LA-UR 81-3140

WM RECORD COPY

NTS #831

TITLE: RADIONUCLIDE MIGRATION: LABORATORY EXPERIMENTS WITH ISOLATED FRACTURES

AUTHOR(S): R. S. Rundberg, J. L. Thompson, and S. Maestas
Los Alamos National Laboratory, Los Alamos, New Mexico

SUBMITTED TO: International Symposium on the Scientific Basis for Nuclear Waste Management, Materials Research Society, Boston, Massachusetts, November 16-19, 1981

Scientific Basis for Nuclear Waste Management, Vol. 4 (in press)

This is a preprint of a paper intended for publication in a journal or proceedings. Because changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos Scientific Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

University of California

HYDROLOGY DOCUMENT NUMBER 238

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545
An Affirmative Action/Equal Opportunity Employer

Form No. 836 R3
St. No. 2629
12/78

UNITED STATES
DEPARTMENT OF ENERGY
CONTRACT W-7405-ENG. 36

RADIONUCLIDE MIGRATION: LABORATORY EXPERIMENTS WITH ISOLATED FRACTURES*

R. S. RUNDBERG, J. L. THOMPSON, AND S. MAESTAS
Los Alamos National Laboratory, MS-514, Los Alamos, New Mexico, USA

ABSTRACT

Laboratory experiments examining flow and element migration in rocks containing isolated fractures have been initiated at the Los Alamos National Laboratory. Techniques are being developed to establish simple fracture flow systems which are appropriate to models using analytical solutions to the matrix diffusion - flow equations, such as those of I. Neretnieks [1]. These experiments are intended to be intermediate steps toward larger scale field experiments where it may become more difficult to establish and control the parameters important to nuclide migration in fractured media.

Laboratory experiments have been run on fractures ranging in size from 1 to 20 cm in length. The hydraulic flow in these fractures was studied to provide the effective apertures. The flows established in these fracture systems are similar to those in the granite fracture flow experiments of Witherspoon et al. [2]. Traced solutions containing ^{85}Sr and ^{137}Cs were flowed through fractures in Climax Stock granite and welded tuff (Bullfrog and Tram members, Yucca Mountain, Nevada Test Site). The results of the elutions through granite agree with the matrix diffusion calculations based on independent measurements of K_d. The results of the elutions through tuff, however, agree only if the K_d values used in the calculations are lower than the K_d values measured using a batch technique. This trend has been previously observed in chromatographic column experiments with tuff.

INTRODUCTION

The study of fracture flow and element transport through flow in fractures is essential to provide a complete understanding of the geologic barriers surrounding a nuclear waste repository. The ability to correctly model radionuclide transport in fractured systems is necessary to the performance assessment of a repository. Fracture flow is an important transport mechanism because fluid velocities in fractures can be many orders of magnitude greater than the fluid velocity in the porous matrix. This is especially true of crystalline rock such as granite.

There are many chemical and physical processes which can affect the transport of radionuclides by flow in fractures. The principal processes studied in the experiments to be discussed are sorption and diffusion into the rock matrix. These experiments are part of a program to examine the mechanisms contributing to the retardation of radionuclides in flow through fractures. The objectives of the program are to test existing theoretical models and to

---

*This work was supported by the U.S. Department of Energy.

perform laboratory experiments with rock samples of varying size to see if the results of small scale laboratory experiments can be extrapolated to the field.

The elution of ^{85}Sr and ^{137}Cs was observed in flow through fractured Climax Stock (CS) granite and through two tuff samples taken from cores from the USW G1 drillhole at the Nevada Test Site. The tuff samples, G1-2335 and G1-2840, are welded tuffs from the Bullfrog and Tram members, respectively.

The breakthrough curves were compared with the curves predicted by the analytic solution to flow through a one-dimensional fracture coupled to diffusion into the matrix [1]. The fracture volume and the fracture aperture, in particular, were determined using Darcy's law [2]. The experiments on Climax Stock Granite were performed on small cores under confining pressure to simulate depth and to close the fracture. The tuff samples were not placed under confining pressure because the samples were found to seal under moderate pressure (~1000 PSI), whereupon the fracture permeability was reduced to the same magnitude as the matrix permeability.

