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ABSTRACT

The most important mechanisms that control the release of contaminants from a repository for nuclear or chemical waste have been studied. For the time scale of interest for the disposal of nuclear and even chemical waste, diffusion into the rock matrix is an important factor which retards and dilutes the contaminants. It is found that the water flow-rate distribution and the flow-wetted surface are the entities that primarily determine the solute transport. If the diffusion in to the rock matrix is negligible, the solute transport is determined by the water flow rate and the flow porosity. This is shown by simulations using analytical solutions obtained for simple geometries, such as the flow in a fracture or a channel. Similar results are obtained for more complex systems, such as flow in a fracture with variable aperture and through a network of channels. It is also found that the use of a retardation factor relating the travel times of interacting and noninteracting solutes is inappropriate and may be misleading.

ABSTRACT (Swedish)

De viktigaste mekanismerna som kontrollerar utsläpp från ett förvar för använt kärnbränsle eller kemiskt avfall har studerats. För den tidskala som är av intresse för ett förvar av kärnavfall och även kemiskt avfall, är diffusionen in i bergmatrisen en viktig faktor som orsakar retardation och utspädning av de förorenande ämnena. Flödesfördelningen och den flödesvätta ytan är de primära storheterna som bestämmer transporten av de lösta ämnena. Om matrisdiffusionen är försumbar så är flödesfördelningen och flödesporositeten de avgörande storheterna för transporten av de lösta ämnena. Detta har visats genom att använda analytiska lösningar för att simulera transporten genom de enkla geometrier som en spricka eller en kanal utgör. Liknande resultat har erhållits från transportsimuleringar i mer komplexa system, som flöde i en spricka med variabel apertur och ett nätverk av kanaler. Användandet av en retardationsfaktor för att relatera transporttiden för de ämnen som diffunderar in i matrisen och sorberar, med de ämnen som inte gör det har funnits ej vara ändamålsenligt och kan därigenom vara missledande.

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SUMMARY

The most important mechanisms that control the release and transport of contaminants from a repository for nuclear or chemical waste is studied. In performance assessment of a repository, we are interested in the release of contaminants over long or very long times. In these circumstances, solutes have time to diffuse into rock, to be sorbed within it and eventually to decay.

Different transport mechanisms in fractured media are reviewed and some calculations are then presented for some simple geometries (flow in a channel in a fracture). The calculations showed that for solutes that do not interact with the matrix, the residence time is determined by the flow porosity and the water flow rate. For solutes that do interact with the matrix, however, the travel time is determined mainly by the flow-wetted surface since matrix diffusion and sorption effects are the determining mechanisms. Since the spreading of a solute is caused by the different velocities with which the solute travels through a medium, the dispersion is determined by the same entities as the mean residence time.

The fact that the transport of solutes in a fractured medium is determined by different mechanisms depending on whether or not the solute interacts with the matrix has important implications when tracer tests are performed to obtain data for models to be used in predictions of solute transport for performance assessment. The main information obtained in tracer tests with noninteracting species, the flow porosity, has no influence on the prediction of solute transport over large distances and on long time scales.

Solute transport models for more realistic cases are also presented. They include calculations of solute transport in a fracture with variable aperture and in a three-dimensional channel network.

When the transport times for sorbing and nonsorbing tracers are compared, a retardation factor is frequently calculated. This retardation factor cannot however be used to compare other tracer tests performed over the same distance, e.g., when the water velocity is changed. The retardation factor therefore has no practical use. Moreover, it may be misleading to use the retardation factor to extrapolate results from tracer tests to longer distances or longer times.

1 INTRODUCTION

The transport of contaminants in the ground is an important issue in the disposal of dangerous waste, e.g. chemical and nuclear wastes. Species escaping from the disposal site will be transported by the flowing groundwater to the biosphere. During the transport, the species may interact with the rock and be sorbed on the fracture surfaces. They may also diffuse into the stagnant water in the pores of the rock and be sorbed onto the inner pore surfaces.

If the contaminants do not interact with the rock and do not significantly diffuse into the rock matrix, they will travel at the velocity of the water. Due to heterogeneities in the system, water moves with different velocities and the contaminant velocity is dispersed around the mean. Diffusion into the rock is not important if the contact time between rock and contaminants is too short or if the diffusion coefficients are too small. On the time scale of interest for the disposal of nuclear and chemical waste, diffusion into the rock matrix is often an important factor in the transport of contaminants (Neretnieks, 1980).

For solutes that interact with the matrix by diffusion and/or sorption, the residence time of the solutes is strongly influenced by these mechanisms. In performance assessment of a repository, we are interested in the release of contaminants over long or very long times. In these circumstances, solutes have time to diffuse into rock, to be sorbed within it and eventually to decay. Other contaminants may be strongly retarded, diluted and dispersed in time. Due to the long time scale involved, no experimental tests can be performed over the distances of interest. Predictive modelling is commonly used to simulate the release of such contaminants. Among other means to gain confidence in simulation results, natural analogues have been used to validate these models.

The use of transport models requires a number of parameter values. Some of them may be determined in the laboratory, e.g. the sorption capacity of the rock, the effective diffusion coefficient, and the matrix porosity. Other parameters may be determined by field measurements, e.g., hydraulic conductivity and hydraulic gradient. Finally, specially designed experiments can be used to determine flow porosity, water flow distribution and flow-wetted surface (Moreno and Neretnieks, 1993). At present, tracer tests with nonsorbing (noninteracting) and sorbing (interacting) species are carried out to determine some of these parameters. These experiments are time-consuming and costly. For these reasons, it is of vital importance to determine which parameters are the most important for the release of contaminants for repository conditions, e.g., long time scales and usually low water flow rates.

The aim of this paper is to discuss the most important mechanisms that control the release and transport of contaminants from a repository for nuclear or chemical waste. Some of these concepts have been addressed by Neretnieks in Olsson et al., (1995). First, the different transport mechanisms in fractured media are reviewed. Some calculations are then presented for some simple geometries (flow in a channel in a fracture). Solute transport models for more realistic cases are then presented. We present calculations of solute transport in a fracture with variable aperture (Moreno et al., 1988) and in a three-dimensional channel network (Moreno and Neretnieks, 1993). The paper addresses the flow and solute transport in fracture media, but the results may in many cases be applied to porous media. The extension to porous media is indicated.

2 CONCEPTS, MECHANISMS, AND MODELS FOR FLOW AND TRANSPORT IN FRACTURED ROCK

One of the simplest transport models is based on the assumption that the dissolved species are carried by the water with the average velocity of the water and that the variation in the rate of species transport around the average velocity is a random process similar to molecular diffusion. The concentration of a species in the water is then described by the so-called advection-dispersion equation:

$$\frac{\partial c}{\partial t} + u \frac{\partial c}{\partial z} = D_L \frac{\partial^2 c}{\partial z^2} \qquad (1)$$

where c is the concentration in the liquid, t the time, and z the distance in the flow direction. The water velocity is denoted by u and the dispersion coefficient by D_L.

Equation (1) can be used to calculate how species that do not interact with the solid material will move with the water. Decay is not accounted for in Equation (1), but is easily added when needed.

