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A FUNDAMENTAL APPROACH TO THE ANALYSIS
OF RADIONUCLIDE TRANSPORT RESULTING FROM
FLUID FLOW THROUGH JOINTED MEDIA

MASTER

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Prepared by Sandia Laboratories, Albuquerque, New Mexico 87185
and Livermore, California 94550 for the United States Department of
Energy under Contract DE-AC04-76DP00789.

February 1981

Sandia National Laboratories

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Printed in the United States of America

Available from:

National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

Price: Printed copy \$6.00, Microfiche A01

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A Fundamental Approach to the Analysis of Radionuclide
Transport Resulting from Fluid Flow Through Jointed Media

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ABSTRACT

A theoretical and experimental basis is being developed for analysis of radionuclide transport in jointed geologic media. Batch equilibration and rate experiments involving samples of Eleana argillite and Tertiary silicic tuffs in contact with solutions containing Cs, Sr or Pm indicated that most radionuclide sorption is associated with the surfaces of very small intergranular regions and that the rate of sorption is controlled by diffusion of the nuclides into such regions. Based on these experimental results, the continuity equations for radionuclides in the mobile and immobile phases were reduced to a model analogous to Rosen's equations for packed beds and were solved similarly to Rosen's solutions. Using the model and experimental data, limited radionuclide transport analyses were made which indicated that important parameters controlling transport include the intergranular porosity and nuclide penetration depth, fracture plate spacing and length, fluid velocity, and sorption distribution coefficient. Many of these parameters represent physical quantities or processes which can be quantified in the laboratory. However, fluid velocities and fracture plate spacings and lengths must be obtained from the field, and methods must be developed to establish reliable bounds for such field-determined parameters.

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INTRODUCTION

Current literature regarding analysis of radionuclide transport in jointed geologic media is very limited. This situation has partly resulted because finite rates of mass transfer between phases must be considered. In this regard, recent works of interest include those of Rickert, Strickert and Seitz [1]; Friedman and Fried [2]; Hinkebein [3]; Nerentnieks [4,5]; and Rasmuson and Nerentnieks [6]. An earlier paper [7] briefly summarized initial work conducted to develop theoretical and experimental approaches which will provide a basis for analyzing radionuclide transport in jointed geologic media. The purpose of this report is to provide additional details and discussion pertaining to analyses and results. In the following discussion, the theoretical and experimental approaches are illustrated and used to develop a transport model for relatively simple systems. The model and experimental data are then used for limited transport analyses which identify important parameters and some possible deficiencies in current methods of parameter evaluation.

THEORETICAL APPROACH

For purposes of this discussion, the solution phase which is subject to bulk motion in the joints is referred to as the mobile phase, and the solid phase, which includes voids containing stagnant solution, is referred to as the immobile phase. The immobile phase is idealized as consisting of distinct permeable and impermeable

regions whose complicated geometries can be represented in terms of characteristic dimensions and surface areas. The permeable regions are defined as the fractures, pores, and intergranular regions containing stagnant solution, and the impermeable regions are defined as the crystal grains. In the mobile phase mass transfer occurs by convection and diffusion, and in the immobile phase, mass transfer is considered to only occur by molecular diffusion in the permeable regions (see Figure 1).

In general, the transport of radionuclides by fluid flow-through jointed media is described by the differential material balances for the nuclides in the mobile and immobile phases and appropriate initial and boundary conditions. The boundary conditions usually contain terms representing equilibrium and/or mass fluxes at the interface between phases. Hence, the material balances for the mobile and immobile phases are coupled and require simultaneous solution.

Assuming the mobile phase is an aqueous solution in laminar flow, then the material balance for a dissolved component is [8]

$$\frac{\partial C}{\partial t} = -\vec{\nabla} \cdot \vec{v}C - \vec{\nabla} \cdot \vec{J} + \sum_{j=1}^n R_j \quad (1)$$

where C is the component's concentration; J is the component's flux relative to the solution's mass average velocity v ; R_j are terms representing n different rates of production and/or depletion due to homogeneous chemical reactions and/or nuclear decay, and t is time.

The initial condition generally used for the mobile phase is

$$C(X,Y,Z,0) = F(X,Y,Z) \quad (2a)$$

Figure 1. Schematic representation of mobile and immobile phases.

where F is some known function of the rectangular cartesian coordinates X, Y, Z . The boundary conditions employed will depend on the particular situation being considered and the simplifying assumptions used. One commonly employed condition is sorption equilibrium at the interface between phases

$$\hat{C}(0, Y, Z, t) = G[0, Y, Z, C(0, Y, Z, t)] \quad (2b)$$

where $\hat{C}$ is the local concentration associated with the immobile phase; G is the sorption equilibrium relationship, and X has been taken as the coordinate parallel to the direction of mass transfer between the mobile and immobile phases. Another boundary condition is continuity of the radionuclide flux across the interface between phases

$$\vec{n} \cdot \vec{J} = \vec{n} \cdot \vec{J} + \sum_{j=1}^m \hat{r}_j \quad (2c)$$

where $\vec{n}$ is the unit outward normal vector at the interface; $\vec{J}$ is the flux in the permeable regions of the immobile phase relative to the mass average velocity of that phase, and $\hat{r}_j$ are terms representing the m number of rates of production and/or depletion due to sorption, heterogeneous chemical reaction, and/or nuclear decay on the impermeable regions of the bounding surface of immobile phase.

