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ANALYSIS OF DATA FROM LEACHING CONCRETE SAMPLES TAKEN FROM THE TMI-2 REACTOR BUILDING BASEMENT

E. D. Collins, W. D. Box, H. W. Godbee, and T. C. Scott

Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831

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Fig. 1. Location of concrete structures sampled.

Fig. 2. Hassler cell apparatus for measurement of permeability.

Fig. 3. Relationship of ^{137}Cs contamination to permeability of the structure (concrete, paint, etc.).

Fig. 4. Apparatus for leach tests.

Fig. 5. Leaching from unpainted concrete block (SUB-3).

Fig. 6. Leaching from 3000-psi painted concrete (C-34).

Fig. 7. Analogy of models for packed-bed and concrete-block systems.

Fig. 8. Predicted elution of cesium and strontium from a concrete block wall.

ABSTRACT

Samples of contaminated concrete from the basement of the reactor building at the Three Mile Island Nuclear Power Station, Unit 2 were tested and analyzed at Oak Ridge National Laboratory to determine the potential for decontamination by diffusion-controlled leaching under conditions of full submergence and by forced flow-through leaching of porous concrete block walls. Pertinent physical characteristics of the concrete were measured, and leaching tests were performed. Data were analyzed by established mass transport principles, and predictions of leaching for several years were made. A numerical algorithm was used to model removal of ^{137}Cs and ^{90}Sr by forced flow-through leaching. Results indicated that forced flow-through leaching would require only a few days, whereas complete decontamination by submerged, diffusion-only methods would require several years.

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Oak Ridge National Laboratory

The accident at the Three Mile Island Nuclear Power Station, Unit 2 (TMI-2) in March 1979 caused ~2500 m^3 of high-activity-level water to spill into the reactor building basement. The water collected (~2.5 m deep) in the basement and remained there until late 1981. A processing period was then begun, and the water (containing 160 μCi/mL of ^{137}Cs and 2.3 μCi/mL of ^{90}Sr) was transferred through an ion-exchange decontamination system into storage tanks.[1]

Following removal of the high-activity-level water, periodic spraying and flushing of accessible portions of the basement floor and walls were carried out. However, radiation sources remained too high (~100 R/h in some areas) to permit entry. Full submergence decontamination was considered, but no data were available to compare potential benefits with costs.

In 1985 and 1986, samples of concrete were obtained from several areas in the basement by means of robotic equipment. Leaching tests on three samples, representing different types of concrete, were made at Idaho National Engineering Laboratory; and the results indicated that such treatment could induce significant decontamination.[2] More extensive leach tests, measurements of pertinent characteristics of the concrete (e.g., density, porosity, and permeability), and analyses of the data were made at Oak Ridge National Laboratory (ORNL) in 1986 and 1987.

The testing, analysis, modeling calculations, and predictions of removal rates for the radionuclides, ^{137}Cs and ^{90}Sr, were carried out for various types of concrete that are present in structures located in the TMI-2 reactor building basement. Both diffusion-controlled and forced flow-controlled leaching models were used in these studies.

DESCRIPTION OF CONCRETE SAMPLES

Samples of six different types of concrete were taken from structures located in the TMI-2 reactor building basement, as illustrated in the plan view diagram shown in Fig. 1. The six types of concrete structures that were sampled included (1) unpainted block wall; (2) painted block wall; (3) unpainted, 3000-psi compressive strength (hereinafter referred to as "3000-psi") wall; (4) painted, 3000-psi wall; (5) painted, 3000-psi floor; and (6) painted, 5000-psi wall.

Initially, four samples were sent to ORNL in late 1986 for measurement of physical characteristics. Two of these samples (SCB-5 and SC5-6) were subsequently used in the leach tests, along with six others that were sent to ORNL in early 1987. All of the samples were cylindrical cores, and each had a diameter of ~1.25 in.; however, their lengths varied. The descriptions of these samples, as determined at TMI-2, are listed in Table I.

Upon receipt at ORNL, samples SCB-5 and SU3-3 were found to be broken into several pieces. Before testing, these samples were reassembled by using paraffin to stick the pieces together. Another observation was that samples SUB-3, SUB-7, SU3-3, C-34, C-31, and Floor/b had been wrapped in duct tape prior to shipment from TMI-2, and this tape was difficult to remove at ORNL using remotely operated handling tools (use of these tools was necessary because several of the

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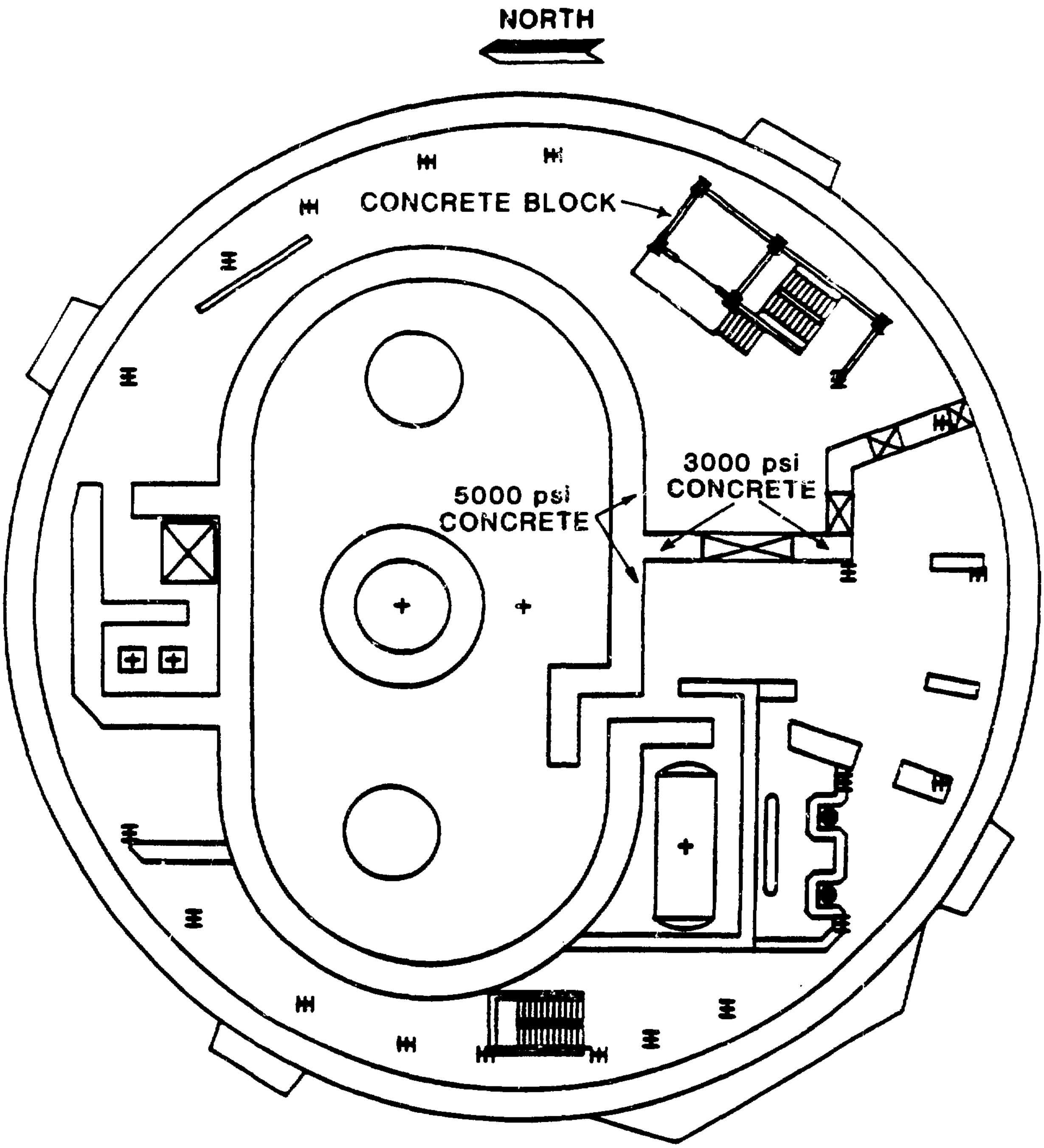