2.1 SORPTION ON SURFACES

When a dissolved species is sorbed onto the fracture surfaces it will be retarded in relation to the water. For a linear instantaneous equilibrium, Equation (1) is modified to become:

$$\frac{\partial c}{\partial t} + K_a \frac{a}{\varepsilon_f} \frac{\partial c}{\partial t} + u \frac{\partial c}{\partial z} = D_L \frac{\partial^2 c}{\partial z^2} \qquad (2)$$

where K_a is the surface sorption coefficient, a the specific area, and ε_f the flow porosity. The specific area is defined as the surface area per volume of rock. For a medium with parallel fractures, the specific area is expressed as $a = 2/S$, and the flow porosity as $\varepsilon_f = \delta/S$, where S is the average spacing between fractures and δ the fracture aperture.

For a given rock with known flow rate per cross section (Darcy velocity, u_O), the velocity "u_{water}" in the fractures is inversely proportional to the fracture aperture, assuming that the number of fractures is constant:

$$u_{water} = \frac{u_O S}{\delta} \qquad (3)$$

The velocity for a solute that is reversibly sorbed onto the fracture surface, may be calculated from Equation (2) as:

$$u_{solute} = \frac{u_{water}}{1 + K_a \frac{a}{\varepsilon_f}} \qquad (4)$$

Inserting Equation (3) and the definitions for specific area and porosity into (4) gives:

$$u_{solute} = \frac{u_O S}{\delta + 2 K_a} \qquad (5)$$

For many of the solutes of interest, especially radionuclides, the value of the surface sorption coefficient is much larger than the fracture aperture, $2K_a >> \delta$. Therefore, the velocity of the solute may be expressed as:

$$u_{solute} \approx \frac{u_O}{\frac{2}{S} K_a} \qquad (6)$$

The term $2/S$ is the flow-wetted surface per volume of rock. Since nonsorbing solutes are transported with the water, their velocity is determined by the water flux, the aperture and the spacing between fractures (Equation 3). For sorbing solutes, usually $K_a 2/\delta >> 1$, the solute velocity is therefore independent of the water velocity in the rock for a given flux (Darcy velocity). The solute velocity is then primarily determined by the flux, the surface sorption coefficient, and the flow-wetted surface in the rock. It may be noted that the flow porosity does

not, under these circumstances, influence the velocity of sorbing solutes for a given Darcy velocity and fracture spacing.

These concepts may be used directly for porous media. For example, for a medium formed by particles of diameter, d_p, the expressions for the water and solute velocities are (Neretnieks et al., 1987):

$$u_{water} = \frac{u_o}{\varepsilon_f} \tag{7}$$

$$u_{solute} = \frac{u_o}{\varepsilon_f + \frac{6(1-\varepsilon_f)}{d_p} K_a} \tag{8}$$

The water velocity is a function of the flux and flow porosity of the bed. For sorbing species with $K_a \gg d_p$, the solute velocity is determined by the water flux, the particle specific area, $6/d_p$, which is assumed to be accessible to water, and only marginally by the porosity.

2.2 MATRIX DIFFUSION

In the case of surface sorption, it is assumed that only the fracture surface in contact with the flowing water is available for sorption. In practice, it may be expected that the rock nearest to the fracture surface will be penetrated by the dissolved diffusing species and that the penetration depth will increase with increasing contact time. The rock volume, which may be expected to be accessed by diffusion during contact times of hundreds or more years, may have a considerably larger retardation effect than surface sorption alone may allow. On the other hand, the penetration depths for strongly sorbing solutes, such as plutonium and neptunium, are not expected to be more than a few tens of centimetres even for contact times of hundreds of thousands of years. Most of the rock matrix will not then be penetrated and will not help in retarding such solutes. The amount of rock accessed will depend on the sorption and diffusion properties of the matrix and on the solutes and also on the area of the surface area in contact with the mobile water from which the solutes may diffuse into the matrix. For radionuclide transport, it was shown by Neretnieks (1980) that the area of the wetted surface per volume of rock is one of the primary variables, whereas the flow porosity will, in practice, have a negligible influence on the velocity of most sorbing solutes.

The concentration of the effluent from a fracture will depend on the retardation of the solutes travelling through the fracture. The main retardation is due to sorption within the rock matrix. For illustration and the sake of simplicity, the hydrodynamic dispersion and sorption onto the fracture surface are neglected in this case. The impact of the dispersion on the solute transport of sorbing species can in most cases be neglected (Maloszewski and Zuber, 1985).

The effluent concentration from a single fracture has been modelled including the porous matrix (Neretnieks et al., 1982). The solute will diffuse into the micropores of the matrix and sorb onto the inner surfaces of the matrix. The differential equation describing the transport in the fracture is:

$$\frac{\partial c_f}{\partial t} + u_f \frac{\partial c_f}{\partial z} = \frac{2D_e}{\delta} \left(\frac{\partial c_p}{\partial x}\right)_{x=0} \tag{9}$$

where c_f is the concentration in the water in the fracture, u_f is the water velocity in the fracture, z is the distance in the direction of the flow, c_p is the concentration in the water in the rock pores, D_e is the effective diffusion coefficient in the rock, and x is the distance into rock from the fracture.

The diffusion in the rock matrix is given by:

$$\frac{\partial c_p}{\partial t} = \frac{D_e}{K_d \rho_p} \frac{\partial^2 c_p}{\partial x^2} \tag{10}$$

where $K_d \rho_p$ is the volume sorption capacity of the rock including the solutes in the pore water.

For a step injection at the fracture inlet and no solute in the fracture and rock at initial time, the solution to the above equations becomes (Carslaw and Jaeger, 1959):

$$\frac{c_f}{c_o} = \text{erfc}\left[\left(\frac{D_e K_d \rho_p}{t - t_{water}}\right)^{0.5} \frac{LW}{Q}\right] \tag{11}$$

where L is the length of the channel and W the width. The product 2LW is the flow-wetted surface area. This is an important entity because it is the surface in contact with the flowing water through which the solutes diffuse into and sorb in the rock matrix. For contact times longer than the water residence time, the water residence time has little influence on the solute transport. The most important entities to assess are: the water flow rate in the channel, Q, the flow-wetted surface area of the channel, 2LW, and the diffusion and sorption properties of the rock, $D_eK_d\rho_p$.

When the solute diffuses into the rock matrix, the mean solute travel time is not a useful entity with wich to describe the solute transport. The solute travel time is determined by all the accessible water volume and will spread over a very long time as illustrated in Figure 1. For this reason we will use the time to reach a given concentration at the outlet equal to 0.5 times the concentration at the inlet. Since erfc(0.48) is equal to 0.5, the time to reach half of the inlet concentration may be written as:

