Journal of Hydrology, 52 (1981) 47—57
Elsevier Scientific Publishing Company, Amsterdam — Printed in The Netherlands

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AN ANALYTICAL SOLUTION FOR SOLUTE TRANSPORT THROUGH FRACTURED MEDIA WITH MATRIX DIFFUSION

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(Received May 1, 1980; accepted for publication June 16, 1980)

ABSTRACT

Grisak, G.E. and Pickens, J.F., 1981. An analytical solution for solute transport through fractured media with matrix diffusion. J. Hydrol., 52: 47—57.

An analytical solution is presented for solute transport in a planar fracture coupled with diffusion into the adjacent matrix. The solution solves for the transient concentration distribution along the fracture as well as into the matrix. The mechanisms for solute transport are assumed to be advection in the fracture and diffusion in the porous matrix. The use of the analytical solution is illustrated by simulation of laboratory data from a column tracer test on fractured till and by simulation of a hypothetical scenario consisting of a single long continuous fracture in a regional groundwater flow system.

INTRODUCTION

Most studies of groundwater flow within fractured media emphasize the dominating influence of fractures on the effective permeability of the rock mass. For nonreactive solutes the matrix porosity between major fractures can provide significant solute storage space which, in the case of radioactive contaminants, provides additional time for decay before release to the biosphere from a hydrogeologic system. The porosity in the matrix which is connected to the major fractures and allows for solute diffusion from the fractures into the matrix has been termed the *diffusion porosity* (Norton and Knapp, 1977) or *matrix porosity* (Grisak and Pickens, 1980).

Garrels et al. (1949) conceptually illustrated that diffusion of a solute from a fracture into a porous matrix could be invoked as an important ore depositional process. Foster (1975) illustrated with a one-dimensional analytical solution that the tritium profile in the unsaturated part of the Chalk aquifer in England could be a consequence of tritium diffusion from the fractures into the porous blocks. The solution was of limited value, however, since it was not suited to handle an advective component in the fracture and empirical corrections were necessary to account for the matrix porosity. Young et al. (1976) used similar reasoning to Foster's in their interpretation

0022-1694/81/0000—0000/$02.50

of the nitrate profile in the unsaturated zone of the Chalk aquifer. In a simulation of solute transport through the Chalk, Oakes (1977) assumed that only 15% of the surface input of nitrates and tritium travelled with the infiltrated water to the water table and did not interact with the matrix pore water. As a result of solute storage in the unfractured chalk blocks, most of the tritium and nitrate remained shallow and lagged behind the bulk of the recharge water.

Barbreau et al. (1979) and Davison et al. (1979) illustrated, using finite-difference modeling techniques, that matrix diffusion can provide significant retardation of the transport of contaminants in fractures. Davison et al. illustrated the effect of the magnitude of the diffusion coefficent D^* in the porous matrix by calculating concentration profiles along the fracture and breakthrough curves at relatively large (202 m) distances away from a continuous source. Barbreau et al. illustrated, with calculated breakthrough curves at a large (1000 m) distance from a source, the significance of matrix porosity and of sorption in the fracture as well as in the matrix. Grisak and Pickens (1980) conducted a sensitivity analysis, using a finite-element model, of the parameters involved in solute transport in a fracture with matrix diffusion. The parameters included fracture aperture size, water velocity in the fracture, matrix (diffusion) porosity, matrix distribution coefficient, and dispersivity. The finite-element model was applied to simulate a laboratory column tracer test conducted on a relatively undisturbed sample of fractured till. The laboratory tracer test results are discussed in Grisak et al. (1980). The breakthrough data were adequately simulated by using a water velocity for a representative fracture calculated from an effective fracture spacing in the sample and effective aperture width.

The purpose of this paper is to present an analytical solution for describing solute transport in fractured media where the solute diffuses from the fracture into the adjacent matrix. The solution is for nonreactive transport in both the fracture and the matrix. The governing conditions include one-dimensional advective transport in the fracture and one-dimensional diffusive transport (normal to the fracture) into the matrix. The utility of the solution is illustrated by applying it to the laboratory data of Grisak et al. (1980) and by illustrating hypothetical cases of transport along a single fracture for large temporal and spatial scales.