LOCATION OF CONCRETE STRUCTURES SAMPLED

Table I. Description of Samples

Sample Identification	Type[a]	Sample Point Elevation on Wall (ft-in.)	Length (in.)	Total ^{137}Cs Activity (μCi)	Dose Rate Gamma (rad/h)	Dose Rate Beta (rad/h)
SUB-1[b]	Unpainted block wall		1-1/4	12,516	1.8	2.0
SCB-5[b,c]	Painted block wall	3-8	3/8	6,980	2.0	32
SC5-3[b]	Painted 5000-psi[d] wall		1/2	34.9	0.005	0.04
SC5-6[b,c]	Painted 5000-psi[d] wall	6-3	7/8	1,829	0.4	4.0
SUB-3[c]	Unpainted block wall	6-0	3/4	70,400	10	16
SUB-7[c]	Unpainted block wall	6-4	1-3/16	62,000	Not Reported	
SU3-3[c]	Unpainted 3000-psi[d] wall	6-3	5/8	30,700	12	600
C-34[c]	Painted 3000-psi[d] wall	2-10	15/16	1,600	Not Reported	
C-31[c]	Painted 3000-psi[d] wall	3-7	5/8	350	0.06	0.8
Floor/b[c]	Painted 3000-psi[d] floor		2-1/2	90	0.03	0.4

[a]All samples were cylindrical cores, and each had a diameter of ~1.25 in.
[b]Samples used at ORNL for measurement of physical characteristics.
[c]Sample used at ORNL for leach tests.
[d]Poured concrete having the indicated compressive strength.

samples emitted relatively high gamma radiation). Some of the tape mastic stuck to the sample faces and could not be totally removed. These factors could have caused some degree of inaccuracy during the testing.

Measurement of Pertinent Physical Characteristics

Several properties of porous media, such as the density, porosity, and permeability, are needed to evaluate mass transport by forced flow of leachant. These properties were measured, using the first four samples (SUB-1, SCB-5, SC5-3, and SC5-6) sent to ORNL, and are listed in Table II.

The solid densities were measured by weighings in air (after vacuum drying) and in water. The porosities could be determined only approximately because of difficulties in determining the volumes of samples, which were irregularly shaped.

Permeabilities were measured by means of a Hassler Cell, as shown in Fig. 2. In using this apparatus, the pressure chamber is first disassembled and the sample is placed on top of the support plug (in the case where one end surface of the cylindrical sample was painted, the painted surface was placed toward the flow of water); then, the chamber is reassembled, and water is pressured through the sample into a collection bottle. The pressure is increased to provide a pressure drop across the sample of ~100 psig (except in the test of the unpainted concrete block sample in which only a maximum pressure of ~10 psig could be attained); then, the flow rate of water through the sample is measured over a ~24-h period. Using the results of the test, the

Table II. Measured Properties of TMI-2 Concrete Samples

Sample Identification	Wall Type and Coating	Sample Length[a] (in.)	^{137}Cs Activity[a] (μCi)	Measured Permeability (ft^2)	Solid Density (g/mL of solid)	Approximate Porosity (mL voids/[mL solids + mL voids])
SUB-1	Unpainted, block	1-1/4	12,515.8	2.2E-11	2.49	~0.35
SCB-5	Painted, block	3/8	6,980	5.3E-15	2.45	~0.35
SC5-3	Painted, 5000-psi	1/2	34.9	1.0E-18	2.42	~0.15
SC5-6	Painted, 5000-psi	7/8	1,828.5	4.3E-16	2.47	~0.15

[a]Measured at TMI-2.

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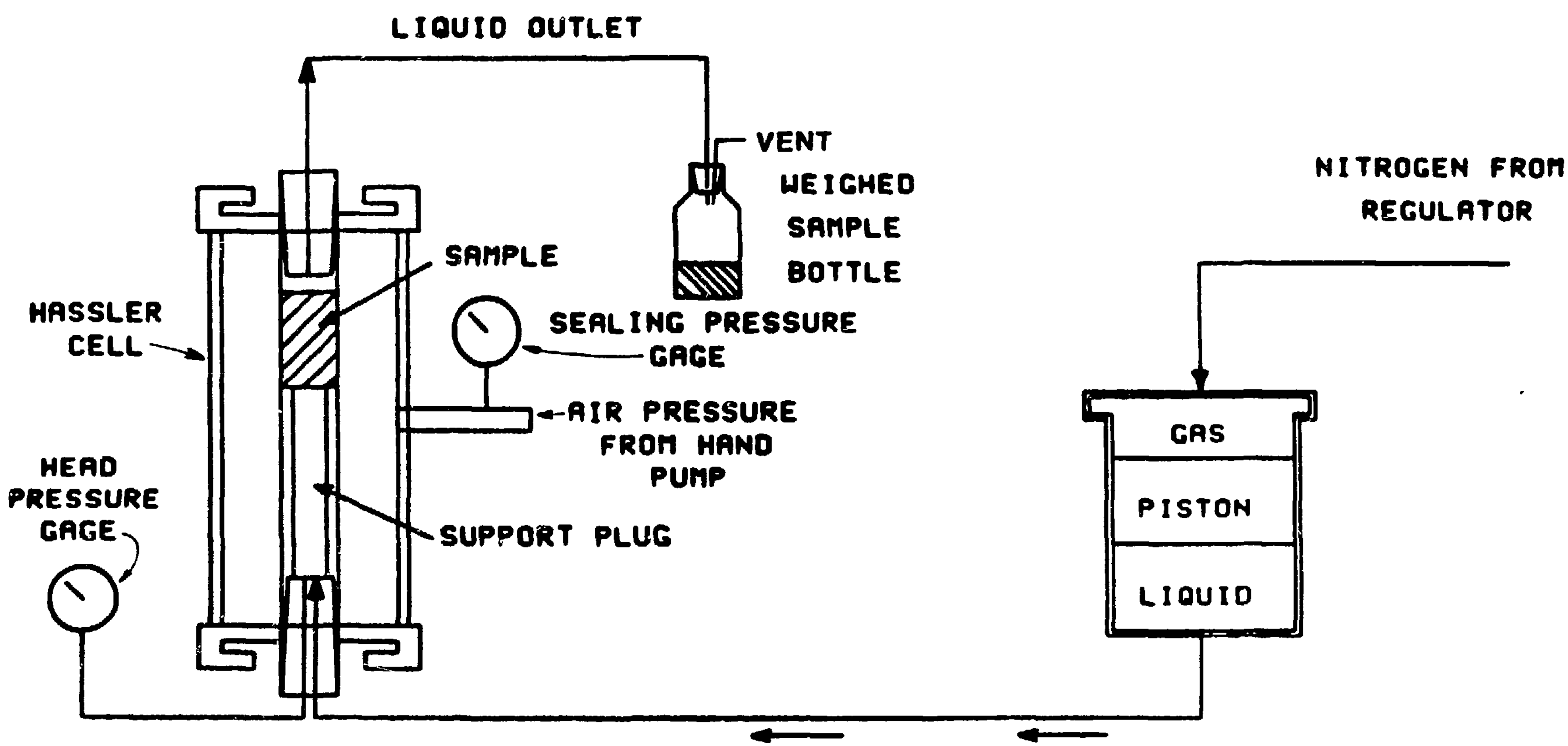


HASSLER CELL APPARATUS FOR MEASUREMENT OF PERMEABILITY

permeability is calculated by means of Darcy's equation, describing viscous flow through porous media:[3]

$$\frac{1}{\alpha} = \frac{\mu \ vL}{\Delta P \ g_c} \quad , \tag{1}$$

where

$1/\alpha$ = permeability coefficient, ft^2;

μ = viscosity of water, lb(mass)/(ft-s);

v = velocity of water, ft/s;

