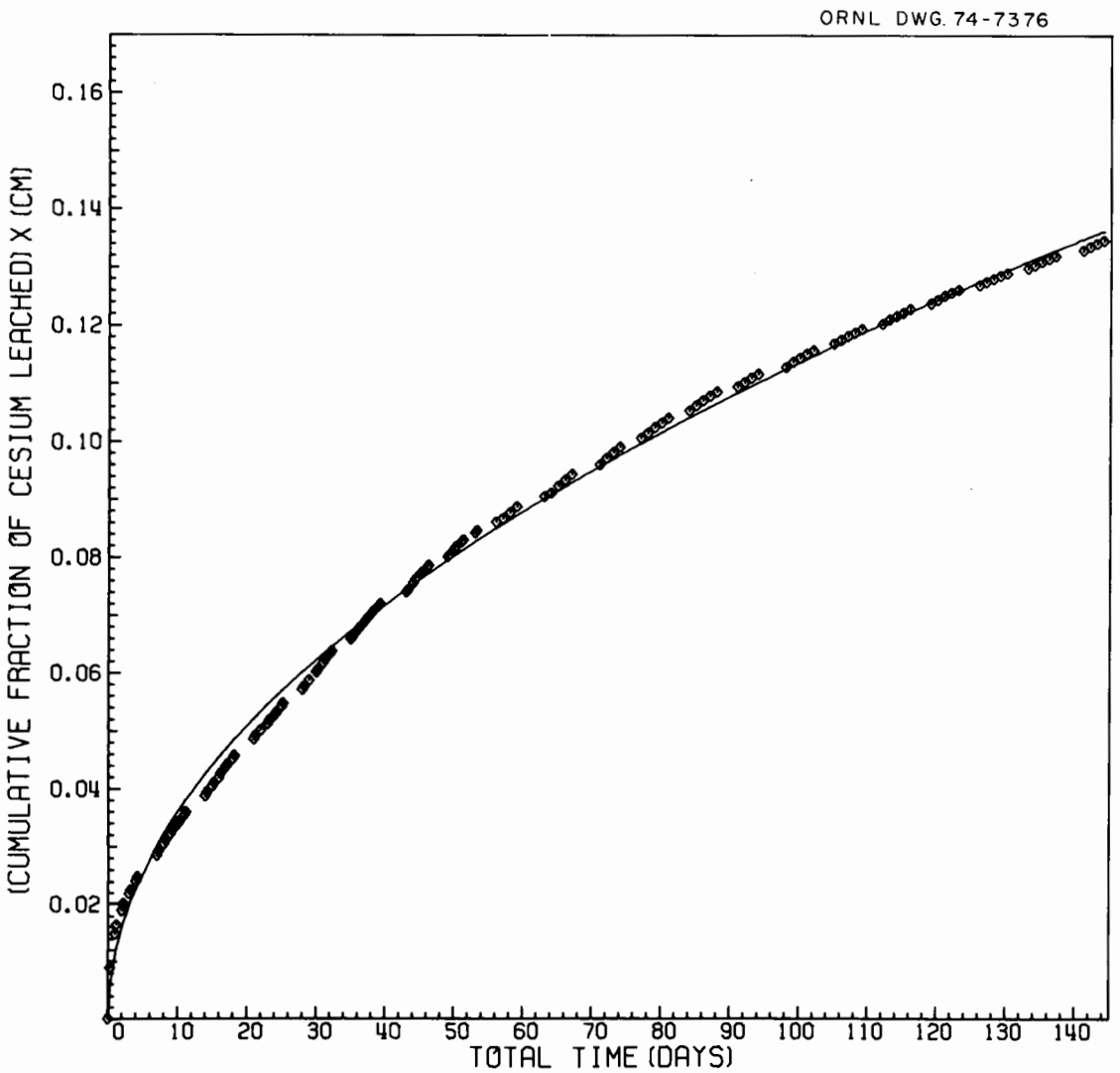

Fig. 26. Comparison of the Cumulative Fraction of Cesium Leached from Cementitious Grout 93C to That Predicted by an Equation Based on Transport Phenomena. [Theoretical curve calculated using Eq. (8) with $D_e = 1.2 \times 10^{-9}$ cm^2/sec.]

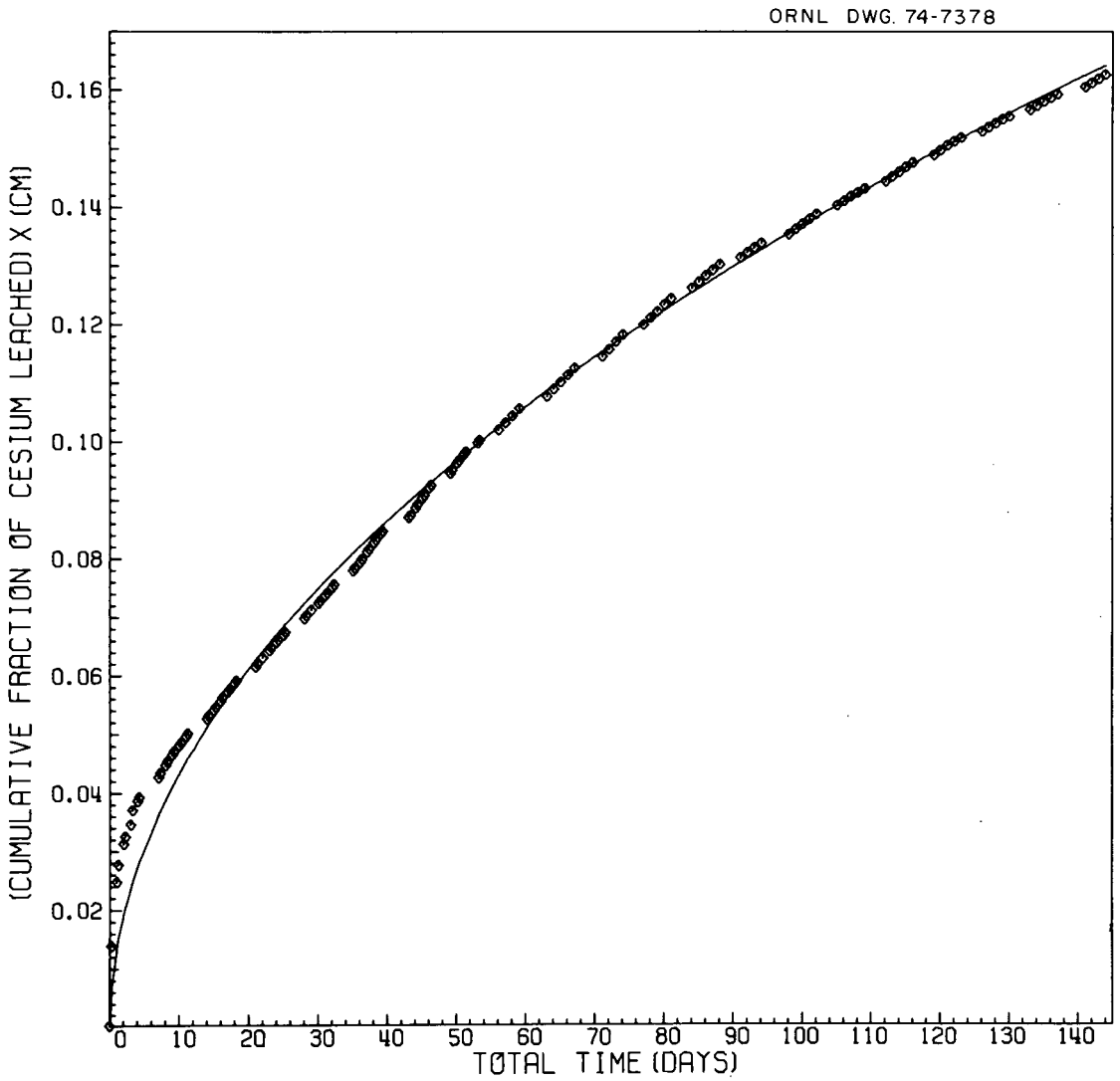

Fig. 27. Comparison of the Cumulative Fraction of Cesium Leached from Cementitious Grout 94C to That Predicted by an Equation Based on Transport Phenomena. [Theoretical curve calculated using Eq. (8) with $D_e = 1.7 \times 10^{-9}$ cm²/sec.]

For the case of diffusion plus instantaneous reaction, this situation with respect to decay can be obtained immediately from Eq. (8) by multiplying by $e^{-\lambda t}$ to give:

$$\left(\frac{\Sigma a_n}{A_o}\right)\left(\frac{V}{S}\right) = 2\left(\frac{D_e}{\pi}\right)^{1/2} t^{1/2}\, e^{-\lambda t}, \tag{9}$$

where

$\lambda = \ln 2/t_{1/2}$ = radioactive decay constant, sec^{-1},

$t_{1/2}$ = half-life of isotope, sec.

Also, in the foregoing considerations, the specimen has been considered to approximate a semi-infinite medium. Solutions of the mass transport equations that consider the actual physical shape of the specimen can be obtained for many simple geometries as shown in standard textbooks such as refs. 20 and 21. The solution of mass transport equations (Appendix C) for a finite cylinder for the case of diffusion with reaction as in Eq. (7) yields:

$$\frac{\Sigma a_n}{A_o} = 1 - \frac{32}{\pi^2 a^2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{e^{-D_e\,[\alpha_m^2 + (2n-1)^2\pi^2/4\ell^2]\,t}}{(2n-1)^2\alpha_m^2}, \tag{10}$$

where D_e is a modified effective diffusivity (cm^2/sec), which is defined as $\mathcal{D}_e/(1 + K)$; a is the radius of the cylinder (cm); ℓ is the half-height of the cylinder (cm); and the α_m's are the positive roots of the zero-order Bessel function of the first kind, $J_o\,(a\,\alpha_m) = 0$. The solution for this case that accounts for radioactive decay (but not production) in the source and in the environment is:

$$\frac{\Sigma a_n}{A_o} = \left\{1 - \frac{32}{\pi^2 a^2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{e^{-D_e\,[\alpha_m^2 + (2n-1)^2\pi^2/4\ell^2]\,t}}{(2n-1)^2\alpha_m^2}\right\} e^{-\lambda t}. \tag{11}$$

The amounts of cesium that Eqs. (10), (8), (11), and (9) predict would be leached from grout products 93C and 94C and would enter the environment are shown in Tables 5 and 6 as a function of time. The waste products are considered to be in the form of a 55-gal drum, that is, a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^5$ cm^3) with all surfaces ($S = 1.99 \times 10^4$ cm^2) continuously exposed to fresh water. Four significant figures are shown in these tables only to emphasize the differences in the calculations for the finite cylinder and those for the semi-infinite medium. The numbers presented in Tables 5 and 6 show that a significant difference between the finite-cylinder and the semi-infinite-medium calculations [Eq. (10) vs (8) and Eq. (11) vs (9)] begins to appear after a few years of leaching with D_e's of 1.2×10^{-9} and 1.7×10^{-9} cm^2/sec. If the cesium is considered to be stable, the finite-cylinder calculation [Eq. (10)] predicts a fraction leached that approaches unity with time, while the semi-infinite-medium calculation [Eq. (8)] predicts a fraction leached that exceeds unity and continues to increase with time (Tables 5 and 6). It should be kept in mind that a solution which assumes the specimen is a semi-infinite medium is valid for a finite specimen only as long as the concentration profiles in the finite specimen approximate those in a semi-infinite medium. Eventually, enough of a species will be leached from a finite specimen so that the concentration profiles do not closely approximate those of a semi-infinite medium. Subsequently, calculations based on the semi-infinite medium will overpredict the amount leached, and consideration must be given to carrying out a solution that accounts for the finite size of the product.

If all the cesium is considered to be ^{137}Cs (half-life, 30 years), the activity of the fraction leached as predicted by Eq. (11) or (9) increases with time, reaches a maximum, and then decreases with time as shown in Tables 5 and 6. The time of maximum activity in the environment as predicted by Eq. (9) can be determined by differentiating the expression with respect to time and setting the result equal to zero. If this procedure is followed with Eq. (9), the time at which the maximum activity is present in the environment is:

$$t_{max} = \frac{1}{2\lambda} = 0.721\ t_{1/2} \,. \tag{12}$$

Table 5. Comparison of the Amounts of ^{137}Cs Leached from a 28-day-cured Cementitious Grout Product[a] and in the Environment as Predicted by Equations Based on Transport Phenomena, Considering a 55-gal Drum as a Finite Source and as a Semi-infinite Medium With and Without Decay

Time		$\left(\frac{\Sigma a_n}{A_o}\right)$, (fraction leached)			
		No Decay in Product or Leachate		Decay In Both Product and Leachate	
(days)	(years)	Finite, Eq. (10)[b,c]	Semi-infinite, Eq. (8)[c,d]	Finite, Eq. (11)[c,e]	Semi-infinite, Eq. (9)[c,f]
91	1/4	1.023×10^{-2}	1.028×10^{-2}	1.017×10^{-2}	1.022×10^{-2}
182	1/2	1.446×10^{-2}	1.453×10^{-2}	1.429×10^{-2}	1.436×10^{-2}
365	1	2.044×10^{-2}	2.058×10^{-2}	1.997×10^{-2}	2.011×10^{-2}
3,650	10	6.381×10^{-2}	6.508×10^{-2}	5.065×10^{-2}	5.165×10^{-2}
5,475	15	7.781×10^{-2}	7.971×10^{-2}	5.502×10^{-2}	5.636×10^{-2}
10,950	30	1.090×10^{-1}	1.127×10^{-1}	5.450×10^{-2}	5.635×10^{-2}
21,900	60	1.519×10^{-1}	1.594×10^{-1}	3.797×10^{-2}	3.985×10^{-2}
32,850	90	1.840×10^{-1}	1.952×10^{-1}	2.300×10^{-2}	2.440×10^{-2}
43,800	120	2.105×10^{-1}	2.254×10^{-1}	1.316×10^{-2}	1.409×10^{-2}
54,750	150	2.335×10^{-1}	2.521×10^{-1}	7.297×10^{-3}	7.878×10^{-3}
65,700	180	2.539×10^{-1}	2.761×10^{-1}	3.967×10^{-3}	4.314×10^{-3}
76,650	210	2.723×10^{-1}	2.982×10^{-1}	2.127×10^{-3}	2.330×10^{-3}
87,600	240	2.892×10^{-1}	3.188×10^{-1}	1.130×10^{-3}	1.245×10^{-3}
98,550	270	3.049×10^{-1}	3.382×10^{-1}	5.955×10^{-4}	6.605×10^{-4}
109,500	300	3.196×10^{-1}	3.565×10^{-1}	3.121×10^{-4}	3.481×10^{-4}
547,500	1,500	6.185×10^{-1}	7.971×10^{-1}	5.493×10^{-16}	7.079×10^{-16}
1,095,000	3,000	7.784×10^{-1}	1.127×10^{0}	6.139×10^{-31}	8.888×10^{-31}

[a]The waste product is considered to be in the form of a 55-gal drum, i.e., a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^5$ cm^3) with all surfaces ($S = 1.99 \times 10^4$ cm^2) continuously exposed to fresh water. The value of the modified effective diffusivity, D_e, was obtained by fitting the leach data of product 93C to Eq. (8).

[b]Equation (10) with $D_e = 1.2 \times 10^{-9}$ cm^2/sec.

[c]Four significant figures are shown only to emphasize the difference in the calculations for the finite cylinder and those for the semi-infinite medium.