The flow through the fracture was straight flow enabling direct comparison with the one-dimensional calculations.

Fluid Flow Through a Single Fracture

The flow of fluid through a fracture can be described by the Darcy equation or cubic law

$$\frac{Q}{\Delta h} = C(2b)^3 \quad , \qquad (1)$$

where

Q = the flow rate in m^3/s,
Δh = the hydraulic head in meters of water,
$2b$ = the aperture, and
C = a constant for a given geometry.

For straight flow

$$C = \frac{W}{L}\frac{\rho g}{12\mu} \quad , \qquad (2)$$

where

L = the length of the fracture,
W = the width of the fracture,
ρ = the density of the fluid,
g = the gravitational constant, and
μ = the viscosity of the fluid.

The validity of Eq. (1) has been demonstrated by Witherspoon et al. [2]. With some fractures a correction factor is required to correct for the effect of surface roughness on the flow. A correction factor f was defined by Witherspoon et al. and inserted into the Darcy equation as follows.

$$\frac{Q}{\Delta h} = \frac{C}{f}(2b)^3 \quad . \qquad (3)$$

Values of f varied from 1.04 to 1.21 in a granite fracture with straight flow. Assuming f = 1.00 rather than 1.21, however, would result in only a 7% error in the aperture.

Two small granite cores, 2.54 cm diam by 1.59 cm long, were used in the experiments. Core 1 had a natural fracture, which appeared to be filled. The fracture was mechanically opened prior to use in the experiment. Core 2 contained no natural fractures but was stressed to induce a fracture. The cores were stressed while in a Teflon sleeve in which the core remained throughout the experiment. The cores were then placed in a modified permeability apparatus similar to that of Brace [4]. The apertures were determined by measuring the flow rate vs the hydraulic head. Table I summarizes the results.

The aperture of Core 1, the one with a natural fracture, was in good agreement with the measurements of Witherspoon et al. [2,3] and Isherwood (D. Isherwood, Lawrence Livermore National Laboratory, personal communication, June 1981). The stress-induced fracture, however, had an unusually large residual aperture. This may be a result of granite grains being lodged in the fracture, preventing proper mating of the rock surfaces. Alternatively, there may be other non-parallel fractures through the core, although these were not apparent prior to the experiment.

Radionuclide Transport by Flow Through A Single Fracture

The transport of radionuclides by flow through a single fracture has been solved analytically for a one-dimensional fracture with matrix diffusion by I. Neretnieks and is described in detail in Reference 1. This model does not include velocity dispersion but should serve well as a first approximation to the experiment and as a benchmark for numerical code development. The effect of matrix porosity on the transport of radionuclides was clearly demonstrated by Neretnieks.

TABLE I
Apertures of Granite Fractures Under Stress

	Pressure (MPa)	$Q/\Delta h$ (m^2/s)	2b (μm)
CS granite core 1	24.8	4.94×10^{-9}	15.6
CS granite core 2	35.9	2.15×10^{-8}	25.4
	27.6	2.01×10^{-8}	24.8
	13.8	2.47×10^{-8}	26.6
Isherwood[a]	21	1.84×10^{-9}	13.6
	16	3.56×10^{-9}	17.0
	10	1.23×10^{-9}	25.6
Witherspoon[b], Run #1	17.0	4.08×10^{-10}	7.7
	12.5	8.3×10^{-10}	9.7
	8.0	1.14×10^{-9}	10.8

---

[a] D. Isherwood, Lawrence Livermore National Laboratory, personal communication, June 1981.

[b] Ref. 2.

Before proceeding to the analysis, some useful definitions are given.

1. The volumetric sorption ratio, $K_d\rho$, is given by

$$K_d\rho = \varepsilon_p + (1 - \varepsilon_p)K_d'\rho_s \quad , \tag{4}$$

where

K_d' = the distribution coefficient ($K_d' = 0$ for a nonsorbing material),

ε_p = the matrix porosity, and

ρ_s = the density of the solid.

2. The effective diffusion coefficient

$$D_{eff} = \varepsilon_p \frac{\alpha}{\tau^2} D^i \quad , \tag{5}$$

where

D^i = the ionic diffusion coefficient,
α = the constrictivity of the pores, and
τ = the tortuosity of the pores.