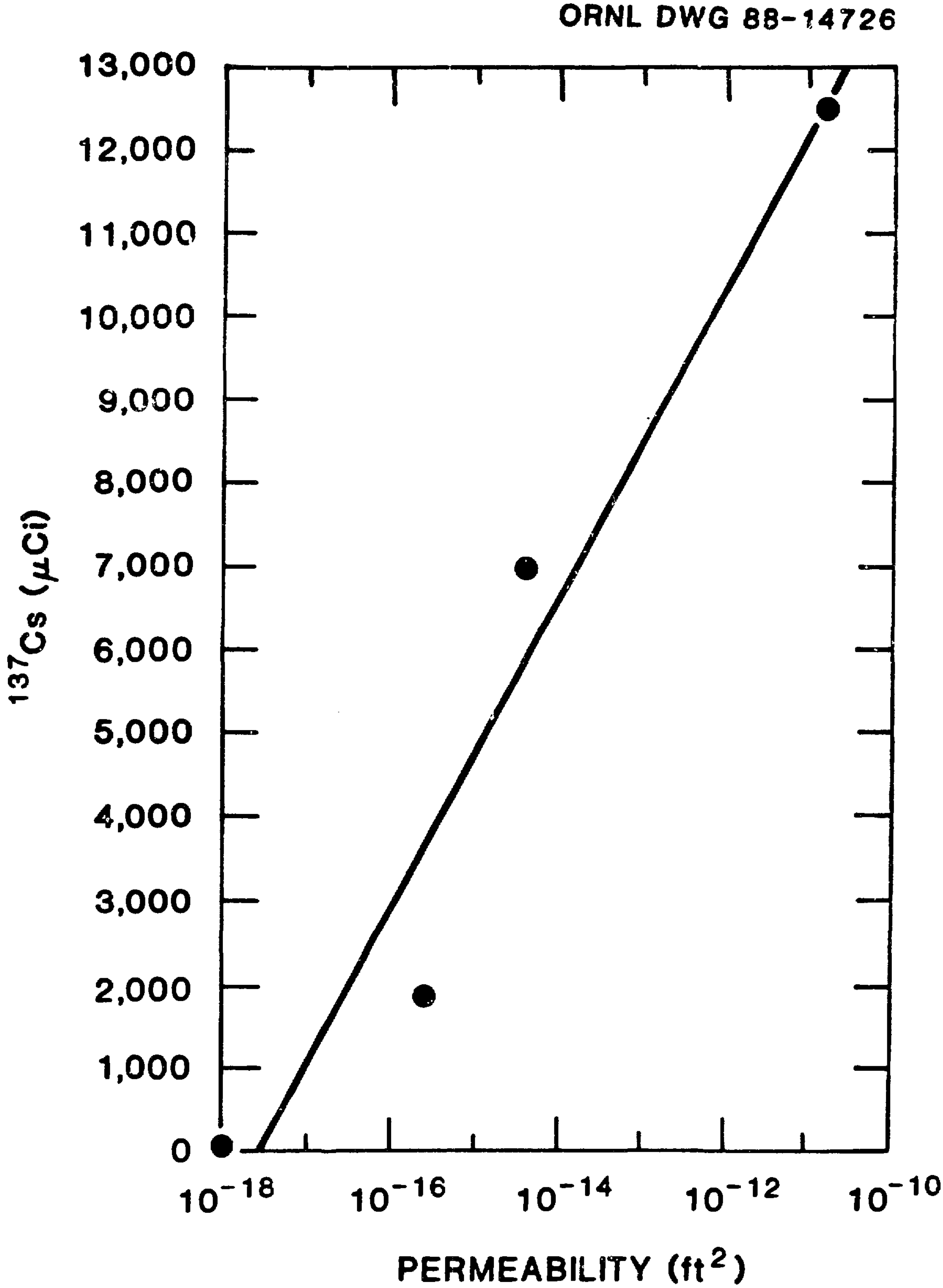
L = sample length, ft;

ΔP = pressure drop, lb(force)/ft^2;

g_c = gravitational coefficient, [lb(mass)-ft]/[lb(force)-s^2].

The calculated permeability coefficients, listed in Table II, show that the unpainted block wall is quite porous. The measured permeability of 10^{-11} ft^2 is similar to that of a bed of filter aid, whereas, for comparison, the permeability of a bed of sand is 10^{-8} ft^2 and a bed of clay is 10^{-13} ft^2. Thus, decontamination of unpainted block by means of flowing water should be quite effective. Since the permeability of the painted concrete block was about four orders of magnitude lower than that for unpainted block, however, the flow-through decontamination would be expected to be much less effective for painted block.

An apparent relationship between the amounts of sorbed ^{137}Cs activity and the permeabilities of the samples was observed (see Fig. 3). These results indicate that the sorption of ^{137}Cs into the concrete structures was directly and linearly proportional to the permeability of the structure (concrete, paint, etc.).



RELATIONSHIP OF ^{137}Cs CONTAMINATION TO CONCRETE PERMEABILITY

Another observation, potentially significant, was observed for samples SCB-5 (painted concrete block wall) and SC5-6 (painted 5000-psi wall), which were subsequently used for the leach tests described below. Prior to the leach tests, the amounts of sorbed ^{137}Cs activity were measured by nondestructive assays (gamma spectrometry) and were compared with the assays previously made at TMI-2, as shown in Table III. The data indicate that much of the activity was removed from the painted concrete block wall sample, apparently by the water flow through the sample in the Hassler Cell apparatus. Also, a significant amount of the activity was removed from the painted 5000-psi wall sample. These results suggest that the activity can be effectively removed by flow-through water. This effect is further demonstrated by the flow-through modeling and calculations described below.

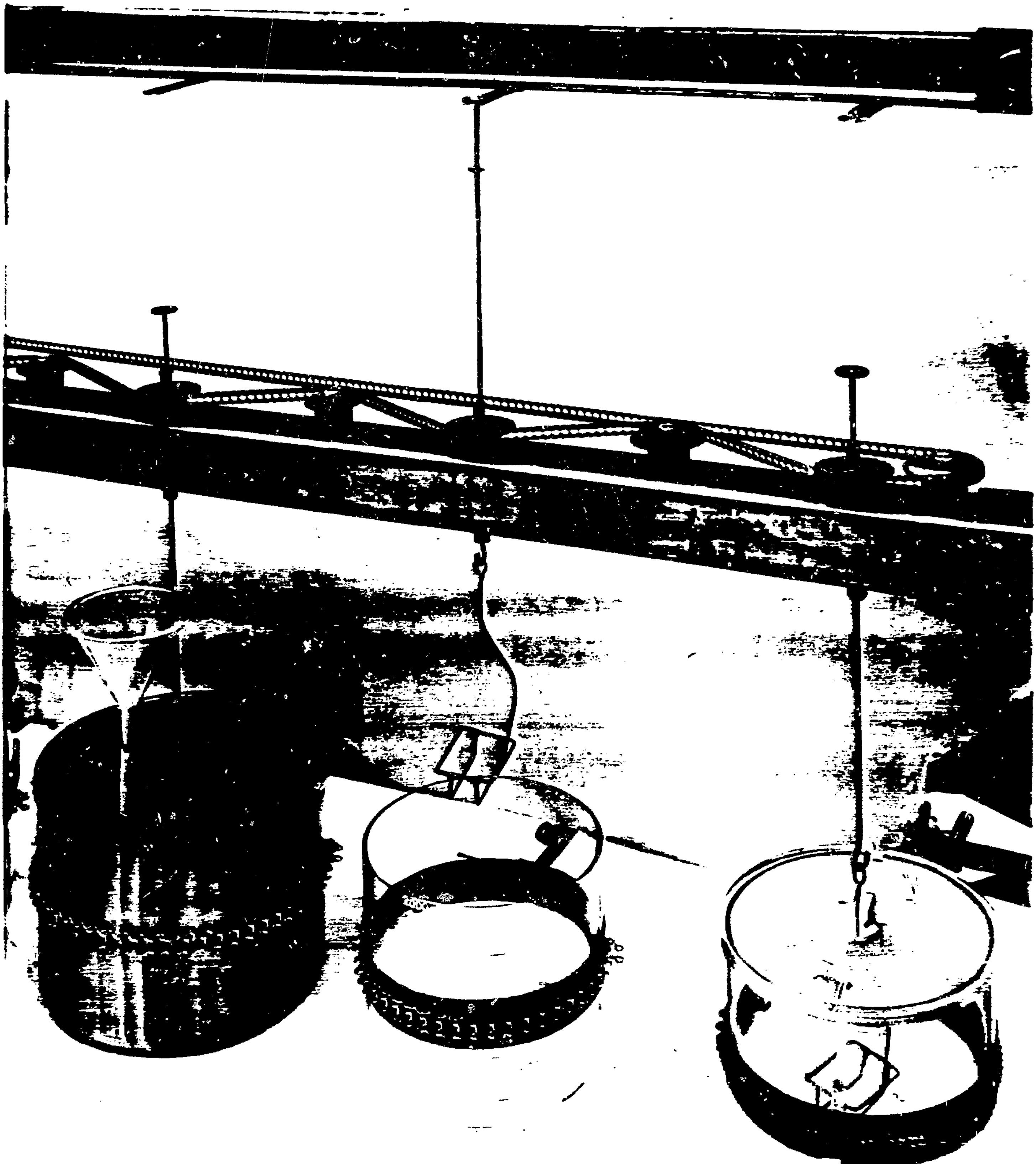
Table III. Comparison of ^{137}Cs Activity in Concrete Samples Before and After Flow-Through Tests in the Hassler Cell