[d]Equation (8) with $D_e = 1.2 \times 10^{-9}$ cm^2/sec.

[e]Equation (11) with $D_e = 1.2 \times 10^{-9}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

[f]Equation (9) with $D_e = 1.2 \times 10^{-9}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

Table 6. Comparison of the Amounts of ^{137}Cs Leached from a 28-day-cured Cementitious Grout Product[a] and in the Environment as Predicted by Equations Based on Transport Phenomena, Considering a 55-gal Drum as a Finite Source and as a Semi-infinite Medium With and Without Decay

Time		$\left(\frac{\Sigma a_n}{A_o}\right)$, (fraction leached)			
		No Decay in Product or Leachate		Decay in Both Product and Leachate	
(days)	(years)	Finite, Eq. (10)[b,c]	Semi-infinite, Eq. (8)[c,d]	Finite, Eq. (11)[c,e]	Semi-infinite, Eq. (9)[c,f]
91	1/4	1.232×10^{-2}	1.238×10^{-2}	1.225×10^{-2}	1.231×10^{-2}
182	1/2	1.741×10^{-2}	1.751×10^{-2}	1.721×10^{-2}	1.731×10^{-2}
365	1	2.460×10^{-2}	2.480×10^{-2}	2.404×10^{-2}	2.423×10^{-2}
3,650	10	7.658×10^{-2}	7.841×10^{-2}	6.078×10^{-2}	6.223×10^{-2}
5,475	15	9.330×10^{-2}	9.604×10^{-2}	6.597×10^{-2}	6.791×10^{-2}
10,950	30	1.304×10^{-1}	1.358×10^{-1}	6.520×10^{-2}	6.790×10^{-2}
21,900	60	1.812×10^{-1}	1.921×10^{-1}	4.530×10^{-2}	4.802×10^{-2}
32,850	90	2.190×10^{-1}	2.352×10^{-1}	2.737×10^{-2}	2.940×10^{-2}
43,800	120	2.501×10^{-1}	2.716×10^{-1}	1.563×10^{-2}	1.697×10^{-2}
54,750	150	2.768×10^{-1}	3.037×10^{-1}	8.650×10^{-3}	9.490×10^{-3}
65,700	180	3.005×10^{-1}	3.327×10^{-1}	4.695×10^{-3}	5.198×10^{-3}
76,650	210	3.219×10^{-1}	3.593×10^{-1}	2.515×10^{-3}	2.807×10^{-3}
87,600	240	3.414×10^{-1}	3.841×10^{-1}	1.334×10^{-3}	1.500×10^{-3}
98,500	270	3.594×10^{-1}	4.075×10^{-1}	7.019×10^{-4}	7.959×10^{-4}
109,500	300	3.762×10^{-1}	4.295×10^{-1}	3.674×10^{-4}	4.194×10^{-4}
547,500	1,500	7.042×10^{-1}	9.604×10^{-1}	6.254×10^{-16}	8.529×10^{-16}
1,095,000	3,000	8.595×10^{-1}	1.358×10^{0}	6.778×10^{-31}	1.071×10^{-30}

[a]The waste product is considered to be in the form of a 55-gal drum, i.e., a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^5$ cm^3) with all surfaces ($S = 1.99 \times 10^4$ cm^2) continuously exposed to fresh water. The value of the modified effective diffusivity, D_e, was obtained by fitting the leach data of product 94C to Eq. (8).

[b]Equation (10) with $D_e = 1.7 \times 10^{-9}$ cm^2/sec.

[c]Four significant figures are shown only to emphasize the difference in the calculations for the finite cylinder and those for the semi-infinite medium.

[d]Equation (8) with $D_e = 1.7 \times 10^{-9}$ cm^2/sec.

[e]Equation (11) with $D_e = 1.7 \times 10^{-9}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

[f]Equation (9) with $D_e = 1.7 \times 10^{-9}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

According to Eq. (12), the maximum ^{137}Cs activity is present in the environment after 22 years of leaching. The fractions of 93C and 94C leached at this time are 5.9×10^{-2} and 7.0×10^{-2} respectively. After this time, decay of the isotope becomes the controlling factor in the amount leached and in the environment.

Calculations similar to those for cesium are given for 28-day-cured grouts containing strontium (^{90}Sr), plutonium (^{239}Pu), or curium (^{244}Cm) in Tables 7-9. A modified effective diffusivity for each of these isotopes was estimated by first obtaining the ratio of its fraction leached to the fraction leached from a corresponding grout with cesium (Sect. 4.4). A value for D_e was then calculated as the product of the square of this ratio and the D_e for cesium obtained in Sect. 6. This treatment assumes that Eq. (8) holds for all isotopes in these specimens over the time span of the data. The numbers presented in Table 7 show that a significant difference between the finite-cylinder and the semi-infinite-medium calculations begins to appear after about 30 years when a D_e of 3.2×10^{-11} cm^2/sec is used. Equation (12) predicts that the maximum activity of ^{90}Sr would be present in the environment after about 20 years of leaching (fraction leached = 9.3×10^{-3}), of ^{239}Pu after about 17,500 years (fraction leached = 1.2×10^{-2}), and of ^{244}Cm after about 13 years (fraction leached = 4.1×10^{-5}). The numbers presented in Tables 8 and 9 show that no significant difference between the finite-cylinder and the semi-infinite calculations appears after leaching up to 3000 years when a D_e of 6.0×10^{-16} or 9.7×10^{-16} cm^2/sec is used.

The calculations presented in Tables 5-9 show that the smaller the modified effective diffusivity, the longer the period of time over which a finite specimen will behave as a semi-infinite medium. It is obvious that the larger the finite specimen, the longer it will behave as a semi-infinite medium. Of course, it should be kept in mind that the fraction of the activity lost, as shown in Tables 5-9, may increase if mechanisms other than diffusion and instantaneous reaction take place in the product, if the D_e increases with time, or if the product does not retain its integrity.

Table 7. Comparison of the Amounts of ^{90}Sr Leached from a 28-day-cured Cementitious Grout Product[a] and in the Environment as Predicted by Equations Based on Transport Phenomena, Considering a 55-gal Drum as a Finite Source and as a Semi-infinite Medium With and Without Decay

Time (days)	Time (years)	$\left(\frac{\Sigma a_n}{A_o}\right)$, (fraction leached)			
		No Decay in Product or Leachate		Decay in Both Product and Leachate	
		Finite, Eq. (10)[b,c]	Semi-infinite, Eq. (8)[c,d]	Finite, Eq. (11)[c,e]	Semi-infinite, Eq. (9)[c,f]
91	1/4	1.713×10^{-3}	1.713×10^{-3}	1.702×10^{-3}	1.702×10^{-3}
182	1/2	2.422×10^{-3}	2.422×10^{-3}	2.392×10^{-3}	2.392×10^{-3}
365	1	3.430×10^{-3}	3.430×10^{-3}	3.346×10^{-3}	3.346×10^{-3}
3,650	10	1.080×10^{-2}	1.085×10^{-2}	8.432×10^{-3}	8.471×10^{-3}
5,475	15	1.322×10^{-2}	1.329×10^{-2}	9.119×10^{-3}	9.168×10^{-3}
10,950	30	1.867×10^{-2}	1.879×10^{-2}	8.884×10^{-3}	8.941×10^{-3}
21,900	60	2.365×10^{-2}	2.657×10^{-2}	5.355×10^{-3}	6.016×10^{-3}
32,850	90	3.222×10^{-2}	3.254×10^{-2}	3.472×10^{-3}	3.506×10^{-3}
43,800	120	3.715×10^{-2}	3.758×10^{-2}	1.905×10^{-3}	1.927×10^{-3}
54,750	150	4.148×10^{-2}	4.210×10^{-2}	1.012×10^{-3}	1.025×10^{-3}
65,700	180	4.538×10^{-2}	4.602×10^{-2}	5.269×10^{-4}	5.343×10^{-4}
76,650	210	4.896×10^{-2}	4.971×10^{-2}	2.705×10^{-4}	2.746×10^{-4}
87,600	240	5.229×10^{-2}	5.314×10^{-2}	1.375×10^{-4}	1.397×10^{-4}
98,550	270	5.541×10^{-2}	5.637×10^{-2}	6.932×10^{-5}	7.052×10^{-5}
109,500	300	5.835×10^{-2}	5.941×10^{-2}	3.473×10^{-5}	3.536×10^{-5}
547,500	1,500	1.276×10^{-1}	1.329×10^{-1}	9.537×10^{-18}	9.933×10^{-18}
1,095,000	3,000	1.775×10^{-1}	1.879×10^{-1}	9.916×10^{-34}	1.050×10^{-33}

[a]The waste product is considered to be in the form of a 55-gal drum, i.e., a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^5$ cm^3) with all surfaces ($S = 1.99 \times 10^4$ cm^2) continuously exposed to fresh water. The value of the modified effective diffusivity, D_e, was estimated as described in Sect. 6.

[b]Equation (10) with an estimated $D_e = 3.2 \times 10^{-11}$ cm^2/sec.

[c]Four significant figures are shown only to emphasize the difference in the calculations for the finite cylinder and those for the semi-infinite medium.

[d]Equation (8) with an estimated $D_e = 3.2 \times 10^{-11}$ cm^2/sec.

[e]Equation (11) with an estimated $D_e = 3.2 \times 10^{-11}$ cm^2/sec and $t_{1/2}$ of ^{90}Sr = 28 years.

[f]Equation (9) with an estimated $D_e = 3.2 \times 10^{-11}$ cm^2/sec and $t_{1/2}$ of ^{90}Sr = 28 years.

Table 8. Comparison of the Amounts of ^{239}Pu Leached from a 28-day-cured Cementitious Grout Product[a] and in the Environment as Predicted by Equations Based on Transport Phenomena, Considering a 55-gal Drum as a Finite Source and as a Semi-infinite Medium With and Without Decay

Time		$\left(\frac{\Sigma a_n}{A_o}\right)$, (fraction leached)			
		No Decay in Product or Leachate		Decay in Both Product and Leachate	
(days)	(years)	Finite, Eq. (10)[b,c]	Semi-infinite, Eq. (8)[c,d]	Finite, Eq. (11)[c,e]	Semi-infinite, Eq. (9)[c,f]
91	1/4	7.341×10^{-6}	7.341×10^{-6}	7.341×10^{-6}	7.341×10^{-6}
182	1/2	1.038×10^{-5}	1.038×10^{-5}	1.038×10^{-5}	1.038×10^{-5}
365	1	1.470×10^{-5}	1.470×10^{-5}	1.470×10^{-5}	1.470×10^{-5}
3,650	10	4.649×10^{-5}	4.649×10^{-5}	4.648×10^{-5}	4.648×10^{-5}
5,475	15	5.694×10^{-5}	5.694×10^{-5}	5.692×10^{-5}	5.692×10^{-5}
10,950	30	8.053×10^{-5}	8.053×10^{-5}	8.046×10^{-5}	8.046×10^{-5}
21,900	60	1.139×10^{-4}	1.139×10^{-4}	1.137×10^{-4}	1.137×10^{-4}
32,850	90	1.395×10^{-4}	1.395×10^{-4}	1.391×10^{-4}	1.391×10^{-4}
43,800	120	1.611×10^{-4}	1.611×10^{-4}	1.606×10^{-4}	1.606×10^{-4}
54,750	150	1.801×10^{-4}	1.801×10^{-4}	1.793×10^{-4}	1.793×10^{-4}
65,700	180	1.972×10^{-4}	1.972×10^{-4}	1.962×10^{-4}	1.962×10^{-4}
76,650	210	2.131×10^{-4}	2.131×10^{-4}	2.118×10^{-4}	2.118×10^{-4}
87,600	240	2.278×10^{-4}	2.278×10^{-4}	2.263×10^{-4}	2.263×10^{-4}
98,550	270	2.416×10^{-4}	2.416×10^{-4}	2.398×10^{-4}	2.398×10^{-4}
109,500	300	2.546×10^{-4}	2.546×10^{-4}	2.524×10^{-4}	2.524×10^{-4}
547,500	1,500	5.694×10^{-4}	5.694×10^{-4}	5.456×10^{-4}	5.456×10^{-4}
1,095,000	3,000	8.053×10^{-4}	8.053×10^{-4}	7.395×10^{-4}	7.395×10^{-4}

[a]The waste product is considered to be in the form of a 55-gal drum, i.e., a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^4$ cm^3) with all surfaces ($S = 1.99 \times 10^5$ cm^2) continuously exposed to fresh water. The value of the modified effective diffusivity, D_e, was estimated as described in Sect. 6.

[b]Equation (10) with an estimated $D_e = 6.0 \times 10^{-16}$ cm^2/sec.

[c]Four significant figures are shown only to emphasize the difference in the calculations for the finite cylinder and those for the semi-infinite medium.

[d]Equation (8) with an estimated $D_e = 6.0 \times 10^{-16}$ cm^2/sec.

[e]Equation (11) with an estimated $D_e = 6.0 \times 10^{-16}$ cm^2/sec and $t_{1/2}$ of $^{239}Pu = 2.44 \times 10^4$ years.

[f]Equation (9) with an estimated $D_e = 6.0 \times 10^{-16}$ cm^2/sec and $t_{1/2}$ of $^{239}Pu = 2.44 \times 10^4$ years.

Table 9. Comparison of the Amounts of ^{244}Cm Leached from a 28-day-cured Cementitious Grout Product[a] and in the Environment as Predicted by Equations Based on Transport Phenomena, Considering a 55-gal Drum as a Finite Source and as a Semi-infinite Medium With and Without Decay

Time		$\left(\frac{\Sigma a_n}{A_o}\right)$, (fraction leached)			
		No Decay in Product or Leachate		Decay in Both Product and Leachate	
(days)	(years)	Finite, Eq. (10)[b,c]	Semi-infinite, Eq. (8)[c,d]	Finite, Eq. (11)[c,e]	Semi-infinite, Eq. (9)[c,f]
91	1/4	9.343×10^{-6}	9.343×10^{-6}	9.251×10^{-6}	9.251×10^{-6}
182	1/2	1.321×10^{-5}	1.321×10^{-5}	1.295×10^{-5}	1.295×10^{-5}
365	1	1.871×10^{-5}	1.871×10^{-5}	1.799×10^{-5}	1.799×10^{-5}
3,650	10	5.917×10^{-5}	5.917×10^{-5}	3.901×10^{-5}	3.901×10^{-5}
5,475	15	7.247×10^{-5}	7.247×10^{-5}	4.014×10^{-5}	4.014×10^{-5}
10,950	30	1.025×10^{-4}	1.025×10^{-4}	3.145×10^{-5}	3.145×10^{-5}
21,900	60	1.449×10^{-4}	1.449×10^{-4}	1.364×10^{-5}	1.364×10^{-5}
32,850	90	1.775×10^{-4}	1.775×10^{-4}	5.128×10^{-6}	5.128×10^{-6}
43,800	120	2.050×10^{-4}	2.050×10^{-4}	1.817×10^{-6}	1.817×10^{-6}
54,750	150	2.292×10^{-4}	2.292×10^{-4}	6.235×10^{-7}	6.235×10^{-7}
65,700	180	2.510×10^{-4}	2.510×10^{-4}	2.095×10^{-7}	2.095×10^{-7}
76,650	210	2.712×10^{-4}	2.712×10^{-4}	6.946×10^{-8}	6.946×10^{-8}
87,600	240	2.899×10^{-4}	2.899×10^{-4}	2.278×10^{-8}	2.278×10^{-8}
98,550	270	3.075×10^{-4}	3.075×10^{-4}	7.416×10^{-9}	7.416×10^{-9}
109,500	300	3.241×10^{-4}	3.241×10^{-4}	2.398×10^{-9}	2.398×10^{-9}
547,500	1,500	7.247×10^{-4}	7.247×10^{-4}	1.608×10^{-29}	1.608×10^{-29}
1,095,000	3,000	1.025×10^{-3}	1.025×10^{-3}	5.049×10^{-55}	5.049×10^{-55}

[a]The waste product is considered to be in the form a 55-gal drum, i.e., a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^5$ cm^3) with all surfaces ($S = 1.99 \times 10^4$ cm^2) continuously exposed to fresh water. The value of the modified effective diffusivity, D_e, was estimated as described in Sect. 6.