3. The apparent diffusion coefficient

$$D_{app} = \frac{D_{eff}}{K_d\rho} \quad . \tag{6}$$

Transport of radionuclides through a single fracture can be described mathematically by the following expressions.

1. Diffusion in the rock matrix is given by

$$\frac{\partial C_p}{\partial t} = D_{app} \frac{\partial^2 C_p}{\partial z^2} - \lambda C_p \quad . \tag{7}$$

2. The sorption and convection are given by

$$\frac{\partial C_f}{\partial t} + U_f \frac{\partial C_f}{\partial x} = \frac{D_{eff}}{b} \frac{\partial C_p}{\partial z}\bigg|_{z=0} - \lambda C_f \quad , \tag{8}$$

where

C = concentration in water in pores,
C_f = concentration in water in fissures,
x = distance along the fracture,
z = distance into the matrix from the fracture surface,
b = half-width of the fracture,
U_f = water velocity, and
λ = decay constant.

The solution to these equations with the appropriate boundary and initial conditions for a concentration step of duration Δt is

$$\frac{C_p}{C_o} = e^{-\lambda t}\left(\operatorname{erfc}\left\{\frac{G}{[t - (t_w + t_o)]^{1/2}}\right\} - \operatorname{erfc}\left\{\frac{G}{[t - (t_w + t_o + \Delta t)]^{1/2}}\right\}\right), \qquad (9)$$

where

$$G = \left\{\left[D_{eff} + \frac{1}{2}\,\frac{U_f(2b)z}{x}\right] \Big/ 2b(D_{app})^{1/2}\right\} t_w ,$$

t_w = the time required for the water to reach x, and
t_o = the initial time.

Results

The breakthrough curves were calculated using the matrix diffusion model. Tables II and III list some of the values used as input to the model. Most of these values were based on previous measurements [5,6] with crushed rock and solid rock cores. The breakthrough curves for granite Core 1 were calculated using the parameters in Table I, which are based on our earlier data [11]. The porosity of the matrix in the core used was not actually determined, and $\varepsilon = 0.005$ was chosen as a nominal value. The range of values of porosities determined for Climax Stock granite varies from 0.0015 to 0.008. Another parameter that has not been determined for the particular granite core used is the constrictivity-tortuosity term α/τ^2, which was somewhat arbitrarily taken as 0.1. The breakthrough curves were calculated for two porosities, 0.001 and 0.005, which served to illustrate the dramatic dependence on porosity. The breakthrough curves along with results of the experiment are shown in Figs. 1 and 2. Fig. 3 shows the calculated curve for a 20 ml pulse of activity along with the experimental results.

The elution curves for tuff core G1-2335 were calculated using the parameters in Table III, which were based on measurements from the same core. The constrictivity-tortuosity term α/τ^2 was again taken to be 0.1. This assumption is supported by a measurement on a sample from G1-2290, which gave a value of 0.098 for α/τ^2.

The elution curve shown in Fig. 4 was calculated using the K_d from batch measurements and an elution curve with the K_d adjusted to fit the experimental breakthrough is shown in Fig. 5.

Discussion

The results of the fracture flow experiments with Climax stock granite are in good agreement with the calculations using the analytic model previously described. These first experiments show that for some simple cations the transport of radionuclides by fracture flow can be predicted with reasonable accuracy by a matrix diffusion model. The hydraulic measurements on the stressed granite fractures gave results similar to the measurements by Witherspoon et al. The tuff samples were not run under stress because it was found that the fracture closed, under moderate pressure, to a point where the permeability of the fracture was approximately equal to that of the matrix.

This does not mean that open fractures do not exist in the field because faulting and slipping could still open up fractures by exerting a stress field that is not simulated in these experiments.