For the permeable regions of the immobile phase, the material balance is

$$\frac{\partial \hat{C}}{\partial t} = -\vec{\nabla} \cdot \vec{J} + \sum_{j=1}^{\bar{n}} \hat{R}_j \quad (3)$$

where $\hat{R}_j$ are terms representing the $\bar{n}$ number of rates of production and/or depletion due to homogeneous chemical reaction and/or nuclear decay in the bulk of the permeable regions. The initial and boundary conditions given by Eq. 2 also apply to Eq. 3. Furthermore, at the interface between permeable and impermeable regions, the following conditions are also applicable to Eq. 3

$$\hat{C}(X,Y,Z,t) = \hat{G}[X,Y,Z,\hat{C}(X,Y,Z,t)] \quad (4a)$$

$$\vec{n} \cdot \vec{\hat{J}} = \sum_{j=1}^{\bar{m}} \hat{r}_j \quad (4b)$$

where $\hat{C}$ is the concentration associated with the surface of the impermeable regions; $\hat{G}$ is the equilibrium relationship between permeable and impermeable regions, and $\hat{r}_j$ are the terms representing the $\bar{m}$ number of rates of production and/or depletion due to sorption, heterogeneous chemical reaction, and/or nuclear decay at the interface between permeable and impermeable regions.

The application of Eqs. 1-4 to the analysis of radionuclide transport requires the identification and investigation of all pertinent physical and chemical phenomena and the development of realistic equilibrium and rate expressions which represent those phenomena. These expressions must then be incorporated into the appropriate terms in Eqs. 1-4, and the resulting models simplified such that subsequent analyses will be sufficiently accurate and computationally feasible. In the following section, an experimental approach which is being developed to identify and study such phenomena is discussed, and some initial results are given.

EXPERIMENTAL APPROACH

General

The objective of the experimental program is to identify the important sorption mechanisms and the important homogeneous and heterogeneous chemical reactions and to obtain sufficient data so that these phenomena can be described quantitatively using appropriate mathematical expressions. A basic approach for achieving this objective is described below.

(1) Using the available literature [9] as well as physical, chemical and mineralogical analyses of the geologic media, pertinent sorption mechanisms and chemical reactions are identified. (2) Sorption equilibrium isotherms and sorption capacities are determined from batch equilibration experiments in which samples of crushed rock are contacted with solutions containing the nuclides of interest. Additional experiments are performed as required to more clearly define the sorption phenomena and any competing chemical reactions. (3) Sorption rate data are obtained from additional independent batch experiments in which monolithic rock samples are contacted with well-mixed solutions containing the nuclides of interest. At the conclusion of each experiment, the final distribution of the nuclides on the monolith is examined using autoradiography and densitometry, collimated radiation detectors, electron microscopy, and the electron microprobe. (4) The post-experiment examinations, the sorption equilibrium isotherms, and material characterizations such as pore size distribution and surface area are used to further analyze the sorption phenomena, to evaluate the effects of any

competing chemical reactions, and to develop appropriate equilibrium and rate expressions. (5) The results of the preceding steps are then evaluated by comparing calculated and experimentally determined batch rate data. The results are further evaluated when incorporated into radionuclide transport models, and data calculated from the models are compared with analogous experimental data.

Experimental Results

Initial batch equilibration and rate experiments have been conducted using samples of Eleana argillite contacted with simulant groundwater solutions containing cesium [10]. The equilibria for the sorption of cesium by the argillite were nonlinear. However, the batch rate experiments involving cesium and argillite have been amenable to analysis, since they were conducted using a solution-phase concentration range in which the isotherm was nearly linear. The results from the cesium-argillite experiments are summarized below, so that development of appropriate rate expressions can be illustrated in the next section.

The Eleana argillite used in the experiments primarily consisted of quartz and layered silicate minerals: kaolinite, pyrophyllite, and mixed layer "illite." The argillite also contained five percent or less by weight of ferrous carbonate and about 1.4% organic carbon, graphitic material. Samples of the argillite generally contained some macroscopic fractures. Scanning electron micrographs indicated that the surface of the samples contained intergranular regions of thin approximately rectangular cross

section having wall spacings on the order of 0.1 microns and less, and the porosity associated with these regions appeared to be between one and ten percent. For 30 to 60 mesh particles, surface areas determined by the BET method were on the order of 9 m²/gm.

