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1. Introduction

2. Transport of tracers was modelled
Retardation of mobile radionuclides in granitic rock fractures by matrix diffusion

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Abstract

Transport of iodide and sodium has been studied by means of block fracture and core column experiments to evaluate the simplified radionuclide transport concept. The objectives were to examine the processes causing retardation in solute transport, especially matrix diffusion, and to estimate their importance during transport in different scales and flow conditions. Block experiments were performed using a Kuru Grey granite block having a horizontally planar natural fracture. Core columns were constructed from cores drilled orthogonal to the fracture of the granite block. Several tracer tests were performed using uranine, 131I and 22Na as tracers at water flow rates 0.7–50 µl min⁻¹. Transport of tracers was modelled by applying the advection-dispersion model based on the generalized Taylor dispersion added with matrix diffusion. Scoping calculations were combined with experiments to test the model concepts. Two different experimental configurations could be modelled applying consistent transport processes and parameters. The processes, advection-dispersion and matrix diffusion, were conceptualized with sufficient accuracy to replicate the experimental results. The effects of matrix diffusion were demonstrated on the slightly sorbing sodium and mobile iodine breakthrough curves.

Keywords:
Block-scale experiments
Crystalline rock
Matrix diffusion
Migration

1. Introduction

In Finland, the repository for spent nuclear fuel will be excavated at a depth of about 500 m in the fractured crystalline bedrock in Olkiluoto at Eurajoki, the site proposed by Posiva Oy. The fractures provide the most effective transport paths even though most of the porosity derives from the pores. The diffusion of mobile radionuclides into the micro-fissures and pores is still regarded as the main mechanisms retarding radionuclide transport in crystalline rock (Neretnieks, 1980). Ground water flow in fractured rock is distributed unevenly causing strong channelling effects, where the water flow occurs mainly over a small proportion of the fracture surface (Tsang et al., 1991), and stagnant non-flowing areas. Only matrix diffusion can cause significant changes in the shape of a breakthrough curve as a function of either elution time or the diffusion coefficient. In the case of crystalline rock, the residence times of tracers have been too short for matrix diffusion to occur in short time scale laboratory experiments. In laboratory-scale experiments, the effects of matrix diffusion have been demonstrated by Callahan et al. (2000). They investigated solute transport in fractured saturated volcanic tuff, which is significantly more porous than crystalline rock, allowing matrix diffusion to occur in a reasonable time. The dominant matrix diffusion behaviour was demonstrated in porous ceramic columns, and demonstration of the effects of matrix diffusion in crystalline rock fracture columns succeeded in a series of experiments where the experimental arrangements enabled very low water flow rates (Hölttä, 2002).

Performance assessment is directly concerned with the contribution of 129I, 36Cl, 79Se, 14C and 99Tc in their long-term exposure risks. The elution times of non-sorbing tracers have been used usually to indicate the flow rate of the groundwater in the fracture. However, this knowledge and understanding about transport and retardation processes can be utilized to evaluate the transport of mobile fission and activation products in the geosphere. Radionuclide transport through a natural fracture has been studied in many block-scale experiments (Drew et al., 1990; Cliffe et al., 1993; Vandergaaf et al., 1996, 1997; Park et al., 1997; Vilkas and Baik, 2001; Vilks et al., 2003). Migration experiments in Kuru Grey granite block fracture and core columns were introduced to evaluate the simplified radionuclide transport concept used in assessing the safety of the underground waste repositories (Hölttä et al., 2004). The objectives were to examine the processes causing retardation in solute transport, especially matrix diffusion, and to estimate the importance of retardation processes during transport in different scales and flow conditions. In this paper, we present the modelling concept, scoping calculations, the results of tracer tests performed and demonstrate the effects of matrix diffusion.
2. Experimental

Block-scale migration experiments were performed using Kuru Grey granite block which was obtained from Kuru Quarry, Tampeereen Kouvakiivy, Oy, Finland. The total porosity and the surface areas of mineral grains available for the migration of species were determined by the $^{14}$C-PMMA method (Siitari-Kauppi, 2002). The pore aperture distribution was evaluated on the basis of Hg-porosimetry determinations. Pore apertures and geometry in the mineral phases were analyzed also by scanning electron microscopy (SEM) and the minerals were quantified by means of energy dispersive X-ray microanalysis (EDX). The specific surface area of the solid rock was determined by the BET Hg impregnation method. Determined values are given in Table 1 and detailed rock matrix characterization is reported in Hölttä et al. (2004, 2007).