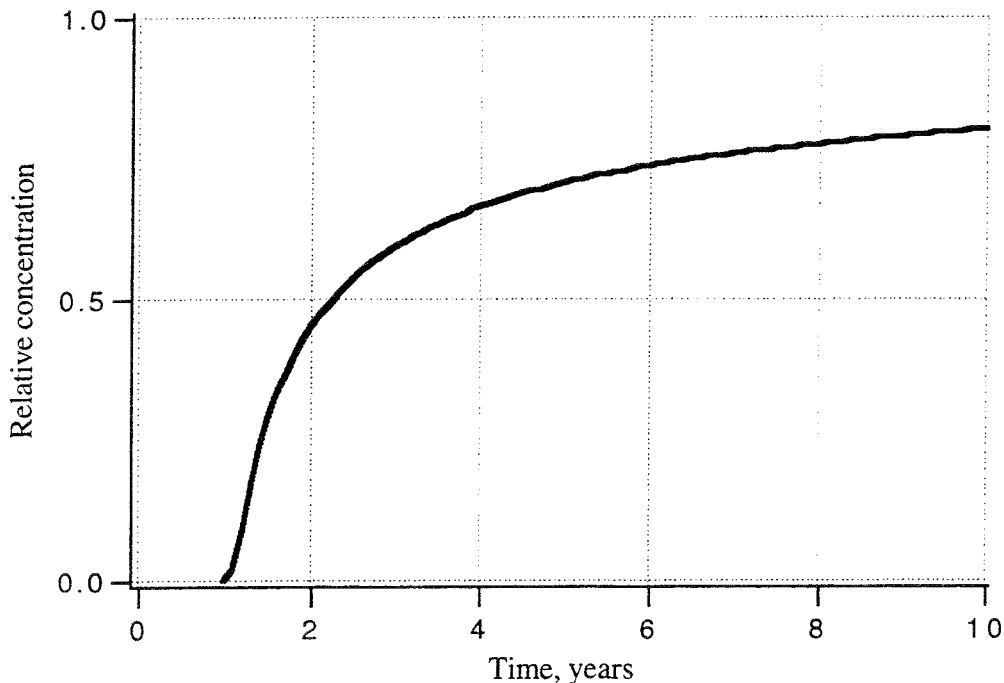$$t_{50} = t_{water} + \frac{(D_eK_d\rho_p)}{0.48^2}\left(\frac{LW}{Q}\right)^2 \tag{12}$$

Figure 1   Calculated breakthrough for a tracer that diffuses into the rock matrix, for a travel distance of 10 m with flow rate of 0.003 m^3/year and a width of 1 m. Here, D_e is $3.15\cdot10^{-6}$ m^2/year, $K_d\rho_p = 0.01$ and the aperture, $\delta = 0.0003$ m.

For the conditions existing in a repository for nuclear or chemical waste (long distance and small flow rate), the water travel time is negligible compared with the travel time for the solute, even for species that are not sorbed in the solid. Equation (12) shows that the solute transport time is determined by the diffusion and sorption, the flow-wetted surface, 2LW, and the flow rate. Since the water travel time is negligible, the impact of the flow porosity may be disregarded.

Equations (9) and (10), which describe the solute transport including matrix diffusion, are applicable if the fractures are sparse. In this case, the solute diffusing into the rock from two parallel fractures never meet each other. The penetration into the rock is determined by the diffusion and sorption in the rock. Nonsorbing species may diffuse long distances into the rock, while strongly sorbing species diffuse only a small distance into the rock, even over long times. The penetration may be defined for different levels of concentration, e.g., for the distance where the concentration in the pore water is 0.01 times the concentration at the surface (Neretnieks, 1980). This may be expressed as:

$$\eta_{0.01} = 4 \left(\frac{D_e}{K_d \rho_p} t \right)^{0.5} \tag{13}$$

where $\eta_{0.01}$ is the distance into the solid with a concentration in the water pore of 0.01 times the concentration at the surface.

Finally, as an extreme case, we may consider the situation where the fracture frequency is high so that all the rock between fractures is reached by the solute. In this case it is assumed that the solid matrix is equilibrated with the solute in the water in the fracture. The velocity for the sorbing solute then becomes, (Neretnieks et al., 1987):

$$u_{solute} = \frac{u_o S}{\delta + (S-\delta) K_d \rho_p} \tag{14}$$

In fractured media, even with a quite high fracture density, the fracture spacing is much larger than the aperture, $S >> \delta$ and for sorbing species Equation (14) becomes,

$$u_{solute} \approx \frac{u_o}{K_d \rho_p} \tag{15}$$

Again, the solute velocity is independent of the flow porosity of the medium.

2.3 DISPERSION

We have shown that for solutes that do not interact with the matrix, the residence time is determined by the flow porosity and the water flow rate. For solutes that do interact with the matrix, however, the travel time is determined mainly by the flow-wetted surface since matrix diffusion and sorption effects are the determining mechanisms. The spreading of a solute, dispersion, is caused by the different velocities with which the solute travels through of the medium. The dispersion may be expressed as a function of the standard deviation of the residence time distribution.

For noninteracting solutes, the water flow rate and the flow porosity determine the residence time. The dispersion of these solutes will, therefore, be a function of the spatial distribution of water flow rate and flow porosity. For solutes that interact with the matrix, the dispersion is determined by the water flow rate and the flow-wetted surface in addition to the diffusion and sorption properties of the rock. For this reason, very weak or in practice no correlation is to be expected between the dispersion of noninteracting solutes and the dispersion of interacting solutes. This implies that a dispersion coefficient determined by tracer experiments with noninteracting tracers cannot be used for solutes that interact with the rock matrix. This applies in situations where the transport time for interacting solutes is considerably longer than the water residence time. If the transport time is short, then the dispersion of the interacting species will also be influenced by the flow porosity.

The fact that the transport of solutes in a fractured medium is determined by different mechanisms depending on whether or not the solute interacts with the matrix has important implications when tracer tests are performed to obtain data for models to be used in predictions of solute transport for performance assessment. The main information obtained in tracer tests with noninteracting species, the flow porosity, has no influence on the prediction of solute transport over large distances and on long time scales.

3 SOME CALCULATIONS OF SOLUTE TRANSPORT IN FRACTURES

In the following, we use some simulations with simple cases to illustrate the above models and to indicate situations where both flow porosity and sorption effects may influence the residence time distribution of solutes. Transport velocities were obtained for the solutes sorbed on the fracture surface and for those that diffuse into the rock matrix, where they may also be sorbed. It was demonstrated that these expressions are independent of the fracture aperture for solutes where diffusion and sorption are important. For slightly sorbing solutes, the fracture aperture may have some importance. Some calculations for simple geometries are made to illustrate this effect.