Based on the solution chemistry of cesium and of the constituents of the groundwater solutions, it was felt that if the surfaces of the immobile phase remained unaltered, no homogeneous and no heterogeneous chemical reactions other than sorption should be expected. The sorption equilibrium isotherm at room temperature determined from batch equilibration experiments using 30 to 60 mesh particles, was found to be represented by the expression

$$\hat{\hat{C}} = \left(\frac{0.05 \text{ cm}}{1 + 5 \times 10^4 \frac{\text{liter}}{\text{mole}} \hat{C}} \right) \hat{C} \quad (5)$$

$$\text{for } 10^{-2} M < \hat{C} < 10^{-10} M$$

The term $\hat{\hat{C}}$ represents the nuclide concentration associated with the surfaces of the impermeable regions, which are considered to have uniform, continuous properties. It has also been assumed that at the interface between phases, $C = \hat{\hat{C}}$. The distribution coefficient K is then defined as $\hat{\hat{C}}/C$.

Sorption rate data were obtained from independent batch experiments using argillite tablets which were approximately 1/4 inch thick by 2-3/8 inch diameter. At the conclusion of the experiments, the final nuclide distributions on the tablets were examined using autoradiography. Based on the autoradiographs from these and the various tuff experiments, it appeared that sorption was primarily associated with the surfaces of the void

regions penetrating the rock rather than with the impermeable regions of the interface between mobile and immobile phases. Furthermore, autoradiographs of fractured tablets indicated that the depth of nuclide penetration into the immobile phase was probably on the order of 0.01 cm or less and that most sorption probably occurred in intergranular regions having comparable effective lengths, which was consistent with the surface area measurements and estimated intergranular porosities. These conclusions were further confirmed by a comparison between calculated and experimentally obtained rate data, as discussed in the following section.

RADIONUCLIDE TRANSPORT MODEL

Based on the preceding experimental results, appropriate rate expressions were developed and incorporated into Eqs. 1-4, and a fundamental radionuclide transport model was obtained from the resulting equations [11]. In the following paragraphs, development of the rate expressions and transport model and the solutions to the various equations are summarized.

Analysis of Rate Data

Based on the literature concerning ion-exchange kinetics [12], the rate of cesium sorption by the argillite should probably have been limited by diffusion rather than by the intrinsic rate of a "sorption reaction." For the batch rate experiments involving well-mixed solutions, the boundary layer thickness in the mobile phase should have been on the order of 10^{-2} to 10^{-3} cm [12], and for

the experiments done using argillite tablets, it appeared reasonable to expect that diffusion into the relatively long, narrow intergranular regions would primarily dominate the rate of mass transfer across the interface between mobile and immobile phases.

Referring to Figure 2, equations describing mass transfer in the intergranular regions were developed by assuming that the rock's physical and chemical properties could be considered continuous; all parameters could be considered constant valued; the ratio of the lengths of the intergranular regions to the wall spacings were sufficiently large so that diffusion could be considered one-dimensional; the $\hat{r}_j$ terms in Eq. 4b were either negligible or sufficiently large so that local equilibrium existed between bulk stagnant solution and impermeable surface; the solution-phase nuclide concentrations were sufficiently dilute so that Fick's law was reasonably valid, and that there were no appreciable effects due to surface diffusion. The resulting flux expression describing mass transfer across the interface between phases was then given by Eq. 2c in the form

$$\vec{n} \cdot \vec{J} = \vec{n} \cdot \vec{\hat{J}} = \left(\frac{-D}{a_t} \right) \frac{\partial \hat{C}}{\partial x} \quad (6a)$$

where a_t is a tortuosity factor; $\hat{C}$ is the nuclide concentration in the solution occupying the intergranular region; D is the diffusion coefficient for the nuclide in the solution, and it has been assumed that the $\hat{r}_j$ terms in Eq. 2c are negligible. An equivalent expression, which later will be more convenient, is

Figure 2. Schematic representation of mass transfer in intergranular regions.

$$\vec{n} \cdot \vec{J} = \left(\frac{-\bar{D}}{a_t} \right) \frac{\partial \bar{C}}{\partial X} \quad (6b)$$

where $\bar{C} = \bar{K}C$; $\bar{D} = D/\bar{K}$; $\bar{K} = 1 + a_c hK$; h is the ratio of the perimeter length to the area of a simple characteristic cross section for the intergranular regions, and a_c is a roughness factor for those regions. The material balance for the bulk of the intergranular regions is given by

$$\frac{\partial \bar{C}}{\partial t} = \left(\frac{\bar{D}}{a_t^2} \right) \frac{\partial^2 \bar{C}}{\partial X^2} \quad (7a)$$

with the initial and boundary conditions given by Eq. 6b and

$$C(X, 0) = 0 = \bar{C}(X, 0) \quad (7b)$$

$$\frac{\partial \hat{C}(\bar{H}, t)}{\partial X} = 0 = \frac{\partial \bar{C}(\bar{H}, t)}{\partial X} \quad (7c)$$

The $\hat{R}_j$ terms in Eq. 3 have been assumed negligible, and $\bar{H}$ is a characteristic depth to which the intergranular regions penetrate the immobile phase and is considered constant. The material balance, represented by Eqs. 7, for the immobile phase is coupled by Eqs. 6 to the material balance, represented by Eqs. 1 and 2, for the mobile phase.

