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Fracture flow and radionuclide transport in block-scale laboratory experiments

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Summary. Block-scale migration experiments were introduced to evaluate the simplified radionuclide transport concept used in assessing the safety of underground spent nuclear fuel repositories. The experiments were aimed to demonstrate visually the fracture flow, and to determine the hydraulic characteristics of a natural planar fracture and the transport behaviour of non-sorbing and sorbing radionuclides. For drill holes orthogonal to the fracture and equipped with injection or sealing packers flow rates in this study were measured as a function of hydraulic head. The outflow positions of water at each four side of the block were determined using uranine dye tracer. Tracer tests were performed using uranine, ^{99m}Tc and ^{22}Na.

Transport of a non-sorbing tracer through one of the flow channels was interpreted using an advection-dispersion model that on the generalised Taylor dispersion.

Characterisation of the hydraulic properties of the fracture indicated that some drill holes were located in the region where the fracture was open and water conductive. No water conductivity was observed in two drill holes indicating closure of the fracture. Reasonably low flow rates obtained from three drill holes indicated their suitability for further radionuclide transport experiments. Elution times of technetium and uranine were fairly similar. Sodium was slightly retarded and was spread over a wider area than uranine and technetium. High water flow rates suggest that advective flow field dominated tracer transport. Experimental and calculated elution curves substantiate the suitability of our experimental set-up for further radionuclide transport experiments.

1. Introduction

Crystalline rock is being considered as a host medium for repository of highly radioactive spent nuclear fuel in Finland and elsewhere. The geosphere would act as the ultimate barrier retarding the migration of radionuclides to the biosphere if radionuclides are released through engineered barriers. In crystalline rock water flows through a fracture network and radionuclide transport is thought to proceed along water-carrying fractures. Retardation occurs both in the fractures and within the rock matrix. In order to understand the transport of dissolved radionuclides through rock it is necessary to consider both the fracture network geometry and the transport properties of the individual fractures. Ground water flow in fractured granite rock is distributed unevenly causing strong channelling effects, where the water flow occurs mainly over a small proportion of the fracture surface [1–3]. Stagnant water is also found in side fractures, micro fissures and in pores within the rock matrix. Block-scale experiments with natural fractures and only a few flow paths are important intermediate stages between small-scale fracture column and field experiments. The knowledge obtained from transport experiments in well-defined cm to m-scale laboratory conditions provides a basis for m- to km-scale field experiments performed to validate the radionuclide transport concept and to test the transferability of laboratory data to *in-situ* conditions.

Radionuclide transport has been studied in numerous laboratory-scale experiments in single fractures using the column method [4–7] and in block-scale fractures [8–14]. The influence of variable fracture aperture on the transport of non-sorbing solutes in a single fracture was investigated numerically by Grenier *et al.* [15], and the influence of specific surface area and fracture aperture on the transport of sorbing solutes in a fracture was investigated experimentally by Wels *et al.* [16]. Rock-block migration experiments were introduced to evaluate the simplified radionuclide transport concept used in assessing the safety of the underground waste repositories. Such experiments demonstrate visually the fracture flow, and determine the hydraulic characteristics of a natural planar fracture as well as the transport behaviour of non-sorbing and sorbing radionuclides. We describe below the experimental design utilising a large granite block and present hydraulic and first tracer test results.

2. Experimental

The $0.9 \times 0.9 \times 0.7$ m block of fine-grained, non-foliated and equigranular Kuru gray granite (Kuru Quarry, Tampereen Kovakivi Oy) composed of 36% potassium feldspar, 35% quartz, 21% plagioclase and 8% amphibole and mi-

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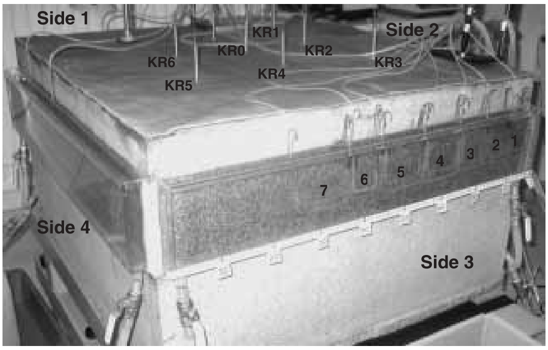

Fig. 1. A photograph of the granite block.

cas, has 0.2% total porosity and 2660 kg m^{-3} density. The single natural horizontal water-conducting planar fracture is located about 17 cm below the top of the block. A drill hole into the centre of the block with 3-cm diameter and eight additional drill holes with 2-cm diameter were drilled orthogonally to the fracture. The drill holes were equipped with injection or sealing packers. An experimental set-up is illustrated in a photograph of the granite block (Fig. 1).