For fractures where there is no diffusion into the matrix and where the solute may be sorbed on the surfaces, the solute velocity is given by Equation (6) if the sorption coefficient is much larger than the fracture aperture, $2K_a >> \delta$. Travel times have been calculated for transport distances of 10 m as a function of the surface sorption coefficient for several fracture apertures using Equation (5). A water flux, u_o, of 0.1 m^3/m^{2}year and a distance between fractures of 5 m are used in the calculations. Figure 2 shows the travel time as function of the surface sorption coefficient. The transport time for a given flux and a given fracture frequency is independent of the fracture aperture if the surface sorption coefficient is much larger than the fracture aperture.
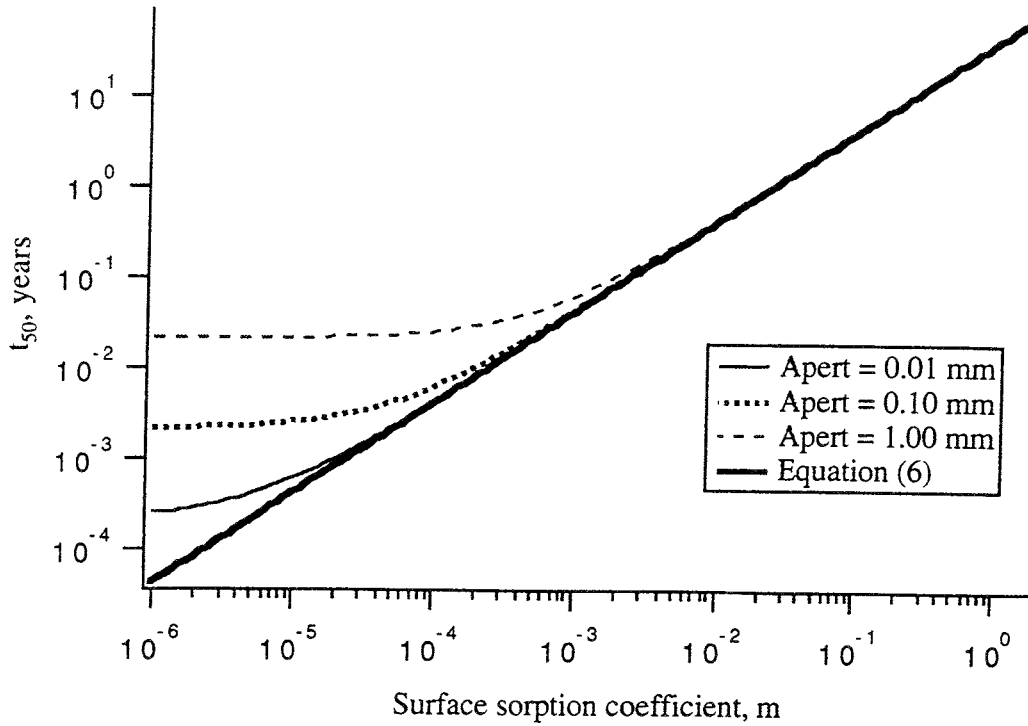
Figure 2 t_{50} as a function of the surface sorption coefficient. A flux of 0.1 $m^3/(m^2 year)$ and a distance between fractures of 5 m are used.

For solutes that diffuse into the rock matrix, the time to reach 50% of the inlet concentration, Equation (12), is used for comparison. For sorbing species, the travel time for the water is in general negligible compared with the travel time for the solute. This equation as a function of the flux and fracture density becomes:

$$t_{50} = \frac{(D_e K_d \rho_p)}{0.23} \left(\frac{L}{u_o S}\right)^2$$
(16)

Calculations have been made for solutes with different sorption properties using Equation (12). The same fracture system has been used in this case, and a value of the effective diffusion coefficient of $3.15 \cdot 10^{-5}$ $m^2/year$ has been assumed. The value of $K_d \rho_p$ is varied over a wide interval. For nonsorbing solutes, the value of $K_d \rho_p$ is equal to the matrix porosity, assumed to be 0.01. For a strongly sorbing solute, the value of $K_d \rho_p$ can be 10 000 and more. The transport time is influenced by the fracture aperture only for nonsorbing or weakly sorbing solutes. If the distances are longer or the flux is less, even the transport of nonsorbing solutes may become independent of the fracture aperture. Results are shown in Figure 3.
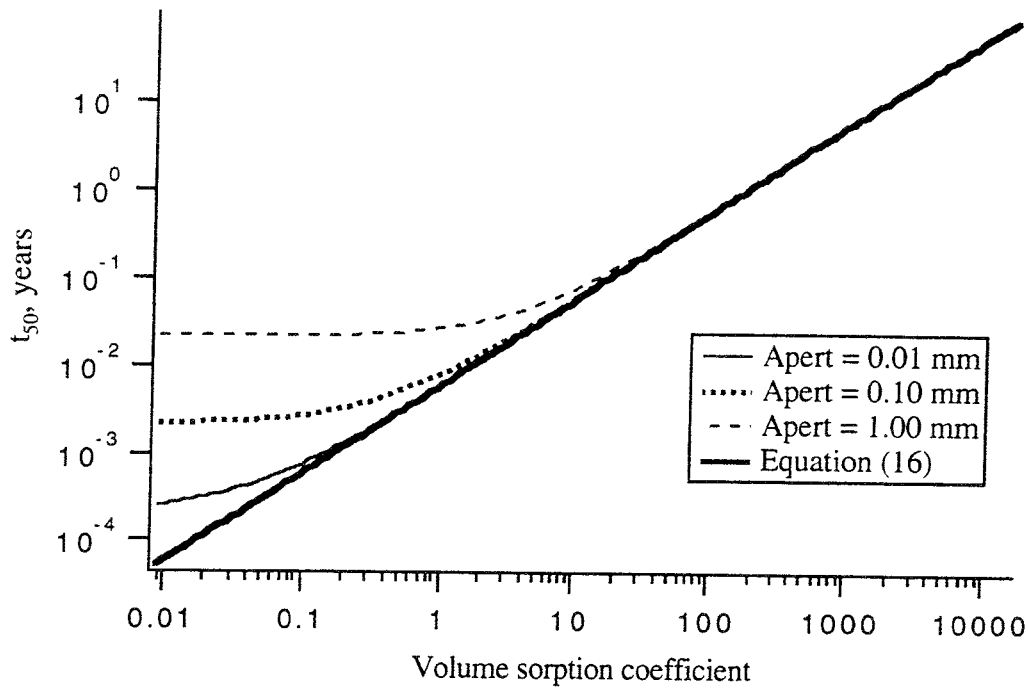
Figure 3 t_{50} as a function of the volume sorption coefficient. A flux of 0.1 $m^3/(m^2 year)$ and a distance between fractures of 5 m are used.

For high fracture frequencies, the penetration length into the matrix must be considered. If the penetration length is longer than the distance between fractures, then the equation for simple fractures cannot be used. In this case, mathematical solutions for solute transport through parallel fractures may be used (Sudicky and Frind, 1982).

4 SOLUTE TRANSPORT IN SYSTEMS WITH COMPLEX FLOW PATTERNS

In the previous sections, solute transport has been studied in simple systems such as fractures and channels. In these calculations, it was found that, for solutes that interact with the rock matrix, the transport is determined by the water flow rate and the flow-wetted surface. It is expected that these relationships may also be applied to more complex systems, for example, solute transport in a fracture with a varying aperture or in a channel network. Both models consider that the solutes diffuse into the rock matrix and may be sorbed on the surface of the microfissures in the rock. A review of both models is here presented. More details may be found elsewhere (Moreno et al., 1988; Moreno and Neretnieks, 1993).

4.1 FRACTURE WITH VARIABLE APERTURE

The fracture surfaces are rough, and the aperture is thus not constant but varies spatially. The spatial variation of the fracture aperture is characterized by a spatial correlation length. This means that, within a range smaller than the correlation length, the aperture values are more likely to be similar but that at separation distances much longer than the correlation length, there is little or no correlation between aperture values. In this model, the apertures are characterized by an aperture density distribution and a spatial correlation length.

The flow between two adjacent nodes is determined by the apertures of both nodes. The flow between square nodes i and j may be expressed as (Moreno et al., 1988):

$$Q_{ij} = \frac{1}{6\mu} \left[\frac{1}{\delta_i^3} + \frac{1}{\delta_j^3} \right]^{-1} (P_i - P_j) \tag{17}$$

where δ is the aperture, μ is the dynamic viscosity, and P is the pressure at the center of the node. Square nodes are considered. Using Equation (17), the mass balance at each node may then be written. The solution of this system of equations yields the pressure at each node. The flow rate between adjacent nodes may then be calculated using Equation (17).