In order to describe a batch rate experiment, Eqs. 7a,b,c are solved simultaneously with Eq. 8 below

$$\frac{v}{apAa_t \bar{K}} = \left(\frac{\bar{D}}{a_t^2} \right) \frac{\partial \bar{C}}{\partial X} \quad (8)$$

where $C(o) = C_o = \text{a constant}$; A is the gross geometrical surface area of the rock tablet; a is a roughness factor for the tablet's surface; p is the interfacial porosity associated with the intergranular regions, and V is the volume of the mobile phase used in the experiment. The solution to Eqs. 7 and 8 can be obtained from Crank [13]. Some typical data from batch rate experiments with argillite tablets and cesium solutions are shown in Figure 3. Analogous data calculated using Eqs. 7 and 8 and the given parameter values are shown by the solid line in the figure.

The agreement between calculated and experimental data is not entirely quantitative, as shown on Figure 3. However, when inhomogeneities in and variations between rock samples are considered, as well as the purely representative nature of some of the parameters given in Figure 3, and when the initial effects due to diffusion through the boundary layer in the solution phase are also considered, the agreement between calculated and experimental results appears sufficiently good to support the validity of Eqs. 6 and 7 and the parameter values given in the figure.

In Figure 3, the range of values for each of the parameters a , a_c , a_t^2 , $\bar{H}$ and p was estimated based on the physical characterization of the tablets' surfaces by SEM, BET surface area measurements, and post-experiment autoradiographs. The value of h was calculated so as to be consistent with an internal and external surface area for the 30 to 60 mesh particles of 9×10^4 and $40 \text{ cm}^2/\text{gm}$, respectively, and with the values estimated for a_c , a_t , $\bar{H}$ and p . The value for K was determined using data from the batch equilibration experiments and surface area measurements, and $\bar{K}$ was calculated from $\bar{K} = 1 + a_c h K$.

BATCH RATE DATA

PARAMETER	VALUE USED	ESTIMATED RANGE
a	1	1 - 2
a_c	$\sqrt{2}$	1 - 4
a_t^2	2	1 - 4
D	$2 \times 10^{-5} \text{ cm}^2/\text{sec}$	---
$\bar{H}$	$5 \times 10^{-3} \text{ cm}$	$10^{-3} - 10^{-2} \text{ cm}$
h	$6 \times 10^6 \text{ cm}^{-1}$	*
K	$5 \times 10^{-2} \text{ cm}$	---
$\bar{K}$	4×10^5	---
p	3×10^{-2}	$10^{-2} - 10^{-1}$

* CALCULATED SO AS TO BE CONSISTENT WITH SURFACE AREA MEASUREMENTS AND VALUES USED FOR a_c , a_t , H , AND p

Figure 3. Comparison of Experimental (circles) and Calculated (solid line) Rate Data Using the Parameter Values Given.

Model Development and Solution of Equations

Based on the preceding analyses, a fundamental radionuclide transport model was developed for systems involving an incompressible solution in laminar, one-dimensional flow through linear fractures having rectangular cross sections and approximately constant, uniform dimensions and continuous physical and chemical properties (see Figure 4). Dilute aqueous solutions of cesium, 10^{-6} M and less, flowing through appropriate fractures in Eleana argillite would appear to be an example of such systems. In order to obtain an analytical solution for the model, it was further necessary to neglect terms representing effects due to nuclear decay, to assume constant-valued parameters, to replace the parabolic velocity profile by an appropriate average velocity, to use a step function for the radionuclide source term, and to assume that the fracture wall spacing H is sufficiently small so that diffusion in the x_1 -direction in the mobile phase can be approximated as a quasi-steady-state process

$$n \cdot J \approx \frac{D(C - C_s)}{H/4} \quad (9)$$

where C_s is the mobile phase concentration evaluated at the interface between phases. Eqs. 1,2,3 and 4 then reduce to the following fundamental model for radionuclide transport

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial z} = - \left(\frac{1}{M} \right) \frac{\partial Q}{\partial t} \quad \left(\begin{array}{l} \text{solution-phase} \\ \text{material balance} \end{array} \right) \quad (10a)$$

with initial and boundary conditions

SIMPLE FRACTURE

Linear

Constant and Uniform Dimensions

Constant and Continuous Properties

Mobile Phase: Laminar, 1-D Motion

Dissolved Nuclides Only

Source Term: Step Function

$\vec{v} \doteq$ A Constant

H Sufficiently Small That:

$$\vec{n} \cdot \vec{J} \doteq \frac{D(C - C_s)}{H/4}$$

Figure 4. Schematic Diagram of Simple Fracture.