[b]Equation (10) with an estimated $D_e = 9.7 \times 10^{-16}$ cm^2/sec.

[c]Four significant figures are shown only to emphasize the difference in the calculations for the finite cylinder and those for the semi-infinite medium.

[d]Equation (8) with an estimated $D_e = 9.7 \times 10^{-16}$ cm^2/sec.

[e]Equation (11) with an estimated $D_e = 9.7 \times 10^{-16}$ cm^2/sec and $t_{1/2}$ of ^{244}Cm = 17.6 years.

[f]Equation (9) with an estimated $D_e = 9.7 \times 10^{-16}$ cm^2/sec and $t_{1/2}$ of ^{244}Cm = 17.6 years.

7. EVALUATION OF CLAY MINERALS OTHER THAN GRUNDITE

Recently it became apparent that large quantities of Grundite would no longer be available. Thus, a series of studies was initiated to evaluate the suitability of various other clay minerals to retain cesium in the grouts. Several of these, in particular Conasauga shale and pottery clay, have given encouraging results. To obtain data more rapidly, the proposed IAEA leach test[15] was not used in these studies. Instead, small cylinders (diam of ~ 1.6 cm and length of ~ 2 cm) of the cementitious grouts were prepared as leach specimens. All surfaces of a leach specimen were continuously exposed to the leachant (200 ml) with the leachant being replaced four times a day.

When fitted to Eqs. (8) and (10), preliminary leach data for grouts containing Conasauga shale or pottery clay (instead of Grundite) give D_e's that range from about 1×10^{-14} to 1×10^{-12} cm^2/sec. The predicted amounts leached from three of these grouts are compared with a borosilicate glass in Table 10. The amounts leached from the grouts are calculated, using Eq. (8) with D_e's of 7.6×10^{-14} and 8.8×10^{-13} cm^2/sec for grouts containing Conasauga shale and a D_e of 4.9×10^{-13} cm^2/sec for a grout containing pottery clay. The data for the grouts indicate that the total fraction leached varies with the square root of time. This is the behavior predicted by Eq. (8). The amounts leached from the borosilicate glass are calculated using an equation presented previously:[12]

$$\left(\frac{\Sigma a_n}{A_o}\right)\left(\frac{V}{S}\right) = (D_e k)^{1/2} \left[\left(t + \frac{1}{2k}\right) \text{erf } (kt)^{1/2} + \left(\frac{t}{\pi k}\right)^{1/2} e^{-kt}\right] e^{-\lambda t}, \quad (13)$$

where erf is the error function as defined in Sect. 11 and k is a dissolution rate constant (sec^{-1}). In the earlier study,[12] D_e and k values of 8.4×10^{-15} cm^2/sec and 1.1×10^{-7} sec^{-1}, respectively, were obtained for the glass by fitting the experimental leach results for the glass (product SS-12 in ref. 22) to Eq. (13). Much of the information available on the leaching of glasses,[23-25] especially silicates, indicates that the total fraction leached varies with the square root of time at short times and becomes linear with time at longer times. This is the behavior predicted by Eq. (13).

Table 10. Comparison of the Amounts of ^{137}Cs Leached from a Borosilicate Glass and Cementitious Grouts and in the Environment As Predicted by Equations Based on Mass Transport Phenomena[a]

Time		$\left(\frac{\Sigma a_n}{A_o}\right)$, (fraction leached)			
			Cementitious Grout		
days	years	Borosilicate Glass[b]	Conasauga Shale I[c,d]	Conasauga Shale II[c,e]	Pottery Clay[c,f]
91	1/4	3.5×10^{-5}	8.3×10^{-5}	2.8×10^{-4}	2.1×10^{-4}
182	1/2	5.6×10^{-5}	1.2×10^{-4}	4.0×10^{-4}	3.0×10^{-4}
365	1	1.0×10^{-4}	1.7×10^{-4}	5.5×10^{-4}	4.1×10^{-4}
3,650	10	7.4×10^{-4}	4.2×10^{-4}	1.4×10^{-3}	1.1×10^{-3}
5,475	15	9.9×10^{-4}	4.6×10^{-4}	1.6×10^{-3}	1.2×10^{-3}
10,950	30	1.4×10^{-3}	4.6×10^{-4}	1.6×10^{-3}	1.2×10^{-3}
21,900	60	1.4×10^{-3}	3.2×10^{-4}	1.1×10^{-3}	8.2×10^{-4}
32,850	90	1.0×10^{-3}	2.0×10^{-4}	6.7×10^{-4}	5.0×10^{-4}
43,800	120	6.9×10^{-4}	1.1×10^{-4}	3.9×10^{-4}	2.9×10^{-4}
54,750	150	4.3×10^{-4}	6.4×10^{-5}	2.2×10^{-4}	1.6×10^{-4}
65,700	180	2.6×10^{-4}	3.5×10^{-5}	1.2×10^{-4}	8.8×0^{-5}
76,650	210	1.5×10^{-4}	1.9×10^{-5}	6.4×10^{-5}	4.8×10^{-5}
87,600	240	8.6×10^{-5}	1.0×10^{-5}	3.4×10^{-5}	2.6×10^{-5}
98,550	270	4.8×10^{-5}	5.3×10^{-6}	1.8×10^{-5}	1.4×10^{-5}
109,500	300	2.7×10^{-5}	2.8×10^{-6}	9.6×10^{-6}	7.1×10^{-6}
547,500	1,500	1.2×10^{-16}	5.7×10^{-18}	2.0×10^{-17}	1.4×10^{-17}
1,095,000	3,000	2.2×10^{-31}	7.2×10^{-33}	2.5×10^{-32}	1.8×10^{-32}

[a]Each waste product is considered to be in the form of a 55-gal drum, i.e., a 56-cm-diam x 85-cm-long right circular cylinder ($V = 2.09 \times 10^5$ cm^3) with all surfaces ($S = 1.99 \times 10^4$ cm^2) continuously exposed to fresh water.

[b]Equation (13) with $D_e = 8.4 \times 10^{-15}$ cm^2/sec, $k = 1.1 \times 10^{-7}$ sec^{-1}, and $t_{1/2}$ of ^{137}Cs = 30 years. The parameters for the glass were obtained by fitting the data for product SS-12 from ref. 22 to Eq. (13).

[c]Clay material added to retain cesium.

[d]Equation (9) with $D_e = 7.6 \times 10^{-14}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

[e]Equation (9) with $D_e = 8.8 \times 10^{-13}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

[f]Equation (9) with $D_e = 4.9 \times 10^{-13}$ cm^2/sec and $t_{1/2}$ of ^{137}Cs = 30 years.

The comparison of fractions leached (see Table 10) shows that, at short times, a slightly smaller fraction of cesium is leached from the glass product than from the grouts; at long times, a smaller fraction of cesium is leached from the grout products than from the glass. For the grout products considered in Table 10, the crossover occurs at about three years for the grout with a D_e of 7.6×10^{-14} cm^2/sec, at about 38 years for the grout with a D_e of 8.8×10^{-13} cm^2/sec, and at about 22 years for the grout with a D_e of 4.9×10^{-13} cm^2/sec. This is an important observation since it indicates, at least from the standpoint of leach rate, that these grouts can be comparable to borosilicate glasses. Leach tests carried out for a sufficient time to establish long-term trends are needed for both products. Equation (8) assumes an instantaneous reaction and diffusion, while Eq. (13) assumes zero- and first-order reactions and diffusion. More long-term data are needed so that solutions of the mass transport equations that take into account diffusion, first-order reaction, a moving boundary (dissolution of the matrix), linear surface transfer (a surface film), a time-dependent surface concentration, and other mechanisms can be more fully evaluated.

8. CONCLUSIONS AND RECOMMENDATIONS

The following conclusions have been reached from this study:

1. The grouts used in this study, which were made by blending simulated ORNL intermediate-level waste with a dry mixture of cement, fly ash, and clays selected to retain cesium, can give leach rates that are comparable to those of wastes incorporated in borosilicate glasses.

2. The rate at which cesium was leached from the grouts was lowered by the addition of Grundite, pottery clay, or Conasauga shale. (These additives are listed in the order of increasing effectiveness with respect to cesium retention.)

3. The leach rate for strontium was lowered by adding small quantities of stable strontium or stable strontium plus

calcium to the waste, and allowing the precipitate formed to age before blending it with the dry ingredients to make a grout. Apparently, this is due to scavenging of strontium by the precipitate formed and by isotopic dilution of radioactive strontium with stable strontium.

4. For the grouts made with Grundite, the leach rates of the isotopes used in this study are in the order: Cs > Sr > Cm > Pu.

5. The leach rates of duplicate specimens of a grout may differ by as much as a factor of 2. The difficulty appears to be in specimen replication and may be caused by variations in mixing speed, mixing time, and curing conditions.

6. Curing a grout specimen (up to 28 days) in humid air prior to leaching decreased the amount leached from the specimen. Cesium leach studies indicated that longer curing periods (up to 365 days) had no significant effect on the amount of cesium leached from a specimen.

7. The standard leach test procedure as proposed by the IAEA has limitations for testing these grouts and can give misleadingly low leach rates. This apparently is caused by both the low ratio of leachant volume to exposed surface area and by the too infrequent replacement of leachant as recommended in the proposed standard.

8. A theoretical relationship that considers diffusion and instantaneous reaction gives good agreement with the data obtained in tests using 28-day-cured specimens and a leachant replacement frequency of twice daily.

9. If the same mechanism or combination of mechanisms is taken into account for both semi-infinite and finite models, the projected amounts leached from a waste product assumed to approximate a semi-infinite medium are conservative (on the side of safety) since they are equal to or greater than the

amounts predicted by a solution based on the actual geometry of the finite product.

The results of this study suggest some interesting areas for further work. Among these are improvements in the grout properties and the leach test method.

Improvements in the properties of the grout would include decreasing the already low leach rates, increasing the mechanical strength, and increasing the percentage of waste incorporated into the grout while maintaining or decreasing the present leach rate. The minerals used to retain cesium in these studies included Grundite, Conasauga shale, or pottery clay and were limited to only one or two particle sizes and size distributions each. Studies with other minerals, over a wide range of particle sizes and size distributions, seem desirable in order to determine whether even lower leach rates for cesium can be achieved. The results obtained when alkaline-earth cations were added to the waste before blending with the dry ingredients to form a grout suggest that this approach can decrease the strontium leach rate and that the effect should be investigated in greater depth. In addition, the effect of adding small quantities of various anions (e.g., PO_4^{3-}) to take advantage of low solubility product of the strontium salt formed should be studied.

The mechanical strengths of the grouts as prepared for hydrofracture are adequate for this method of disposal. However, if products based on these grouts are considered for interim surface storage and subsequent transport to an off-site waste repository, a higher strength may be desirable. Efforts toward this goal would encompass: (1) investigation of the dry-mix recipe with variations in the amounts of fly ash and minerals (clays) added to the cement, use of other types of Portland cement, and use of other types of cement (e.g., alumina); (2) investigation of conditions such as speed and duration of mixing as they affect the properties of the product; and (3) investigation of curing conditions such as humidity, curing time, temperature, wet vs dry curing, steam curing, pressure curing, curing under CO_2, etc., as they affect the properties of the product. Work toward increasing the percentage of waste in the grout would include

approaches such as (1) concentrating the waste further by evaporation (e.g., with a thin-film evaporator) before blending with the dry ingredients, and (2) treating the waste by a scavenging-precipitation-ion exchange process and then incorporating the resulting sludge and loaded ion exchange material into grouts.

Improvements in the leach test method would include carrying out the test in such a manner that, for a given ratio of leachant volume to exposed surface area, the results are not dependent on leachant replacement frequency. For a given leachant (tap water, seawater, groundwater, etc.), the maximum amount of a species is removed from a specimen if the leachant is replaced at such frequent intervals (either by successive decantations or continuous flow) that the concentrations of all species from the specimen are essentially negligible in the leachate. In mass transport theory, this case is sometimes described as loss into an infinite medium. Leach tests carried out in this manner give a maximum upper bound (MUB) for the amount of a species which can be removed by the leachant as a function of time. Another case frequently studied in mass transport theory is loss into a well-stirred solution of limited volume. In such an experimental setup, the leachant is simply analyzed as a function of time instead of being replaced. The results obtained from this type of test give the rate of approach to an equilibrium, saturation, or steady-state concentration in the leachate. For waste products leached by a limited volume of water, it is important to know whether the concentration in the leachate is less than the maximum permissible concentration (MPC) allowed. Many leach tests, including some in this study, are carried out in such a manner that the results fall somewhere between those for the two cases described above. Thus, it is recommended that leach tests on products being considered for disposal (not necessarily those being screened in the laboratory) be carried out so that both MUB and percent MPC values are obtained. The MUB values are needed to evaluate the maximum amount that might be leached from a waste product by rapidly flowing water, while the percent MPC serves to evaluate the maximum concentration that might be expected in slowly flowing water or a limited volume of stagnant water. Intrinsic properties of the product, which are independent of the leach conditions, can be

derived from the data obtained in either the MUB or MPC type leach test. Systematic application of mass transport theory to leach data (from well-characterized systems and tests carried out for sufficient time to establish long-term trends) can provide significant information concerning the suitability of these products in waste management programs. The properties determined are useful when comparisons are made of several products. Coupled with solutions of mass transport equations, they provide a way to estimate the amount of an isotope escaping from a product as a function of time, taking into account actual geometries and decay characteristics of the radioactive species.

9. ACKNOWLEDGMENTS

Gratitude is expressed to D. O. Campbell for his many valuable suggestions and helpful discussions during the preparation of this manuscript. Comments and suggestions on the preparation of the grouts by H. O. Weeren were sincerely appreciated. P. C. Arwood gave significant assistance in carrying out the experiments. The authors are indebted to W. J. McDowell and B. S. Weaver for their advice concerning the analytical method for plutonium and curium. Special thanks are extended to L. M. Ferris, who was a source of encouragement during the work.

10. REFERENCES

1. W. de Laguna, T. Tamura, H. O. Weeren, E. G. Struxness, W. C. McClain, and R. C. Sexton, Engineering Development of Hydraulic Fracturing as a Method for Permanent Disposal of Radioactive Wastes, ORNL-4259 (August 1968).

2. U.S. Atomic Energy Commission, Radioactive Waste Facilities, Environmental Statement (Oak Ridge National Laboratory, Oak Ridge, Tenn.), WASH-1532 (January 1974).

3. W. de Laguna, H. O. Weeren, F. T. Binford, E. J. Witkowski, and E. G. Struxness, Safety Analysis of Waste Disposal by Hydraulic Fracturing at Oak Ridge, ORNL-4665 (September 1971).