2.1 Hydraulic characterisation of the fracture

Preliminary estimates of water conductivity in the fracture were obtained by observation of water consumption in tubes connected to the drill holes. The total water transmissivity of the fracture was determined by weighing the water consumption as a function of time. A hydraulic head controlled the water flow rate into the fracture through a drill hole. Hydraulic head values were determined as a difference in altitude between a fracture level and a water surface level in a container on scales. To determine channelling in the fracture, water was fed into the fracture from one drill hole at a time while the fracture in the other drill holes was plugged up with packers. Experiments were performed at different hydraulic head values. The outflow positions of water at all four sides of the block were detected and recorded by a video. The water mass flow distribution in different channels was determined by collecting the out flowing water from the main channels.

2.2 Tracer experiments

After hydraulic characterisation of the fracture, the block was surrounded with polymetacrylate pools filled with water (Fig. 1). A 5-mm wide water collection slit near the fracture was separated with a partitioning wall having openings at the bottom of the pool. The pools were constructed to maintain equal pressure conditions all around the fracture area and to avoid external disturbances. The outflow positions of water at all four sides of the block were determined using uranine dye tracer. Water was fed into the fracture from the drill holes labelled KR0, KR1, KR2 and KR5 using three different water flow rates, conrolled by hydraulic heads. A short uranine tracer pulse (5 μl) was injected into the water flow using an injection loop. The outflow points of the tracer were located by following the experiment with a video camera and later with digital camera.

Tracer experiments were performed using drill hole KR1 as the injection point and the tracer was collected from the opposite Side 3 in order to get as long flow path as possible. The fracture area in Side 2 was sealed up with rubber insulation in order to prevent tracer leakage from Side 2. Owing to no uranine leakage Sides 1 and 4 were not sealed. In Side 3 the water collection slit was separated into channels for collection of tracer. A peristaltic pump controlled water flow rate in the tracer transport experiments. Water was fed into the fracture from the drill hole KR1 using different flow rates of 0.2–0.5 ml min^{-1}. A short tracer pulse (50 μl) was injected into the water flow using an injection loop (Rheodyne). Out flowing tracer was collected by pumping and flushing collection channel areas. Uranine and ^{99m}Tc were used as non-sorbing tracers and ^{22}Na as a slightly sorbing tracer. Absorbance of uranine was measured by UV/VIS spectrophotometer and gamma activities of ^{99m}Tc and ^{22}Na were detected using a Wizard gamma counter.

3. Results and discussion

3.1 Hydraulic characterisation of the fracture

No water was consumed by drill holes KR7 and KR8 indicating that these holes were located in an area where the fracture was closed. Table 1 presents the measured water flow rates as a function of the hydraulic head. Hydraulic characterisation of the fracture is based on these measurements.

Pumping of the fracture by applied over-pressure in the drill holes was approximated by a two-dimensional radial flow field. The hydraulic head at distance r from the drill hole is given by Eq. (1)

$$h(r) = h_{\mathrm{w}} - \frac{Q}{2\pi T} \ln\left(\frac{r}{r_{\mathrm{w}}}\right) , \tag{1}$$

where h_{w} is the hydraulic head in the drill hole, Q is the flow rate, T is the fracture transmissivity and r_{w} is the ra-

Table 1. Water flow rates (ml min^{-1}) measured from different drill holes using different hydraulic heads (cm).