TABLE II
Parameter Values Used to Calculate Breakthrough Curves for Climax Stock Granite

Parameter	^{85}Sr	^{137}Cs
D^i	7.75×10^{-6} cm^2/s	2.02×10^{-5} cm^2/s
K_d	8 ml/g	400 ml/g
ρ_s	3.01 g/cm^3	3.01 g/cm^3
α/τ^2	0.1	0.1
U_f	1.41×10^{-2} cm/s	1.41×10^{-2} cm/s
2b	15.6 μm	15.6 μm
x	1.59 cm	1.59 cm

TABLE III
Parameter Values Used to Calculate ^{85}Sr Elution Curves for G1-2335 Tuff

Parameter	Value
D^i	7.75×10^{-6} cm^2/s
K_d	148 ml/g and 30 ml/g
ρ_s	1.71
α/τ^2	0.1
U_f	2.85×10^{-2} cm/s
2b	30.7 μm
x	4.76 cm
ε	0.312

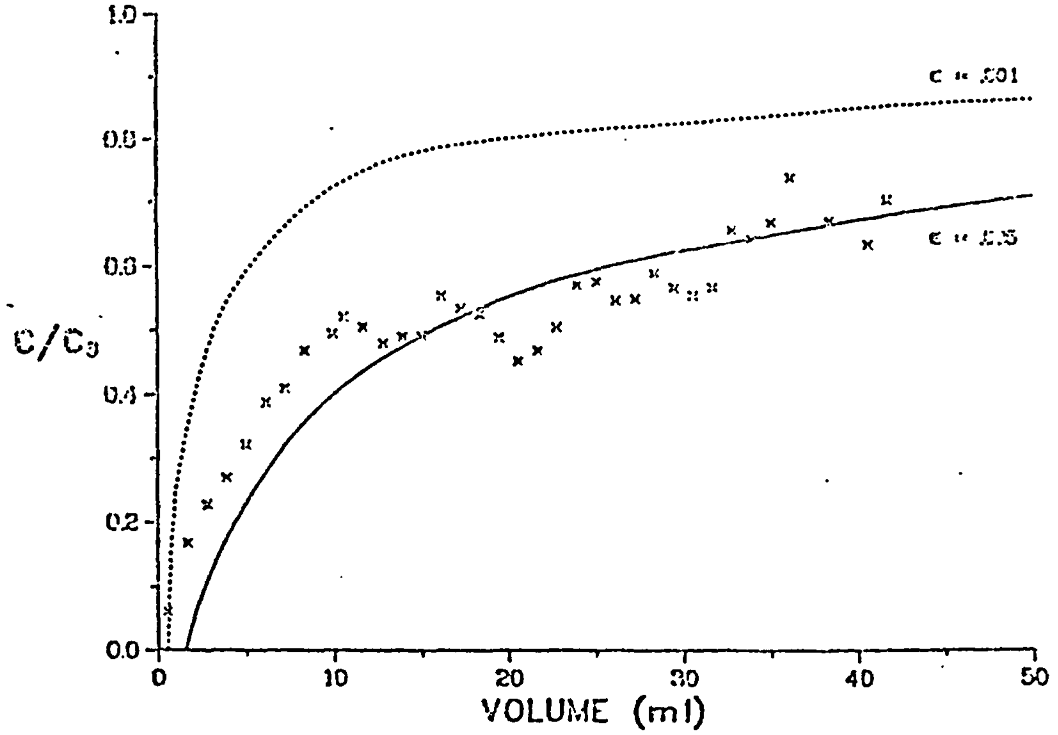

Fig. 1. Breakthrough curves for ^{137}Cs calculated using porosities $\varepsilon = 0.001$ and $\varepsilon = 0.005$. The points (x) represent experimental data.