CONCEPTUAL FRAMEWORK AND ANALYTICAL SOLUTION

The development presented below, for solute transport in fractured geologic media, is based on the conceptualizations described by Carslaw and Jaeger (1959, pp. 391—396) for a heat exchanger and by Jost (1960, pp. 55 and 56) for surface treatment of metals. Carslaw and Jaeger (1959) describe a case in which a uniformly flowing fluid is in contact with a solid wall. Heat is transferred from one to the other across the fluid—wall interface. Jost (1960) describes a case in which a streaming gas, containing a component

NOTATION

List of symbols

b	fracture half-aperture width
C	concentration
C_f	concentration in the fracture
C_m	concentration in the matrix
C_0	input concentration
C/C_0	relative concentration
D^*	effective molecular diffusion coefficient of the solute in the porous matrix
erfc	complementary error function
t	time
V	water velocity in the fracture
x	direction of the fracture
y	direction normal to the fracture
∂	partial derivative
θ_m	matrix porosity

which is to diffuse into a metal, is passed over the metal surface. The concentration of the component in the streaming gas decreases as a result of diffusion into the metal.

Fig. 1 schematically illustrates the conditions on which the mathematical derivation is based. See also the Notation for symbols used in this paper. Groundwater is considered to be flowing at a constant velocity V in the x-direction in a planar fracture having a half-aperture width b. Solute is introduced to the fracture at $x = 0$ at a constant concentration C_0. The solute is transported in the fracture by advection only and into the matrix in the y-direction by diffusion. The fracture water is considered well mixed so that the concentration is constant in the fracture in any plane in the y-direction perpendicular to the direction of flow. The depth to which the solute penetrates into the matrix is assumed to be small compared to the thickness of the matrix; this implies that diffusion in the x-direction in the matrix is negligible compared with that in the y-direction. The solute con-

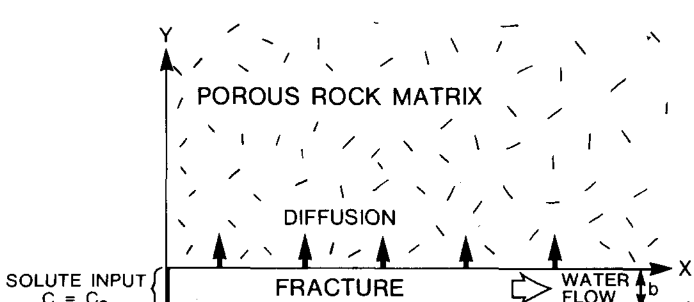

Fig. 1. Schematic illustration of solute transport in a fracture of half-aperture width b with a constant source input concentration C_0. Solute diffuses from the fracture into the matrix.

centration (mass per unit volume of liquid) in the fracture C_f and in the matrix C_m are assumed to be in equilibrium at the fracture—matrix interface.

The flux of solute from the fracture into the matrix is controlled by the porosity of the matrix θ_m, the effective molecular diffusion coefficient D^* of the solute in the porous matrix and the concentration gradient into the matrix. The governing equations for solute transport in the matrix and in the fracture, respectively, become:

$$\frac{\partial C_m}{\partial t} = D^* \frac{\partial^2 C_m}{\partial y^2} \quad \text{and} \quad b\left(\frac{\partial C_f}{\partial t} + V\frac{\partial C_f}{\partial x}\right) = \theta_m D^* \frac{\partial C_m}{\partial y} \qquad (1), (2)$$

where t is time.

The initial and boundary conditions for the present study are written:

$$C_f(x,y) = C_m(x,y) = 0, \qquad t = 0 \qquad (3)$$

$$C_f(0,0) = C_m(0,0) = C_0, \qquad t > 0 \qquad (4)$$

$$C_f(x,0) = C_m(x,0), \qquad x > 0; \quad t > 0 \qquad (5)$$

The initial concentration in the matrix and the fracture is zero. The water entering the fracture at $x = 0$ is of constant concentration C_0. The concentrations C_f and C_m are assumed to be equal at the fracture—matrix interface ($y = 0$). The matrix is assumed to extend to infinity in the region $y > 0$.