An experimental set-up for a block is illustrated in Fig. 1a. The block contains a natural hydraulically conducting fracture (0.9 m × 0.9 m) intersected by nine vertical boreholes, which were equipped with injection or sealing packers. Water pools were installed on the vertical sides and top of the block in order to ensure the saturation of the block and to stabilize the hydraulic head on the vertical faces. The block was instrumented also at the outer vertical boundary of the block where the horizontal fracture intersects the faces of the block for the collection of the tracer. Hydrological properties of the fracture were characterized and flow paths are described in Hölttä et al. (2004). Estimated transmissivities are illustrated in Fig. 1b, which shows clearly the increase in transmissivity towards side 3. Evaluation of the water consumption tests from drill holes KRO to KRB show transmissivities that vary between $9 \times 10^{-8} \text{m}^2\text{s}^{-1}$ and $2 \times 10^{-6} \text{m}^2\text{s}^{-1}$, and the average parallel plate aperture of the fracture was about 0.1 mm. In parallel with the block-scale experiments core column experiments were performed to estimate the diffusion properties of Kuru Grey granite. Core columns (Fig. 1c) were constructed from cores drilled to the fracture and were placed inside a tube to form a flow channel representing an artificial fracture formed by the 0.5 mm gap between the core and the tube (Hölttä et al., 2007).

In a block, flow path tests with uranine dye tracer showed that migration took place through distinct channels (Hölttä et al., 2004). Drill hole KR1 was chosen for the tracer transport experiments because hydraulic characterization and qualitative uranine dye tracer tests indicated that it had the longest flow path of about 0.7 m. The water pool at the side 3 was divided into eight adjacent tracer collection cells based on the outflow positions of the main transport channels. The out flowing tracer was collected by pump-

![Fig. 1a. Kuru Grey granite block having a natural hydraulically conducting fracture (0.9 m × 0.9 m). The vertical bore-holes are equipped with injection or sealing packers. Water pools equipped with adjacent tracer collection channels ensure saturation and stabilize the hydraulic head around the vertical faces.](image1)

![Fig. 1b. Local transmissivities in a natural fracture determined from the water pumping tests.](image2)