$$U(0,t) = C(0,t)/C_0 = \begin{cases} 0, & t \leq 0 \\ 1, & t > 0 \end{cases} \quad (10b)$$

$$U(z,0) = 0 \quad t \leq 0 \quad (10c)$$

and where for a characteristic penetration depth $\bar{H}$

$$Q(z,t) = \frac{1}{\bar{H}} \int_0^{\bar{H}} \bar{C}(x,z,t) dx \quad (11a)$$

with initial conditions

$$Q(z,0) = 0 \quad (11b)$$

$$\frac{\partial \bar{C}}{\partial t} = \bar{D}_c \frac{\partial^2 \bar{C}}{\partial x^2} \quad \left(\begin{array}{l} \text{material-balance for} \\ \text{the intergranular regions} \end{array} \right) \quad (12a)$$

with initial and boundary conditions

$$\bar{C}(x,z,0) = 0 \quad (12b)$$

$$\frac{\partial \bar{C}}{\partial x}(\bar{H},z,t) = 0 \quad (12c)$$

$$\frac{\partial Q}{\partial t} = \frac{1}{R_M} (C - \bar{C}_s/K) \quad (13)$$

where C is now the average concentration for the cross section of the fracture; C_0 is the constant mobile phase concentration entering the fracture; $\bar{C}_s$ is the value of $\bar{C}$ at the interface between mobile and immobile phases; $\bar{D}_e = \bar{D}/a_t^2$; $M = H/2ap\bar{H}$; $R_M = \bar{H}H/4D$, and z is the spatial coordinate parallel to the direction of bulk fluid motion. Eqs. 10, 11, 12 and 13 can be solved analogously

to Rosen's [14] development for packed beds. The result is

$$U(\bar{Z}, \bar{t}) = \frac{1}{2} + \frac{2}{\pi} \int_0^{\infty} e^{-\bar{X}E_1(\ell, g)} \sin [\bar{Y}\ell^2 - \bar{X}E_2(\ell, g)] \frac{d\ell}{\ell} \quad (14)$$

where $\bar{t} = t - Z/v$; $U = C/C_0$, $\bar{Z} = Z/Mv$, and the remaining terms are defined in Table 1. Eq. 14 is identical in form to Rosen's solution except for relatively small differences in the numerical values for the functions E_1 and E_2 . Such differences should be masked by the inherent uncertainties associated with the values of the various parameters, and for purposes of making preliminary calculations, the numerical results given by Rosen should be applicable [15]. For values of $\bar{X}$ on the order of 50 and greater, Eq. 14 reduces to the following asymptotic expression

$$U(\bar{Z}, \bar{t}) = \frac{1}{2} + \frac{1}{2} \operatorname{erf} \left[\frac{\frac{2\bar{Y}}{\bar{X}} - 1}{2 \left(\frac{1 + 3g}{3\bar{X}} \right)^{1/2}} \right] \quad (15)$$

Discussion

Consider Eq. 15, the right side of which is symmetrical about the value of $U = 0.5$. If for a given value of t , $t_{0.01}$ is defined as the elapsed time required for U to reach a value of 0.01, and if $\bar{t}_{0.5}$, $t_{0.5}$ and $t_{0.99}$ are defined analogously, then from Eq. 15 and appropriate values of the error function, it can be shown that

$$\frac{t_{0.99} - t_{0.01}}{\bar{t}_{0.5}} = \frac{6.6}{\left(\frac{2ap}{H} \cdot \frac{Z}{v} \right)^{1/2}} \left(\frac{\frac{3\bar{D}_e}{H} \bar{K} + \frac{4D}{H}}{\frac{3\bar{D}_e}{H} \bar{K} - \frac{4D}{H}} \right)^{1/2} \quad (16)$$

Table 1. Definitions of Terms in Eq. 14.

B	=	parameter in the limits of integration of the inversion integral for the Laplace transform
$E_1(\ell, g)$	=	$(\bar{E}_1 + g\bar{E}_1^2 + g\bar{E}_2^2)/E'$
$E_2(\ell, g)$	=	$\bar{E}_2/E'$
$\bar{E}_1$	=	$\ell(\sinh 2\ell - \sin 2\ell)/E''$
$\bar{E}_2$	=	$\ell(\sinh 2\ell - \sin 2\ell)/E''$
E'	=	$(\ell + g\bar{E}_1)^2 + (g\bar{E}_2')^2$
E''	=	$\cosh 2\ell + \cos 2\ell$
f	=	$\bar{D}_e \bar{K}/\bar{H}^2 = D/a_t^2 \bar{H}^2$
g	=	$fR_M = H/4\bar{H}$
ℓ	=	$(BW)^{1/2}$
w	=	$\bar{D}_e/2\bar{H} a_t^2 \bar{H}$
$\bar{X}$	=	$f\bar{z} = \frac{\bar{D}_e}{\bar{H}} \cdot \frac{2ap\bar{K}}{\bar{H}} \cdot \frac{z}{v}$
$\bar{X}/q$	=	$\frac{4D}{\Pi} \cdot \frac{2ap}{\bar{H}} \cdot \frac{z}{v}$
Y	=	$w\bar{t} = (D/2\bar{H} a_t^2 \bar{K}) (t - z/v)$
Y/X	=	$\frac{v}{z} \cdot \frac{M}{2\bar{K}} (t - z/v)$