The solution for the concentration in the matrix is given by [adapted from Carslaw and Jaegar (1959, p. 396) and Jost (1960, p. 56)]:

$$\frac{C_m}{C_0} = \begin{cases} \operatorname{erfc}\left(\dfrac{(\theta_m D^*/Vb)\,x + y}{2[D^*(t - x/V)]^{\frac{1}{2}}}\right), & t > x/V \qquad (6) \\[2ex] 0, & t < x/V \qquad (7) \end{cases}$$

The concentration C_f in the fracture is evaluated from eq. 6 by setting $y = 0$. This is written:

$$\frac{C_f}{C_0} = \begin{cases} \operatorname{erfc}\left(\dfrac{(\theta_m D^*/Vb)\,x}{2[D^*(t - x/V)]^{\frac{1}{2}}}\right), & t > x/V \qquad (8) \\[2ex] 0, & t < x/V \qquad (9) \end{cases}$$

For calculations where time is variable, the solutions are therefore restricted in the x-direction to distances less than the piston-flow distance; if distance is variable, the solution is restricted to times greater than the piston flow time.

The analytical solutions given by eqs. 6 and 8 exhibit certain advantages over numerical solutions including their ability to handle extremes of spatial and temporal scales, their lack of susceptibility to numerical dispersion problems and their economy and utility since they can be evaluated using a calculator and a table of complementary error function values. However, the analytical solutions are limited to constant values of the system parameters (homogeneous systems) and the assumptions and initial conditions must be carefully considered before drawing conclusions.

APPLICATIONS

Laboratory

Grisak et al. (1980) presented a laboratory study of a chloride and calcium tracer test conducted with a large, relatively undisturbed cylindrical column of fractured till. The column diameter and length were 0.67 and 0.76 m, respectively. There were two sets of orthogonal fractures oriented vertically in the column parallel to the long axis. The fracture spacing in each set was ~4 cm. The test was conducted by passing the tracer solution vertically through the column and measuring the tracer breakthrough in each of four effluent quadrants at the outlet end of the apparatus. A composite breakthrough curve for chloride and calcium was calculated and the breakthrough curves were analyzed with a finite-element numerical model. The model simulated an "equivalent" fracture and matrix which were considered to be representative of the column conditions.

The chloride breakthrough data for the column effluent is shown in Fig. 2. Grisak et al. (1980) numerically simulated the data, using the following parameters: velocity in the fracture $V = 29.7$ m/day; half-aperture size $b = 20\,\mu$m; longitudinal dispersivity $\alpha_L = 0.04$ m; matrix (diffusion) porosity $\theta_m = 0.35$; and solute diffusion coefficient D^* in the matrix $= 5.0 \cdot 10^{-7}$ cm^2/s. The data were simulated with the analytical solution given by eq. 8,

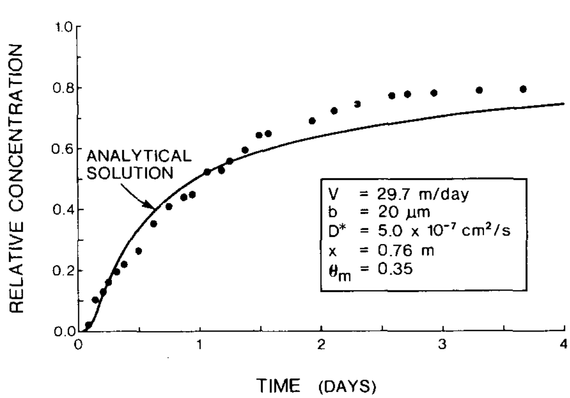

Fig. 2. Chloride breakthrough (effluent) data and analytical solution results for column of fractured till. The piston flow time is ~0.02 days.

using the parameters listed above with the exception of dispersivity. The analytical solution assumes zero longitudinal dispersion in the fracture. The results of the simulation using eq. 8 and the laboratory-measured effluent data are plotted on Fig. 2. It can be seen, from the relatively good data fit shown in Fig. 2, that the assumption of negligible dispersion in the fracture is a reasonable approximation in this case.

