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Modelling of nonreactive tracer dipole tests in a shear zone at the Grimsel test site

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Abstract

The investigation of the migration of a high pH plume in a fractured shear zone is foreseen by a long-term experiment at the Grimsel rock laboratory. In order to characterise the initial conditions for the long-term experiment and to evaluate an optimal hydraulic in situ set-up, several dipole experiments with nonreacting tracers have been performed. The dipole experiments differ in geometry, pumping rates and orientation to the background water flow. Several single and double-porosity models have been applied to fit the results of these dipole tracer tests in order to extract values for some transport parameters and discriminate for certain transport processes. A two-dimensional porous medium approach was successfully used to fit tracer breakthrough curves measured for a dipole experiment. A model based on a one-dimensional dual porous medium approach was also successful, although the applied hydraulic dipole, with similar injection and extraction rates, suggests the existence of an extended two-dimensional flow field. For the two-dimensional porous medium approach, tracer breakthrough could only be fitted with a complex flow field geometry within the heterogeneous fractured shear zone. The heterogeneity was generated by heterogeneous porosity and hydraulic permeability distributions. Predictions for further dipole geometries and a sorbing tracer have been calculated by means of both models using the flow and transport parameters deduced from fits for a single dipole experiment. This allows for comparison with the measured breakthrough of sorbing tracers. The foreseen experiment with sorbing (radionuclide) tracers will help decide on the appropriate approach that should be used to describe such dipole experiments in this shear zone. Additionally, the migration and spreading of a solution with high pH has been calculated taking into account mineral dissolution and precipitation in a two-dimensional porous medium approach in order to estimate the amount and character of the mineral reactions induced by the interaction between the high pH solution and the rock.

Keywords: Dipole experiment; Modelling; Reactive transport; Heterogeneous media

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1. Introduction

At the Grimsel test site, tracer (radionuclide) migration experiments have been performed to improve the understanding of transport processes in fractured rocks. Transport models that describe these experiments and contribute to performance assessment studies of nuclear waste repositories have been developed and tested. The migration shear zone (MI) served as an ideal place for such investigations for more than a decade (Frick et al., 1992). In the parallel, neighbour shear zone (AU126), the HPF shear zone, a reactive transport experiment, is planned. The interaction of a high pH solution with the fractured rock, especially important for a cementitious repository, and its influence on radionuclide migration are investigated within a long-term in situ experiment, in which minerals will be dissolved and secondary mineral will be precipitated, changing the porosity, hydraulic conductivity and mineral specific sorption sites. Although a lot of laboratory experiments and modelling have been performed on this topic (e.g., Bateman et al., 1998, 1999; Berner, 1992, 1998; Grindrod and Takase, 1996; Haworth and Noy, 1993; Lichtner and Eikenberg, 1995; Lichtner et al., 1998; Neall, 1994; Pfingsten and Shiotsuki, 1998; Pfingsten, 2001; Steefel and Lichtner, 1994), their results show that an up-scaling from laboratory scale to field scale is difficult, as with the up-scaling from simplifying model assumption to complex near-field heterogeneity. The chosen experimental conditions in the laboratory and the model assumptions allow for a variety of consequences for the migration of a high pH solution. Depending on hydraulic conditions, flow path geometry (open fracture or heterogeneous porous medium), and groundwater and mineral composition, the results and predictions range from increased movement to completely stationary pH front and related radionuclide migration. To decrease this uncertainty, in situ experiments and their successful modelling are necessary. Before the long-term reactive transport experiment, it is essential to characterise the hydrogeological and transport properties of the fractured shear zone and to identify the relevant transport processes. Hydraulic dipoles between six different boreholes within a single shear zone have been established to perform independent tracer dipole experiments between boreholes using different dipole geometries, pumping rates and orientation to the background water flow to deduce its transport properties and test the applicability of the models. Additional site specific data included analysis of bore cores, local hydraulic transmissivity values deduced from pumping tests and hydraulic head measurements from the boreholes before and during the dipole experiments. The different model approaches (dual porous and heterogeneous porous medium) and their results indicate that, due to the relatively low permeability in the HPF shear zone and the applied hydraulic dipole, a complex flow field and several transport paths have to be assumed. This puts an extra demand on the modelling of radionuclide migration in the shear zone, since mineral reactions, changing mineral composition and hydraulic conditions will change transport parameters, such as the amount of available sorption sides for radionuclides that has to be taken into account in a heterogeneous shear zone. These changing conditions will become especially important for the modelling of the long-term behaviour of a cementitious repository near-field; a challenging venture which will be tested in situ at rock laboratory scale.