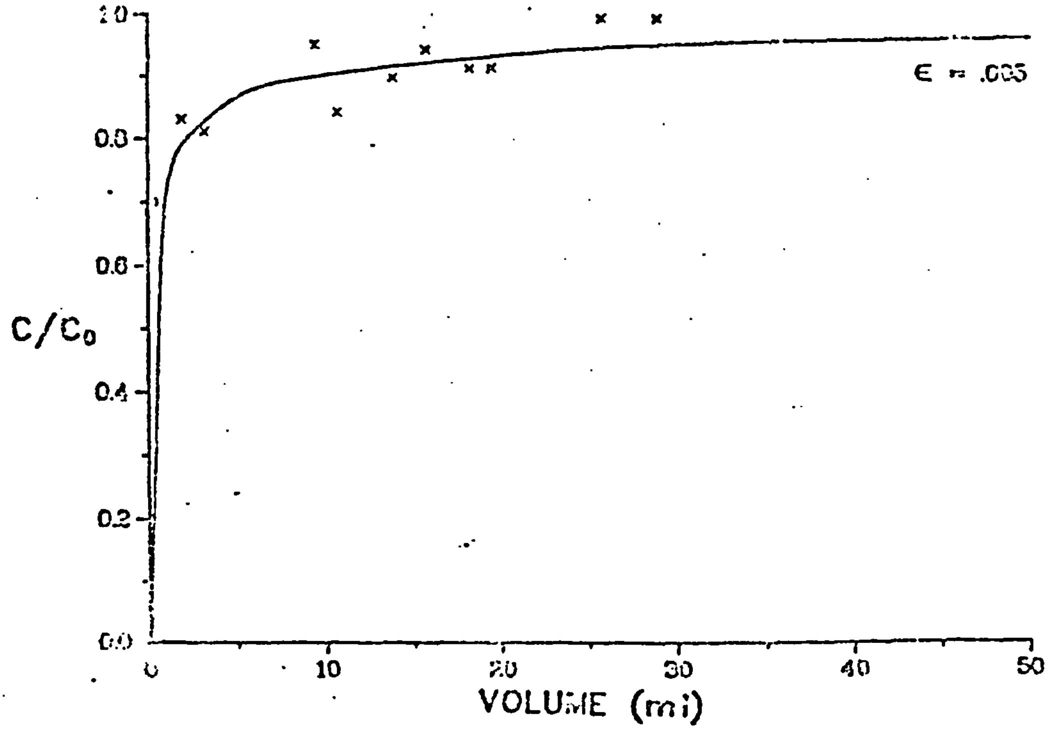

Fig. 2. Breakthrough curves for ^{85}Sr calculated using a porosity $\varepsilon = 0.005$. The points (x) represent experimental data.

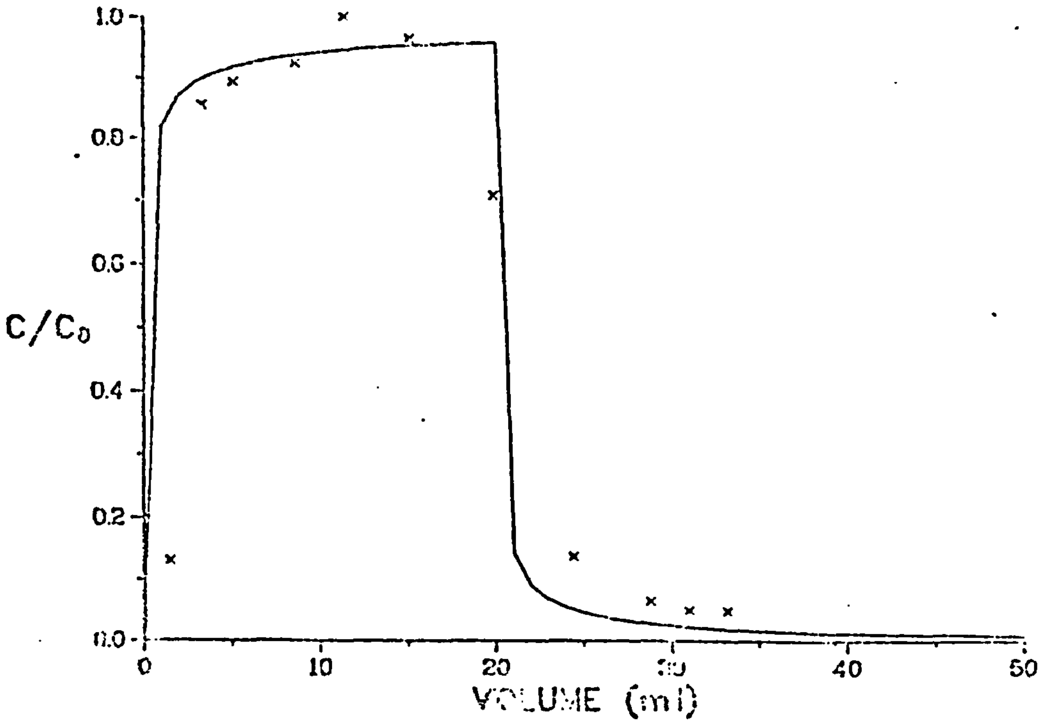

Fig. 3. Elution of a 20 ml slug of ^{85}Sr through a granite fracture. The solid line is a theoretical curve and the points (x) represent experimental data.