Sample Identification	Type of Concrete	^{137}Cs Activity Before[a] (μCi)	^{137}Cs Activity After[b] (μCi)
SCB-5	Painted block wall	6980	20.6
SC5-6	Painted 5000-psi wall	1830	1450

[a]Measured at TMI-2.
[b]Measured at ORNL.

Leaching Tests

Leaching tests were made at ORNL to (1) corroborate the results obtained at Idaho National Engineering Laboratory, (2) expand the data base to other types of concrete in the TMI-2 reactor building basement, and (3) conduct experiments in a manner that would more closely match conditions of an actual leaching operation at TMI-2. The leachant used in the tests was demineralized water containing ~4350 ppm of boron, in the form of boric acid, buffered with sodium hydroxide to a pH of 7.5 to 7.7. Before the tests began, the exterior surfaces of each concrete sample (which were not exposed directly to the contaminated liquid at TMI-2) were coated with paraffin wax to prevent exposure to the leaching solution. Thus, only the concrete surfaces that are exposed at TMI-2 were exposed to the leach solution. During the tests, each sample was suspended, by means of a wire basket (Fig. 4), in a 4-L beaker containing 135 mL of leach solution per square centimeter of exposed sample surface. This amounted to about 2200 mL for the concrete block wall samples, which had both ends exposed, and to about 1100 mL for the other samples, which had only one end exposed. Each sample was placed in its basket with the long axis in a horizontal position, and the basket was rotated continuously at 0.5 revolution per minute. These conditions were chosen to simulate (1) immersion of the concrete in water in the TMI-2 reactor building basement and (2) a slight flow of water past the concrete surfaces. Also during the test, approximately 4% of the leach solution was replaced each day with fresh solution; this was done to simulate continual removal, decontamination, and return of a portion of the solution. Samples of the leach solutions (those in contact with the concrete samples as well as the cumulative solutions removal) were sampled after leaching times of 1, 2, 4, 8, 16, 32, 64, and ~120 days and were analyzed for ^{137}Cs and ^{90}Sr to determine the rates of leaching. Because the gamma radiations from several of the samples were too high



for direct handling, all of the leach tests were made in a shielded hot cell, using remotely handled tools. The ^{137}Cs activity contained in each sample was measured by nondestructive analysis (NDA; gamma spectrometry) before starting the leach tests. At periodic intervals (after 16, 32, 64, and ~120 days), each sample was removed from the leach solution and the residual ^{137}Cs activity was again measured by NDA. The decreases of activity in the concrete samples were generally consistent with the increases of activity in the leach solutions. The leach tests were concluded after ~120 days, and the amounts of ^{137}Cs activity remaining in the concrete samples were measured by NDA and by destructive radiochemical analyses. The total amounts of ^{137}Cs found (the sum of the amounts in the leach solution and that remaining in the concrete, as measured by both NDA and destructive analysis) are compared with the initial amounts, as measured by NDA at ORNL and at TMI-2, in Table IV.

The NDA results obtained at ORNL for sample SUB-7 were substantially higher (consistently for the initial measurement and those made at intervals during the leach tests) than that measured at TMI-2 by NDA and that measured at ORNL by destructive radiochemical analyses. The reason for this difference is not known. Also, the destructive analysis of the residue from sample SC5-6 gave an extremely lower result than the NDA measurement for ^{137}Cs. The destructive analysis data were assumed to be inaccurate and were not used for the subsequent modeling and predictions. The initial amount of ^{90}Sr in this sample was assumed to be a factor of 35 less than the amount of ^{137}Cs determined by the NDA analysis.

The amounts of ^{90}Sr activity remaining in the concrete samples after conclusion of the leach tests were determined by the destructive

Table IV. Total Amounts of ^{137}Cs Found

Sample Identification	Type of Concrete	Total Leached in ~120 Days (μCi)	Residue (μCi)	Total Found (μCi)	Initial ORNL NDA (μCi)	Initial TMI-2 NDA (μCi)
SUB-3	Unpainted block wall	14,864	49,101[a]	63,965	66,400	65,300
			51,239[b]	66,103		
SUB-7	Unpainted block wall	23,594	54,183[a]	77,777	106,500	62,000
			89,369[b]	112,963		
SCB-5	Painted block wall	4.4	11.6[a]	16.0	20.6	6,980[c]
			18.5[b]	22.9		
SU3-3	Unpainted 3000-psi wall	12,981	16,885[a]	29,866	35,100	30,700
			19,144[b]	32,125		
C-34	Painted 3000-psi wall	427	739[a]	1,166	1,280	1,600
			929[b]	1,356		
C-31	Painted 3000-psi wall	45.2	217[a]	262	295	350
			256[b]	301		
Floor/b	Painted 3000-psi floor	37.0	139[a]	176	--	90
			85.4[b]	122	--	
SC5-6	Painted 5000-psi wall	345	32.9[a]	378	1,450	1,830[c]
			1,112[b]	1,457		

[a]Determined by destructive radiochemical analysis.
[b]Determined by nondestructive analysis (gamma spectrometry).
[c]Analysis made before permeability test in which some ^{137}Cs was washed out of the concrete specimen.

radiochemical analysis method. The total amount of ^{90}Sr in each concrete sample before leaching was calculated from the sum of the amount remaining in the concrete sample and that found in the leach solutions. Table V lists the total amounts of ^{137}Cs and ^{90}Sr found in each sample and the fractions of each of these radionuclides that were leached after ~120 days. The fractions leached after 1, 2, 4, 8, 16, 32, and 64 days were also determined.

Diffusion Models and Predictions of Leach Rates

The results of the leaching tests were analyzed by established mass transport principles. Data for the unpainted samples (SUB-3, SUB-7, and SU3-3) and the painted concrete block sample (SCB-5), which was tested with its unpainted end exposed to the leach solution, were fitted to an equation describing diffusion from the region between parallel planes (viz. a slab).[4] The equation used (with the assumption of a negligible or zero surface concentration) was

$$F_T = \left(\frac{A_{ow} - W}{A_{ow}}\right)\left\{1 - \frac{8}{\pi^2} \sum_{n=o}^{\infty} \exp\left[-(2n + 1)^2 \pi^2 Dt/4\ell^2\right]\right\} + \frac{W}{A_{ow}} \quad . \qquad (2)$$

where

A_{ow} = total amount of material in the specimen and loosely bound to the surface,

D = apparent diffusion coefficient,

F_T = total fraction leached by washoff of the surface and diffusion,

ℓ = length (specimen volume/exposed surface),

t = leaching time,

W = loosely bound surface material.