2. Experiments

The dipole experiments have been performed in the Grimsel granodiorite, which is characterised by the presence of ductile shear zones that include mylonite bands. Brittle fractures developed in the shear zone, mainly in the mylonite bands, and include a highly porous fault gouge. A detailed description can be found in Bossart and Mazurek (1991). Main differences between the MI and the HPF shear zones are the lower transmissivity of the HPF shear zone (10^{-8} m^2/s and less) compared to that of MI (about 10^{-6} m^2/s) and a higher degree of fracturing (number of fractures and their distribution across the shear zone) in the HPF shear zone. The fracture apertures or their connectivity should be lower for the HPF shear zone, since transmissivity in the HPF shear zone was measured to be two orders of magnitude lower than in the MI shear zone. Borehole imaging showed a highly fractured HPF shear zone—up to 10 fractures with apertures of about 1 mm and more. Ten boreholes (about 7–17 m long) drilled from the access tunnel through the HPF shear zone could be used to get information on its hydrogeological properties. Six boreholes intersect the shear zone within a 2×2 m^2 area, which is used for tracer and reactive transport experiments. The distances between the boreholes range from 0.77 to 1.8 m. High pH resistant packer systems have been installed to divide the individual boreholes into several intervals. One interval includes the water-conducting shear zone. Step-pulse injection of different dye tracers was performed (Table 1) by an injection loop, parallel to a pumping loop that preconditioned a stationary flow field during the whole dipole experiment. The tracer is monitored by a fluorescence signal at the injection and extraction sides of the dipole. Due to low permeability in the HPF shear zone, extraction could not be done by active pumping but through free outflow. A constant hydraulic head was adjusted to fix the outflow rate. Low outflow rates have to be used to avoid desaturation (degassing) at the outlet and to allow investigation of mineral reactions in the long-term experiments, which should not be dominated by a fast water flow. The main difference of the HPF tracer experiments compared to the MI experiments is that narrow flow fields may no longer exist from injection to extraction. In the MI experiments, the ratio of extraction rate to injection rate, β, was generally greater than 10 with rates of about 150 ml/min for extraction and 10 ml/min for injection. In the HPF experiment, β was about 1 and even below, with rates of about 1 ml/min extraction and 1 ml/m injection, due to hydraulic constrains in the HPF shear zone. A wide dipole flow field is assumed to be generated in the HPF dipole experiments. In total, seven dipole tracer experiments have been performed (see Table 1, NAGRA, 2001) to investigate tracer transport in the HPF shear zone and to estimate the flow path geometry and relevant processes. During these experiments, the experimental set-up has been improved to avoid problems during the planned long-term reactive transport experiment and the excavation of the dipole area after the experiments.

3. Modelling of tracer dipole experiments

Two different approaches are used to model the dipole tracer experiments. First, we used the double-porosity codes RANCHMD (Jakob, 1990) and PICNIC (Barten and Robinson, 2001), which were successfully applied to all tracer and radionuclide transport experiments

Table 1
Summary of the dipole tracer experiments performed in the HPF shear zone (NAGRA, 1999, 2000, 2001)

Nomenclature	Dipole experiment						
	1	2	3	4	5	6	7
	March 1999	June 1999	July 1999	HPF1/2000	HPF2/2000	HPF3/2000	HPF4/2000
Borehole	BOHP003 to BOHP001	BOHP003 to BOHP001	BOHP002 to BOHP001	BOHP003 to BOHP001	BOHP003 to BOHP009	BOHP010 to BOHP008	BOHP001 to BOHP002
(Distance [m])	(0.77)	(0.77)	(1.02)	(0.77)	(1.23)	(1.19)	(1.02)
Assumed orientation with respect to background flow	~ parallel	~ parallel	~ 45°	~ parallel	~ 45°	~ 45°	~ 45°
Tracer[a]	Uranine (0.5 ppm, 0.126 mg)	Pyranine (0.5 ppm, 0.66 mg) {and T3 (5 ppm, 6.6 mg)}	Perylene (2 ppm, 1.58 mg) {and UV-1 (10 ppm, 7.7 mg)}	Uranine (0.368 ppm, 0.060 mg)	Sulforhodamin (2.469 ppm, 0.33 mg)	Uranine (379.9 ppb, 0.13 mg)	Uranine (283.8 ppb, 0.038 mg)
Pumping rate (injection/ extraction [ml/min]	~ 7.5 ~ 2.1	~ 1.1 ~ 0.9	~ 1 ~ 0.9	~ 0.92 ~ 0.76	~ 0.75 ~ 1.4	~ 0.95 ~ 1.74	~ 0.75 ~ 0.86
Tracer injection Δt [h]	0.75	24	12	~ 3	~ 3	~ 3	~ 3
Time for experiment [h]	100	160	300	670	596	457	813
Peak arrival time [h]	6.1	36	23	18	33	158	11.3
Max. conc., extracted [ppb]	16	92 {~ 920}	126 {~ 630}	8.4	8.9	1.4	7.4
Recovered tracer	~ 15% during experiment	~ 40% { ~ 40%}	47% {40%}	52%	33%	48%	59%

[a] All tracers that have been used are fluorescent dyes, whereas UV-1 and T3 (recently developed) have their excitation and emission wavelength in the UV-range.