Since Eq. 15 is symmetrical about the value of $U = 0.5$, Eq. 16 implies that for given values of Z and $(2ap/H)(3\bar{D}_e\bar{K}/\bar{H})(4D/H)$, as Z/v becomes sufficiently large, the time interval by which the value of $U = 0.01$ precedes the value of $U = 0.5$ becomes negligible compared with the interval required for the value of $U = 0.5$ to arrive at the given value of Z . The value of $t_{0.5}$ is then given by

$$t_{0.5} = (1 + \bar{K}/M)Z/v \quad (17)$$

and the velocity $v_{0.5}$ at which the value of $U = 0.5$ travels is given by

$$v_{0.5} = vM/(M + \bar{K}) \quad (18)$$

Conversely, as Z/v becomes small, the time interval by which the value of $U = 0.01$ precedes the value of $U = 0.5$ becomes large relative to the interval required for the value of $U = 0.5$ to arrive at the given value of Z , which implies that $v_{0.01}$ approaches v . The terms $4D/H$ and $\bar{D}_e\bar{K}/\bar{H}$ in Eq. 16 essentially reflect the extent to which the shape of the concentration profile $U(\bar{Z}, \bar{t})$ is influenced by diffusion of radionuclides in the mobile and immobile phases, respectively.

First, consider the effect of diffusion through the mobile phase. If $t = Z/v$, then $\bar{t}$ is zero, and as shown by Rosen

$$U(\bar{Z}, 0) = \exp(-Z/vMR_M) \quad (19)$$

If the value of the right side of the above equation is equal to or greater than 0.01, then the value of U equal to or greater

than 0.01 essentially travels with the mobile phase velocity v . This results because the ratio, represented by vMR_M/Z , of the mobile phase velocity to the rate of diffusion of radionuclides through that phase is sufficiently large so that in the distance Z , the concentration C_0 entering the fracture does not have sufficient residence time to be reduced to a value of 0.01 or less. Therefore, Eq. 19 can be considered to quantitatively define conditions for which the rate of diffusion of radionuclides through the mobile phase is sufficiently slow so as to significantly influence the shape of the concentration profile. In particular, from Eq. 19 it can be shown that the value of $t_{0.01}$ will be less than Z/v only when

$$Z/v > \frac{2.3}{\frac{4D}{H} \cdot \frac{ap}{H}} \quad (20a)$$

As $t_{0.01}$ becomes much less than Z/v , the effect which diffusion through the mobile phase has on the concentration profile becomes small. For the effect to be considered negligible, Z/v should be much larger than the right side of Eq. 20a, say by a factor of 10, or

$$\frac{Z}{v} > \frac{23}{\frac{4D}{H} \cdot \frac{ap}{H}} \quad (20b)$$

Now consider the effect of diffusion into the immobile phase. If $\bar{D}_e \bar{K}/\bar{H} \gg 4D/H$, which is rather uncommon, then if Eq. 20b is satisfied, Eqs. 17 and 18 apply, and $v_{0.01}$ and $t_{0.01}$ approach $v_{0.5}$ and $t_{0.5}$ respectively. If $\bar{D}_e \bar{K}/\bar{H} \leq 4D/H$, which is more common,

then as the initial effects of diffusion through the mobile phase become small, so that $v_{0.01} < v$, the shape of the concentration profile is determined by diffusion into the immobile phase. Referring to Rosen's numerical results, as the value of $\bar{X}$ increases, the velocity at which the value of $U = 0.01$ or greater travels becomes increasingly less than v . At values of $\bar{X}$ greater than 50 and $g/\bar{X}$ much less than 0.2, the velocities at which various values of U travel become small compared with the velocity $v_{0.5}$ given by Eq. 18, and

$$v_{0.01} \approx v_{0.5} = vM/(M + \bar{K})$$

If $t_{0.5}$ is to be given by Eq. 17 and $(t_{0.99} - t_{0.01})$ is to be negligible relative to $t_{0.5}$, then the value of $\bar{X}$ should be much greater than 50, say on the order of 500. Therefore, when $\bar{D}_e \bar{K}/\bar{H} \leq 4D/H$, Eq. 17 should be applicable if

$$\frac{z}{v} > \frac{250}{\frac{\bar{D}_e \bar{K}}{\bar{H}} \frac{ap}{H}} \quad (21)$$

Since Eqs. 20b and 21 quantitatively define conditions for when the concentration profile is negligibly affected by rates of diffusion through the mobile and immobile phases and when the ratio $(t_{0.99} - t_{0.01})/\bar{t}_{0.5}$ is small, those two equations essentially determine the conditions for which the assumption of local equilibrium between bulk phases would be a reasonable approximation to be applied to the analysis of radionuclide transport in jointed geologic media.

The application of Eqs. 14-21 is discussed in the next section. However, some cautionary remarks are first in order.

1) The model was derived assuming continuous, constant-valued parameters and does not account for fluctuations in parameter values due to variations in the geologic media or to functional relationships between parameters. 2) When terms for chemical reactions or nuclear decay are needed, use of the model would require numerical computations. 3) The equation used to describe diffusion through the mobile phase was very approximate, and in the case of large fractures, may not be sufficiently accurate.

RADIONUCLIDE TRANSPORT ANALYSIS

Model Evaluation

Some initial experiments have been conducted in which stable cesium solutions, having initial concentrations of about $10^{-3}M$, have been pumped vertically upward through artificially prepared fractures in Eleana argillite core samples. These experiments have primarily been designed to develop sample preparation techniques, and quantitative analysis of the data has been complicated by the nonlinear nature of the sorption isotherm for the cesium concentrations used. However, qualitative analysis of the data has been encouraging. In particular, the equations developed in the preceding section have successfully been used to calculate bounds for the experimentally obtained breakthrough curves.