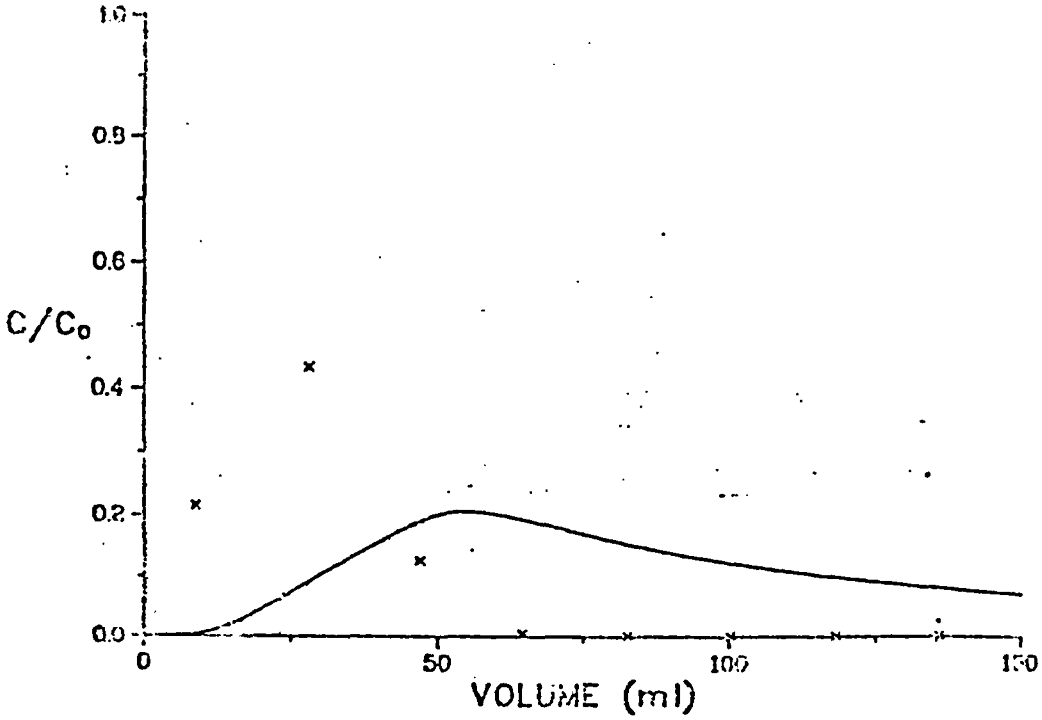

Fig. 4. Elution of a 40 ml slug of ^{85}Sr through a tuff (G1-2335) fracture. Theoretical curve (solid line) assumes the batch K_d. The points (x) represent experimental data.