performed in the MI shear zone. And second, we used the two-dimensional code MCOTAC (Pfingsten, 1996, 2001) describing reactive transport in a (heterogeneous) porous medium. For both modelling approaches, the experimental tracer injection is approximated by step-pulse injection of a tracer during a certain time (top-hat) under steady-state hydraulic conditions. A priori, it is not obvious which approach is more appropriate, although, the double porous medium approach was successful in the experiments in the MI shear zone. The application of both model concepts—dual porous and heterogeneous porous medium—allows for a complementary description of such experiments and an estimation on their applicability to different dipole flow fields (narrow or wide, depending on the pumping rates). For modelling exercises, the last four dipole experiments (4–7, Table 1) have been used. They are characterised by a long duration and a tracer step input of about 3 h. The dipole experiment 4 provides a breakthrough curve, which was fitted by RANCHMD/PICNIC and MCOTAC. The parameters obtained by fitting are used to reproduce the remaining dipoles 5, 6 and 7. Both conceptual models are applied, changing only the distance between the individual boreholes and the applied pumping rates. Parameters like diffusion coefficient, dispersion length, fracture aperture or the porosity (hydraulic conductivity) distribution have been kept constant in order to investigate how representative the transport parameters from single dipole experiments are.

3.1. Double-porosity approach

Following the modelling of the MI experiments performed at Grimsel test site (Hadermann and Heer, 1996; Heer and Hadermann, 1994; Heer and Smith, 1998) and several tracer dipole experiments at Äspö Hard Rock Laboratory (Jakob and Heer, 2001), the RANCHMD and PICNIC codes are used for modelling the HPF dipole tracer experiment. RANCHMD approximates tracer transport (advection and dispersion) to take place in the shear zone in planar open fractures (open channels in the fault gouge material, or in veins) in one dimension. Water flow velocity in the fracture is a mean value for a stream tube within the dipole flow field. Longitudinal mechanical dispersion reflects the variability of the fracture aperture and fracture connectivity along the stream tube. In addition, diffusion into the accessible rock matrix is considered perpendicular to the fracture. Reversible sorption on the surfaces of the fracture wall and on surfaces within the matrix can be taken into account. In addition, the PICNIC code allows for fracture network transport calculations, including multiple flow paths. These two codes do not include a flow model. Flow velocities in the fracture(s) have to be given in advance of the transport calculation and only a few independent parameters can be modified for each single flow path. The calculated breakthrough curves can be adjusted to fit the measured ones taking into account characteristic times typical for the dual porous medium approach. These are the advection time, $t_{\text{adv.}}$, the matrix diffusion time, τ_0, and the time for the tail end perturbation, γ, which is visible in the breakthrough curve (Jakob, 1997).

$$t_{\text{adv.}} = \frac{R_{\text{f}} L}{v_{\text{f}}} \qquad \tau_0 = \left(\frac{L}{v_{\text{f}}}\right)^2 \left(\frac{\varepsilon_{\text{p}}}{2b}\right)^2 D_{\text{p}} R_{\text{p}} \qquad \gamma = \frac{1}{5} d^2 \frac{R_{\text{p}}}{D_{\text{p}}}.$$

Table 2
Comparison of transport parameters[a] used to model dipole experiments performed in the MI and the neighbour HPF shear zone at Grimsel Test Site

Experiment	Transport parameter						
	v_f [m/year]	Q_{ex}/Q_{in} [ml/min]	$2b$ [m]	Pe [−]	D_p [m^2/s]	ε_p [−]	d [m]
MI (Uranine)	17 181	10/150	9.3×10^{-5}	19.6	2.5×10^{-11}	0.062	5×10^{-2}
HPF tracer, PICNIC (two flow paths with different transport properties)	500, 53	~ 0.76/ ~ 0.92	5×10^{-5}, 1.43×10^{-4}	30	2.5×10^{-11}	0.062	0.25×10^{-2}, 0.6×10^{-2}
HPF tracer, MCOTAC	variable flow field	~ 0.76/ ~ 0.92	porosity distribution	Pe_{grid}>10	2.5×10^{-11}	porosity distribution	–

[a] Q_{ex} and Q_{in} are the extraction and the injection rates, respectively. Pe ($= v_f L/D$) is the Peclet number defined by the pore velocity, v, the length of the model domain, L, the dispersion coefficient, D ($D = \alpha_L v_f$, α_L is the dispersion length). The grid Peclet number is given by $Pe_{grid} \cong \Delta x/\alpha_L$. $2b$ is the fracture aperture, D_p is the pore diffusion constant, ε_p is the diffusion-accessible matrix porosity and d is the matrix thickness.

R_f is the distribution coefficient for surface sorption at the fracture walls, L is the fracture length, v_f is the pore velocity in the fracture, ε_p is the matrix porosity, $2b$ is the fracture aperture, D_p is the pore diffusion coefficient, R_p is the retardation factor and d is the thickness of the matrix accessible by diffusion. In case of several independent flow paths, i, with individual transport parameters, a superposition of the individual paths will contribute to the calculated breakthrough. Additional parameters can be used to weight the individual flow paths. It should be noted that a single good fit is not unique because several combinations of parameter sets for L/v_f, D_p, b, ε_p and d may lead to equivalent fits. Although knowing that these modelling approaches are a simplification with respect to the real flow field of the dipoles, modelling in a manner analogue to the MI experiments allows for parameter comparison. Parameters were chosen in analogy to those used in the MI experiments and modified by the hydraulic properties of dipole experiment 4 (smaller pumping rates for the HPF tracer experiments generally cause smaller pore water velocities in the fractures, see Table 2). The results are shown in Fig. 2a.