General Analyses

For analysis of a specific system, either Eq. 14 or 15 should be used. However, for more general cause-and-effect type analyses, the equations and discussion of the preceding section are conveniently summarized in Figure 5, which represents the superposition of independent plots of Eq. 20b and Eq. 21. For values of $4apD/H^2$ and Z/v in regions 3 and 4, the breakthrough time t_B at which the value of U equal to 0.01 or greater appears at distance Z is equal to Z/v . For values of $4apD/H^2$ and Z/v in regions 1 and 6 and values of $ap\bar{D}_e\bar{K}/H\bar{H}$ and Z/v in regions 1, 2 and 3, t_B is given by

$$t_B \approx [1 + (2ap\bar{H}/H)\bar{K}] \frac{Z}{v} \quad (22)$$

and the assumption of local equilibrium between bulk mobile and immobile phases can be applied to radionuclide transport models for jointed media. For other combinations of parameters values, the time t_B is between Z/v and that given by Eq. 22, and the local equilibrium assumption is not valid. For example, if $4apD/H^2$ were equal to 10 day^{-1} , and z/v were equal to 10^{-2} day , then a concentration $C \geq 0.01 C_0$ would occur at position Z at 10^{-2} day , regardless of the value of $ap\bar{D}_e\bar{K}/H\bar{H}$. If $4apD/H^2$ were again equal to 10, but z/v were equal to 10, and $ap\bar{D}_e\bar{K}/H\bar{H}$ were greater than 10^3 day^{-1} , then a concentration of $C \geq 0.01 C_0$ would occur at time t_B given by Eq. 22.

Figure 5. Effect of mobile- and immobile-phase diffusion rates on breakthrough time.

Most of the parameters shown on Figure 5 represent physical quantities or processes which can be quantified in the laboratory. However, parameters such as the fluid velocity, v and the fracture plate spacing H must be determined from field measurements. Current measurement techniques are crude and introduce large uncertainties into the parameter values. The importance of these uncertainties can be illustrated by choosing a typical hydraulic gradient $\Delta P/Z$ and then calculating the maximum value of H which will allow some minimum value for time to breakthrough t_B at selected distances L from a radionuclide source. For purposes of illustration, a simple fracture such as that shown on Figure 4, or a system of joints having approximately uniform dimensions and connected in a manner such that fluid motion is laminar and one-dimensional is considered; parameter values are taken from Figure 3; t_B is taken as 10^6 years, and $\Delta P/Z$ as 10^{-3} psi/ft or about 10 feet of water per mile. Approximate values of v as a function of H are then given by

$$v \approx 10^4 \frac{M}{\text{cm}^2 \text{ day}} H^2 \quad (23)$$

where H is in units of centimeters. The largest maximum values of H will occur when Eq. 22 is valid, and if the value of $2ap\bar{H}\bar{K}/H$ is much greater than unity. In which case the maximum value of H , as illustrated on Figure 6, is given by

$$H_{\max} = L^{1/3} \left(\frac{2ap\bar{H}\bar{K}}{t_{0.01} 10^4 \frac{m}{\text{cm}^2 \text{ day}}} \right)^{1/3} \quad (24)$$

For values of $4apD/H^2$ and z/v in regions 3 and 4 of Figure 5 or for values of $2ap\bar{H}\bar{K}/H$ much less than unity, the maximum value of H , as illustrated on Figure 6, is given by

$$H_{\max} = L^{1/2} \left(\frac{1}{t_{0.01} 10^4 \frac{m}{cm^2 day}} \right)^{1/2} \quad (25)$$

For example, if $4apD/H^2$ were equal to 10 day^{-1} , z/v were equal to 10 day, and $ap\bar{D}\bar{e}\bar{K}/H\bar{H}$ were greater than 10^3 day^{-1} , then $H_{\max}$ would be given by Eq. 24. However, if $4apD/H^2$ were again equal to 10, but z/v were equal to 10^{-2} day , then $H_{\max}$ would be given by Eq. 25, regardless of the value of $ap\bar{D}\bar{e}\bar{K}/H\bar{H}$. Some values of $H_{\max}$ calculated using Eqs. 24 and 25 are shown in Table II.

Table II. Maximum Fracture Widths for $\Delta P/Z = 10^{-3} \text{ psi/ft}$ and $t_{0.01} = 10^6 \text{ years}$

$(2ap\bar{H}/H)\bar{K}$	$L \text{ (m)}$	$H_{\max} \text{ (cm)}$
120	30	1×10^{-3}
	10^4	7×10^{-3}
0	30	3×10^{-6}
	10^4	5×10^{-5}

The values of $H_{\max}$ given in Table II are for ideal fractures. In natural systems, constrictions and debris would probably cause lower mobile phase velocities than those given by Eq. 23 and would, therefore, result in larger maximum fracture widths. However, the nature and extent of such effects currently appear uncertain.