Deep disposal: long-range transport

The analysis of contaminant transport over large distances from deep disposal zones is often conducted in the context of geologic disposal of radioactive or toxic wastes. The solutions given by eqs. 6 and 8 can provide the basis for a parameter sensitivity analysis for long-range transport in a single idealized fracture. An example of such an analysis is presented below based on the simple conceptualization shown in Fig. 3. It is assumed that a contaminant source is located in a geologic formation in the vicinity of a discrete continuous fracture having an aperture of 100 μm, and extending 4000 m from the disposal zone to the exit point at ground surface. The groundwater velocity in the fracture is assumed to be a constant 100 m/yr., which provides a piston-flow rate of transport of 40 yr. from disposal zone to ground surface.

Fig. 4 illustrates the effect of the matrix porosity on breakthrough curves at ground surface for a nonreactive contaminant introduced continuously to the fracture at the disposal zone. The breakthrough curves are calculated using eq. 8. The solute diffusion coefficient in the matrix is assigned a value of 10^{-6} cm^2/s which is considered to be an average within the range of diffusion coefficients in geologic media. The matrix porosities for the three calculated breakthrough curves are chosen to represent a range of matrix porosities in geologic deposits. A matrix porosity of 0.003 is likely the lower limit of granitic or other crystalline rocks; a matrix porosity of 0.03 could apply to a cemented sandstone/siltstone or a welded tuff (ignimbrite); and a matrix porosity of 0.3 could be considered representative of larger porosity materials such as shales or clay.

Significant delay in contaminant breakthrough at the exit point is shown

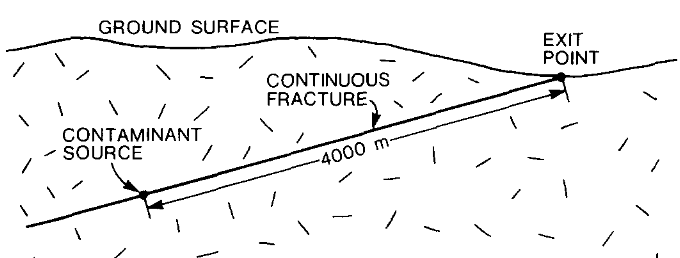

Fig. 3. Schematic illustration of contaminant transport from a source in a deep fractured geologic formation to an exit point at ground surface at 4000 m distance.

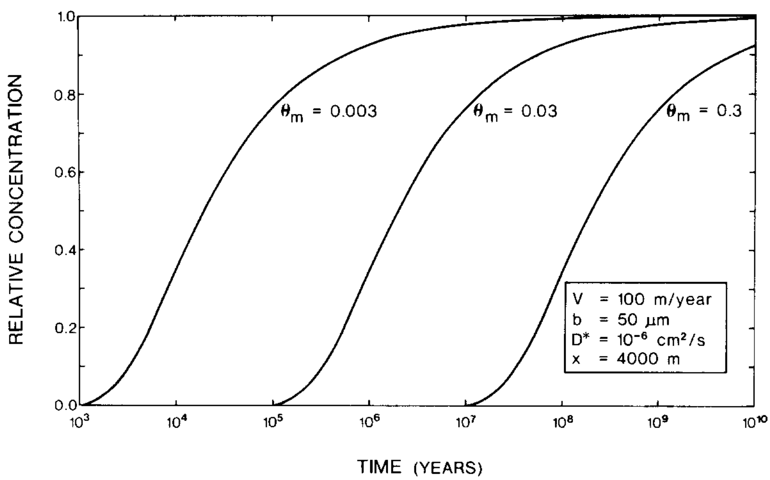

Fig. 4. The effect of matrix porosity θ_m on contaminant breakthrough curves at the exit point (4000 m).

to be provided by even granitic or crystalline rocks with low matrix porosities (0.003). The arrival of the average concentration ($0.5\,C_0$) at $\sim 2 \cdot 10^4$ yr. illustrates that the average contaminant transport velocity in the fracture is 0.2% of the piston-flow velocity. Earliest arrivals (for example, $0.001\,C_0$) of contaminants are seen at ~800 yr. or 5% of the flow velocity. Over 99% of the total mass that has entered the fracture is in storage in the matrix after 800 yr.