The solute transport is simulated using a particle-following technique (Robinson, 1984; Moreno et al., 1990). A given number of particles is introduced into the flow field at the

injection node. The residence time distribution is obtained from the residence times of a large number of individual particle runs. From the residence time distribution (RTD), the mean residence time and variance can be calculated.

When diffusion from the moving water into and out of the rock matrix takes place, a particle may reside in the matrix for some time, in addition to its residence time in the water in the fracture. For a flat channel from which the diffusion is perpendicular to the channel surface, a simple analytical solution is available for the RTD. The cumulative curve, F, for the residence times is obtained (Carslaw and Jaeger, 1959) as:

$$F = erfc \left(\frac{(D_e \, K_d \, \rho_p)^{0.5} \, L \, W}{(t - t_w)^{0.5} \, Q} \right) \tag{18}$$

for times greater than the water-plug-flow residence time t_w. Otherwise the value is zero. Equation (18) considers only advection in the channel and diffusion into the rock matrix. Longitudinal dispersion is neglected. For particle following, we use the same technique as that used by Yamashita and Kimura (1990). The travel time for each particle in a channel member is determined by choosing a uniform random number in the interval [0,1]. The travel time for the particle, t, is then calculated by solving for t in Equation (19):

$$[R]_0^1 = erfc \left(\frac{(D_e \, K_d \, \rho_p)^{0.5} \, L \, W}{(t - t_w)^{0.5} \, Q} \right) \tag{19}$$

4.2 CHANNEL NETWORK MODEL

In this model, it is assumed that the flow paths make up a three-dimensional channel network in the rock. All properties of the channel members used in the model are assumed to have a stochastic nature. The flow calculations need only the information on the conductances of the channel members and the boundary conditions. The conductance is defined as the flow in a channel member divided by the pressure difference between its ends. When solute transport is included, the volume of the member has to be known. An accurate estimation of the channel volume is important only for simulation of noninteracting solute transport. If sorption onto the fracture surface or diffusion into the matrix is to be included in the model, the area of the flow-wetted surface must also be included. Some properties of the rock are also needed, such as rock matrix porosity, diffusivity and sorption capacity for sorbing species.

In the present simulations, the conductances of the channel members are assumed to be lognormally distributed and not correlated in space. For simulations of noninteracting or weakly interacting solutes, the channel volume is estimated by assuming that the conductance of the channels is proportional to the cubed channel aperture, owing to the lack of data.

For laminar conditions, the flow through a channel member is proportional to the pressure gradient. The flow between two points "i" and "j" may be written as:

$$Q_{ij} = C_{ij}(P_j - P_i) \tag{20}$$

where C_{ij} is the conductance connecting the nodes "i" and "j." The pressure field is calculated by writing the mass balance at each intersection point. The solution of this system of equations yields the pressure at each node. Flow between adjacent nodes is then calculated using Equation (20).

The solute transport is simulated by using a particle-following technique.The diffusion into the rock matrix is determined using the procedure described for transport in a fracture. This is described in more detail in Moreno and Neretnieks (1993).

4.3 SIMULATIONS OF SOLUTE TRANSPORT IN A FRACTURE WITH VARIABLE APERTURE.

In the fracture with variable aperture, simulations were made using solutes that do not interact with the matrix and solutes that diffuse into the rock matrix. In the latter case, the solutes may also be sorbed within the rock matrix. In the simulations, water is pumped up from the centre of a fracture and a tracer is injected into one of eight holes located in a circle around the center of the fracture. The fracture is a square with six-metre sides and the distance between the point of injection and the withdrawal hole is 1.8 m. The boundary conditions are: a specified head at the four sides and given withdrawal and injection rates. The injection is carried out at one point at a time.

Two sets of simulations are presented. In the first, the influence of the sorption coefficient is studied with the mean aperture kept constant. In the second set of simulations, the effect of increasing the mean aperture is studied for various sorption values. In these simulations, the mean aperture of the fracture is chosen to be 0.1 and 1.0 mm and the standard deviation in the aperture distribution to be 1.2 (natural logarithm). A correlation length of 0.3 m is used. The pumping flow-rate used is 10 ml/h. This means that the mean residence time would be 101 h if the fracture had a constant aperture of 0.1 mm and the rock was impervious. As the fracture is created by a stochastic process, the results for a given fracture correspond to only one of an infinite number of possible realizations. Three different tracers were considered: a nonsorbing, a slightly sorbing ($K_d\rho_p = 1.0$) and a strongly sorbing species ($K_d\rho_p = 100$).

From the travel time distribution for the particles, the residence time and dispersion (Peclet number) are evaluated. Because the mean travel time and dispersion are strongly influenced by particles with a very long residence time, we have chosen another way to illustrate these entities. The time to reach 50% of the initial concentration, t_{50}, is used instead of the mean travel time. For the dispersion, the ratio between the time to increase the concentration from 10 to 90%, $(t_{90} - t_{10})$, and t_{50} is used (Neretnieks et al., 1982). We define this ratio as D_{159}.

In a first set of simulations, a fracture aperture of 0.1 mm was chosen and the sorption properties of the tracer were varied. Figure 4 shows the travel time, t_{50}, obtained for a nonsorbing species and the time, t_{50}, for a slightly sorbing species for the eight simulated tracer tests. A point in the plot represents injection in a given injection point in the same fracture realization, the time t_{50} for the nonsorbing tracer being on the abscissa and the t_{50} for the slightly sorbing tracer on the ordinate. A slight correlation is found for the tracer tests with a short travel time, but no correlation is found for the tests with a long travel time. The

relationship between the time t_{50} for the two sorbing species with $K_d\rho_p$ values of 1.0 and 100 is shown in Figure 5, where a clear correlation may be observed as expected. In Figures 6 and 7, the ratio D_{159}, which is a measurement of the dispersion, is shown for the same cases. Similar results are found, a good correlation in the case of the two sorbing species but only a slight or no correlation between the data for nonsorbing and sorbing species.

The lack of correlation between the travel times for a nonsorbing and sorbing species is because the residence time for a nonsorbing species with travel time of the order of hours is determined mainly by the flow rate distribution and the volume of the paths through which the water flows, whereas, for the sorbing species, the travel time is determined by the flow rate distribution and the flow-wetted surface. In the case of the two sorbing species, the travel time is in both cases determined by the same parameters, namely flow rate and flow-wetted surface, and a good correlation is found between its travel times. Similar results are found when the dispersion in the simulated tracer tests is compared. In summary, we found that the flow rate distribution is the only common factor in tracer tests with sorbing tracers and nonsorbing tracers.
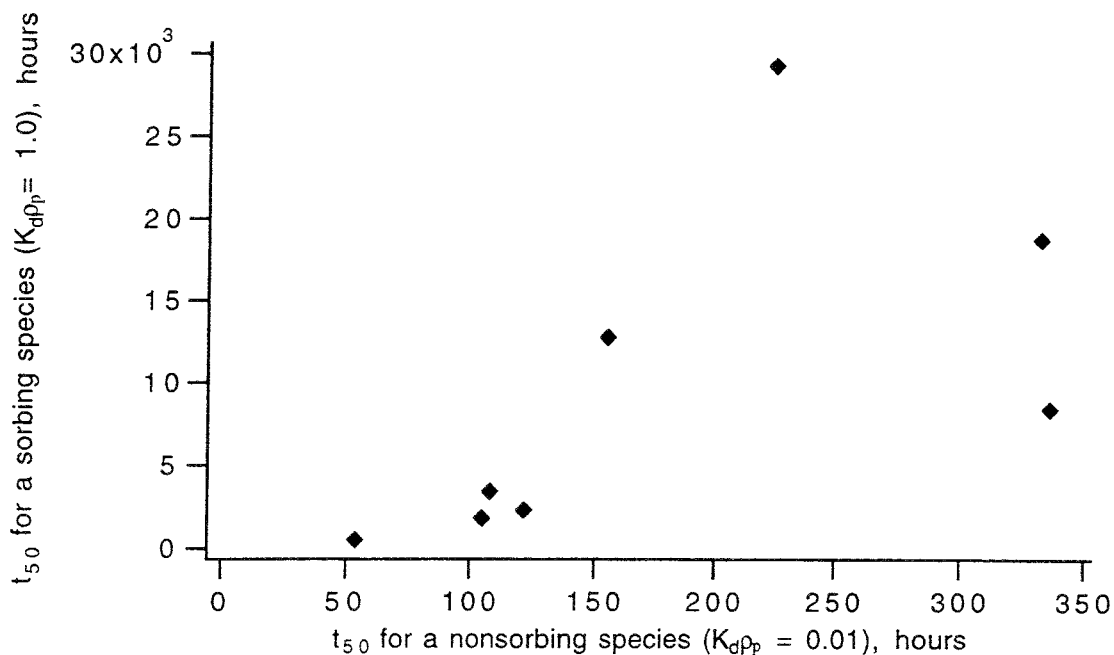