4. (a) R. M. Wallace, H. L. Hull, and R. F. Bradley, Solid Forms for Savannah River Plant High-Level Waste, DP-1335 (December 1973).
(b) R. F. Bradley, W. H. Hale, and R. M. Wallace, "Solid Forms for Savannah River Plant Radioactive Waste," paper presented at the 167th National Meeting of the American Chemical Society at Los Angeles, Calif., Mar. 31-Apr. 5, 1974 (CONF-740428--4), DP-MS-73-58 (1974).

5. International Atomic Energy Agency, Treatment of Low- and Intermediate-Level Radioactive Waste Concentrates, Technical Reports Series No. 82, STI/DOC/10/82, IAEA, Vienna, 1968.

6. International Atomic Energy Agency, Management of Low- and Intermediate-Level Radioactive Wastes, STI/PUB/264, IAEA, Vienna, 1970.

7. R. H. Burns, "Solidification of Low- and Intermediate-Level Wastes," At. Energy Rev. 9(3), 547-99 (1971).

8. A. H. Kibbey and H. W. Godbee, A Critical Review of Solid Radioactive Waste Practices at Nuclear Power Plants, ORNL-4924 (March 1974).

9. J. O. Blomeke and F. E. Harrington, Management of Radioactive Wastes at Nuclear Power Stations, ORNL-4070 (January 1968).

10. International Atomic Energy Agency, Management of Radioactive Wastes at Nuclear Power Stations, Safety Series No. 28, STI/PUB/208, IAEA, Vienna, 1968.

11. M. J. Bell, An Analysis of the Diffusion of Radioactivity from Encapsulated Wastes, ORNL-TM-3232 (February 1971).

12. H. W. Godbee and D. S. Joy, Assessment of the Loss of Radioactive Isotopes from Waste Solids to the Environment, Part I: Background and Theory, ORNL-TM-4333 (February 1974).

13. J. E. Mendel, A Review of Leaching Test Methods and the Leachability of Various Solid Media Containing Radioactive Wastes, BNWL-1765 (July 1973).

14. A. J. Smith, Leaching of Radioactive Sludges Incorporated in Cement and Bitumen, AERE M. 2223 (May 1969).

15. International Atomic Energy Agency, "Leach Testing of Immobilized Radioactive Waste Solids, A Proposal for a Standard Method," ed. by E. D. Hespe, At. Energy Rev. 9(1), 195-207 (1971).

16. ASTM C: 150-72, "Standard Specifications for Portland Cement," Annual Book of ASTM Standards: 1972, Part 9, American Society for Testing and Materials, Philadelphia, Pa.

17. H. F. W. Taylor, The Chemistry of Cements, Vol. 1, Academic, London and New York, 1964.

18. W. Czernin, Cement Chemistry and Physics for Civil Engineers, Chemical Publishing Co., Inc., New York, 1962.

19. R. H. Bogue, The Chemistry of Portland Cement, 2nd ed., Reinhold, New York, 1955.

20. H. S. Carslaw and J. C. Jaeger, Conduction of Heat in Solids, 2nd ed., Oxford University Press, London, 1959.

21. J. Crank, The Mathematics of Diffusion, Oxford University Press, London, 1970.

22. J. E. Mendel and J. L. McElroy, Waste Solidification Program, Volume 10, Evaluation of Solidified Products, BNWL-1966 (July 1972).

23. R. W. Douglas and T. M. M. El-Shamy, "Reactions of Glasses with Aqueous Solutions," J. Am. Ceram. Soc. 50, 1-8 (1967).

24. M. N. Elliot and D. B. Auty, "The Durability of Glass for the Disposal of Highly Radioactive Waste. Discussion of Method and Effect of Leaching Conditions," Glass Technol. 9, 5-13 (1968).

25. W. Heimerl, H. Heine, L. Kahl, H. W. Levi, W. Lutze, G. Malow, E. Schiewer, and P. Schubert, Research on Glasses for Fission Product Fixation, Hahn-Meitner-Institut, Berlin GmbH, Report No. HMI-B 109 (September 1971).

26. H. S. Carslaw and J. C. Jaeger, Conduction of Heat in Solids, 2nd ed., pp. 58, 59, Oxford University Press, London, 1959.

27. H. S. Carslaw and J. C. Jaeger, Conduction of Heat in Solids, 2nd ed., pp. 297-305, Oxford University Press, London, 1959.

28. M. Abramorwitz and I. A. Stegun (Eds.), Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables, National Bureau of Standards, Applied Mathematics Series 55, 9th printing, U.S. Govt. Printing Office, Washington, D.C., 1970.

29. F. B. Hildebrand, Advanced Calculus for Applications, 3rd printing, pp. 293, 294, Prentice-Hall, Inc., Englewood Cliffs, N.J., 1963.

30. H. S. Carslaw and J. C. Jaeger, Conduction of Heat in Solids, 2nd ed., pp. 225-27, Oxford University Press, London, 1959.

11. NOMENCLATURE

ENGLISH ALPHABET

A = amount of a species present in a specimen at any time t

A_o = amount of a species present in a specimen at the beginning of the leach test

a = radius, cm

a_n = amount of a species transferred to the leachate during leaching period n

Σa_n = sum of the amounts of a species transferred to the leachates during all leaching periods

C = concentration (amount/cm^3) of the more-mobile forms of a species

Ci = curie, the quantity of any radioactive material in which the number of disintegrations per sec is 3.7×10^{10}

C_o = initial concentration (amount/cm^3) of the more mobile forms of a species

C_T = initial total concentration of a species (A_o/V)

cm = centimeter

c_f = net counts min^{-1} ml^{-1} of a radionuclide in the feed sample at time t

c_ℓ = net counts min^{-1} ml^{-1} of a radionuclide in the leachate at time t

$\mathcal{D}_e$ = effective diffusivity, cm^2/sec

D_e = modified effective diffusivity, defined as $\mathcal{D}_e/(1 + K)$, cm^2/sec

d = differential operator

diam = diameter, cm

e = base of natural logarithms, 2.7182818 . . .

erf (u) = error function, defined as

$$\frac{2}{\pi^{1/2}} \int_0^u e^{-\beta^2} \, d\beta$$

F = concentration (amount/cm^3) of the less-mobile forms of a species

F_o = initial concentration (amount/cm^3) of the less-mobile forms of a species

ft = foot, 30.480 . . . cm

g = gram

gal = gallon, 3785.434 cm^3

h = number of radioactive atoms of a chemical element per ml

hr = hour

h_f = number of radioactive atoms of a chemical element per ml of feed sample at time t

h_ℓ = number of radioactive atoms of a chemical element per ml of leachate at time t

in. = inch

J_o = zero-order Bessel function of the first kind

J_1 = first-order Bessel function of the first kind

K = proportionality or equilibrium constant, dimensionless

k = dissolution rate constant, sec^{-1}

ℓ = half-height of a cylinder, cm

lb = pound, 453.592 . . . g

M = molar, g-moles/liter

m = a nonnegative integer

mCi = millicurie, 1×10^{-3} curie

min = minute

ml = milliliter

n = a nonnegative integer

ppm = parts per million

r = a radial coordinate, cm

rpm = revolutions per minute

S = surface area exposed to the leachant, cm^2

s = number of stable atoms of a chemical element per ml

sec = second

s_f = number of stable atoms of a chemical element per ml of feed sample

s_ℓ = number of stable atoms of a chemical element per ml of leachate

t = elapsed time in sec, days, or years as required for dimensional consistency

t_{max} = time after leaching commences when the maximum activity of an isotope is in the environment

t_n = duration of leachant renewal period, sec or days

Σt_n = sum of all leaching periods

$t_{1/2}$ = half-life of an isotope in sec, days, or years as required for dimensional consistency

u = a variable

V = volume of a specimen, cm^3

V_f = volume of waste solution used to prepare a specimen, ml

V_ℓ = volume of leachate, ml

x = length (cm) and a space coordinate (x-axis)

y = length (cm) and a space coordinate (y-axis)

z = length (cm) and a space coordinate (z-axis)

GREEK ALPHABET

α_m = the positive roots of J_o (a α_m) = 0, cm^{-1}

β = a variable

∂ = partial differential operator

$\partial/\partial N$ = differentiation along the outward-drawn normal

λ = ln $2/t_{1/2}$, defined as radioactive decay constant (sec^{-1})

π = 3.141592 . . .

Σ = summation

APPENDIX A: COMPILATION OF DATA ON LEACHING OF CEMENTITIOUS GROUTS

Table A-1. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 13A[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 30 mg
^{85}Sr at time t_o = 0.0435 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 0 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^3$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	>14	4.01[e,f]	0.401[e]
1	2	g	4.32	0.833
1	3	g	4.83	1.316
1	4	12.30	4.07	1.723
3	7	12.35	6.73	2.396
1	8	12.00	3.08	2.704
1	9	11.55	3.19	3.023
1	10	11.95	2.64	3.287
1	11	11.85	2.51	3.538
3	14	12.75	4.35	3.973
1	15	11.75	2.17	4.189
1	16	11.55	2.13	4.403
1	17	11.48	1.99	4.602
1	18	11.25	1.47	4.749
4	22	11.95	3.44	5.093
1	23	11.08	1.59	5.252
1	24	11.00	1.32	5.384
1	25	10.80	1.62	5.546
3	28	11.45	1.98	5.744
1	29	10.65	1.22	5.866
1	30	10.55	1.01	5.967
1	31	10.40	0.885	6.056
1	32	10.45	0.812	6.137
3	35	11.20	1.19	6.256
7	42	11.60	1.50	6.406
7	49	11.30	1.21	6.527
7	56	11.15	1.24	6.651
7	63	11.45	1.23	6.774
7	70	11.00	1.03	6.877
7	77	11.00	0.850	6.962
7	84	10.70	0.757	7.037
7	91	10.80	0.662	7.104
21	112	10.66	0.853	7.190
28	140	10.07	0.313	7.220
28	168	10.61	0.162	7.236
28	196	10.55	0.286	7.265

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 4.01×10^{-3}.

[g]pH not measured.

Table A-2. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 13B[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 30 mg
^{85}Sr at time t_o = 0.0435 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	12.45	24.2[e,f]	0.242[e]
1	2	12.35	17.1	0.413
1	3	11.90	16.2	0.575
1	4	11.95	12.3	0.698
3	7	12.80	17.7	0.875
1	8	11.75	8.54	0.960
1	9	11.25	8.25	1.043
1	10	11.15	7.73	1.120
1	11	10.85	5.76	1.178
4	15	11.85	11.2	1.290
1	16	10.75	4.45	1.332
1	17	10.65	5.47	1.389
1	18	10.65	4.76	1.437
3	21	11.25	6.45	1.501
7	28	11.60	9.32	1.594
7	35	11.55	9.72	1.692
7	42	11.30	6.21	1.754
7	49	11.25	5.15	1.805
7	56	11.30	7.53	1.880
7	63	11.00	4.35	1.924
7	70	10.20	3.97	1.964
7	77	9.10	0.735	1.971
28	105	8.24	0.822	1.979
28	133	8.16	0.987	1.989
28	161	8.17	2.45	2.014
28	189	8.46	1.23	2.026

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 24.2×10^{-4}.

Table A-3. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 13C[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 30 mg
^{85}Sr at time t_o = 0.0435 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^4$
1	1	11.75	0.912[e,f]	0.912[e]
6	7	12.00	1.08	1.992
7	14	11.70	1.05	3.042
7	21	11.20	0.030	3.072
7	28	11.10	1.16	4.232
7	35	11.25	2.13	6.363
7	42	11.30	1.61	7.972
7	49	10.95	0.836	8.808
7	56	10.40	0.662	9.470
28	84	11.03	1.03	10.50
28	112	11.08	2.73	13.23
28	140	11.00	2.00	15.23
28	168	11.05	1.93	17.16

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 0.912×10^{-4}.

Table A-4. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 13D[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 30 mg
^{85}Sr at time t_o = 0.0435 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 0 days

Leachant 150 ml tap water (initial pH = 7.96 – 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^3$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	>14	2.90[e,f]	0.290[e]
1	2	g	3.21	0.611
1	3	g	3.32	0.943
1	4	12.40	2.47	1.190
3	7	12.65	4.43	1.633
1	8	11.35	2.35	1.868
1	9	12.05	1.77	2.045
1	10	11.75	2.06	2.251
1	11	11.75	1.87	2.438
3	14	12.80	3.07	2.745
1	15	11.90	1.62	2.907
1	16	11.65	1.54	3.061
1	17	11.40	1.43	3.204
1	18	11.35	1.25	3.329
4	22	11.95	2.66	3.595
1	23	11.05	1.14	3.709
1	24	11.00	1.02	3.811
1	25	10.95	1.03	3.914
3	28	11.45	1.46	4.060
1	29	10.70	0.792	4.138
1	30	10.50	0.790	4.217
1	31	10.55	0.656	4.283
1	32	10.68	0.665	4.349
3	35	11.30	0.883	4.438
1	36	10.40	0.470	4.485
6	42	11.48	1.04	4.589
7	49	11.40	1.10	4.699
7	56	11.40	1.09	4.808
7	63	11.50	0.851	4.893
7	70	11.35	0.870	4.980
7	77	11.35	0.687	5.048
7	84	10.90	0.671	5.116
7	91	11.20	0.579	5.173
21	112	11.07	0.863	5.260
28	140	11.14	0.735	5.335
28	168	10.92	0.325	5.366
28	196	11.14	0.512	5.417

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 2.90×10^{-3}.

[g]pH not measured.

Table A-5. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 13E[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 30 mg
^{85}Sr at time t_o = 0.0435 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$
1	1	12.50	16.3[e,f]	1.63[e]
1	2	12.25	12.1	2.84
1	3	11.85	10.1	3.85
1	4	11.95	8.49	4.699
3	7	12.20	11.3	5.829
1	8	11.65	6.95	6.524
1	9	11.15	5.38	7.062
1	10	11.15	4.49	7.511
1	11	10.95	3.65	7.876
4	15	11.80	7.81	8.657
1	16	10.78	3.69	9.026
1	17	10.50	3.78	9.404
1	18	10.50	3.58	9.762
3	21	11.35	4.71	10.233
7	28	11.60	6.69	10.902
1	29	11.20	1.31	11.033
6	35	11.45	6.00	11.633
7	42	11.30	5.07	12.140
7	49	11.25	4.49	12.589
7	56	11.25	3.76	12.965
7	63	11.05	3.34	13.299
7	70	10.45	1.83	13.482
7	77	9.45	0.604	13.542
28	105	9.14	0.488	13.591
28	133	8.56	0.897	13.681
28	161	8.44	0.635	13.744
28	189	8.72	0.232	13.768

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 16.3×10^{-4}.

Table A-6. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 13F[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 30 mg
^{85}Sr at time t_o = 0.0435 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^5$	Cumulative $(\Sigma a_n/A_o) \times 10^4$
1	1	11.75	1.73[e,f]	0.173[e]
6	7	11.85	7.28	0.901
7	14	11.85	16.9	2.591
7	21	11.35	0.857	2.677
7	28	11.30	4.31	3.108
7	35	11.40	5.91	3.699
7	42	11.20	2.49	3.948
7	49	10.80	8.00	4.748
7	56	10.45	5.32	5.280
28	84	11.20	10.2	6.300
28	112	11.25	8.37	7.137
28	140	11.17	9.82	8.119
28	168	10.67	8.11	8.930

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 1.73×10^{-5}.