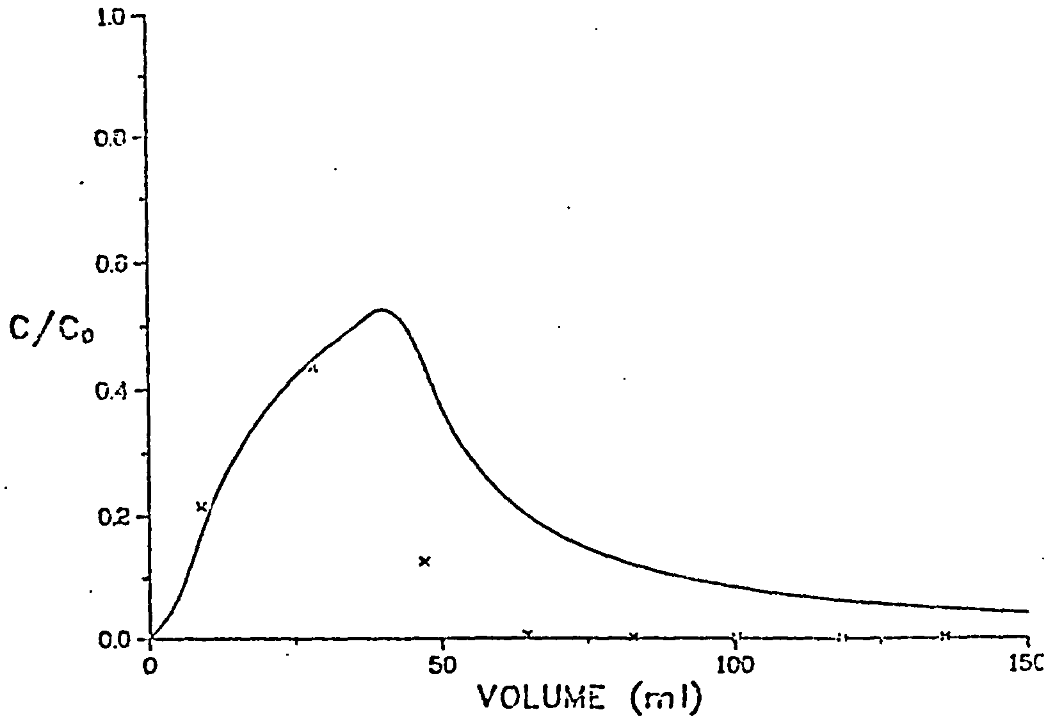

Fig. 5. Elution of a 40 ml slug of ^{85}Sr through a tuff (G1-2335) fracture. The K_d was adjusted to fit the breakthrough. The solid line is a theoretical curve and the points (x) represent experimental data.

The results of the fracture for experiments with tuff samples from G1-2335 and G1-2840 were not in agreement with the calculation if the K_d values determined from batch experiments were used (see Fig. 4). The K_d values which gave a best fit to the breakthrough portion of the strontium elution were 30 ml/g and 16 ml/g for G1-2335 and G1-2840, respectively. The batch measurements yielded 148 ml/g and 160 ml/g for G1-2335 and G1-2840, respectively. A general trend which has been observed in sorption experiments on tuff is that batch measurements yield K_d values that are 3 to 5 times larger than the K_d values that are determined by column experiments. These experiments are consistent with that trend. In addition, the shape of the elution calculated for the tuffs is not in agreement with the observed elution. The activity desorbs more slowly than one would expect for reversible, diffusion-controlled sorption. This observation is also consistent with previous measurements of sorption on tuff. In general the K_d values determined by desorbing activity from tuff are considerably larger than those determined from the sorption process.

REFERENCES

1. I. Neretnieks, "Diffusion in the Rock Matrix: An Important Factor in Radionuclide Retardation?" J. Geophys. Res. 85, 4379 (1980).

2. P. A. Witherspoon, J. S. Y. Wang, K. Iwai, and J. E. Gale, "Validity of Cubic Law for Fluid Flow in a Deformable Rock Fracture," Lawrence Berkeley Laboratory report LBL-9557 (October 1979).

3. P. A. Witherspoon, C. H. Amick, J. E. Gale, and K. Iwai, "Observations of a Potential Size-Effect in Experimental Determination of the Hydraulic Properties of Fractures," Lawrence Berkeley Laboratory report LBL-8571 (May 1979).

4. W. R. Brace, J. B. Walsh, and W. T. Frangos, "Permeability of Granite under High Pressure," J. Geophys. Res. 73, 2225 (1968).

5. B. R. Erdal, R. D. Aguilar, B. P. Bayhurst, W. R. Daniels, C. J. Duffy, F. O. Lawrence, S. Maestas, P. Q. Oliver, and K. Wolfsberg, "Sorption-Desorption Studies on Granite. I. Initial Studies of Strontium, Technetium, Cesium, Barium, Cerium, Europium, Uranium, Plutonium, and Americium," Los Alamos Scientific Laboratory report LA-7456-MS (February 1979).

6. B. R. Erdal, W. R. Daniels, D. T. Vaniman, and K. Wolfsberg, Eds., "Research and Development Related to the Nevada Nuclear Waste Storage Investigations, April 1 - June 30, 1981," Los Alamos National Laboratory report LA-8959-PR (in press, 1981).