Figure 4 t_{50} for a nonsorbing species and a slightly sorbing species. Each point represents results from injection at one location.
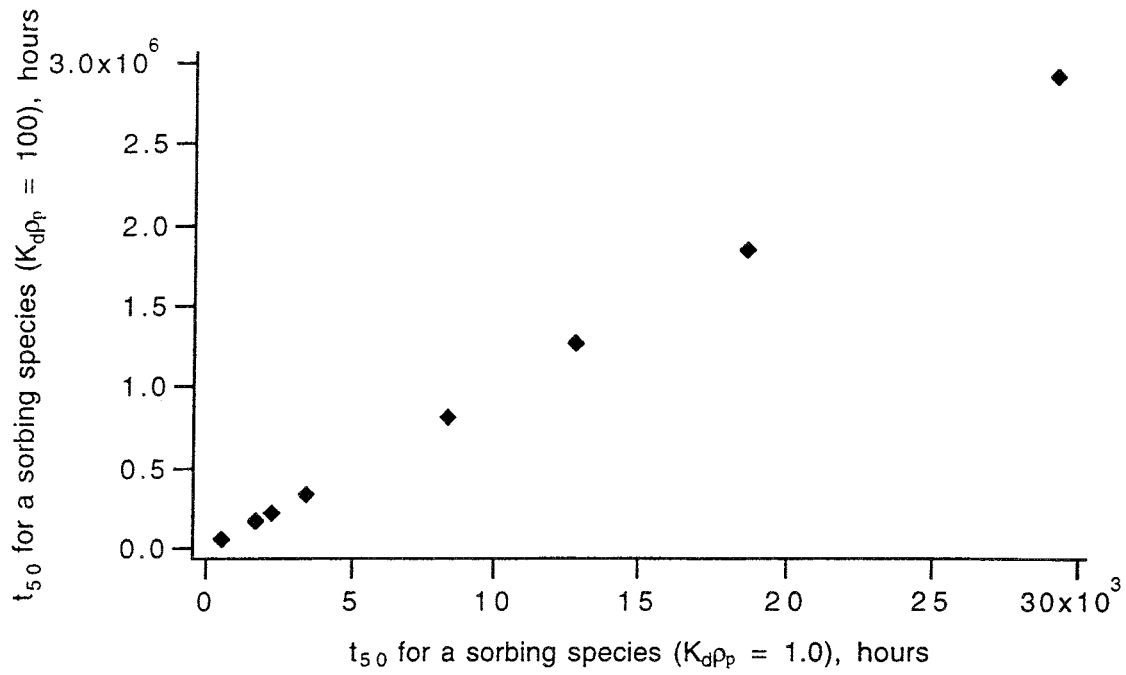
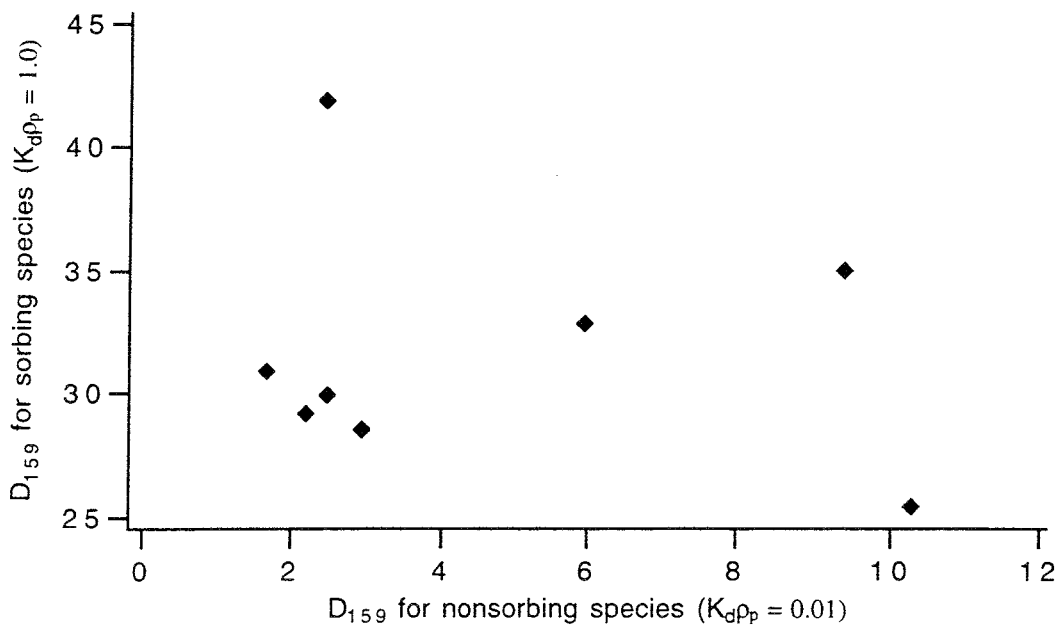Figure 5 t_{50} for two sorbing species, with $K_d\rho_p$ values of 1.0 and 100.