Table V. Results of Leaching Tests

Sample Identification	Type of Concrete	A_{ow}[a] ^{137}Cs (μCi)	A_{ow}[a] ^{90}Sr (μCi)	F_T[b] ^{137}Cs	F_T[b] ^{90}Sr
SUB-3	Unpainted block wall	63,965[c]	2,688[c]	0.232	0.571
		66,103[d]		0.225	
SUB-7	Unpainted block wall	77,777[c]	4,056[c]	0.303	0.615
		112,963[d]		0.209	
SCB-5	Painted block wall	16.0[c]	1.99[c]	0.275	0.623
		22.9[d]		0.192	
SU3-3	Unpainted 3000-psi wall	29,866[c]	1,314[c]	0.435	0.696
		32,155[d]		0.404	
C-34	Painted 3000-psi wall	1,166[c]	32.1[c]	0.366	0.617
		1,356[d]		0.315	
C-31	Painted 3000-psi wall	262[c]	30.5[c]	0.173	0.305
		301[d]		0.150	
Floor/b	Painted 3000-psi floor	176[c]	12.1[c]	0.210	0.167
		122[d]		0.303	
SC5-6	Painted 5000-psi wall	378[c]	21.5[c]		
		1,457[d]		0.237	0.471[e]

[a]Total amount of material found in the sample and loosely bound to the surface.

[b]Fraction leached by washoff of the surface and diffusion after ~120 days of leaching. Data were also obtained after leaching for 1, 2, 4, 8, 16, 32, and 64 days.

[c]Determined by radiochemical analysis of leachate and destructive analysis of residue.

[d]Determined by radiochemical analysis of leachate and nondestructive analysis of residue.

[e]Based on calculated A_{ow} for ^{90}Sr, assuming it was a factor of 35 less than that for ^{137}Cs.

Data from the samples (C-34, C-31, Floor/b, and SC5-6), which had only a painted surface exposed to the leach solution, were fitted to an equation describing diffusion from a slab with two-stage sorption/desorption on the surface (i.e., in the paint film).[4] The equation used was:

$$F_T = \left(\frac{A_{ow} - W}{A_{ow}}\right) \Bigg\{ 1 - \frac{8}{\pi^2} \sum_{n=o}^{\infty} \exp\left[-(2n + 1)^2 \pi^2 Dt/4\ell^2\right] \tag{3}$$

$$+ \exp(-\beta_2 t)(D/\beta_2 \ell^2)^{\frac{1}{2}} \tan (\beta_2 \ell^2/D)^{\frac{1}{2}}$$

$$- \exp(-\beta_1 t)(D/\beta_1 \ell^2)^{\frac{1}{2}} \tan (\beta_1 \ell^2/D)^{\frac{1}{2}}$$

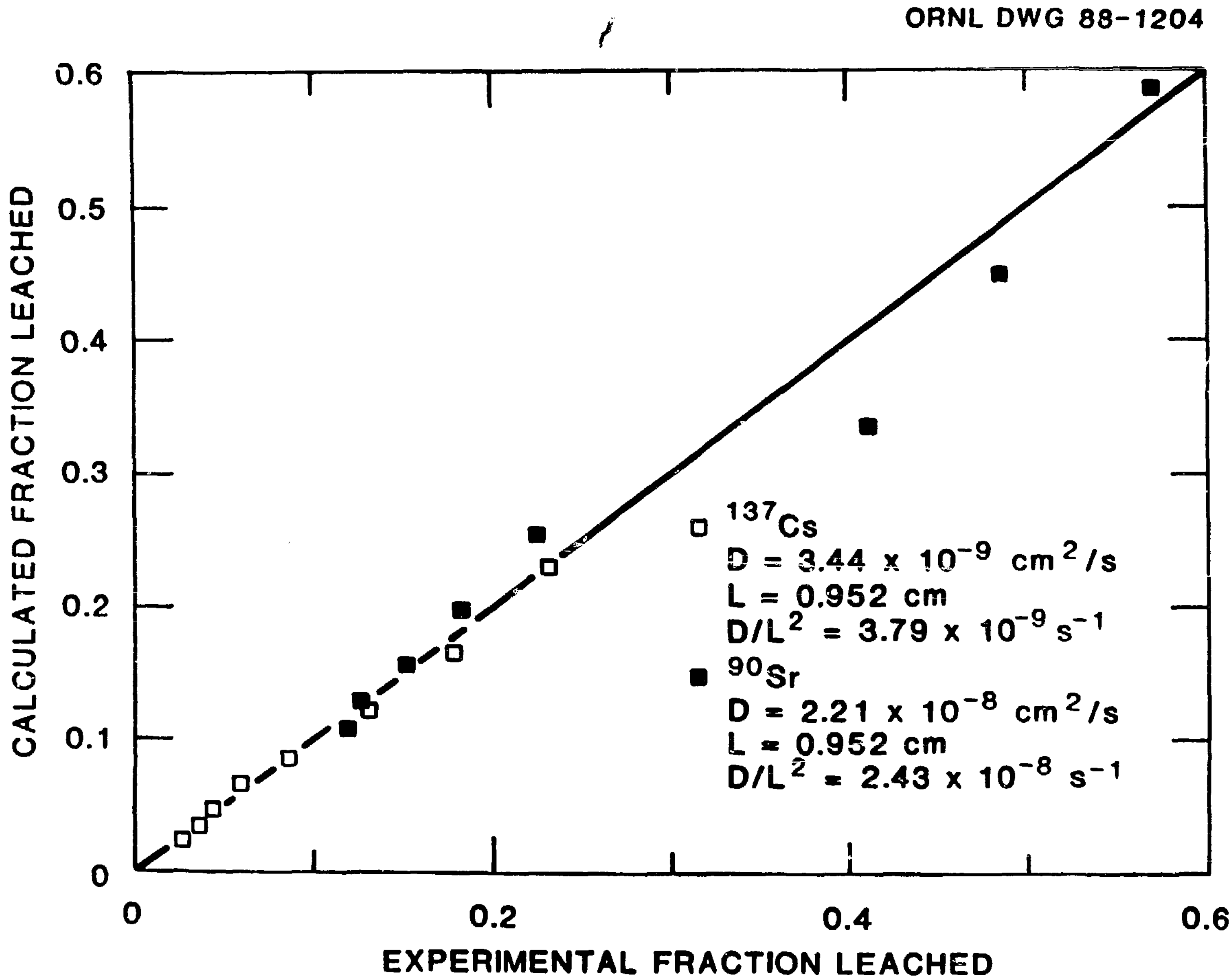
$$+ \frac{8}{\pi^2} \sum_{n=o}^{\infty} \frac{\exp[-(2n+1)^2 \pi^2 Dt/4\ell^2]}{(2n+1)^2[1 - (2n+1)^2 \pi^2 Dt/4\beta_2 \ell^2]}$$

$$- \frac{8}{\pi^2} \sum_{n=o}^{\infty} \frac{\exp[-(2n+1)^2 \pi^2 Dt/4\ell^2]}{(2n+1)^2[1 - (2n+1)^2 \pi^2 Dt/4\beta_1 \ell^2]} \Bigg\} + \frac{W}{A_{ow}} \; .$$