Hydraulic head/cm	Drill hole water flow rate/ml min^{-1} KR0	KR1	KR2	KR3	KR4	KR5	KR6
24	4.9	1.9	16	35		26	2.2
23							1.2
22	3.9	1.6			19		1.2
21							1.1
20	3.5	1.5	13	27	16	17	1.1
18	2.9	1.5	12			15	1.0
16	2.2	1.3		27	12		0.8
14	1.8	1.1	9.1			12	
12	1.3	1.0	6.7	20	7.5		
10	0.7	0.8				7.9	
8		0.6	5.5	13	3.8	1.8	
6		0.4	4.1			0.6	
4		0.3	2.9	6.7			
2			1.8	3.2			
1			1.2				

dius of the drill hole. The hydraulic head is zero at the outer boundary of the rock block. Radial flow analysis is applied to interpret the tests by assuming that $h(r_0 = 0)$, where r_0 is the distance from the drill hole to the nearest side of the rock block. This may slightly underestimate the transmissivity for the drill holes that are located asymmetrically near to one of the sides. However, a radial flow field corresponds to a linear dependency between the flow rate and the corresponding hydraulic head. This allows a straightforward estimation of the local transmissivity using Eq. (2)

$$T = C\frac{\ln\left(\frac{r_0}{r_w}\right)}{2\pi}\,, \tag{2}$$

where C is the slope of the (h_w, Q)-plot.

Estimated local transmissivities were consistent with those detected visually, *i.e.* the fracture opens towards Side 3 and is closed in the corner between Sides 1 and 4. Transmissivities are between $9\times10^{-8}\,m^2\,s^{-1}$ and $2\times10^{-6}\,m^2\,s^{-1}$ and they show the pattern illustrated in Fig. 2. Fracture aperture contours in Fig. 3 were determined by a least squares fit of the linear extrapolation between the data points. Parallel plate aperture corresponding to the mean transmissivity was approximately 0.1 mm.

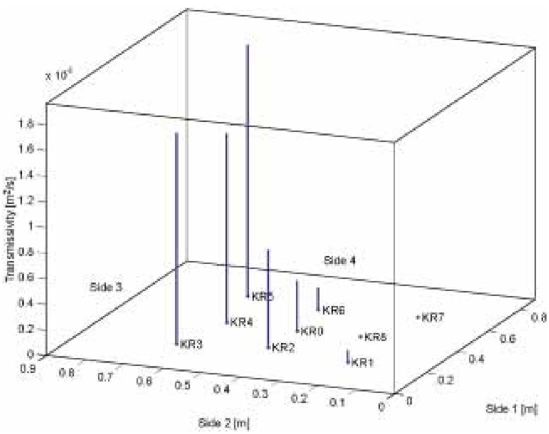

Fig. 2. Local transmissivities determined from the water consumption tests in the drill holes.

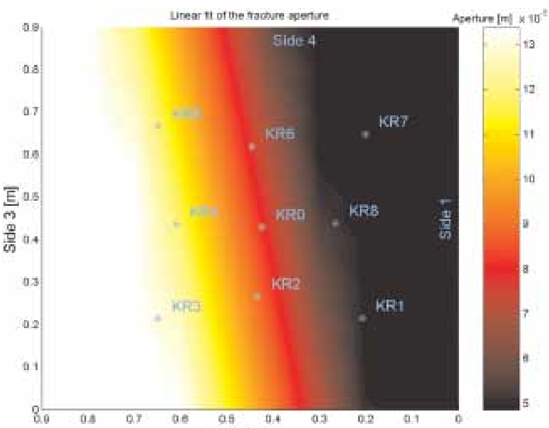

Fig. 3. Fracture aperture contours calculated from the transmissivities (linear fit).

3.2 Tracer tests

Drill hole KR1 was chosen for the tracer transport experiments because hydraulic characterisation and qualitative uranine dye tracer tests indicated that it had the longest flow path. Two different tracer tests were performed. The first test was performed using uranine and technetium. The injection flow rate to the drill hole was approximately $0.35\,ml\,min^{-1}$. The second test was performed using uranine, technetium and sodium with an injection flow rate of approximately $0.23\,ml\,min^{-1}$. The distance from the injection drill hole KR1 to the collection channels at Side 3 was 70 cm. Both tests showed breakthrough in collection channels 1–6 at the Side 3 (Fig. 1) and in total 80% of injected uranine and technetium was collected from Side 3. In both tests the elution times of technetium and uranine were similar. In some elution curves inadequate consistency arose from measurement problems due to the short half-life of ^{99m}Tc (6 h). In the second test only a slight retardation on sodium was found. The proportional recoveries from different collection channels are presented in Table 2.