3.2. Two-dimensional heterogeneous porous medium approach

The two-dimensional reactive transport code, MCOTAC (Pfingsten, 1996, 2001), was applied to fit the tracer experiments and to allow for predictions for dipole set-ups of the long-term reactive transport experiment. Its application seemed necessary because initial RANCHMD modelling of dipole experiment 4 by a single flow path was not very successful, and secondly but more importantly, because an extended dipole flow field is expected for the applied dipole. MCOTAC takes into account advection, dispersion and diffusion in a two-dimensional heterogeneous porous medium of variable thickness in the third dimension in space corresponding to the plane of the shear zone. In addition, it includes a finite difference flow model. Matrix diffusion is not explicitly taken into account.

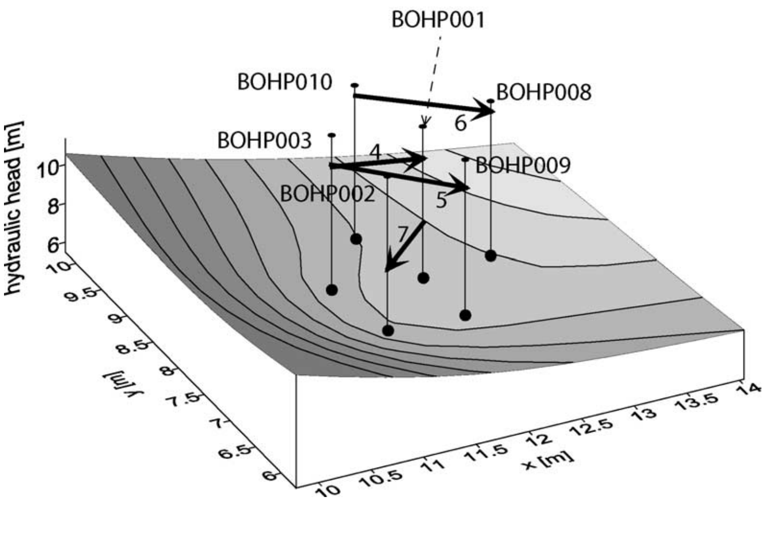


Fig. 1. Hydraulic head distribution within the HPF shear zone by kriging interpolation from values measured in the packer intervals of the boreholes.

Several approximations have been made to fix the model domain and the hydraulic and transport boundary conditions. The interpolation of the hydraulic heads that have been measured in 10 boreholes intersecting the shear zone (Fig. 1) provides an estimate for the hydraulic background flow, showing a general water flow to the tunnel. Similarly, the thickness of the shear zone has been interpolated from 10 measurements in the boreholes to the finite grid, representing the thickness of the two-dimensional heterogeneous water conducting shear zone. In the domain of the dipole experiments, the hydraulic gradient is in the direction from borehole BOHP003 to BOHP001, which correspond to injection and extraction for dipole 4, respectively. The model domain was chosen to include an area larger than the dipole dimension, assuming a constant hydraulic head boundary condition upstream and downstream of the dipole. Pumping rates have to be given at the locations of the boreholes. It is also assumed that the dipole has negligible influence on the hydraulic heads at the domain boundaries.

The hydraulic conductivity was assumed to be related to the porosity by a nonlinear function (Kozeny–Carman equation; Bear, 1972). Starting from a homogeneous porosity (and hydraulic conductivity) distribution in the model domain, which was justified by the transmissivity measurements in the six boreholes in the vicinity of the dipoles, the porosity