Table A-7. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 70[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 106 mg
^{85}Sr at time t_o = 0.508 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^5$	Cumulative $(\Sigma a_n/A_o) \times 10^4$
1	1	11.80	2.15[e,f]	0.215[e]
1	2	11.54	2.61	0.476
1	3	11.35	3.25	0.801
3	6	11.71	11.42	1.943
1	7	10.62	3.73	2.316
1	8	10.34	4.23	2.739
1	9	10.32	5.69	3.308
1	10	10.18	5.80	3.888
3	13	11.10	9.48	4.836
1	14	10.37	7.74	5.610
1	15	10.28	9.03	6.513
1	16	10.46	6.79	7.192
1	17	10.21	5.67	7.759
3	20	10.69	6.00	8.359
1	21	9.71	2.83	8.642
1	22	9.12	1.34	8.776
5	27	10.01	2.11	8.987
7	34	9.59	1.43	9.130
8	42	9.49	1.08	9.238
6	48	9.03	0.72	9.310
29	77	8.86	0.25	9.335
27	104	8.74	0.46	9.381
70	174	8.89	0.13	9.394
80	254	8.81	1.79	9.572

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 2.15×10^{-5}.

Table A-8. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 87[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 34 mg
^{85}Sr at time t_o = 0.90 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 112 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^5$	Cumulative $(\Sigma a_n/A_o) \times 10^5$
1	1	10.10	1.21[e,f]	1.21[e]
5	6	11.39	0.741	1.951
7	13	11.36	0.479	2.430
7	20	11.12	0.517	2.947
21	41	11.41	1.644	4.591
14	55	11.20	1.881	6.472
14	69	10.92	1.465	7.937
28	97	10.66	3.950	11.887
14	111	10.45	4.293	16.180
14	125	10.05	4.746	20.926
29	154	9.91	6.804	27.730
23	177	9.84	5.825	33.555
26	203	10.02	4.959	38.514

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 1.21×10^{-5}.

Table A-9. Experimental Results for the Leaching of Strontium from Cementitious Grout No. 92[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Strontium content: stable = 34 mg
^{85}Sr at time t_o = 0.90 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 168 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25).

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^5$	Cumulative $(\Sigma a_n/A_o) \times 10^5$
1	1	10.04	1.778[e,f]	1.778[e]
1	2	10.11	0.500	2.278
11	13	11.52	0.127	2.405
28	41	11.62	1.513	3.918
28	69	11.33	2.260	6.178
52	121	11.29	2.718	8.896

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 1.778×10^{-5}.

Table A-10. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 16A[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: Stable = 27 mg
^{137}Cs at time t_o = 0.0276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 0 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^3$	Cumulative $(\Sigma a_n/A_o) \times 10^2$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^3$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	12.60	6.34[e,f]	0.634[e]	3	28	11.45	1.45[e]	4.307[e]
1	2	12.25	4.10	1.044	7	35	11.65	2.56	4.563
1	3	12.45	2.64	1.308	7	42	11.50	1.99	4.762
1	4	11.95	2.77	1.585	7	49	11.45	1.80	4.942
3	7	13.05	5.26	2.111	7	56	11.50	1.65	5.107
1	8	11.75	2.12	2.323	7	63	11.55	1.59	5.266
1	9	11.95	1.78	2.501	7	70	11.35	1.15	5.381
1	10	11.90	1.77	2.678	7	77	11.05	1.14	5.495
1	11	11.85	1.73	2.851	7	84	11.25	0.991	5.594
4	15	12.40	4.01	3.252	21	105	11.16	1.92	5.786
1	16	11.80	1.64	3.416	28	133	11.26	2.26	6.012
1	17	11.60	1.10	3.526	28	161	11.17	1.79	6.191
1	18	11.60	1.06	3.632	28	189	11.09	1.33	6.324
3	21	11.90	2.14	3.846	22	218	11.07	1.21	6.445
1	22	11.15	1.04	3.950	69	280	11.11	1.66	6.611
1	23	11.00	0.764	4.026	136	416	10.85	1.248	6.736
1	24	10.80	0.663	4.093	14	430	10.57	0.993	6.835
1	25	10.85	0.692	4.162	19	449	10.30	0.946	6.930

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 6.34×10^{-3}.

Table A-11. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 16B[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	12.65	28.7[e,f]	0.287[e]
1	2	12.30	16.6	0.453
1	3	12.10	13.7	0.590
1	4	11.95	12.5	0.715
4	8	12.35	25.1	0.966
1	9	11.65	6.82	1.034
1	10	11.40	6.48	1.099
1	11	11.20	5.06	1.150
3	14	11.70	11.1	1.261
2	16	11.35	7.23	1.333
5	21	11.60	12.5	1.458
7	28	11.80	15.8	1.616
7	35	11.40	12.0	1.736
7	42	11.45	10.4	1.840
7	49	11.35	8.42	1.924
7	56	11.35	7.45	1.999
7	63	11.10	7.44	2.073
7	70	10.60	6.48	2.138
7	77	10.45	6.12	2.199
21	98	9.66	6.65	2.266
28	126	8.56	3.26	2.298
28	154	8.47	1.06	2.308
28	182	8.62	0.982	2.318
22	204	8.74	2.06	2.339
69	273	8.83	2.20	2.361

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 28.7×10^{-4}.

Table A-12. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 16C[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$
1	1	11.65	7.96[e,f]	0.796[e]
1	2	10.95	3.18	1.114
5	7	11.82	5.02	1.616
7	14	11.45	3.93	2.009
7	21	11.30	4.41	2.450
7	28	11.45	4.44	2.894
7	35	11.25	6.21	3.515
7	42	11.10	4.48	3.963
7	49	10.40	3.20	4.283
7	56	10.55	3.28	4.611
21	77	10.99	6.57	5.268
28	105	11.08	6.48	5.916
28	133	11.02	6.22	6.538
28	161	11.08	3.20	6.858
22	183	10.92	4.67	7.325
69	252	11.08	6.28	7.953
136	388	10.74	6.30	8.583
14	402	10.15	1.82	8.765
19	421	9.62	3.69	9.134

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 7.96×10^{-4}.

Table A-13. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 16D[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 0 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^3$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	12.55	6.51[e,f]	0.651[e]
1	2	12.25	4.13	1.064
1	3	12.25	2.81	1.345
1	4	11.35	2.72	1.617
3	7	13.10	5.33	2.150
1	8	12.25	2.25	2.375
1	9	12.05	2.11	2.586
1	10	11.95	1.98	2.784
1	11	12.05	1.68	2.952
4	15	12.40	4.48	3.400
1	16	11.70	1.43	3.543
1	17	11.75	1.19	3.662
1	18	11.50	1.10	3.772
3	21	11.90	2.24	3.996
1	22	11.20	1.11	4.107
1	23	10.95	0.867	4.194
1	24	10.80	0.656	4.259
1	25	10.80	0.740	4.333
3	28	11.40	1.66	4.499
7	35	11.70	2.73	4.772
7	42	11.65	2.10	4.982
7	49	11.55	1.69	5.153
7	56	11.55	1.65	5.316
7	63	11.60	1.43	5.459
7	70	11.45	1.37	5.596
7	77	10.65	1.22	5.783
7	84	11.10	0.966	5.815
21	105	11.02	2.09	6.024
28	133	11.05	2.56	6.280
28	161	10.83	1.95	6.480
28	189	11.07	1.44	6.619
22	211	11.09	1.35	6.754
69	280	9.80	1.91	6.945

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 6.51×10^{-3}.

Table A-14. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 16E[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^3$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	12.55	3.25[e,f]	0.325[e]
1	2	12.35	2.03	0.528
1	3	12.10	1.66	0.694
1	4	12.05	1.23	0.817
4	8	12.30	2.88	1.105
1	9	11.65	0.815	1.187
1	10	11.45	0.675	1.254
1	11	11.20	0.566	1.311
3	14	11.75	1.09	1.420
2	16	11.40	0.810	1.501
5	21	11.60	1.37	1.638
7	28	11.80	1.59	1.797
7	35	11.45	1.14	1.911
7	42	11.40	1.10	2.021
7	49	11.50	0.882	2.109
7	56	11.35	0.835	2.192
7	63	11.25	0.729	2.265
7	70	9.65	0.570	2.322
7	77	9.10	0.247	2.347
21	98	8.43	0.557	2.403
28	126	8.45	0.761	2.479
28	154	8.33	0.176	2.496
28	182	8.56	0.290	2.525
22	204	8.53	0.102	2.536
69	273	8.69	0.415	2.577
136	409	8.65	0.469	2.624
14	423	8.31	0.407	2.665
19	442	8.23	0.161	2.681

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 3.25×10^{-3}.

Table A-15. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 16F[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$
1	1	11.70	10.53[e,f]	1.053[e]
1	2	8.70	3.97	1.450
5	7	11.85	8.66	2.316
7	14	11.70	6.46	2.962
7	21	11.40	5.98	3.560
7	28	11.40	4.78	4.038
7	35	11.45	4.84	4.522
7	42	11.25	5.46	5.068
7	49	10.60	5.24	5.592
7	56	8.45	0.42	5.634
21	77	11.14	10.8	6.714
28	105	11.10	8.95	7.609
28	133	11.09	8.14	8.423
28	161	11.16	4.87	8.910
22	183	11.03	5.43	9.453
69	252	11.05	9.18	10.371

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 10.53×10^{-4}.

Table A-16. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 86[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.136 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 84 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	10.66	18.18[e,f]	0.1818[e]	1	143	8.36	1.514[e]	1.5977[e]
1	2	10.41	4.906	0.2309	1	144	8.33	1.908	1.6168
1	3	9.87	2.252	0.2534	3	147	8.69	2.676	1.6426
1	4	9.55	1.625	0.2696	1	148	8.39	1.664	1.6602
4	8	11.02	4.892	0.3186	1	149	8.38	1.706	1.6773
6	14	11.20	6.444	0.3831	1	150	8.35	1.594	1.6932
29	43	11.69	20.279	0.5859	1	151	8.33	1.480	1.7080
6	49	11.06	6.517	0.6511	3	154	8.57	2.677	1.7348
7	56	10.84	6.483	0.7158	2	156	8.60	2.607	1.7609
7	63	10.68	6.414	0.7800	1	157	8.40	1.777	1.7786
7	70	10.50	6.113	0.8410	1	158	8.04	1.342	1.7920
7	77	10.20	5.597	0.8970	3	161	8.37	2.253	1.8146
7	84	10.28	6.137	0.9584	2	163	8.36	2.219	1.8368
8	92	10.29	6.061	1.0190	1	164	8.19	1.633	1.8531
1	93	9.45	2.700	1.0460	1	165	8.18	1.408	1.8672
1	94	9.00	2.660	1.0726	3	168	8.54	2.332	1.8905
1	95	8.72	1.937	1.0920	1	169	8.22	1.610	1.9066
3	98	9.40	3.291	1.1249	1	170	8.17	1.335	1.9200
1	99	8.91	2.373	1.1486	1	171	8.17	1.468	1.9346
1	100	8.45	2.272	1.1713	1	172	8.13	1.421	1.9488
1	101	8.55	2.203	1.1934	4	176	8.43	2.534	1.9742
1	102	8.57	1.992	1.2133	6	182	8.63	3.341	2.0076
4	106	9.37	3.499	1.2483	7	189	8.82	3.086	2.0385
2	108	8.87	3.335	1.2816	7	196	8.91	3.657	2.0751
1	109	8.54	2.304	1.3047	8	204	9.01	3.643	2.1115
3	112	8.93	2.980	1.3345	6	210	9.09	3.615	2.1476
1	113	8.48	2.129	1.3558	3	213	8.71	3.080	2.1784
1	114	8.38	1.610	1.3718	4	217	8.70	2.858	2.2070
1	115	g	1.891	1.3908	3	220	8.67	2.682	2.2338
19	134	9.68	5.673	1.4475	4	224	8.41	2.414	2.2580
1	135	8.99	0.906	1.4566	3	227	8.30	2.643	2.2844
1	136	9.38	2.060	1.4772	4	231	8.37	2.435	2.3088
1	137	8.91	2.014	1.4973	3	234	8.31	2.371	2.3325
3	140	8.60	4.394	1.5412	4	238	8.39	2.624	2.3587
1	141	8.43	2.208	1.5633	3	241	8.35	2.214	2.3809
1	142	8.39	1.929	1.5826	4	245	8.38	2.387	2.4047

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 18.18×10^{-4}.

[g]pH not measured.

Table A-17. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 91[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.136 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 168 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	9.64	16.91[e,f]	0.1691[e]	1	67	8.82	1.552[e]	0.6943[e]
7	8	11.37	13.47	0.3038	3	70	9.02	2.264	0.7169
1	9	10.11	1.526	0.3191	2	72	8.95	2.262	0.7396
1	10	9.87	1.043	0.3295	1	73	8.66	1.801	0.7576
1	11	9.78	0.911	0.3386	1	74	8.24	1.617	0.7737
3	14	10.46	2.332	0.3619	3	77	8.54	2.205	0.7958
1	15	9.64	0.721	0.3691	2	79	8.59	2.323	0.8190
1	16	9.54	0.776	0.3769	1	80	8.34	1.802	0.8370
1	17	9.50	0.789	0.3848	1	81	8.23	1.613	0.8532
1	18	9.45	0.610	0.3909	3	84	8.51	2.413	0.8773
4	22	10.26	2.281	0.4137	1	85	8.32	1.502	0.8923
2	24	9.81	1.439	0.4281	1	86	8.24	1.514	0.9074
1	25	9.29	0.627	0.4344	1	87	8.23	1.557	0.9230
3	28	9.82	1.490	0.4493	1	88	8.22	1.421	0.9372
1	29	9.69	1.249	0.4617	4	92	8.35	2.673	0.9640
1	30	9.72	1.210	0.4738	6	98	8.50	3.062	0.9946
1	31	9.20	0.589	0.4797	7	105	8.64	3.478	1.0294
19	50	11.07	6.531	0.5450	7	112	8.82	3.371	1.0631
1	51	9.55	1.078	0.5558	8	120	8.88	3.603	1.0991
1	52	9.47	0.948	0.5653	6	126	9.07	3.364	1.1327
1	53	9.32	0.481	0.5701	3	129	8.76	2.895	1.1617
3	56	9.42	1.533	0.5855	4	133	8.64	3.182	1.1935
1	57	9.20	0.870	0.5914	3	136	8.71	2.803	1.2216
1	58	9.40	0.871	0.6028	4	140	8.42	2.767	1.2492
1	59	9.31	0.789	0.6107	3	143	8.32	2.651	1.2757
1	60	9.15	0.905	0.6198	4	147	8.31	2.562	1.3013
3	63	9.61	2.068	0.6405	3	150	8.23	2.724	1.3286
1	64	9.11	0.791	0.6484	4	154	8.27	2.781	1.3564
1	65	9.10	1.365	0.6620	3	157	8.24	2.076	1.3772
1	66	9.02	1.674	0.6788	4	161	8.35	2.868	1.4058

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 16.91×10^{-4}.