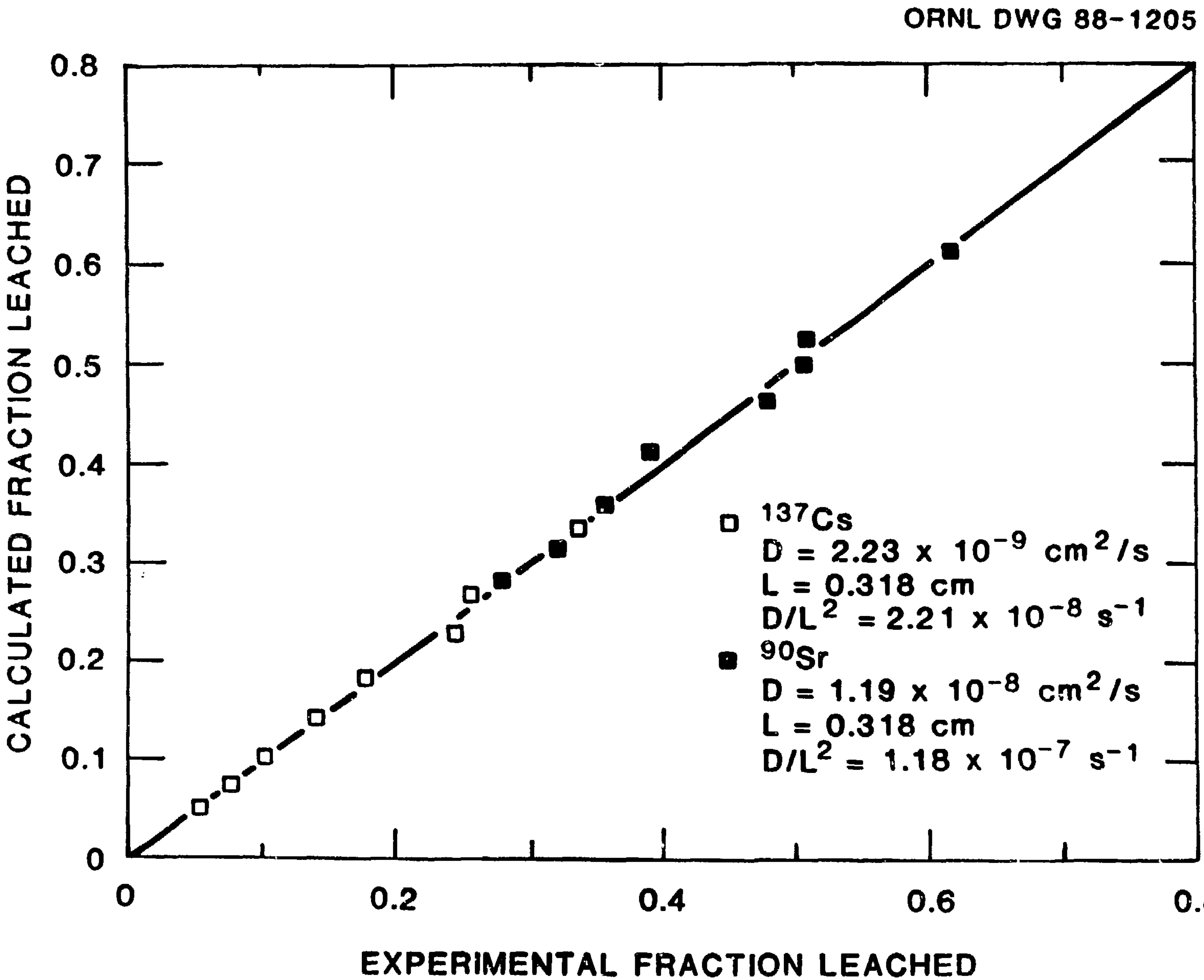
where the symbols A_{ow}, D, F_T, ℓ, t, and W have the same definitions as in equation (2) and β_1, β_2 = sorption/desorption rate constants.

Estimates of values for D and W were obtained by fitting the equations through use of a computer program based on a direct-search method of constrained optimization.[4,5] The values of W (which generally represents nondescript material generated during sampling, storage, transport etc.) were in all instances negligible — usually one or two percent of A_{ow}, but often much less.

Comparisons of the calculated fractions of ^{137}Cs and ^{90}Sr leached as a fraction of time with those measured in the leach tests were in good agreement for all of the samples. As examples, Figs. 5 and 6 show



LEACHING FROM UNPAINTED CONCRETE BLOCK (SUB-3)



LEACHING FROM 3000 PSI PAINTED CONCRETE (C-34)

the agreement for sample SUB-3 (unpainted concrete block) and sample C-34 (painted 3000-psi concrete), respectively. The transport parameters obtained by fitting the data to equations (2) and (3) are presented in Table VI. These values were used with the appropriate equation to predict the percentages of ^{137}Cs and ^{90}Sr that would be leached in extended times (Table VII). These results show that ^{90}Sr is two to three times more leachable from the TMI-2 concrete than is ^{137}Cs, until appreciable amounts (>40 to 50%) have been leached. Further, the results indicate that in 18 months, ~45% of the ^{137}Cs could be expected to be leached from the concrete block, ~75% from the unpainted 3000-psi concrete, and ~60% from the painted 3000-psi and 5000-psi concrete.

Note should be taken of the fact that the concrete samples did not have uniform initial concentrations of ^{137}Cs and ^{90}Sr even though uniform initial concentration was a condition assumed in solving the differential equations leading to equations (2) and (3). As applied, the equations fit the data but yield a diffusion coefficient that is a function of length over which the nuclide of interest is assumed to be distributed. Thus, D/ℓ^2 is a characteristic parameter for these materials and not D alone (as it would be if all initial and boundary conditions were met). For example, if all the ^{137}Cs in Sample SUB-3 were assumed to be in the first 0.159 cm (instead of 0.318 cm, as was assumed here), a fit of the data to equation (2) would give a $D = 2.52 \times 10^{-10}$ cm^2/s (instead of 1.10×10^{-9} cm^2/s as was obtained with 0.318 cm). However, the D/ℓ^2 would be the same in each instance; that is, 1.00×10^{-8} s^{-1} [viz., $(1.01 \times 10^{-9})/(0.318)^2 = (2.52 \times 10^{-10})/(0.159)^2$].

Table VI. Transport Parameters for the Leaching of Cesium and Strontium from Core Samples from the TMI-2 Containment Building Basement

Sample Identification	Type of Concrete	Cesium			Strontium		
		Diffusion Coefficient, D (cm^2/s)	Diffusion Length, ℓ (cm)	Diffusion Parameter, D/ℓ^2 (s^{-1})	Diffusion Coefficient, D (cm^2/s)	Diffusion Length, ℓ (cm)	Diffusion Parameter, D/ℓ^2 (s^{-1})
SUB-3	Block (unpainted)	3.44×10^{-9}	0.952	3.79×10^{-9}	2.21×10^{-8}	0.952	2.43×10^{-8}
SUB-7	Block (unpainted)	7.23×10^{-9}	1.51	3.18×10^{-9}	7.68×10^{-8}	1.51	3.38×10^{-8}
SCB-5[a]	Block (painted)	6.43×10^{-10}	0.476	2.84×10^{-9}	6.81×10^{-9}	0.476	3.00×10^{-8}
SU3-3	3000-psi (unpainted)	1.01×10^{-9}	0.318	1.00×10^{-8}	4.39×10^{-9}	0.318	4.36×10^{-8}
C-34[b]	3000-psi (painted)	2.23×10^{-9}	0.318	2.21×10^{-8}	1.19×10^{-8}	0.318	1.18×10^{-7}
C-31[c]	3000-psi (painted)	1.01×10^{-9}	0.318	1.00×10^{-8}	3.44×10^{-9}	0.318	3.41×10^{-8}
Floor/b[d]	3000-psi (painted)	1.05×10^{-9}	0.318	1.04×10^{-8}	3.18×10^{-9}	0.318	3.15×10^{-8}
SCS-6[e,f]	5000-psi (painted)	5.92×10^{-10}	0.318	5.87×10^{-9}	2.43×10^{-9}	0.318	2.41×10^{-8}

[a]Specimen was used in permeability test before being used in leaching test.

[b]For cesium, $\beta_1 = 2.78 \times 10^{-8}\ s^{-1}$ and $\beta_2 = 1.37 \times 10^{-7}\ s^{-1}$. For strontium, $\beta_1 = 8.43 \times 10^{-8}\ s^{-1}$ and $\beta_2 = 4.66 \times 10^{-7}\ s^{-1}$.

[c]For cesium, $\beta_1 = 7.83 \times 10^{-6}\ s^{-1}$ and $\beta_2 = 2.44 \times 10^{-5}\ s^{-1}$. For strontium, $\beta_2 = 2.83 \times 10^{-4}\ s^{-1}$ and $\beta_2 = 4.26 \times 10^{-2}\ s^{-1}$.

[d]For cesium, $\beta_1 = 5.55 \times 10^{-7}\ s^{-1}$ and $\beta_2 = 1.40 \times 10^{-1}\ s^{-1}$. For strontium, $\beta_1 = 2.74 \times 10^{-6}\ s^{-1}$ and $\beta_2 = 2.41 \times 10^{-5}\ s^{-1}$.

[e]Specimen was used in permeability test before being used in leaching test.

[f]For cesium, $\beta_1 = 2.70 \times 10^{-6}\ s^{-1}$ and $\beta_2 = 1.52 \times 10^{0}\ s^{-1}$. For strontium, $\beta_1 = 1.03 \times 10^{-5}\ s^{-1}$ and $\beta_2 = 7.88 \times 10^{-5}\ s^{-1}$.