Table 1

Parameters applied in the modelling of the tracer experiments through the Kuru Grey granite borehole core and the natural fracture (0.9 m × 0.9 m)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Core column</th>
<th>Block channel II</th>
<th>Block channel IV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Channel length (m)</td>
<td>0.685</td>
<td>0.75</td>
<td>0.80</td>
</tr>
<tr>
<td>Channel width (m)</td>
<td>0.044</td>
<td>0.06</td>
<td>0.035</td>
</tr>
<tr>
<td>Channel aperture (m)</td>
<td>$7 \times 10^{-4}$</td>
<td>$6.5 \times 10^{-4}$</td>
<td>$5 \times 10^{-9}$</td>
</tr>
<tr>
<td>Matrix porosity (%)</td>
<td>0.04</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Specific surface area (m$^2$g$^{-1}$)</td>
<td>0.03</td>
<td>0.03</td>
<td>0.03</td>
</tr>
<tr>
<td>Density (kg m$^{-3}$)</td>
<td>2660</td>
<td>2660</td>
<td>2660</td>
</tr>
<tr>
<td>Average grain size (mm)</td>
<td>0.5-1.5</td>
<td>0.5-1.5</td>
<td>0.5-1.5</td>
</tr>
<tr>
<td>Average pore diameter (mm)</td>
<td>300-400</td>
<td>300-400</td>
<td>300-400</td>
</tr>
<tr>
<td>Flow rates ($\mu$L min$^{-1}$)</td>
<td>Uranine: 3, 3, 6, 20</td>
<td>Uranine: 7 (10)</td>
<td>Uranine: 3 (10)</td>
</tr>
<tr>
<td></td>
<td>$^{22}$Na: 3, 3, 6, 20</td>
<td>$^{131}$I: 3.0</td>
<td>$^{23}$Na: 3 (10)</td>
</tr>
<tr>
<td></td>
<td>$^{22}$Na: 3, 3, 6, 20</td>
<td>$^{131}$I: 3.0</td>
<td>$^{23}$Na: 3 (10)</td>
</tr>
<tr>
<td></td>
<td>$^{131}$I: 0.7, 4.2</td>
<td>$^{131}$I: 0.7, 4.2</td>
<td>$^{23}$Na: 3 (10)</td>
</tr>
<tr>
<td>Width of the velocity profile (m)</td>
<td>0.022</td>
<td>0.03</td>
<td>0.024</td>
</tr>
<tr>
<td>Diffusivity in free water (m$^2$s$^{-1}$)</td>
<td>$2 \times 10^{-9}$</td>
<td>$2 \times 10^{-9}$</td>
<td>$2 \times 10^{-9}$</td>
</tr>
<tr>
<td>Pore diffusivity (m$^2$s$^{-1}$)</td>
<td>Uranine: $6.5 \times 10^{-11}$</td>
<td>Uranine: $6.6 \times 10^{-11}$</td>
<td>Uranine: $6.6 \times 10^{-11}$</td>
</tr>
<tr>
<td></td>
<td>$^{22}$Na: $6.6 \times 10^{-11}$</td>
<td>$^{22}$Na: $6.6 \times 10^{-11}$</td>
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<td></td>
<td>$^{131}$I: $6 \times 10^{-11}$</td>
<td>$^{131}$I: $6 \times 10^{-11}$</td>
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<tr>
<td></td>
<td>$^{22}$Na: $6 \times 10^{-11}$</td>
<td>$^{22}$Na: $6 \times 10^{-11}$</td>
<td>$^{22}$Na: $6 \times 10^{-11}$</td>
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<tr>
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<td>$^{22}$Na: $6 \times 10^{-4}$</td>
<td>$^{22}$Na: $2 \times 10^{-5}$</td>
<td>$^{22}$Na: $2 \times 10^{-5}$</td>
</tr>
<tr>
<td>$K_r$ (m)</td>
<td>$^{22}$Na: $2 \times 10^{-5}$</td>
<td>$^{22}$Na: $2 \times 10^{-5}$</td>
<td>$^{22}$Na: $2 \times 10^{-5}$</td>
</tr>
</tbody>
</table>
ing and flushing collection channel areas. Several tracer tests were performed both in a block and core columns using uranine, HTO, $^{36}$Cl, $^{131}$I and $^{22}$Na as tracers with injection flow rates of 0.7–50 µL min$^{-1}$. Flow channel dimensions, tracers and range of volumetric flow rate used in the experiments are summarized in Table 1.

3. Modelling

Transport of tracers through the flow channels in a core column or in a natural fracture was modelled using an advection–dispersion model based on the generalized Taylor dispersion. It was assumed that a linear velocity profile existed across the flow channel, from zero velocity to some maximum flow velocity, and that the flow field and molecular diffusion perpendicular to the flow dominate the transport of the tracer particles in the mobile pore space of the fracture (Hautojärvi and Taivassalo, 1994; Poteri et al., 2002). The exact shape of the velocity profile is important in the case of purely advective transport which is not the case in the present experiments and is not of interest in this. More essential is to describe the variation of the flow velocities. It is assumed that velocity variation exist also in the experiments carried out with the cores. The aperture between a core and tube is quite small and it is difficult to centre the rough surfaced core on the middle which can easily generate variable flow velocity for different streamlines in the circular slit around the core.