Table A-18. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 93A[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$
1	1	11.31	18.66[e,f]	1.866[e]	1	37	9.03	2.07[e]	9.506[e]
1	2	11.10	5.92	2.458	1	38	8.79	1.98	9.704
1	3	10.79	3.94	2.852	1	39	8.96	1.86	9.890
1	4	10.53	3.03	3.155	4	43	9.76	3.21	10.211
3	7	11.10	6.07	3.762	1	44	9.44	2.24	10.435
1	8	10.24	2.46	4.008	1	45	9.09	1.95	10.630
1	9	10.17	2.30	4.238	1	46	8.77	1.72	10.802
1	10	10.07	2.21	4.459	3	49	9.62	2.47	11.049
1	11	10.01	2.46	4.705	1	50	9.13	1.88	11.237
3	14	10.70	4.64	5.169	1	51	8.94	1.70	11.407
1	15	9.90	1.99	5.368	2	53	9.28	2.116	11.618
1	16	9.79	2.02	5.570	3	56	9.54	2.632	11.882
1	17	9.91	2.88	5.858	7	63	10.09	3.819	12.264
1	18	9.75	2.76	6.134	8	71	10.23	4.832	12.747
3	21	10.42	5.18	6.652	6	77	10.13	4.033	13.150
2	23	10.09	3.10	6.962	7	84	10.08	4.017	13.552
1	24	9.71	2.15	7.177	7	91	10.15	3.239	13.876
1	25	9.53	2.87	7.464	7	98	9.68	3.706	14.246
3	28	9.94	3.99	7.863	7	105	9.77	3.757	14.622
2	30	9.92	3.62	8.225	7	112	9.71	3.477	14.970
1	31	9.38	2.59	8.484	7	119	9.68	3.415	15.311
1	32	9.12	2.37	8.721	7	126	9.71	3.226	15.634
3	35	9.68	3.34	9.055	7	133	9.26	3.298	15.964
1	36	9.30	2.44	9.299	8	141	9.54	3.540	16.318

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 18.66×10^{-4}

Table A-19. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 93C[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.276 mCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water, 2 changes/day (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
0.25	0.25	11.01	17.87[e,f]	0.1787[e]
0.75	1.0	11.29	11.59	0.2946
0.25	1.25	10.28	3.30	0.3276
0.75	2.0	10.73	5.08	0.3784
0.25	2.25	9.85	2.19	0.4003
0.75	3.0	10.55	3.57	0.4360
0.25	3.25	9.58	1.52	0.4512
0.75	4.0	10.26	3.12	0.4824
0.25	4.25	9.21	1.33	0.4957
2.75	7.0	11.15	7.36	0.5693
0.25	7.25	9.70	1.45	0.5838
0.75	8.0	10.02	2.49	0.6087
0.25	8.25	9.48	1.16	0.6203
0.75	9.0	10.05	2.34	0.6437
0.25	9.25	9.38	1.13	0.6550
0.75	10.0	9.92	2.10	0.6760
0.25	10.25	9.29	1.03	0.6863
0.75	11.0	9.81	2.21	0.7084
0.25	11.25	9.10	1.09	0.7193
2.75	14.0	10.76	5.51	0.7744
0.25	14.25	9.44	1.29	0.7873
0.75	15.0	9.71	2.15	0.8088
0.25	15.25	9.18	1.03	0.8191
0.75	16.0	9.61	1.84	0.8375
0.25	16.25	9.25	1.52	0.8527
0.75	17.0	9.51	2.37	0.8764
0.25	17.25	8.84	0.817	0.8846
0.75	18.0	9.48	1.903	0.9036
0.25	18.25	8.82	1.093	0.9145
2.75	21.0	10.36	5.625	0.9708
0.25	21.25	9.22	1.208	0.9829
0.75	22.0	9.47	1.910	1.0020
1.0	23.0	9.74	2.247	1.0244
0.25	23.25	9.15	1.066[e]	1.0351[e]
0.75	24.0	9.48	1.788	1.0530
0.25	24.25	8.70	0.900	1.0620
0.75	25.0	9.18	2.211	1.0841
0.25	25.25	8.62	1.070	1.0948
2.75	28.0	10.03	4.658	1.1414
0.25	28.25	8.96	1.050	1.1519
0.75	29.0	9.37	2.293	1.1748
1.0	30.0	9.38	2.676	1.2016
0.25	30.25	8.54	0.947	1.2110
0.75	31.0	9.07	2.293	1.2340
0.25	31.25	8.67	1.196	1.2459
0.75	32.0	8.92	2.084	1.2667
0.25	32.25	8.54	0.934	1.2761
2.75	35.0	9.81	4.053	1.3166
0.25	35.25	8.82	0.872	1.3254
0.75	36	9.29	2.328	1.3486
0.25	36.25	8.37	0.922	1.3578
0.75	37.0	9.91	1.958	1.3774
0.25	37.25	8.36	0.930	1.3867
0.75	38.0	8.77	1.962	1.4063
0.25	38.25	8.32	0.803	1.4144
0.75	39.0	8.75	1.701	1.4314
0.25	39.25	8.56	0.860	1.4400
3.75	43.0	9.94	3.972	1.4797
0.25	43.25	8.99	0.800	1.4877
0.75	44.0	9.35	2.396	1.5117
0.25	44.25	8.65	1.068	1.5223
0.75	45.0	8.85	1.832	1.5407
0.25	45.25	8.51	0.703	1.5477
0.75	46.0	8.89	1.665	1.5643
0.25	46.25	8.58	0.775	1.5721
2.75	49.0	9.60	2.936	1.6014

Table A-19 (continued)

Leaching Time (days) Incremental t_n	Leaching Time (days) Cumulative Σt_n	Final pH of Leachate	Fraction Leached Incremental[d] $(a_n/A_o) \times 10^4$	Fraction Leached Cumulative $(\Sigma a_n/A_o) \times 10^2$	Leaching Time (days) Incremental t_n	Leaching Time (days) Cumulative Σt_n	Final pH of Leachate	Fraction Leached Incremental[d] $(a_n/A_o) \times 10^4$	Fraction Leached Cumulative $(\Sigma a_n/A_o) \times 10^2$
0.25	49.25	8.86	0.715[e]	1.6086[e]	4.0	98.0	9.23	2.294[e]	2.2536[e]
0.75	50.0	9.13	2.102	1.6296	1.0	99.0	8.92	1.757	2.2712
0.25	50.25	8.60	0.758	1.6372	1.0	100.0	8.66	1.478	2.2860
0.75	51.0	8.87	1.466	1.6519	1.0	101.0	8.45	1.407	2.3000
0.25	51.25	8.51	0.728	1.6591	1.0	102.0	8.59	1.240	2.3124
1.75	53.0	9.28	2.364	1.6828	3.0	105.0	9.05	2.219	2.3346
0.25	53.25	8.65	0.844	1.6912	1.0	106.0	8.75	1.342	2.3480
2.75	56.0	9.54	2.984	1.7211	1.0	107.0	8.63	1.303	2.3611
1.0	57.0	9.20	1.220	1.7333	1.0	108.0	8.54	1.143	2.3725
1.0	58.0	9.25	2.076	1.7540	1.0	109.0	8.54	1.249	2.3850
1.0	59.0	9.00	2.064	1.7747	3.0	112.0	9.06	1.915	2.4041
4.0	63.0	9.80	3.481	1.8095	1.0	113.0	8.58	1.492	2.4190
1.0	64.0	9.27	1.220	1.8217	1.0	114.0	8.57	1.216	2.4312
1.0	65.0	9.26	2.483	1.8465	1.0	115.0	8.43	1.116	2.4424
1.0	66.0	8.90	2.126	1.8678	1.0	116.0	8.61	1.296	2.4553
1.0	67.0	8.91	1.943	1.8872	3.0	119.0	8.98	1.921	2.4745
4.0	71.0	9.73	3.250	1.9197	1.0	120.0	8.70	1.303	2.4876
1.0	72.0	9.42	2.204	1.9417	1.0	121.0	8.58	1.375	2.5013
1.0	73.0	9.09	1.988	1.9616	1.0	122.0	8.48	1.009	2.5114
1.0	74.0	8.94	1.868	1.9803	1.0	123.0	8.48	1.033	2.5217
3.0	77.0	9.52	2.968	2.0100	3.0	126.0	8.78	1.601	2.5378
1.0	78.0	8.99	1.824	2.0282	1.0	127.0	8.58	1.115	2.5489
1.0	79.0	8.89	1.876	2.0470	1.0	128.0	8.52	1.063	2.5595
1.0	80.0	8.95	1.618	2.0632	1.0	129.0	8.40	1.099	2.5705
1.0	81.0	8.88	1.575	2.0789	1.0	130.0	8.44	0.882	2.5793
3.0	84.0	9.40	2.519	2.1041	3.0	133.0	8.64	1.637	2.5957
1.0	85.0	9.14	1.836	2.1224	1.0	134.0	8.20	1.144	2.6072
1.0	86.0	8.85	1.667	2.1391	1.0	135.0	8.38	1.207	2.6192
1.0	87.0	8.79	1.550	2.1546	1.0	136.0	8.43	1.017	2.6294
1.0	88.0	8.74	1.458	2.1692	1.0	137.0	8.45	1.021	2.6396
3.0	91.0	9.04	1.737	2.1866	4.0	141.0	8.87	1.838	2.6580
1.0	92.0	8.71	1.516	2.2017	1.0	142.0	8.75	1.253	2.6705
1.0	93.0	8.63	1.492	2.2166	1.0	143.0	8.46	1.164	2.6822
1.0	94.0	8.62	1.400	2.2306	1.0	144.0	8.40	0.995	2.6921

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 17.87×10^{-4}

Table A-20. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 94A[a]

Specimen Solid-to-waste ratio: 7 lb dry mix[b]/gal W-7 waste[c] (839 g/liter)
Cesium content: stable = 29 mg
^{137}Cs at time t_o = 0.276 mCi
Mass: 143 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
1	1	11.49	29.51[e,f]	0.2951[e]	1	37	9.24	2.35[e]	1.0883[e]
1	2	11.37	9.26	0.3877	1	38	9.15	2.20	1.1103
1	3	11.26	4.54	0.4331	1	39	9.09	2.22	1.1325
1	4	11.06	3.60	0.4691	4	43	9.97	4.52	1.1777
3	7	11.45	5.75	0.5266	1	44	9.38	2.62	1.2039
1	8	10.71	2.63	0.5529	1	45	9.14	2.25	1.2264
1	9	10.73	2.31	0.5760	1	46	8.96	2.01	1.2465
1	10	10.54	2.10	0.5970	3	49	9.69	3.30	1.2795
1	11	10.40	1.90	0.6160	1	50	9.08	2.42	1.3037
3	14	11.13	4.13	0.6573	1	51	8.92	1.77	1.3214
1	15	10.26	2.19	0.6792	2	53	9.28	2.67	1.3481
1	16	10.19	1.95	0.6987	3	56	9.55	3.37	1.3818
1	17	10.23	2.31	0.7218	7	63	10.09	4.91	1.4309
1	18	10.03	1.91	0.7409	8	71	10.15	6.17	1.4926
3	21	10.77	4.23	0.7832	6	77	9.95	5.51	1.5477
2	23	10.44	3.36	0.8168	7	84	9.91	5.29	1.6006
1	24	9.90	2.25	0.8393	7	91	9.80	4.60	1.6466
1	25	9.66	1.90	0.8583	7	98	9.43	4.88	1.6954
3	28	10.41	4.55	0.9038	7	105	9.39	4.74	1.7429
2	30	10.12	3.62	0.9400	7	112	9.28	4.54	1.7883
1	31	9.61	2.53	0.9653	7	119	9.17	4.38	1.8320
1	32	9.57	2.71	0.9924	7	126	9.05	3.84	1.8704
3	35	10.16	4.38	1.0362	7	133	8.84	3.64	1.9068
1	36	9.52	2.86	1.0648	8	141	8.87	3.97	1.9466

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 29.51×10^{-4}

Table A-21. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 94C[a]

Specimen Solid-to-waste ratio: 7 lb dry mix[b]/gal W-7 waste[c] (839 g/liter)
Cesium content: stable = 29 mg
^{137}Cs at time t_o = 0.276 mCi
Mass: 143 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water, 2 changes/day (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^2$
0.25	0.25	10.77	27.78[e,f]	0.2778[e]	0.25	23.25	9.29	1.118[e]	1.2970[e]
0.75	1.0	11.28	21.74	0.4952	0.75	24.0	9.68	1.757	1.3146
0.25	1.25	10.66	5.86	0.5538	0.25	24.25	8.84	0.847	1.3231
0.75	2.0	11.18	7.15	0.6253	0.75	25.0	9.34	1.659	1.3397
0.25	2.25	10.39	2.56	0.6509	0.25	25.25	8.93	1.027	1.3500
0.75	3.0	11.02	4.13	0.6922	2.75	28.0	10.17	4.529	1.3952
0.25	3.25	10.11	4.92	0.7414	0.25	28.25	9.06	1.086	1.4061
0.75	4.0	10.80	3.05	0.7719	0.75	29.0	9.34	1.941	1.4255
0.25	4.25	9.59	1.48	0.7867	1.0	30.0	9.51	2.231	1.4478
2.75	7.0	11.41	6.56	0.8523	0.25	30.25	8.76	0.808	1.4559
0.25	7.25	9.85	1.56	0.8679	0.75	31.0	9.26	1.878	1.4747
0.75	8.0	10.46	2.65	0.8944	0.25	31.25	8.62	0.769	1.4824
0.25	8.25	9.91	1.22	0.9066	0.75	32.0	9.24	1.965	1.5020
0.75	9.0	10.42	2.35	0.9301	0.25	32.25	8.61	1.038	1.5124
0.25	9.25	9.75	1.10	0.9411	2.75	35.0	9.95	4.457	1.5570
0.75	10.0	10.28	2.00	0.9611	0.25	35.25	8.70	1.081	1.5678
0.25	10.25	9.62	0.992	0.9710	0.75	36.0	9.20	1.923	1.5870
0.75	11.0	10.14	2.11	0.9921	0.25	36.25	g	1.	1.5970
0.25	11.25	9.57	0.978	1.0019	0.75	37.0	9.20	2.479	1.6218
2.75	14.0	10.96	4.836	1.0503	0.25	37.25	8.37	1.010	1.6319
0.25	14.25	9.63	1.212	1.0624	0.75	38.0	8.98	2.128	1.6532
0.75	15.0	9.99	1.898	1.0814	0.25	38.25	8.39	1.037	1.6636
0.25	15.25	9.51	1.026	1.0916	0.75	39.0	8.92	2.175	1.6853
0.75	16.0	9.94	1.995	1.1116	0.25	39.25	8.48	0.872	1.6940
0.25	16.25	9.53	1.340	1.1250	3.75	43.0	9.85	4.476	1.7388
0.75	17.0	9.81	1.925	1.1442	0.25	43.25	8.75	0.894	1.7477
0.25	17.25	9.24	0.847	1.1527	0.75	44.0	9.31	2.428	1.7720
0.75	18.0	9.80	2.003	1.1727	0.25	44.25	8.61	1.196	1.7840
0.25	18.25	9.28	1.030	1.1830	0.75	45.0	9.00	2.241	1.8064
2.75	21.0	10.52	4.725	1.2303	0.25	45.25	8.43	1.033	1.8167
0.25	21.25	9.36	1.180	1.2421	0.75	46.0	8.93	2.424	1.8409
0.75	22.0	9.59	2.031	1.2624	0.25	46.25	8.46	1.042	1.8514
1.0	23.0	9.88	2.349	1.2859	2.75	49.0	9.62	3.872	1.8901

Table A-21 (continued)