Figure 6   Dispersion, expressed by the ratio D_{159}, for a nonsorbing species and a slightly sorbing species.
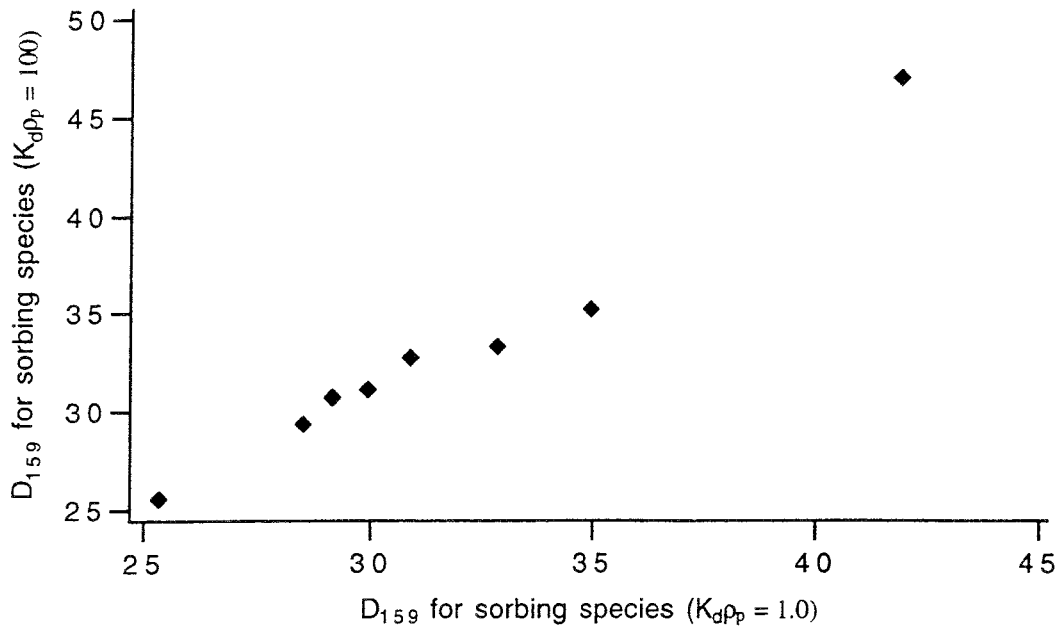
Figure 7   Dispersion, expressed by the ratio D_{159}, for two sorbing species, with $K_d\rho_p$ values of 1.0 and 100.

In a second set of simulations, a fracture aperture of 1.0 mm was used and the results were compared to the results for the first set using a fracture aperture of 0.1 mm. If diffusion into the matrix is not active, then the travel time should be increased by a factor 10 since the fracture volume was increased by a factor 10. The ratio between the travel times, t_{50}, for a fracture aperture of 0.1 mm and the travel times for a fracture of 1.0 mm is used to study the influence of the fracture volume on the travel time. These ratios are shown in Table 1 for different tracers. The results show that for a nonsorbing tracer, the ratio between travel times is close to the value of 10 (for an impervious matrix). This means that, in this case, the travel time is determined by the fracture volume. For slightly sorbing tracers, the ratio is about 1.5, which means that there is a small influence of the fracture aperture. For sorbing tracers with $K_d\rho_p$ = 100, the ratio is 1.0, which means that the fracture aperture has no influence on the travel time.

Table 1   Ratio between travel times, t_{50}, for fractures with apertures of 0.1 and 1.0 mm for the different injection locations.

Run	Nonsorbing	Sorbing $(K_d\rho_p = 1.0)$	Sorbing $(K_d\rho_p = 100.0)$
1	8.92	1.46	1.01
2	8.30	1.14	1.00
3	8.99	1.34	1.01
4	8.51	1.25	1.01
5	7.26	1.08	1.00
6	6.39	1.04	1.00
7	9.20	1.52	1.01
8	9.34	1.94	1.01

4.4   SIMULATIONS OF SOLUTE TRANSPORT IN A CHANNEL NETWORK

To test the influence of flow porosity in a more complex situation, solute transport through a network of channels has been studied. The tracer tests performed at the Swedish ÄSPÖ Hard Rock Laboratory and analysed using the Channel Network model (Gylling et al., 1994) are used in this discussion. First, one of the tracer tests with a nonsorbing tracer is evaluated and predictions are then made for sorbing species for different flow porosities.

The experiments were done by first establishing a constant flow field by pumping for two weeks. The pumping hole was 600 m deep. In other boreholes at distances ranging between 100 and 250 m tracers were injected. In the rock volume studied, 1000x700x700 m, there were 13 identified fracture zones. The zones are much more permeable than the rock and most of the water pumped is estimated to flow through the network of zones. The zones and the rock are both modelled by the Channel Network model. The channel frequency, conductivity distribution and flow-wetted surface are obtained by interpretations of hydraulic packer tests using the method described in Moreno and Neretnieks (1993). The obtained values may be summarized as follows. The flow wetted surface is 0.76 m^2/m^3 rock. The standard deviation of the channel conductance is 1.4 on the ^{10}log scale. These data were obtained from the packer

tests. As there are no observations on the channel widths or lengths we assumed for the network that L = 20 m. This may not be realistic but earlier studies (Gylling et al., 1994) have shown that as long as the flow wetted surface is kept the same, the breakthrough curves are not very sensitive to the choice of L and W.

In the main experiment, the "straight" distance from injection point to collection region is about 200 m. This tracer test was predicted using assumed flow porosities of 0.001 and 0.0001. No data are available on flow porosities at this site. The experimental result and the predicted curves are shown in Figure 8, and these show a reasonable agreement considering that no adjustable parameters were used and that some of the data used are estimated and are not site-specific.
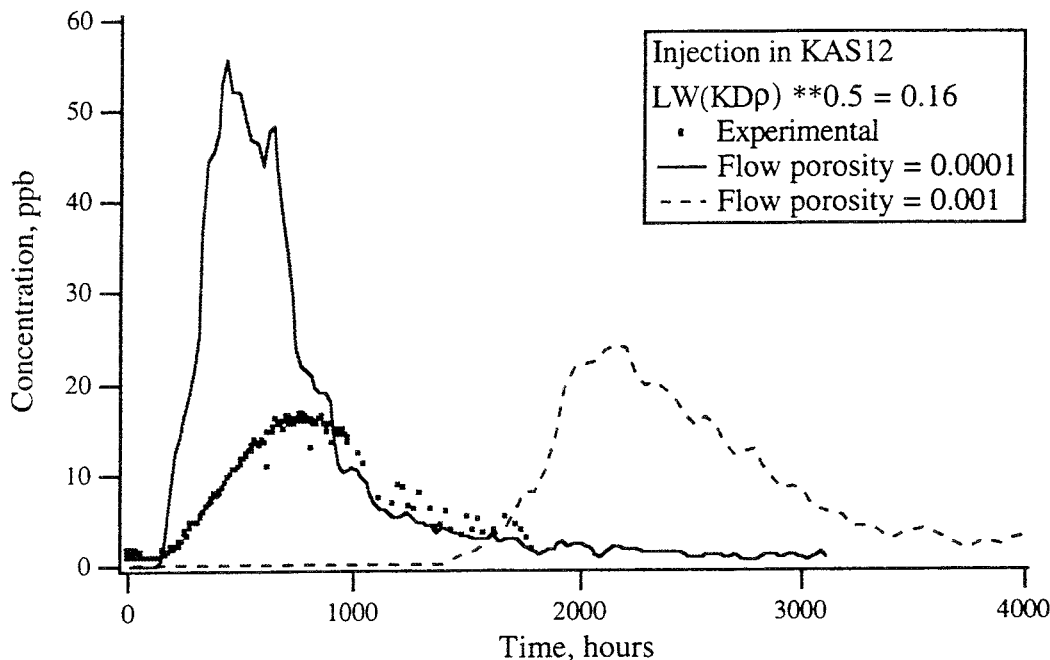

Figure 8   Experimental breakthrough curve for the tracer test with injection in KAS12 and the simulated curves for flow porosities of 0.001 and 0.0001.

From Figure 8 it is seen that the flow porosity should be somewhat larger than 0.0001. In some other travel paths with other tracers the predictions showed much higher recovery than the experimental. This indicates that there either was more flow wetted surface that made the tracers better enter the rock matrix or that there was a higher flow porosity.