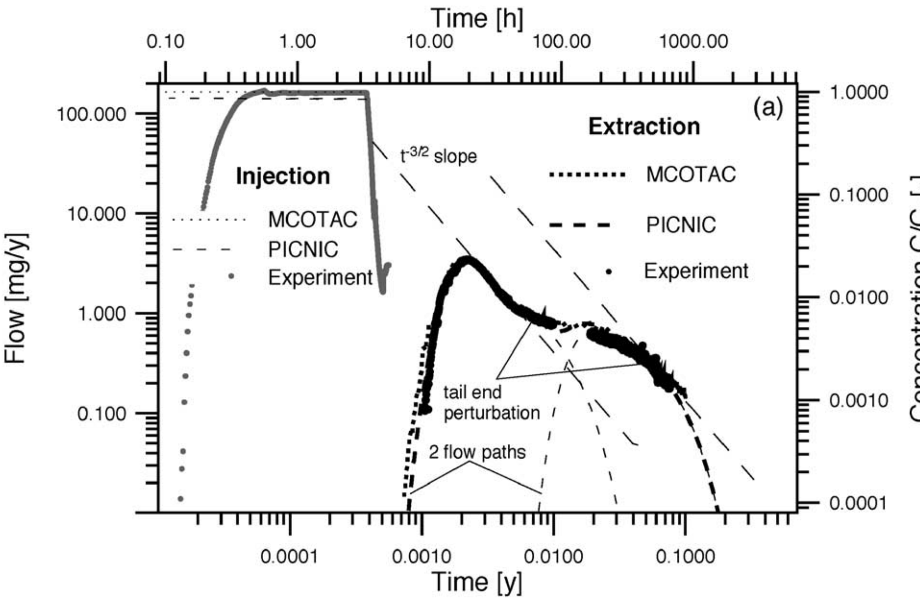


Fig. 2. Measured and calculated breakthrough curves for tracer dipole experiments performed in the HPF shear zone. (a) Fit of PICNIC (superposition of two flow paths) and MCOTAC to dipole experiment 4. (b) PICNIC (solid lines) and MCOTAC (dashed lines) calculations for dipole experiments 5 (b1), 6 (b2) and 7 (b3) using calibrated transport parameters from the fit to dipole experiment 4 (indicated by square symbols). The transport parameters have been modified according to the differences for the geometry of the individual dipoles, the pumping rates applied and the injected tracer masses. (c) Prediction for the breakthrough of a sorbing tracer (Retardation factor $R = 2$) for dipole 4 hydraulic conditions calculated by PICNIC and MCOTAC in comparison to calculations for the non-sorbing tracer breakthrough.

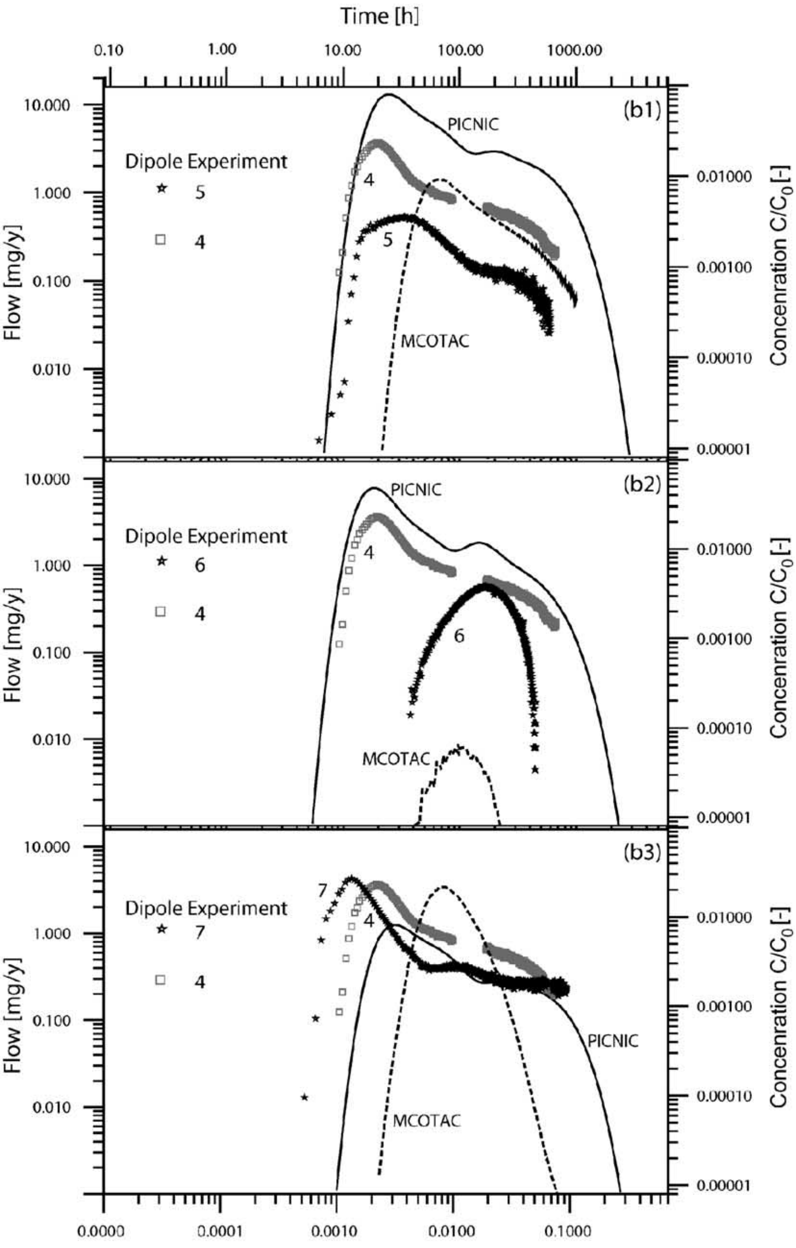


Fig. 2 (*continued*).