Leaching Time (days) Incremental t_n	Leaching Time (days) Cumulative Σt_n	Final pH of Leachate	Fraction Leached Incremental[d] $(a_n/A_o) \times 10^4$	Fraction Leached Cumulative $(\Sigma a_n/A_o) \times 10^2$	Leaching Time (days) Incremental t_n	Leaching Time (days) Cumulative Σt_n	Final pH of Leachate	Fraction Leached Incremental[d] $(a_n/A_o) \times 10^4$	Fraction Leached Cumulative $(\Sigma a_n/A_o) \times 10^2$
0.25	49.25	8.60	1.252[e]	1.9026[e]	4.0	98.0	8.92	2.934[e]	2.7044[e]
0.75	50.0	8.94	2.392	1.9265	1.0	99.0	8.54	1.873	2.7232
0.25	50.25	8.44	0.834	1.9349	1.0	100.0	8.55	1.769	2.7409
0.75	51.0	8.85	2.078	1.9556	1.0	101.0	8.49	1.654	2.7574
0.25	51.25	8.32	0.950	1.9651	1.0	102.0	8.48	1.682	2.7742
1.75	53.0	9.27	2.925	1.9944	3.0	105.0	8.77	2.984	2.8041
0.25	53.25	8.49	1.032	2.0047	1.0	106.0	8.46	1.565	2.8197
2.75	56.0	9.46	3.476	2.0395	1.0	107.0	8.48	1.667	2.8364
1.0	57.0	9.11	2.341	2.0629	1.0	108.0	8.43	1.228	2.8487
1.0	58.0	8.90	2.380	2.0867	1.0	109.0	8.42	1.450	2.8631
1.0	59.0	8.88	2.464	2.1113	3.0	112.0	8.69	2.317	2.8863
4.0	63.0	9.67	4.066	2.1520	1.0	113.0	8.43	1.768	2.9040
1.0	64.0	8.97	2.495	2.1769	1.0	114.0	8.43	1.605	2.9201
1.0	65.0	8.93	2.357	2.2005	1.0	115.0	8.42	1.674	2.9368
1.0	66.0	8.79	2.511	2.2256	1.0	116.0	8.46	1.551	2.9523
1.0	67.0	8.80	2.313	2.2487	3.0	119.0	8.71	2.472	2.9770
4.0	71.0	9.38	3.939	2.2881	1.0	120.0	8.52	1.649	2.9935
1.0	72.0	8.90	2.491	2.3130	1.0	121.0	8.45	1.689	3.0104
1.0	73.0	8.87	2.589	2.3389	1.0	122.0	8.46	1.387	3.0243
1.0	74.0	8.78	2.376	2.3627	1.0	123.0	8.43	1.315	3.0374
3.0	77.0	9.20	3.447	2.3972	3.0	126.0	8.62	2.015	3.0576
1.0	78.0	8.68	2.352	2.4207	1.0	127.0	8.50	1.397	3.0716
1.0	79.0	8.72	2.158	2.4423	1.0	128.0	8.43	1.434	3.0859
1.0	80.0	8.69	2.380	2.4661	1.0	129.0	8.37	1.301	3.0989
1.0	81.0	8.65	2.050	2.4866	1.0	130.0	8.37	1.071	3.1096
3.0	84.0	9.08	3.513	2.5217	3.0	133.0	8.55	2.104	3.1306
1.0	85.0	8.55	2.200	2.5437	1.0	134.0	8.24	1.399	3.1446
1.0	86.0	8.61	2.080	2.5645	1.0	135.0	8.22	1.473	3.1594
1.0	87.0	8.59	1.879	2.5833	1.0	136.0	8.39	1.293	3.1723
1.0	88.0	8.59	1.926	2.6025	1.0	137.0	8.44	1.250	3.1848
3.0	91.0	8.72	2.310	2.6256	4.0	141.0	8.84	2.351	3.2083
1.0	92.0	8.48	1.776	2.6434	1.0	142.0	8.43	1.369	3.2220
1.0	93.0	8.48	1.604	2.6594	1.0	143.0	8.42	1.493	3.2369
1.0	94.0	8.50	1.566	2.6751	1.0	144.0	8.39	1.416	3.2511

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 27.78×10^{-4}.

[g]pH not measured.

Table A-22. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 3A[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-9/10 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0554 mCi
Mass: 134 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$
1	1	11.22	9.828[e,f]	0.9828[e]
1	2	10.68	5.206	1.5034
1	3	10.44	4.027	1.9061
1	4	10.19	3.600	2.2661
3	7	11.04	8.124	3.0785
1	8	10.02	3.856	3.4641
1	9	9.83	3.146	3.7787
1	10	9.67	2.856	4.0643
1	11	9.55	2.589	4.3232
3	14	10.44	4.459	4.7691
1	15	9.38	2.253	4.9944
1	16	9.30	2.539	5.2483
1	17	9.23	2.096	5.4579
4	21	10.31	4.755	5.9334
1	22	9.39	2.563	6.1897
1	23	9.08	1.723	6.3620
1	24	8.97	2.020	6.5640
1	25	8.90	1.837	6.7477
3	28	9.64	2.861	7.0338
1	29	8.86	1.808	7.2146
1	30	8.89	1.448	7.3594
1	31	8.71	1.720	7.5314
1	32	8.76	1.246	7.6560
3	35	9.51	2.710	7.9270
1	36	8.84	1.360[e]	8.0630[e]
1	37	8.81	1.163	8.1793
1	38	8.90	0.828	8.2621
1	39	8.76	1.340	8.3961
3	42	9.27	2.396	8.6357
1	43	8.55	1.167	8.7524
1	44	8.55	0.958	8.8482
1	45	8.42	1.000	8.9482
1	46	8.52	1.042	9.0524
3	49	9.05	1.708	9.2232
1	50	8.53	0.944	9.3176
1	51	8.49	1.028	9.4204
1	52	8.51	1.091	9.5295
1	53	8.48	0.797	9.6092
3	56	8.83	1.685	9.7777
1	57	8.36	0.963	9.8740
1	58	8.24	0.901	9.9641
1	59	8.43	1.087	10.0728
1	60	8.57	0.942	10.1670
4	64	9.09	2.295	10.3965
1	65	8.52	0.960	10.4925
1	66	8.49	1.000	10.5925
1	67	8.49	0.638	10.6563
3	70	8.74	1.694	10.8257

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-9/10 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 9.828×10^{-4}.

Table A-23. Experimental Results for the Leaching of Cesium from Cementitious Grout No. 3C[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-9/10 waste[c] (719 g/liter)
Cesium content: stable = 27 mg
^{137}Cs at time t_o = 0.0554 mCi
Mass: 134 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached		Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$	Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^4$	Cumulative $(\Sigma a_n/A_o) \times 10^3$
1	1	11.19	22.12[e,f]	2.212[e]	1	36	8.92	1.915[e]	10.4703[e]
1	2	10.76	9.199	3.1319	1	37	8.85	1.495	10.6198
1	3	10.54	5.589	3.6908	1	38	8.98	1.117	10.7315
1	4	10.27	4.531	4.1439	1	39	8.82	1.381	10.8696
3	7	11.10	8.907	5.0346	3	42	9.44	2.375	11.1071
1	8	10.21	4.000	5.4346	1	43	8.69	2.417	11.3488
1	9	9.96	3.312	5.7658	1	44	8.68	1.313	11.4801
1	10	9.92	3.411	6.1069	1	45	8.62	0.854	11.5655
1	11	9.17	3.637	6.4706	1	46	8.63	1.104	11.6759
3	14	10.41	4.601	6.9307	3	49	9.20	1.979	11.8738
1	15	9.48	3.113	7.2420	1	50	8.67	1.301	12.0039
1	16	9.36	2.457	7.4877	1	51	8.65	1.259	12.1298
1	17	9.43	2.404	7.7281	1	52	8.77	1.196	12.2494
4	21	10.30	5.143	8.2424	1	53	8.74	0.692	12.3186
1	22	9.52	2.746	8.5170	3	56	9.09	2.014	12.5200
1	23	9.20	1.824	8.6994	1	57	8.54	1.416	12.6616
1	24	9.11	1.980	8.8974	1	58	8.50	0.942	12.7558
1	25	9.06	1.633	9.0607	1	59	8.68	1.087	12.8645
3	28	9.81	3.587	9.4194	1	60	8.73	0.963	12.9608
1	29	9.12	2.034	9.6228	4	64	9.36	1.966	13.1574
1	30	8.97	1.966	9.8194	1	65	8.74	1.202	13.2776
1	31	8.95	1.238	9.9432	1	66	8.64	1.182	13.3958
1	32	8.86	1.349	10.0781	1	67	8.75	0.940	13.4898
3	35	9.62	2.007	10.2788	3	70	8.90	1.714	13.6612

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-9/10 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 22.12×10^{-4}.

Table A-24. Experimental Results for the Leaching of Plutonium from Cementitious Grout No. 35A[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Plutonium content: stable = none
^{239}Pu at time t_o = 10 μCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 0 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^6$	Cumulative $(\Sigma a_n/A_o) \times 10^4$
1	1	12.70	90.0[e,f]	0.900[e]
1	2	12.70	5.61	0.956
4	6	12.85	3.51	0.991
1	7	12.25	5.00	1.041
1	8	12.10	2.39	1.065
1	9	12.10	1.11	1.076
5	14	12.45	1.45	1.091
9	23	11.67	1.94	1.110
12	35	11.60	1.20	1.122
27	62	11.70	0.904	1.131
28	90	11.57	0.884	1.140
28	118	11.48	0.260	1.143

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 90.0×10^{-6}.

Table A-25. Experimental Results for the Leaching of Plutonium from Cementitious Grout No. 35B[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Plutonium content: stable = none
^{239}Pu at time t_o = 10 μCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^6$	Cumulative $(\Sigma a_n/A_o) \times 10^6$
1	1	12.55	2.99[e,f]	2.99[e]
1	2	12.35	1.27	4.26
5	7	12.35	1.92	6.18
9	16	11.54	2.62	8.80
12	28	11.60	0.556	9.356
27	55	11.69	0.490	9.846
28	83	11.53	0.188	10.034
28	111	11.43	0.159	10.193

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 2.99 as 10^{-6}.

Table A-26. Experimental Results for the Leaching of Plutonium from Cementitious Grout No. 35C[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Plutonium content: stable = none
^{239}Pu at time t_o = 10 μCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^6$	Cumulative $(\Sigma a_n/A_o) \times 10^6$
1	1	11.30	0.14[e,f]	0.14[e]
6	7	11.56	1.67	1.81
27	34	11.72	1.02	2.83
28	62	11.60	0.65	3.484
28	90	11.31	1.12	4.604

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 0.14×10^{-6}.

Table A-27. Experimental Results for the Leaching of Curium from Cementitious Grout No. 36A[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Curium content: stable = none
^{244}Cm at time t_o = 10 μCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 0 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^6$	Cumulative $(\Sigma a_n/A_o) \times 10^4$
1	1	12.70	655.[e,f]	6.55[e]
1	2	12.60	83.2	7.382
4	6	12.80	58.4	7.966
1	7	12.25	12.9	8.095
1	8	12.10	9.69	8.192
1	9	11.95	4.31	8.235
1	10	12.00	6.02	8.295
4	14	12.30	6.49	8.360
9	23	11.62	5.38	8.414
12	35	11.62	4.16	8.456
27	62	11.66	3.31	8.489
28	90	11.50	2.21	8.511
28	118	11.52	1.51	8.526

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 655. x 10^{-6}.

Table A-28. Experimental Results for the Leaching of Curium from Cementitious Grout No. 36B[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Curium content: stable = none
^{244}Cm at time t_o = 10 μCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 7 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^6$	Cumulative $(\Sigma a_n/A_o) \times 10^4$
1	1	12.55	69.0[e,f]	0.69[e]
1	2	12.30	16.0	0.85
1	3	12.20	11.1	0.961
4	7	12.35	20.0	1.161
9	16	11.55	4.43	1.205
12	28	11.60	3.86	1.244
27	55	11.59	2.34	1.267
28	83	11.52	0.418	1.271
28	111	11.33	0.186	1.273

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 69.0×10^{-6}.

Table A-29. Experimental Results for the Leaching of Curium from Cementitious Grout No. 36C[a]

Specimen Solid-to-waste ratio: 6 lb dry mix[b]/gal W-7 waste[c] (719 g/liter)
Curium content: stable = none
^{244}Cm at time t_o = 10 μCi
Mass: 137 g (as prepared)
Volume: 98 cm^3
Exposed surface area: 19.6 cm^2

Curing Time 28 days

Leachant 150 ml tap water (initial pH = 7.96 - 8.25)

Leaching Time (days)		Final pH of Leachate	Fraction Leached	
Incremental t_n	Cumulative Σt_n		Incremental[d] $(a_n/A_o) \times 10^6$	Cumulative $(\Sigma a_n/A_o) \times 10^6$
1	1	11.16	0.137[e,f]	0.137[e]
6	7	11.60	1.91	2.047
27	34	11.68	1.55	3.597
28	62	11.63	1.60	5.197
28	90	11.47	0.856	6.053

[a]Composition of specimen given in Table 3.

[b]Composition of dry mix given in Table 1.

[c]Composition of waste given in Table 2.

[d]An equivalent sample of the original W-7 waste solution with tracer is used as the standard at each leaching time (see Sect. 2).

[e]To obtain the experimental value, multiply by the reciprocal of the power of 10 appearing in the column heading.

[f]For example, read as 0.137×10^{-6}.

APPENDIX B: SOLUTION OF MASS TRANSPORT EQUATIONS FOR THE CASE OF DIFFUSION WITH INSTANTANEOUS REACTION IN A SEMI-INFINITE MEDIUM

Consider a semi-infinite medium with a uniform initial concentration in which the concentration of the less-mobile forms of a species, F, is directly proportional to the concentration of the more-mobile forms of the species, C; that is,

$$F = KC, \tag{B-1}$$

where F is the proportionality or equilibrium constant (dimensionless). From a mass balance across a differential section, the relationship to be solved is:

$$\frac{\partial C}{\partial t} = \mathcal{D}_e \frac{\partial^2 C}{\partial x^2} - \frac{\partial F}{\partial t} \quad . \tag{B-2a}$$

If the surface concentration of mobile species is maintained at zero for time greater than zero, the initial and boundary conditions are:

$$x > 0, \; t = 0, \; C = C_o, \tag{B-2b}$$

$$x = 0, \; t > 0, \; C = 0, \tag{B-2c}$$

$$x = \infty, \; t > 0, \; C = C_o, \tag{B-2d}$$

where x is a space coordinate (cm), $\mathcal{D}_e$ is an effective diffusivity (cm^2/sec), and t is time (sec). The solution (e.g., by application of the method given in ref. 26 or that given in ref. 27) of Eqs. (B-2) is:

$$C = C_o \operatorname{erf}\left[\frac{x}{2}\left(\frac{1 + K}{\mathcal{D}_e t}\right)^{1/2}\right] , \tag{B-3}$$

where

$$\operatorname{erf}(u) = \frac{2}{\pi^{1/2}} \int_0^u e^{-\beta^2} d\beta \; . \tag{B-4}$$

Properties and tabulated values of the error function (erf) are given in comprehensive mathematical handbooks such as ref. 28.

A material balance for a finite specimen of the material being leached gives:

$$\frac{dA}{dt} = \frac{d}{dt}\left[\iiint C\ dx\ dy\ dz + \iiint F\ dx\ dy\ dz\right]; \tag{B-5}$$

or, since F = KC from Eq. (B-1),

$$\frac{dA}{dt} = \frac{d}{dt}\left[(1 + K)\iiint C\ dx\ dy\ dz\right], \tag{B-6}$$

where A is the amount of a species present in the specimen at any time t. Through the use of Green's theorem (ref. 29), Eq. (B-6) may be written as:

$$\frac{dA}{dt} = (1 + K)\iint\left(\frac{\mathcal{D}_e}{1 + K}\right)\left(\frac{\partial C}{\partial N}\right) dS, \tag{B-7}$$

where $\partial C/\partial N$ is the concentration gradient measured in the outward direction along the normal to the surface S.