In the following simulations with sorbing tracers, the value 0.0001 of the flow porosity is used in the prediction of migration of solutes that are sorbed in the rock matrix. Two values for the

sorption coefficient are used, $K_d\rho_p$ = 1.0 and 100, for the same injection point, water flow and travel distance as in the tests with nonsorbing species. For a flow porosity of 0.0001, the times t_{50} are about 5 and 500 years. For the nonsorbing species, this time is about 400 hours or 0.05 year. Simulations have also been carried out for values of porosities covering a wide interval. The results are shown in Figure 9. The time t_{50} for the injected tracer is plotted versus flow porosity, from a very small value of 10^{-5} to a very large value of 0.1. For the nonsorbing tracer the time is a function of the flow porosity. For large porosities, the relationship is almost linear. For weakly sorbing species, where $K_d\rho_p$ = 1.0, the dependence on the porosity is small and only begins to matter for porosities larger than about 0.01. For strongly sorbing tracers, $K_d\rho_p$ = 100, the travel time is independent of the flow porosity. The same arrival time is obtained over the whole range of flow porosities used in these simulations.
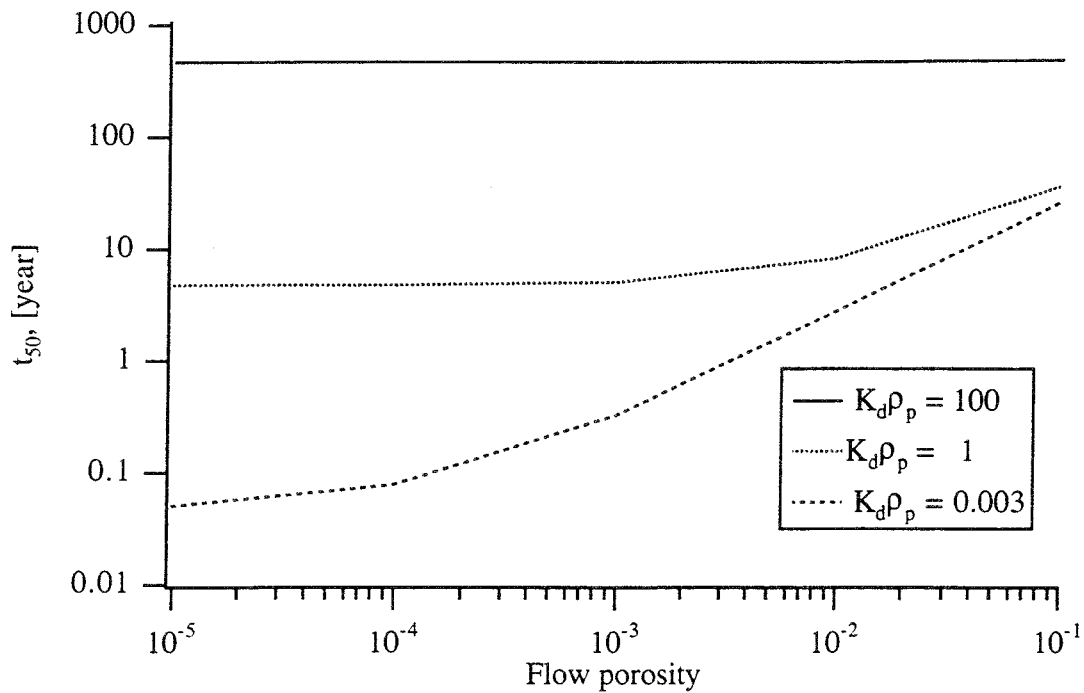

Figure 9 t_{50} as a function of flow porosity for different values of $K_d\rho_p$.

The results show that to predict the arrival time for a strongly sorbing tracer, it is not necessary to know the flow porosity. For weakly sorbing species, the flow porosity may be important if it is large. For nonsorbing tracers, the flow porosity must be known. For smaller flow rates, the influence of the flow porosity will be smaller because the travel time will to a larger extent be determined by the effect of matrix diffusion.

5 DISCUSSION AND CONCLUSIONS

These results have some implications in the planning of tracer tests to obtain information to be used in the prediction of release from a nuclear waste repository. Here, the interest has been centred on the transport of solutes by the water flowing in the rock over long periods of time. The results show that if the solutes have access to the rock matrix, the transport pattern is determined mainly by the water flow rate distribution and the surface area in contact between the flowing water and the rock matrix, the flow-wetted surface. This information cannot be obtained from tracer tests where the interaction with the rock matrix is insignificant. This is true for tracer tests using nonsorbing tracers during short times. Such tracer tests may, however, be used to obtain a preliminary characterization of the flow paths where tracer tests with sorbing species might be performed.

The prediction of the transport of solutes escaping from a repository for nuclear waste requires information of how the flow rate is distributed in the fractures in the rock and the magnitude of the flow-wetted surface. Therefore efforts must be made to determine these entities. Possible ways would be by hydraulic tests with short packer distances to ensure that all fractures are measured.

When the transport times for sorbing and nonsorbing tracers are compared, a retardation factor is frequently calculated. For a given water flux and transport distance, we have shown that the travel time for solutes that do not interact with the solid is determined by the flow porosity. On the other hand, for sorbing solutes, the travel time is in practice independent of the flow porosity. It is determined mainly by the flow-wetted surface and the diffusion and sorption properties of the solid. This means that there is no known relation between the residence times of noninteracting and interacting solutes. A porosity change would influence the former but not the latter if the flow rate is the same. Furthermore diffusion and sorption mechanisms change over time. The longer the surface is exposed to the solute the greater are the diffusion and sorption effects. A retardation factor determined between two tracer tests only relates these specific tests. This retardation factor cannot be used to compare other tracer tests performed over the same distance, e.g., when the water velocity is changed. The retardation factor therefore has no practical use. Moreover, it may be misleading to use the retardation factor to extrapolate results from tracer tests to longer distances or longer times.

For tracers that have access to the porous matrix and for long residences times, the flow volume (flow porosity) has a negligible influence on the residence time distribution. This is determined solely by the flow-rate distribution, the "flow-wetted surface" and the diffusion

and sorption properties of the rock matrix. Only for tracers where the effect of matrix diffusion is small or negligible must the volume accessible to flow be known in order to determine the RTD of the tracer.

NOTATION

a	Specific area	m^2/m^3
c	Solute concentration	$moles/m^3$
c_f	Solute concentration in the water in the fracture	$moles/m^3$
c_p	Solute concentration in the water in the pores in the matrix	$moles/m^3$
D_e	Effective diffusion coefficient	m^2/s
D_L	Dispersion coefficient	m^2/s
d_p	Particle diameter	m
K_a	Surface sorption coefficient	m
K_d	Volume sorption coefficient	m^3/kg
L	Length	m
P	Pressure	kg/ms^2
Q	Water flow-rate	m^3/s
S	Spacing between fractures	m
t	Time	s
t_w	Water residence time	s
u	Water velocity	m/s
u_o	Darcy velocity	m^3/m^2s
W	Channel width	m
x	Distance perpendicular to the flow in the matrix	m
z	Distance along the flow direction	m

δ	Aperture	m
ε_f	Flow porosity	-
μ	Dynamic viscosity	kg/m s
ρ_p	Rock bulk density	kg/m^3

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