Table VII. Predicted Percentage of ^{137}Cs and ^{90}Sr Leached from Concrete Samples as a Function of Time Without Forced Flow[a]

Time (months)	Unpainted Concrete Block[b]		Painted Concrete Block[c]		3000-psi Unpainted Concrete[d,e]		3000-psi Painted Concrete[d,f]		5000-psi Painted Concrete[d,g]	
	Cs	Sr	Cs	Sr	Cs	Sr	Cs	Sr	Cs	Sr
6	28	70	26	77	46	87	36	73	35	68
12	39	88	35	93	63	98	48	92	49	88
18	48	96	43	98	75	99	61	98	60	95
24	55	98	49	99	83	100	73	99	68	98
36	67	100	59	100	92		88	100	80	100
48	75		67		97		95		87	
60	82		74		98		98		92	
120	96		91		100		100		99	

[a]Samples identified as SCB-3, SUB-5, SU3-3, C-34, and SCS-6 in Table 1. Predictions were made with apparent diffusion coefficients obtained by fitting the leaching data summarized in Table 1 to established mass transport equations [e.g., eq. (1) and Ref. 3].

[b]The value of D for Cs = 3.4×10^{-9} cm^2/s and for Sr = 2.2×10^{-8} cm^2/s.

[c]The value of D for Cs = 6.4×10^{-10} cm^2/s and for Sr = 6.8×10^{-9} cm^2/s.

[d]Poured concrete having the indicated compressive strength.

[e]The value of D for Cs = 1.0×10^{-9} cm^2/s and for Sr = 4.4×10^{-9} cm^2/s.

[f]The value of D for Cs = 2.2×10^{-9} cm^2/s and for Sr = 1.2×10^{-8} cm^2/s.

[g]The value of D for Cs = 5.9×10^{-10} cm^2/s and for Sr = 2.4×10^{-9} cm^2/s.

Flow-Through Modeling and Predictions

Concrete block walls, such as those surrounding the elevator and closed stairwell in the TMI-2 reactor building basement, may be amenable to enhanced decontamination rates by using a forced flow of leachant through the concrete. An indication of this possibility was the removal of activity from sample SCB-5 during the permeability tests described previously. Mathematical equations describing forced-flow through porous media can be written but involve partial differential equations that cannot be solved analytically. However, an analogy of flow through a concrete block wall can be made to flow through a packed sorbent bed, as illustrated in Fig. 7. Then, a numerical algorithm, which has been developed[6] to describe convection/adsorption phenomena in packed-bed systems, can be utilized to model the removal of ^{137}Cs and ^{90}Sr from the concrete block walls that exist in the TMI-2 reactor building basement.

The numerical algorithm was derived to simultaneously solve equations describing the liquid phase and the adsorbed-phase mass balances, including (1) an overall mass transfer coefficient that considers parallel resistances from liquid mass transfer and pore diffusion and (2) adsorption equilibria related by the distribution coefficient. A method called orthogonal collocation was used to convert the system of partial differential equations into a system of algebraic expressions and ordinary differential equations, which were then solved by a computer program utilizing numerical integration techniques.

Assumptions were made that (1) the center of the block was filled with leachant, (2) the block behaved as a packed bed consisting of

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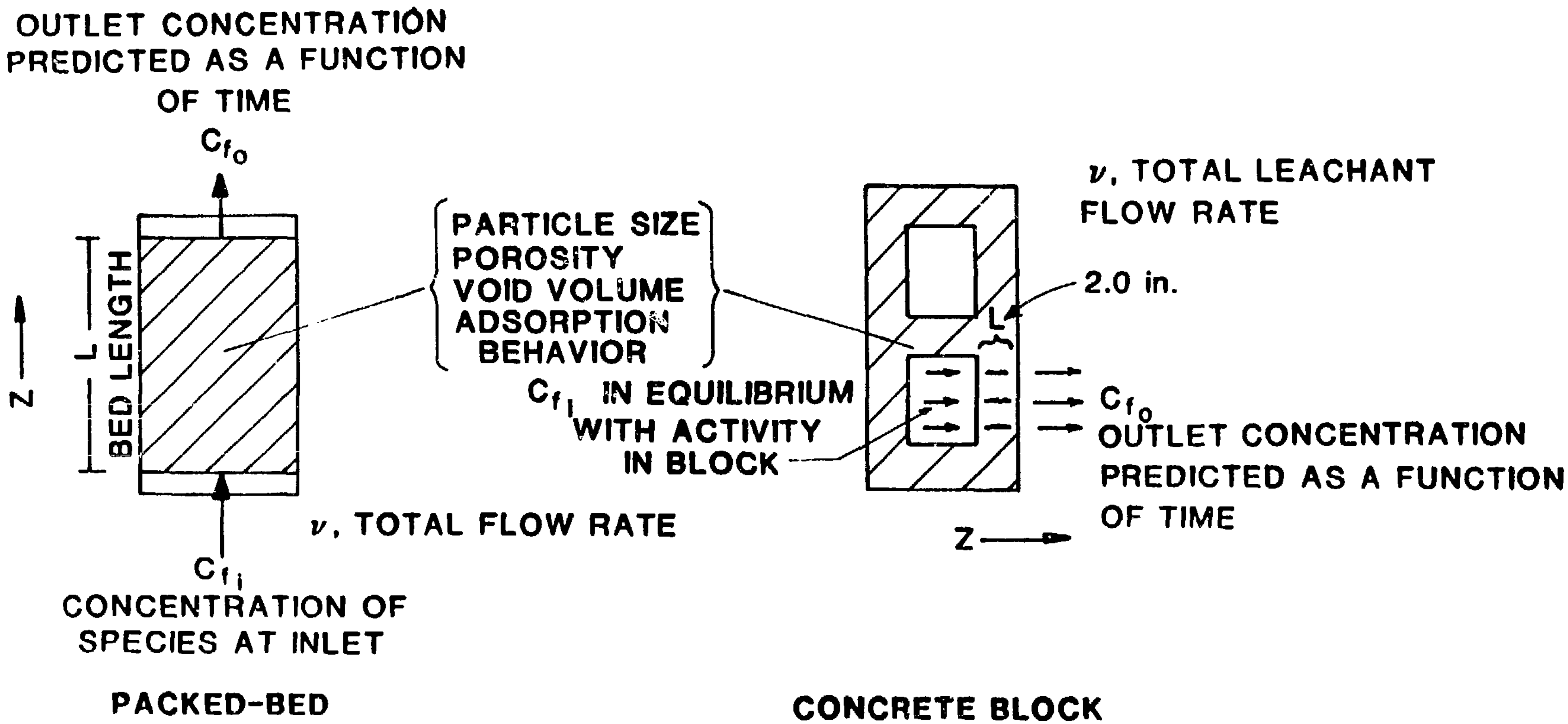

ANALOGY OF MODELS FOR PACKED-BED AND CONCRETE BLOCK SYSTEMS

1-mm-diam spherical particles, and (3) the leachant flow was induced by pressure on the central cavity and traversed the "bed" (block wall) in a "plug flow" fashion. These assumptions were reasonable but were not conservative. They were needed to use the model as it exists to obtain a relative comparison of the benefits of decontamination by means of flow through leaching versus that obtained by diffusion only. The initial use of the model did not consider effects of nonuniform flow paths which could be created by a variety of causes.

Parameters needed for solution of the model include the void-free density ($\rho = 2.5$ g/cm^3) and porosity ($\varepsilon = 0.35$) of the concrete block, which were determined by the measurements described previously, and the distribution coefficients, K([μCi/mL of solid]/[μCi/mL of liquid]) that were estimated using the diffusion coefficients (D) determined from the leach test data (Table 6) and the simplified relationship[7]

$$K = \left(\frac{\varepsilon}{1-\varepsilon}\right) \left(\frac{D_s}{Dg} - 1\right) . \qquad (4)$$

Values for the dimensionless geometry factor, g (20 for cesium and 8.9 for strontium), and for the diffusion coefficient at infinite dilution, D_s (2.1×10^{-5} cm^2/s for cesium and 7.9×10^{-6} cm^2/s for strontium), were taken from available literature.[7-9] Estimates of K are given in Table VIII.