As long as the finite specimen approximates a semi-infinite medium, Eq. (B-3) can be used to approximate the concentration profile in the finite specimen. Thus, differentiation of Eq. (B-3) with respect to x, evaluation of this gradient at x = 0, and insertion of this gradient into Eq. (B-7) give:

$$dA = S\ (1 + K)\ C_o\left[\frac{\mathcal{D}_e}{\pi\ (1 + K)}\right]^{1/2} t^{-1/2}\ dt. \tag{B-8}$$

Integration of Eq. (B-8) over time t yields:

$$\frac{\Sigma a_n}{S} = \frac{2(1 + K)\ C_o}{\pi^{1/2}}\left(\frac{\mathcal{D}_e}{1 + K}\right)^{1/2} t^{1/2}, \tag{B-9}$$

where Σa_n is the total amount of the species which has left the medium in time t. For the finite specimen,

$$C_T = \frac{A_o}{V}, \tag{B-10}$$

where C_T is the initial total concentration of the species, A_o is the initial amount of the species in the specimen, and V is the volume of the specimen (cm^3). For both the finite specimen and the semi-infinite medium,

$$C_T = C_o + F_o = (1 + K)C_o. \tag{B-11}$$

By definition, let

$$D_e = \frac{\mathcal{D}_e}{1 + K} . \tag{B-12}$$

The substitution of Eqs. (B-10), B-11), and (B-12) into Eq. (B-9) gives:

$$\left(\frac{\Sigma a_n}{A_o}\right)\left(\frac{V}{S}\right) = 2 \left(\frac{D_e}{\pi}\right)^{1/2} t^{1/2} \tag{B-13}$$

for the $\left[\text{(cumulative fraction) x (cm)}\right]$ of the species which has left the specimen in time t.

Equation (B-13) is the same as that obtained for the corresponding case of diffusion only, except that a modified effective diffusivity (which includes the proportionality or equilibrium constant) is used.

It should be kept in mind that Eq. (B-13) is valid for the finite specimen only as long as it approximates a semi-infinite medium. Eventually, enough of a species will be leached from a finite specimen to invalidate the boundary condition given by Eq. (B-2d). Subsequently, Eq. (B-13) will overpredict the cumulative fraction leached and consideration must be given to solving the law of conservation of mass, taking into account the finite size of the specimen (e.g., see Appendix C and Sect. 6).

APPENDIX C: SOLUTION OF MASS TRANSPORT EQUATIONS FOR THE CASE OF DIFFUSION WITH INSTANTANEOUS REACTION IN A FINITE CYLINDER

Consider a finite cylinder of radius, a, and length, 2ℓ, with a uniform initial concentration in which the concentration of the less-mobile forms of a species, F, is directly proportional to the concentration of the more-mobile forms of the species, C, that is:

$$F = KC, \tag{C-1}$$

where K is the proportionality or equilibrium constant (dimensionless). From a mass balance across a differential section, the relationship to be solved is:

$$\frac{\partial C}{\partial t} = \mathcal{D}_e \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial z^2} \right) - \frac{\partial F}{\partial t} \quad . \tag{C-2a}$$

If the surface concentration of mobile species is maintained at zero for time greater than zero, the initial and boundary conditions are:

$$t = 0,\ r < a,\ C = C_o, \tag{C-2b}$$

$$t = 0,\ -\ell < z < \ell,\ C = C_o, \tag{C-2c}$$

$$t > 0,\ r = a,\ C = 0, \tag{C-2d}$$

$$t > 0,\ z = \pm\ell,\ C = 0, \tag{C-2e}$$

$$t > 0,\ r = 0,\ \partial C/\partial r = 0, \tag{C-2f}$$

$$t > 0,\ z = 0,\ \partial C/\partial z = 0, \tag{C-2g}$$

where r is a radial coordinate (cm), z is an axial coordinate (cm), $\mathcal{D}_e$ is an effective diffusivity (cm^2/sec), and t is time (sec). The solution of Eq. (C-2) can be obtained as the product of the solution for an infinite cylinder and a slab as shown in ref. 30. This solution is:

$$C = \frac{8C_o}{\pi a} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{(-1)^n J_o(r\alpha_m)}{(2n-1)\alpha_m J_1(a\alpha_m)} \cos \frac{(2n-1)\pi z}{2\ell}$$

$$\times\ e^{-[\mathcal{D}_e/(1+K)]\,[\alpha_m^2 + (2n-1)^2\pi^2/4\ell^2]t}, \tag{C-3}$$

where J_o and J_1 are the zero-order and first-order Bessel functions of the first kind and the α_m's are the positive roots of the zero-order Bessel function $J_o\ (a\alpha_m) = 0$.

A material balance over the cylinder for the material being leached gives:

$$\Sigma a_n = A_o - \int_o^a \int_{-\ell}^{\ell} C(2\pi r)\ dr\ dz - \int_o^a \int_{-\ell}^{\ell} F(2\pi r)\ dr\ dz; \qquad \text{(C-4)}$$

or since F = KC from Eq. (C-1), Eq. (C-4) may be expressed as:

$$\Sigma a_n = A_o - (1 + K) \int_o^a \int_{-\ell}^{\ell} C(2\pi r)\ dr\ dz, \qquad \text{(C-5)}$$

where Σa_n is the total amount of the species which has left the cylinder in time t and A_o is the total amount of the species in the cylinder at time t = 0. For the specimen,

$$C_T = \frac{A_o}{V} = \frac{A_o}{\pi a^2 (2\ell)}, \qquad \text{(C-6)}$$

and

$$C_T = C_o + F_o = (1 + K)\ C_o, \qquad \text{(C-7)}$$

where C_T is the total concentration of the species in the cylinder at time t = 0, and V is the volume of the specimen (cm^3). By definition, let

$$D_e = \frac{\mathcal{D}_e}{1 + K}. \qquad \text{(C-8)}$$

The integration of Eq. (C-5) with C expressed by Eq. (C-3) and the substitution of Eqs. (C-6), (C-7), and (C-8) into the resulting expression yield:

$$\frac{\Sigma a_n}{A_o} = 1 - \frac{32}{\pi^2 a^2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{e^{-D_e\ [\alpha_m^2 + (2n-1)^2 \pi^2/4\ell^2]t}}{(2n - 1)^2\ \alpha_m^2} \qquad \text{(C-9)}$$

for the cumulative fraction of the species which has left the cylinder in time t.

Equation (C-9) is the same as that obtained for the corresponding case of diffusion only, except that a modified effective diffusivity (which includes the proportionality or equilibrium constant) is used. In other words, for diffusion only, $K = 0$ and all of the species is mobile at $t = 0$.

ORNL-4962

UC-70 - Waste Management

INTERNAL DISTRIBUTION

1. P. C. Arwood
2. F. T. Binford
3. R. E. Blanco
4. J. O. Blomeke
5. W. D. Bond
6. R. E. Brooksbank
7. K. B. Brown
8. F. R. Bruce
9. D. O. Campbell
10. H. C. Claiborne
11. W. E. Clark
12. C. F. Coleman
13. L. T. Corbin
14. J. A. Cox
15. A. G. Croff
16. F. L. Culler
17. W. Davis, Jr.
18. G. J. Dixon
19. R. B. Evans III
20. D. E. Ferguson
21. L. M. Ferris
22. E. J. Frederick
23-32. H. W. Godbee
33. R. F. Hibbs
34. D. G. Jacobs
35. G. H. Jenks
36-40. D. S. Joy
41. C. W. Kee
42-46. A. H. Kibbey
47. L. C. Lasher
48. R. E. Leuze
49. K. H. Lin
50. R. S. Lowrie
51. A. P. Malinauskas
52. W. C. McClain
53-62. J. G. Moore
63. J. P. Nichols
64. K. J. Notz
65. H. Postma
66. R. A. Robinson
67. T. H. Row
68. A. D. Ryon
69. C. D. Scott
70. J. W. Snider
71. E. G. Struxness
72. T. Tamura
73. I. L. Thomas
74. D. B. Trauger
75. W. E. Unger
76. H. O. Weeren
77. E. J. Witkowski
78. R. G. Wymer
79. J. C. Frye (consultant)
80. C. H. Ice (consultant)
81. J. J. Katz (consultant)
82. E. A. Mason (consultant)
83. Ken Davis (consultant)
84. R. B. Richards (consultant)
85. Biology Library
86-87. Central Research Library
88. ORNL - Y-12 Technical Library Document Reference Section
89. MIT Practice School
90-114. Laboratory Records Department
115. Laboratory Records, R.C.
116. ORNL Patent Office
117. Nuclear Safety Information Center

EXTERNAL DISTRIBUTION

118. E. H. Hardison, ERDA-ORO
119. C. B. Bartlett, Directorate of Regulatory Standards, U.S. Nuclear Regulatory Commission, Washington, D.C. 20545
120. M. J. Bell, Directorate of Licensing, U.S. Nuclear Regulatory Commission, Washington, D.C. 20545
121. V. Benaroya, Directorate of Licensing, U.S. Nuclear Regulatory Commission, Washington, D.C. 20545

122. W. K. Eister, ERDA, Division of Waste Management and Transportation, Washington, D.C. 20545
123. A. F. Kluk, ERDA, Division of Waste Management and Transportation, Washington, D.C. 20545
124. G. H. Daly, ERDA, Division of Waste Management and Transportation, Washington, D.C. 20545
125. F. K. Pittman, ERDA, Division of Waste Management and Transportation, Washington, D.C. 20545
126. R. W. Ramsey, Jr., ERDA, Division of Waste Management and Transportation, Washington, D.C. 20545
127. R. D. Walton, Jr., ERDA, Division of Waste Management and Transportation, Washington, D.C. 20545
128. R. K. Heusser, ERDA, Division of Production and Materials Management, Washington, D.C. 20545
129. K. K. Kennedy, ERDA, Idaho Operations Office, Idaho Falls, Idaho 83401
130. R. B. Goranson, ERDA, Richland Operations Office, Richland, Washington 99352
131. N. T. Karagianes, ERDA, Richland Operations Office, Richland, Washington 99352
132. H. B. Saucier, ERDA, Savannah River Operations Office, Aiken, South Carolina 29801
133. L. E. Brownell, Atlantic Richfield Hanford Company, Richland, Washington 99352
134. M. H. Campbell, Atlantic Richfield Hanford Company, Richland, Washington 99352
135. R. E. Isaacson, Atlantic Richfield Hanford Company, Richland, Washington 99352
136. M. J. Kupfer, Atlantic Richfield Hanford Company, Richland, Washington 99352
137. W. W. Shulz, Atlantic Richfield Hanford Company, Richland, Washington 99352
138. A. E. Smith, Atlantic Richfield Hanford Company, Richland, Washington 99352
139. D. E. Griffing, Atlantic Richfield Hanford Company, Richland, Washington 99352
140. D. W. Rhodes, Allied Chemical Company, Idaho Falls, Idaho 83401
141. C. M. Slansky, Allied Chemical Company, Idaho Falls, Idaho 83401
142. W. J. Mecham, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439
143. J. L. McElroy, Battelle Pacific Northwest Laboratories, Richland, Washington 99352
144. J. E. Mendel, Battelle Pacific Northwest Laboratories, Richland, Washington 99352
145. A. M. Platt, Battelle Pacific Northwest Laboratories, Richland, Washington 99352
146. Peter Colombo, Brookhaven National Laboratory, Upton, New York 11973
147. Meyer Steinberg, Brookhaven National Laboratory, Upton, New York 11973
148. C. W. Christenson, Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87544
149. L. A. Emility, Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87544

150. A. S. Jennings, E. I. du Pont de Nemours & Co., Inc., Savannah River Plant, Aiken, South Carolina 29801
151. R. M. Wallace, E. I. du Pont de Nemours & Co., Inc., Savannah River Plant, Aiken, South Carolina 29801
152. J. L. Mahloch, Office for Environmental Services, Waterways Experiment Station, P. O. Box 631, Vicksburg, Mississippi 39180
153. M. E. Wadsworth, Department of Metallurgy, University of Utah, Salt Lake City, Utah 84112
154. F. L. Parker, Environmental & Water Resources Engineering, Vanderbilt University, Nashville, Tennessee 37235
155. C. W. Angell, Vice President - Sales, Artisan Industries, Inc., 73 Pond Street, Waltham, Massachusetts 02154
156. Ken Gablin, Vice President - Marketing, Protective Packaging, Inc., P. O. Box 1192, Tacoma, Washington 98401
157. Paul C. Williams, Stock Equipment Company, Research and Development Department, 16490 Chillicothe Road, Chagrin Falls, Ohio 44022
158. D. H. Curtiss, United Nuclear Industries, Incorporated, 1201 Jadwin Avenue, Richland, Washington 99352
159. Lawrence Rutland, Vice President, ATCOR, Incorporated, 5 Westchester Plaza, Elmsford, New York 10523
160. A. A. Aikens, Jr., PRS Systems, Incorporated, 63 East Sanford Boulevard, Mt. Vernon, New York 10550
161. L. T. Lakey, EXXON Nuclear Company, 2101 Horn Rapids Road, Richland Washington 99352
162. S. M. Goldberg, Office of Radiation Programs, Technology Assessment Division, U.S. Environmental Protection Agency, Washington, D.C. 20460
163. R. T. Meyer, Chemical Technology Division 5824, Sandia Laboratories, Albuquerque, New Mexico 87115
164. J. T. Collins, Environmental Consultants, Inc., 14325 Proton Rd., Dallas, Texas 75240
165. F. Girardi, Chemistry Division, J.R.C. EURATOM, 21020, Ispra, Italy
166. Ferruccio Gera, C.N.E.N., Viale Regina Margherita 125, 00198 Rome, Italy
167. H. O. G. Witte, NUKEM GmbH, Wolfgang bei Hanau/Main, West Germany
168. H. W. Levi, Hahn-Meitner-Institut, 1 Berlin 39, Glienickerstr. 100, West Germany
169. W. Lutze, Hahn-Meitner-Institut, 1 Berlin 39, Glienickerstr. 100, West Germany
170. H. Krause, Nuclear Research Center, Waste Management Dept., Weberstr. 5, Karlsruhe, West Germany
171. Nicola Zaghini, Agip Nucleare, Medicina (Bologna), Italy
172. K. T. Thomas, Bhabha Atomic Research Centre, Waste Treatment Division, Trombay, Bombay, India
173. N. S. S. Rajan, Bhabha Atomic Research Centre, Waste Treatment Division, Trombay, Bombay, India
174. K. D. B. Johnson, Atomic Energy Research Establishment, Harwell, Berkshire, England
175. J. R. Grover, Atomic Energy Research Establishment, Harwell, Berkshire, England
176. D. W. Clelland, United Kingdom Atomic Energy Authority, Risley, England

177. S. G. Amarantos, Greek Atomic Energy Commission, Nuclear Research Center "Democritus", Aghia Paraskevi-Attikis, Athens, Greece
178. J. H. Petropoulos, Greek Atomic Energy Commission, Nuclear Research Center "Democritus", Aghia Paraskevi-Attikis, Athens, Greece
179. E. D. Hespe, Australian Atomic Energy Commission Research Establishment, Sutherland, N.S.W., Australia
180. Research and Technical Support Division, ERDA, ORO
181-395. Given distribution as shown in TID-4500 under UC-70, Waste Management category

☆U. S. GOVERNMENT PRINTING OFFICE: 1974-748-189/188