Transport of a non-sorbing tracer through one of the flow channels was also modelled. Modelling was performed for channel 2 for both tracer tests. Both tests used an advection-dispersion model based on the generalised 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. Hautojärvi and Taivassalo [17] give a more detailed discussion of the problem. The mean concentration across the flow channel for a narrow box-function

Table 2. Proportional recovery of injected uranine, technetium and sodium from different outflow channels in Side 3 with flow rates of $0.35\,ml\,min^{-1}$ and $0.23\,ml\,min^{-1}$.

Outflow channel	Tracer Uranine	^{99m}Tc	^{22}Na
$0.35\,ml\,min^{-1}$			
1	0.01	0.03	–
2	0.19	0.19	–
3	0.25	0.24	–
4	0.23	0.24	–
5	0.12	0.11	–
6	0.03	0.02	–
7	0.01	0.002	–
$0.23\,ml\,min^{-1}$			
1	0.19	0.20	0.12
2	0.22	0.21	0.14
3	0.23	0.24	0.10
4	0.17	0.09	0.12
5	0.01	0	0.04
6	0.01	0	0.05
7	0.001	0	0.04

release is given by Eq. (3).

$$C_{\mathrm{m}} = \frac{1}{2}\left(\operatorname{erf}\left[\frac{\frac{1}{2}x_s + x + \xi_1}{2\sqrt{\xi_2}}\right] + \operatorname{erf}\left[\frac{\frac{1}{2}x_s - x - \xi_1}{2\sqrt{\xi_2}}\right]\right);$$

$$\xi_1 = \frac{1}{2}\tau; \quad \xi_2 = \left(\frac{1}{(\mathrm{Pe})^2} + \frac{1}{120}\right)\tau - 8\sum_{n=0}^{\infty}\frac{1-\mathrm{e}^{-(2n+1)^2\pi^2 r}}{(2n+1)^8\pi^8};$$

$$\tau = \frac{Dt}{a^2}; \quad x = \frac{Dx}{a^2 v_0}; \quad x_s = \frac{Dx_s}{a^2 v_0}; \quad \mathrm{Pe} = \frac{a^2 v_0}{D}, \qquad (3)$$

where D is the molecular diffusion coefficient in water (10^{-9} m^2 s^{-1} was used in the present analysis), a is the correlation length of the velocity variation (this is approximated here as half of the flow channel width), x_s is the initial width of the tracer plume (for a pulse release this is selected to be small compared to the length of the flow path), v_0 is the maximum flow velocity, is the time and is the position along the channel. The advection component dominated the advection-dispersion model with the flow rates used in this study. The advection-dispersion model of the present study is a purely advective transport characterised by a linear velocity profile over the flow channel.

The flow rate along the flow path that discharged to collection channel 2 was quantified by measuring the recoveries. It was assumed that the tracer mass flux in the different parallel flow channels was proportional to the flow rates of these flow channels. This means that well mixed conditions were assumed close to the injection drill hole KR1 where the different flow paths diverged. Under these assumptions the flow rates of the different flow channels were proportional to the recoveries collected from these channels. Measured recoveries from the collection channel 2 were 19% and 22% for the first and second tracer test, respectively. This yielded flow rates through the flow channel of 4 ml h^{-1} (0.19×0.35 ml min^{-1}) in the first tracer test and 3 ml h^{-1} (0.22×0.23 ml min^{-1}) in the second tracer test. The measured mean breakthrough times for the uranine were 2.5 hours for the first tracer test and 8.8 hours for the second tracer test. This means that the two tests gave slightly different volumes for transport channel 2, *i.e.* about 10 ml and 26 ml using data from the first and the second test, respectively. The variation in the parameters may indicate that the flow path itself may have been slightly different in the first test and in the second test.