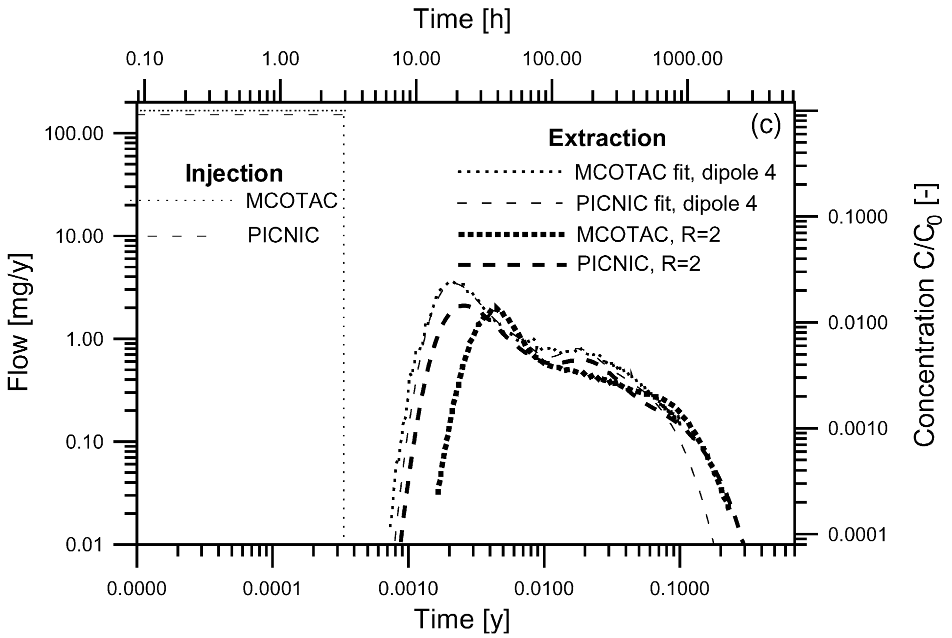


Fig. 2 (*continued*).

distribution in the model domain and the grid-specific dispersion coefficients are used to fit the measured breakthrough curves. The values for the local porosity vary from less than 0.1% to more than 20% at the location of the packer intervals of the boreholes. The hydraulic modelling results in a hydraulic head distribution and a related steady state pore velocity field. Local pore velocities vary over several orders of magnitude in the model domain up to more than 100 m/year for the fit shown in Figs. 2 and 3. Tracer transport is calculated by random walk of multispecie particles (Pfingsten, 1996, 2001) allowing for chemical reactions and sorption at equilibrium. The latter can be described by a retardation factor R.

$$R = 1 - \frac{1 - \varepsilon_{\mathrm{p}}}{\varepsilon_{\mathrm{p}}} \rho K_{\mathrm{D}},$$

where ρ is the solid density and K_{D} the distribution coefficient for sorption [l/kg]. In addition, more complex sorption processes can be taken into account by MCOTAC if the sorption processes are known. Also here, it should be noted that the fit of dipole experiment 4 is not unique. The discretisation of the model domain and the introduction of local, grid-specific fit parameters for the hydraulic and transport modelling allows for a huge number of variables. Depending on the discretisation of the model domain and the distribution of local values, e.g., the porosity, several porosity distributions would fit the measured breakthrough of dipole experiment 4. However, values or the distribution of these variables are not easy to deduce from experiments.

4. Discussion

Fig. 2a shows the model fits of the two different model concepts for dipole experiment 4 (Table 1). As mentioned above, the PICNIC calculation does not include a hydraulic model. Therefore, the injection of the tracer pulse has to be modified in advance. Due to the ratio of extraction to injection rate, $\beta \cong 0.8$, the tracer injection has been scaled by a factor of 0.8 compared to the experiment. It takes into account that for steady state hydraulic conditions not all injected fluid and tracer mass can be extracted—tracer will be lost. The parameters used to fit the breakthrough curves are in the same range as reported in Heer and Smith (1998) for the MI experiments, except that the water flow velocity in the open fracture, v_f, was assumed to be much smaller here (see Table 2). For the MCOTAC calculation, including hydraulic calculations, this scaling is done "automatically" because in the two-dimensional heterogeneous flow field some tracer will not reach the extraction borehole—it will bypass it probably in the direction of the tunnel. However, it was not possible to model the tracer experiment 4 by a fracture matrix double-porosity approach with a single flow path. At least a superposition of two flow paths is necessary for an acceptable fit, neglecting the overshooting at the peak arrival time for the second flow paths (Fig. 2a). The introduction of additional flow paths would improve the model fit by PICNIC calculations but it would also introduce an additional set of parameters. With MCOTAC, a similar acceptable fit was produced, even though matrix diffusion was not explicitly taken into account. The hydraulic model was set-up as described above. Variation of the porosity distribution (and related hydraulic conductivity) in the near-field of the applied dipole leads to the fit shown in Fig. 2a, where the dipole flow field has been adjusted to achieve the fit. The flow field is shown in Fig. 3. Although the boreholes used to set-up the dipole 4 are close together (0.77 m) and the hydraulic testing, performed in advance of the tracer dipole experiments, did not show large heterogeneity for the transmissivity of the shear zone, only a quite heterogeneous porosity and related hydraulic conductivity distribution leads to a reasonable fit. Here, it should be noted that the hydraulic tests were integrated measurements over a certain volume within the shear zone, averaging the properties over the measurement volume and, therefore, did not show large heterogeneity in the 4-m^2 dipole near field. The tracer experiments, however, indicate a quite heterogeneous shear zone with respect to porosity, hydraulic conductivity or fracture flow paths. Unfortunately, it was still not possible to discriminate between these two transport processes. For a double-porosity model approach, matrix diffusion is characterised by a $t^{-3/2}$ slope in the breakthrough curve (Hadermann and Heer, 1996), as well as by a tail end perturbation in case of limited matrix diffusion indicated in Fig. 2a. The $t^{-3/2}$ slope in the tracer breakthrough curve is obvious in dipole experiments 4, 5 and 7. But this characteristic feature for transport in a double-porosity medium is also reproducible by a two-dimensional heterogeneous porous medium approach as shown in Fig. 2a. Also, the tail end perturbation, calculated by the double porous medium approach, could not be observed or deduced from the experiments to get an estimate for the matrix thickness. The question, if the extended heterogeneous dipole flow field and/or matrix diffusion are responsible for the tracer breakthrough, is still open. Experiment 4 shows a complex tracer breakthrough. The same is valid for experiments 5, 6 and 7 keeping in mind the overlap of their dipole flow fields and similar pumping rates. Due to an increasing background signal at the end of the experiment, the measured