The mean concentration across the flow channel for a narrow box-function release is given by Eq. (1)

$$C_m = \frac{1}{2} \left( \text{erf} \left[ \frac{X - X_t + \zeta_1}{2\sqrt{\zeta_2}} \right] + \text{erf} \left[ \frac{X - X_t - \zeta_1}{2\sqrt{\zeta_2}} \right] \right) \; ; \; \zeta_1 = \frac{1}{2} \tau; \; \zeta_2 = \left( \frac{1}{Pe^2} + \frac{1}{120} \right) \tau - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1 - e^{-(2n+1)^2\pi^2 \tau}}{(2n+1)^2\pi^2} \; ; \; \tau = \frac{D \Delta t}{u \cdot W} \; ; \; X = \frac{Dx}{u \cdot W}; \; X_t = \frac{Dx}{u \cdot W}; \; Pe = \frac{a v_0}{D}$$

where $D$ is the molecular diffusion coefficient in water, $a$ is the correlation length of the velocity variation approximated here as half of the flow channel width, $x_t$ is the initial width of the tracer plume, $v_0$ is the maximum flow velocity, $t$ is the time and $x$ is the position along the channel. The solute discharge at the end of the transport channel for the delta function release and taking into account the matrix diffusion and sorption can be written as

$$j(t, t_w, u, R_0) = H(t - R_0 t_w) \frac{u}{\sqrt{\pi (t - R_0 t_w)^2}} e^{-u^2}$$

where parameter $u$ determines the strength of the matrix diffusion, $t_w$ is the groundwater transit time and $R_0$ is the surface retardation coefficient. $H$ is the Heaviside step function. The matrix diffusion property ($u$) is defined as

$$u = \epsilon \frac{D_p R_p}{Q} \frac{W}{L} = \epsilon \frac{D_p R_p}{2b} t_w$$

where $t_w$ is the groundwater transit time, $2b$ is the channel aperture, $D_p$ is the matrix pore diffusivity, $\epsilon$ is the matrix porosity and $R_p$ is the retardation coefficient in the matrix. The last factor, $t_w/ (2b)$, in Eq. (3) is also presented as $WL/Q$. This parameter represents the coupling of the matrix diffusion to the flow field by ratio $Q/W$, i.e. the flow rate per width and to the length of the channel $L$. Sorption was modelled as linear equilibrium sorption, both in the pore space of the rock matrix and on the outer surface of the core. Solute transport with matrix diffusion and sorption was calculated by integrating over the solute mass flux distribution according to Eq. (4)

$$k(t) = \int_0^t j(t, t_w, U_t tw, R_0) b(t_w) \, dt_w$$

where $b(t_w)$ is the solute mass flux distribution in the mobile pore space and $j(t, t_w, U_t tw, R_0)$ is the corresponding matrix diffusion breakthrough curve, Eq. (2).

The shape of the tracer breakthrough curve is affected, at least, by velocity variation over the flow channel, molecular diffusion in the flow channel, possible sorption and matrix diffusion. It could be very difficult to assess from an individual breakthrough curve whether matrix diffusion shows up in the experiment. A more robust approach is to analyse the dynamic behaviour of the tracer discharge when flow rate and tracer sorption properties are changed. Tests with the cores offer an opportunity to carry out a test with the same tracers and with the nearly identical rock matrix but under well-constrained flow geometry. A minor difference in the diffusion properties of the rock matrix could arise for example from possible alteration on the surface of the natural fracture.