After investigating the alternative flow channel geometry it appeared evident that the transport of uranine through flow channel 2 is best described by pure advection. The present analysis applies a linear velocity profile over the channel. This means that the tracer particles have an equal probability for any flow velocity between the minimum and the maximum values. Selecting the minimum flow velocity to be zero and the maximum flow velocity so that the mean flow velocity coincides with the measured average flow velocity, means that in the model the maximum flow velocity is about 0.56 m h^{-1} in the first test and 0.16 m h^{-1} in the second test. In both tests the length of the flow path was 0.7 m.

Modelling results for the uranine breakthrough curves are presented in Fig. 4. Experimental breakthrough curves

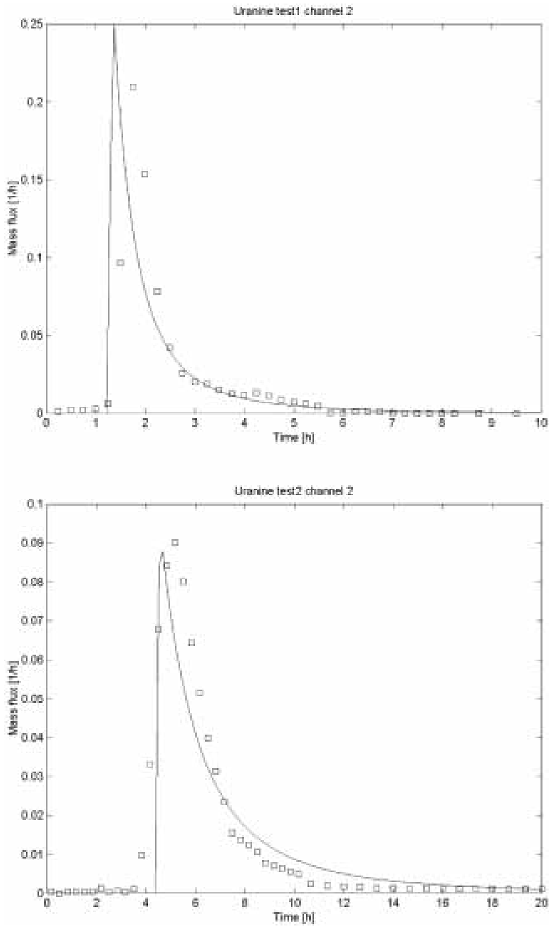

Fig. 4. Measured and modelled uranine breakthrough curves for the collection channel 2 in Side 3 with flow rates of 0.35 ml min^{-1} (upper) and 0.23 ml min^{-1} (lower).

were corrected by taking into account the time that the tracer resided in the tubing: 23 minutes in the first tracer test and 20 minutes in the second tracer test. This very simple model seems to be in quite good agreement with the measured breakthrough curves.

4. Conclusions

The hydraulic properties of the fracture have been characterised. Drill holes KR2-KR5 are located in the area where the fracture is open and water conductive. When the hydraulic head is used to control the water flow rate, than the flow rates from these drill holes are high and the residence times in the fracture are too short for tracer tests. No water conductivity was found in drill holes KR7 and KR8, indicating closure of the fracture in that area. Evaluation of the water consumption tests from drill holes KR0 to KR6 show transmissivities that vary between 9×10^{-8} m^2 s^{-1} and 2×10^{-6} m^2 s^{-1}. The average parallel plate aperture of the fracture was about 0.1 mm. Reasonable low flow rates were obtained from drill holes KR0, KR1 and KR6. For further radionuclide transport experiments the longest flow paths can be obtained from the drill hole KR1 to Side 3.

Two sets of tracer tests were performed using flow rates of $0.35\,\mathrm{ml\,min^{-1}}$ and $0.23\,\mathrm{ml\,min^{-1}}$. Tracer breakthrough was detected from seven collection channels along Side 3. Elution times of technetium and uranine were quite similar and only slight retardation of sodium was found. It was also noted that sodium spreads over a wider area at Side 3 than do uranine and technetium. The reason for this behaviour is not yet known. However, the obtained elution curves indicate that the experimental set-up is suitable for radionuclide transport experiments. In these experiments the water flow rates were quite high. Modelling results indicate that in both tracer tests the transport was dominated by advective flow field. The study of interaction processes such as matrix diffusion requires lower flow rates in future experiments. The present experimental set-up can be modified for flow rates at least an order of magnitude lower.

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