The flow-through model was applied to predict the time required to remove ^{137}Cs and ^{90}Sr from a 40-ft^2 area of concrete block wall, using the flow rate (20 and 100 gal/min), initial concentrations of ^{137}Cs

Table VIII. Distribution Coefficients for Cesium and Strontium in Concrete Block Samples Taken from the TMI-2 Reactor Building Basement

		Cesium		Strontium	
		Diffusion Coefficient[a]	Distribution Coefficient[b]	Diffusion Coefficient[a]	Distribution Coefficient[c]
Sample Identification	Block Type	$D(cm^2/s)$	$K\ \frac{\mu Ci/cm^3\ solid}{\mu Ci/cm^3\ liquid}$	$D(cm^2/s)$	$K\ \frac{\mu Ci/cm^3\ solid}{\mu Ci/cm^3\ liquid}$
SUB-3	Unpainted	3.44×10^{-9}	164	2.21×10^{-8}	21
SUB-7	Unpainted	7.23×10^{-9}	78	7.68×10^{-8}	5.7
SCB-5	Painted	6.43×10^{-10}	879	6.81×10^{-9}	70

[a]Given in Table 6.
[b]Calculated by use of equation (4) with $D_s = 2.1 \times 10^{-5}$ cm^2/s, $g = 20$, and $\varepsilon = 0.35$.
[c]Calculated by use of equation (4) with $D_s = 7.9 \times 10^{-6}$ cm^2/s, $g = 8.9$, and $\varepsilon = 0.35$.

(1500 and 7000 µCi/mL) and ^{90}Sr (60 and 280 µCi/mL) in the concrete, and distribution coefficients (75, 225, and 1000 for ^{137}Cs and 15, 60, and 150 for ^{90}Sr) as variable parameters. The results obtained when using a flow rate of 20 gal/min, initial concentrations of 7000 and 280 µCi/mL for ^{137}Cs and ^{90}Sr, and distribution coefficients of 1000 for ^{137}Cs and 150 for ^{90}Sr are shown in Fig. 8. These results indicate that an elapsed time of 240 h, or 10 days, was required to remove the ^{137}Cs. All other conditions resulted in decreased time requirements. Thus, forced flow-through decontamination appears to require significantly less time (a few days instead of a few years) than decontamination, which depends only on diffusion.

Summary

Ten cylindrical core samples of contaminated concrete were taken from various types of structures in the TMI-2 reactor building basement and were sent to Oak Ridge National Laboratory for analysis. Tests were performed to determine the potential for decontamination of the samples by means of diffusion-controlled leaching under conditions of full submergence and by means of forced flow-through leaching of porous concrete block walls. Pertinent physical properties of the concrete were measured, and the results showed that the amount of sorbed ^{137}Cs activity was indirectly and linearly proportional to the permeability of the structure (concrete, paint, etc.). These measurements also showed that painted concrete block was less permeable than unpainted concrete block by a factor of $\sim 10^4$. The permeability measurements, in which water was forced through the concrete samples, indicated that significant amounts of the ^{137}Cs activity could be removed by flow-through leaching.

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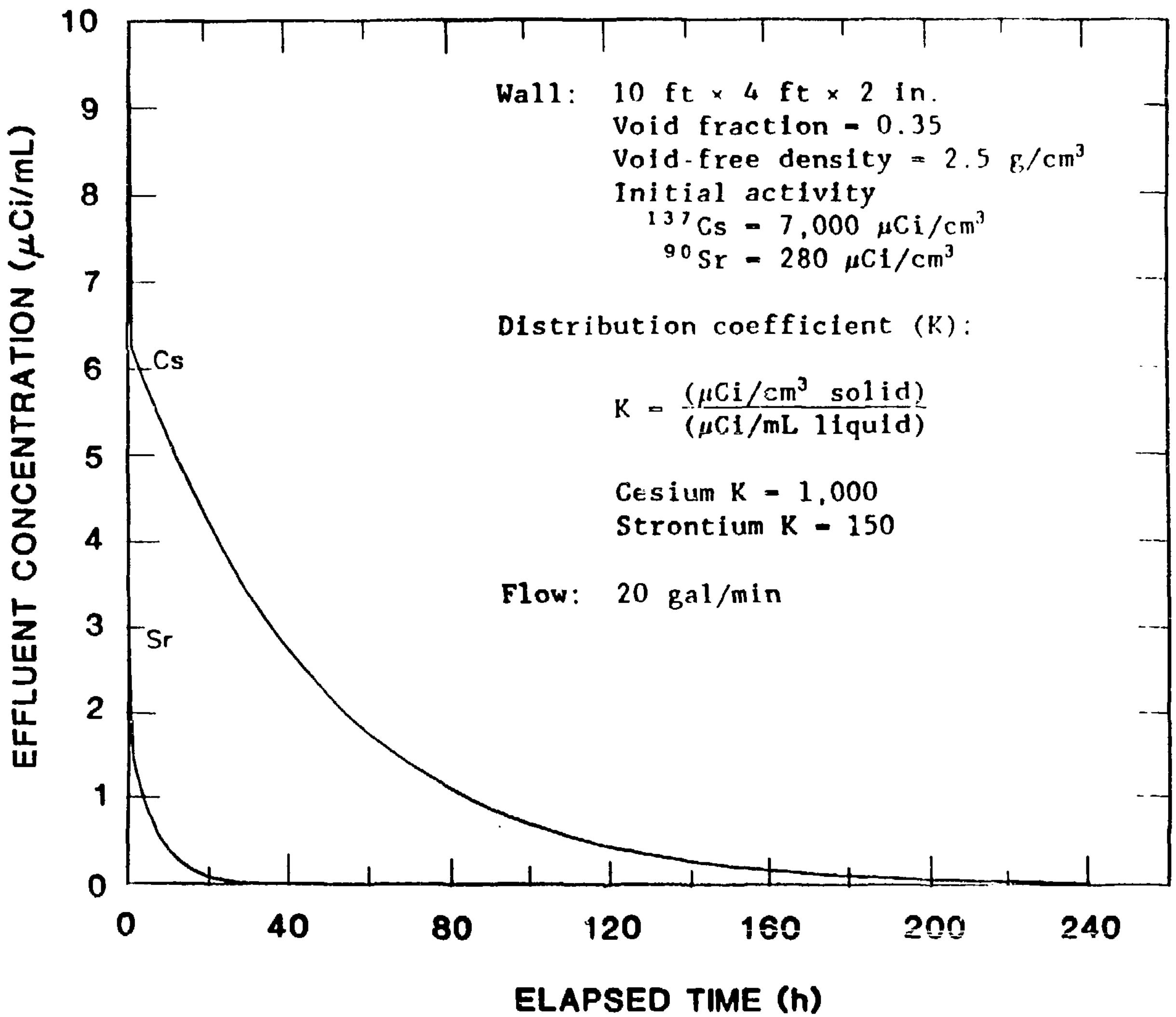

PREDICTED ELUTION OF CESIUM AND STRONTIUM FROM A CONCRETE BLOCK WALL

Leaching tests were made on eight samples that represented various types of concrete structures located in the TMI-2 reactor building basement. The results from these tests were analyzed by established mass transport principles. The data from leaching unpainted samples were fitted to an equation describing diffusion from the region between parallel planes (slab), and the data from leaching through painted surfaces were fitted to an equation describing diffusion from a slab with two-stage sorption/desorption on the surface (i.e., in the paint film). The fitted equations were then used to predict leaching rates at extended times, and the values obtained indicate that ^{90}Sr is two to three times more leachable than ^{137}Cs. Further, although the results indicate that total leaching would be possible, a period of several years would be required. For example, after 18 months, ~45% of the ^{137}Cs could be leached from the concrete block, ~75% from the unpainted 3000-psi concrete, and ~60% from the painted 3000 and 5000-psi concrete.

A numerical algorithm that has been developed to describe convection/adsorption phenomena in packed-bed systems was used to model the removal of ^{137}Cs and ^{90}Sr by forced flow-through leaching from porous concrete block walls. The results of calculations made with this model indicated that forced flow-through decontamination would require significantly less time (a few days instead of a few years) than decontamination, which depends only on diffusion.

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