The ratios of the time of first arrival and average arrival to the piston-flow time at the exit point are given in Table I for matrix porosities of 0.003, 0.03 and 0.3. It can be seen from Table I that matrix diffusion may provide increasingly significant contaminant retardation, under the illustrated condition, for rocks with larger matrix porosities.

Fig. 5 illustrates the contaminant concentration profiles along the fracture with an adjacent matrix porosity of 0.003 at various times under the same conditions given for Fig. 4. The curves are calculated for a total flow path length of 4000 m. They do not extend beyond the time or distance restric-

TABLE I

Effect of matrix porosity on the travel time of the contaminant to the exit point

Matrix porosity	Ratio of first ($0.001\,C_0$) and average ($0.5\,C_0$) arrival time to piston-flow arrival time	
	first arrival	average arrival
0.003	$2 \cdot 10$	$5 \cdot 10^2$
0.03	$2 \cdot 10^3$	$5 \cdot 10^4$
0.3	$2 \cdot 10^5$	$5 \cdot 10^6$

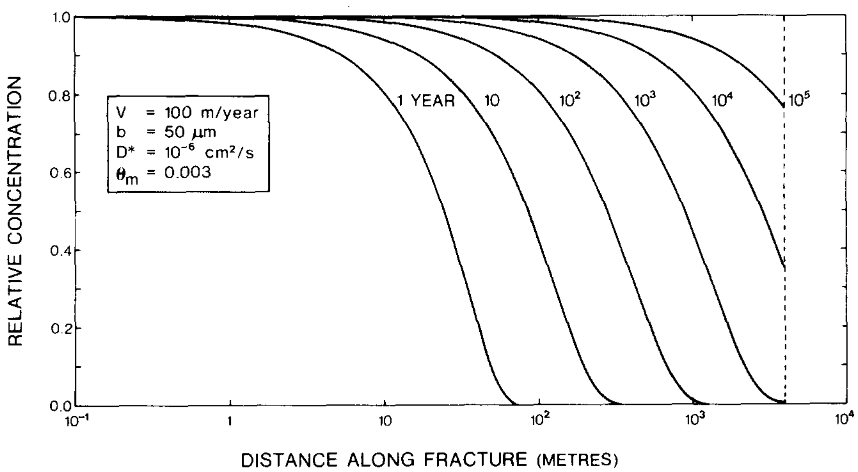

Fig. 5. Concentration profiles along the fracture between the source and the exit point (4000 m) at various times.

tions given in eqs. 6 and 8. The significant retardation that can be provided by even the low matrix porosity of 0.003 is well illustrated. It should be recalled that Figs. 4 and 5 are subject to the assumptions which include no dispersion in the fracture and an infinite matrix in the y-direction. With longitudinal dispersion in the fracture, the first arrivals would be earlier while the transition zone between first arrival and full concentration (for example, 0.999 C_0) would be wider.

The assumption of an infinite matrix in the y-direction can be somewhat restrictive, since fracture sets in natural geologic media can show spacings of the order of a metre or less. However, a fracture with an aperture of 100 μm can be considered a major feature in the analysis of groundwater flow and it may be a relatively large distance between fractures of this size. In order to provide some idea of the depth of penetration in the y-direction of the diffusion front and to evaluate the possible influence of the matrix thickness assumption, concentration profiles into the matrix were calculated with eq. 6 at x-distances of 0, 100 and 1000 m from the source at a time of 1000 yr. and matrix porosity of 0.003 (Fig. 6). The diffusion profile (for $C/C_0 \geqslant 0.001$) extends as far as 8 m into the matrix at the source. Although the diffusion profiles are very much dependent on the selected parameters, it can be seen from inspection of Fig. 6 that if the spacing of major features is of the order of tens of metres, the matrix thickness assumption can be considered reasonable. The effect of fracture spacing on breakthrough curves is discussed in detail in Grisak and Pickens (1980). They utilize a boundary condition of $\partial C_m / \partial y = 0$ midway between the fractures.

The effect of the magnitude of the diffusion coefficient D^* on the breakthrough curves is shown in Fig. 7. The illustrated range of diffusion coefficients is meant to be representative of the likely range in geologic media.