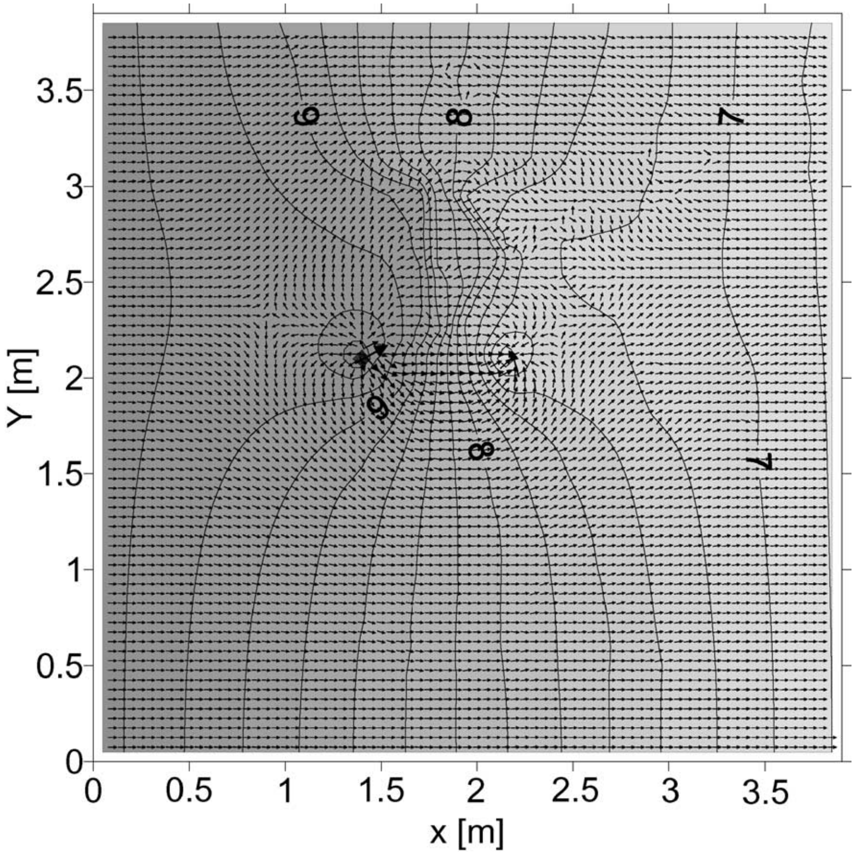
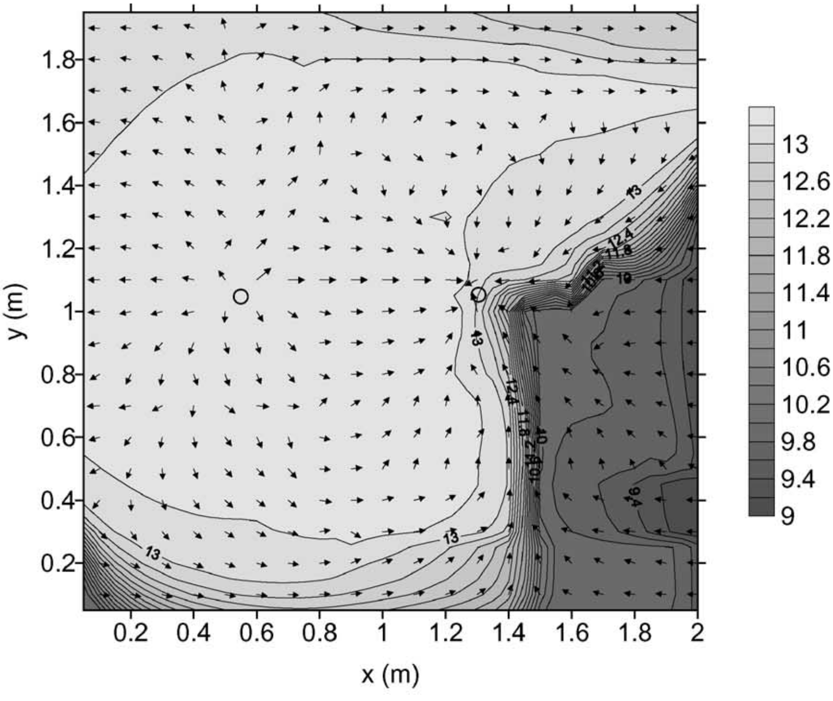