Figure 6. Effect of mobile and immobile phase diffusion rates on the maximum allowable fracture width $H_{\max}$.

Furthermore, for the axial distances considered, 30 and 10^4 meters, the values of $H_{\max}$ calculated from Eq. 24 are about 10^{-3} and 10^{-2} cm, respectively, and are on the same order as some values estimated for actual joint widths. Therefore, the uncertainties in the values of such estimated parameters must be reliably determined.

CONCLUSIONS

The initial work conducted to develop a theoretical and experimental approach for analysis of radionuclide transport in jointed geologic media has been encouraging. The transport model developed for relatively simple systems indicated that important parameters controlling radionuclide transport include the intergranular porosity p and penetration depth $\bar{H}$, the fracture plate spacing H and length Z , the fluid velocity v , and the sorption distribution coefficient $\bar{K}$. Values determined for these parameters can be used to determine when the assumption of local equilibrium between bulk phases can be applied to radionuclide transport in jointed geologic media. Furthermore, many of the parameters represent physical quantities or processes which can be characterized in the laboratory. However, parameters such as joint widths H and fluid velocities v must be obtained from the field. Current techniques for evaluating these parameters involve considerable uncertainties, particularly with regard to values for H , and methods must be developed to establish reliable bounds on the uncertainties in the values of such field-determined parameters.

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NOMENCLATURE

The following parameters are defined in Table I: B , $E_1(l, g)$, $E_2(l, g)$, $\bar{E}_1$, $\bar{E}_2$, E' , E'' , f , g , l , w , $\bar{X}$ and Y . Other parameters are defined as follows:

- A = gross geometrical surface area of rock tablet
- a = a roughness factor for the external surface of a tablet or fracture
- a_c = a roughness factor for the intergranular regions of the immobile phase
- a_t = a tortuosity factor for the permeable regions of the immobile phase
- C_s = mobile-phase concentration evaluated at the interface between mobile and immobile phases
- C = mobile-phase concentration
- C_o = mobile-phase concentration at entrance of fracture
- $\bar{C}$ = $\bar{K}C$
- $\bar{C}_s$ = the value of $\bar{C}$ at the interface between phases
- $\hat{C}$ = the local concentration associated with the immobile phase
- $\hat{C}_i$ = concentration associated with the surface of the impermeable regions of the immobile phase
- D = diffusion coefficient for a dissolved component
- $\bar{D}$ = $D/\bar{K}$
- $\bar{D}_e$ = $\bar{D}/a_t^2$
- F = initial distribution of a dissolved component in the mobile phase, a known function
- G = local sorption equilibrium relationship between mobile and immobile phases
- $\hat{G}$ = local sorption equilibrium relationship between permeable and impermeable regions of the immobile phase
- H = spacing between fracture walls
- $\bar{H}$ = characteristic depth to which intergranular regions penetrate the immobile phase

- h = ratio of the perimeter length to the area of a simple characteristic cross section for the intergranular regions (ratio of the area of the bounding surface to the volume of an intergranular region)
- $\vec{J}$ = flux of a dissolved component relative to the mass average velocity of the mobile phase
- $\star J$ = diffusive flux in the immobile phase
- K = sorption equilibrium distribution coefficient (units of length) defined using immobile phase concentrations in units of mass per unit surface area and mobile phase concentrations in units of mass per unit volume
- $\bar{K} = 1 + a_c h K$
- $M = H/2ap\bar{H}$
- $\vec{n}$ = the unit outward normal vector from a given surface
- p = the interfacial porosity associated with the intergranular regions
- R_j = terms representing rates of production and/or depletion due to homogeneous chemical reactions and/or nuclear decay in the mobile phase
- $R_M = \bar{H}H/4D$
- $\hat{R}_j$ = terms representing rates of production and/or depletion due to homogeneous chemical reaction or nuclear decay in the bulk of the permeable regions of the immobile phase
- $\hat{r}_j$ = terms representing rates of production and/or depletion due to sorption, heterogeneous chemical reaction, and/or nuclear decay on the impermeable regions of the bounding surface of the immobile phase
- $\hat{\hat{r}}_j$ = terms representing rates of production and/or depletion due to sorption, heterogeneous chemical reaction, and/or nuclear decay at the interface between permeable and impermeable regions in the bulk of the immobile phase
- t = time
- $t_{0.01}, t_{0.5}, \bar{t}_{0.5}, t_{0.99}$ = value of t (or $\bar{t}$) required for U to reach a value of 0.01, 0.5 and 0.99, respectively

$$\bar{t} = t - z/v$$

$$U = C/C_0$$

$$V = \text{volume of solution used in a batch rate experiment}$$

$$v = \text{mass average velocity of the mobile phase}$$

$$v_{0.01}, v_{0.5} = \text{the velocity at which the value of } U = 0.01 \text{ and } 0.5, \text{ respectively, travels along the fracture}$$

$$x_i = \text{general coordinates}$$

$$z = \text{spatial coordinate parallel to direction of bulk fluid motion}$$

$$\bar{z} = z/Mv$$

$$\Delta P/z = \text{hydraulic gradient}$$

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