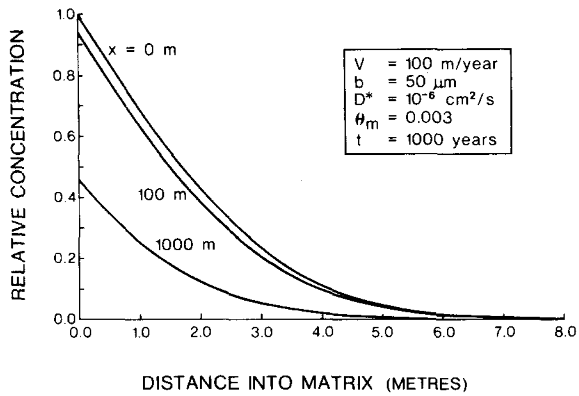

Fig. 6. Concentration profiles from the fracture into the matrix at x-distances of 0, 100 and 1000 m along the fracture.

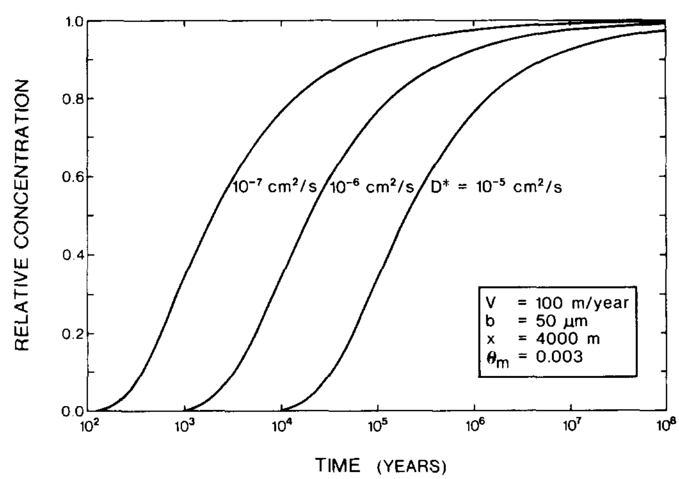

Fig. 7. The effect of the magnitude of the solute diffusion coefficient D^* in the porous matrix on the breakthrough curves at the exit point (4000 m).

The lower limit of the magnitude of D^* is unclear since there are very few published data concerning D^* in crystalline rocks or highly compacted clays and silts. The upper limit of D^* is clearly the free-water molecular diffusion coefficient; generally of the order of 10^{-5} cm^2/s for most solutes.

SUMMARY AND DISCUSSION

An analytical solution is presented for one-dimensional advective transport of solute in a planar fracture with diffusion into the porous matrix adjacent to the fracture. Although the assumptions of no dispersion in the fracture and large matrix thickness may be somewhat limiting in some situations, the solution is useful and cost effective for conducting sensitivity analyses of the effects on solute transport of water velocity, fracture aperture, matrix porosity and solute diffusion coefficients.

The use of the solution and the effect of matrix diffusion is illustrated by simulation of the effluent breakthrough curve data from a laboratory study of chloride transport through a large, relatively undisturbed column of fractured till. Long-range transport through a single discrete fracture is illustrated with a hypothetical example in the context of deep geologic disposal of radioactive or toxic wastes. The range of diffusion or matrix porosities considered is between 0.003 and 0.3, representative of the range typical of most geologic media. For a fracture water velocity, aperture and matrix diffusion coefficient of 100 m/yr., 100 μm and 10^{-6} cm^2/s, respectively, matrix diffusion reduces the average contaminant velocity to an exit point at 4000 m to ~0.2% of the water velocity for media with even the least porous matrix (e.g., granite). The retardation effect of the diffusion of the contaminant from the fracture into the matrix has important implications with regard to contaminant releases from the deep disposal or storage of toxic wastes in rock formations. Further retardation in the transport rate of some contaminants can be provided by decay (in the case of radioactive contaminants), by sorption of the contaminant onto the fracture surfaces resulting in a reduction in the effective value of V and by sorption of the contaminant within the matrix (reactive diffusion) resulting in a higher concentration gradient, a larger mass flux and a smaller penetration distance into the matrix.

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