The transport problem is characterized in the present modelling approach by parameters: the volume of the transport channel ($V_t$), correlation length of the velocity variation in the flow field ($W_0$), molecular diffusion in free water ($D$) and immobile pores of the rock matrix ($D_p$), distribution of flow velocities (linear velocity profile from zero velocity to $v_{\text{max}}$), rock matrix porosity ($\phi$), flow rate distribution in the flow channel ($Q/W$) and the channel length ($L$). Sorption properties of sodium were estimated from the $K_d$ values determined for SyrRY mica gneiss and unaltered tonalite (Hölttä, 2002). Commonly used literature value has been applied for the molecular diffusion coefficient in free water and pore diffusivity in a rock matrix is based on Archie’s law for the formation factor $F = 0.71 e^{1.58}$ (Cheng and Cvetkovic, 2005). Measured data was

\( F = 0.71 e^{1.58} \)
Scoping calculations for a core column tracer test based on the dimensions of an artificial flow channel and preliminary tracer experiments. Dotted lines are modelled breakthrough curves for sodium. In the case of core column experiments with the non-sorbing tracer, the sorption of sodium into account, show very good agreement with the measured breakthrough curves (Fig. 3b). Sorption of the sodium and uranine breakthrough curves are used to constrain volumes of the transport channels and correlation lengths of the velocity variation. Well mixed tracer concentration was assumed close to the injection drill hole KR1 where the transport paths diverged. Under this assumption, the flow rates of the transport channels were proportional to the recovery collected from these channels. Division of the total flow rate between the channels was also subject to minor calibration. Tracer recoveries indicate that flow rate through the channel II is 73–75% of the injection flow rate. Calibrated value is that 70% of the total flow rate goes through the channel II. The only difference between modelled sodium and uranine breakthrough curves is the application of the measured $K_d$ for sodium. In the case of core experiments the correlation length of the velocity variation was selected to be half of the channel width, i.e. 2.2 cm, to take into account that there are different flow velocities at different locations around the core circumference. The only calibration parameter in the core experiment was the aperture. The measured aperture, $2b = 0.5\text{ mm}$, do not account for the roughness of the surface of the core and the actual transport aperture may be larger. The calibrated aperture used is $2b = 0.7\text{ mm}$. Calibration of the transport aperture aimed at reasonable fit for different flow rates and tracers with emphasis to more retarded breakthrough curves, i.e. smaller flow rates. The modelled breakthrough curves for the tracer transport were convoluted with a response function of the tubing before being compared with the measured experimental breakthrough curves. The response function of tubing and other experimental equipment was determined by performing tracer tests without a core column or a natural fracture.

4. Results and discussion

First tracer tests in a block fracture were performed for uranine and $^{99m}$Tc using injection flow rates of 350 $\mu\text{l min}^{-1}$ and 230 $\mu\text{l min}^{-1}$ (Hölttä et al., 2004). Obtained breakthrough curves were dominated by the advective field and the processes like diffusional mixing. Scoping calculations were carried out based on the geometrical dimensions and the hydraulic characterization of the fracture and preliminary tracer tests. In the model simulations for the sorbing tracer, the $K_d$ value used, $7.1 \times 10^{-6} \text{ m}^3 \text{ kg}^{-1}$, was much smaller than the value of $6 \times 10^{-4} \text{ m}^3 \text{ kg}^{-1}$ extracted from fits to experiments. In an ideal fracture, matrix diffusion is clearly observable for a non-sorbing tracer when the flow rate is $0.1 \mu\text{l min}^{-1}$ in a core column (Fig. 2a) and $1 \mu\text{l min}^{-1}$ in a block fracture (Fig. 2b).

Tracer tests analyzed in the present paper were performed for uranine and slightly sorbing sodium ($^{23}$Na) using flow rates 3–20 $\mu\text{l min}^{-1}$ in a core column and 10–50 $\mu\text{l min}^{-1}$ in a block. Measured and modelled breakthrough curves through the core column are presented for uranine in Fig. 3a and for $^{22}$Na in Fig. 3b. Measured and modelled breakthrough curves of uranine and $^{22}$Na through the block fracture are presented in Fig. 3c. Effects of the matrix diffusion in the breakthrough curves can be observed when results for non-sorbing and sorbing tracers are compared side by side. As explained in the model calibration the only difference between modelled breakthrough curves for uranine and sodium in the core column experiments is the non-zero $K_d$ of the sodium. It should also be noted that $K_d$ of the sodium has not been calibrated but it is estimated based on the measured data. The model explains the transport of the uranine for the lower flow rates well, the test

![Fig. 2a](image-url) Scoping calculations for a core column tracer test based on the dimensions of an artificial flow channel and preliminary tracer experiments. Dotted lines are model results for advection-dispersion and matrix diffusion. Solid lines are for the advection-dispersion only.
with the highest flow is dominated by the velocity field in the experimental results and by the velocity profile in the modelled results and is not subject of main interest in the present study. Especially, it is observed that the model predicts matrix diffusion effects in the tailings of the breakthrough curves \( \frac{C_2}{C_0} \) tailing) beyond the range of measured data (Fig. 3a). Using exactly the same model to sodium breakthrough curves, only taking the sorption of the sodium into account, show very good agreement with the measured breakthrough curves (Fig. 3b). Sorption of the sodium enhances matrix diffusion showing clear difference in the tailings of the breakthrough curves. Fig. 3b includes also breakthrough curves that are modelled without matrix diffusion in order to facilitate the identification of the matrix diffusion effects. Results give strong evidence that sodium breakthrough curves are affected by matrix diffusion because: (i) the same model is able to reproduce advection–dispersion dominated the breakthrough of the uranine and even predicts that matrix diffusion effects are beyond the range of measured data, (ii) the same model is able to reproduce the breakthrough curves of the sodium; the only difference between these models being the sorption of sodium, even...
breakthrough very accurately when sorption properties are changed (Fig. 3c). There are strong indications that effects of the matrix diffusion were observed in the fracture flow experiment because:

(i) when sorption properties are changed the dynamic behaviour of the breakthrough curves is well explained by the matrix diffusion model for both channels, i.e. for different flow rates, (ii) the same sorption properties for sodium as in the core column experiment explain the difference in breakthrough curves between uranine and sodium and (iii) the tailings of the sodium breakthrough curves follow $C_2/C_0^{3/2}$ which is typical for matrix diffusion. Note that the difference in the modelled breakthrough curves of uranine and sodium comes solely from the sorption properties of the sodium and that the same measured sorption values were applied as in the core column experiment.

A third set of tracer tests were performed for $^{131}$I using flow rates $0.7–10 \text{ l min}^{-1}$ in a core column and $3–10 \text{ l min}^{-1}$ in a block. Examples of the measured and modelled breakthrough curves through the core column are presented in Fig. 4a and through the block fracture in Fig. 4b. The model used earlier for fracture column experiments (Hölttä, 2002) and the same transport parameterization that were used to examine uranine and sodium experiments were applied to independently interpret iodide results. In a core column, a $D_e$ value of $6 \times 10^{-10} \text{ m}^2 \text{s}^{-1}$ gave the best overall fit between measured and calculated iodine break through.
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through curves. In a block fracture, a $D_v$ value of $2 \times 10^{-11}$ m$^2$ s$^{-1}$ gave the best overall fit between measured and calculated iodine breakthrough curves. This relatively high $D_v$ value for low porosity granite possibly indicates diffusion also into the stagnant water areas of wide and ragged surfaces of a natural fracture. Comparing modelled results for iodide with and without matrix diffusion supports the conclusion that tailings of the iodide breakthrough curves are affected by matrix diffusion.

5. Conclusions

Block and core column migration experiments were performed to evaluate the simplified radionuclide transport concept used in assessing the safety of the underground waste repositories. Two different experimental configurations could be modelled applying consistent transport processes and parameters. The processes, advection–dispersion and matrix diffusion, were conceptualized with sufficient accuracy to reproduce the experimental results. The results provided show that it is possible to investigate matrix diffusion in low porosity crystalline rock at the laboratory scale. The effects of matrix diffusion were demonstrated on the slightly sorbing sodium and mobile iodine breakthrough curves. The modelled experiment builds confidence on the model predictions of the solute retention in groundwater flow. This understanding is transferable from the laboratory scale to in situ conditions though specific parameters can not be transferred directly to the spatial and temporal repository scale.

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References