Fig. 3. Hydraulic head distribution (labels 7, 8, 9 are in m) and flow field calculated by MCOTAC. The porosity distribution was adjusted on a 80×0.05 m by 80×0.05 m grid to fit dipole experiment 4 (directed from BOHP003 to BOHP001).

breakthrough becomes too scattered to definitely identify a tail end perturbation. It becomes obvious that it is difficult to deduce transport parameters within a small range of uncertainty with this experiment. Furthermore, the uncertainty in geometrical parameters leaves an open question: Does transport in the HPF shear zone take place in open fractures or does it take place in a heterogeneous porous medium? (Borehole images show a highly fractured HPF shear zone that might be looked at as a porous medium too.) Depending on these different transport properties, the results could be quite different. This becomes already obvious when looking at the dipole experiments 5, 6 and 7. Although their experimental set-up is similar to dipole experiment 4, their measured breakthrough curves are not

Fig. 4. pH distribution in the fracture after 1 year since the start of injection of the high pH solution. The circles show the location of the injection and extraction boreholes. The arrows show the flow field in the fracture. The two figures correspond to initial surface areas for primary minerals of the order of 10^6 m^2/m^3 rock (top) and 10^7 m^2/m^3 rock (bottom).


y (m)
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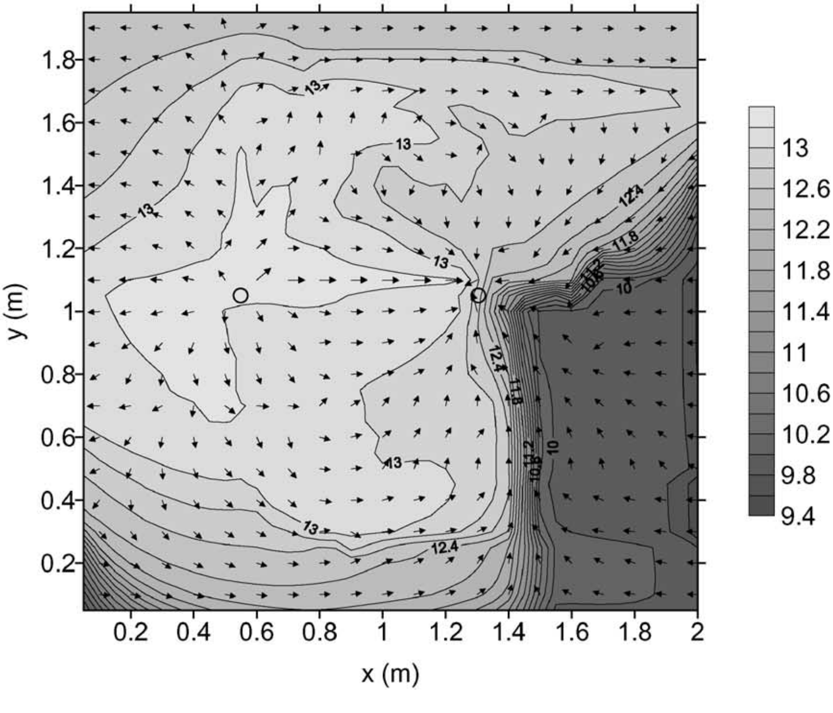
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predictable with the parameter set that fits dipole experiment 4. Both model concepts failed to reproduce, even approximately, the measured breakthrough curves with the parameter set used to fit experiment 4 (see Fig. 2b). Certainly, it would be possible to produce additional acceptable fits by both models; however, this would not enable us to predict another dipole more accurately. The shear zone seems to be too heterogeneous. Therefore, additional dipole experiments with other dipole geometry may increase the information about the heterogeneity but, and more importantly, dipole experiments with an additional sorbing tracer should be performed. As shown in Fig. 2c, the dual porous and heterogeneous porous medium approach give different breakthrough curves for the sorbing tracer. Peak breakthrough is calculated for a sorbing tracer ($R = 2$) by PICNIC at about 20 h and for MCOTAC at about 30 h, whereas there is no such remarkable difference for the rest of the breakthrough; especially, there is no difference in the tailing. In summary, the one-dimensional double-porosity (open fracture and adjacent rock matrix) and the two-dimensional heterogeneous porous medium approach have different characteristics in the breakthrough curves for sorbing tracers. This will allow the discrimination between dominant and negligible processes, e.g., heterogeneous flow field versus matrix diffusion. Eventually, the excavation and the analysis of the rock will provide additional information.

5. Predictive modelling

Two-dimensional reactive transport simulations have been performed making use of the calculated heterogeneous flow field for dipole experiment 4. The domain of the simulations is a square region of the fracture plane including the injection and extraction boreholes (Fig. 3). The discretisation has been changed to a coarser grid due to computing resources. A modified version of the GIMRT software package (Steefel and Yabusaki, 1996) has been used for the reactive transport calculations including kinetically controlled mineral reactions. Mineral reaction rates are calculated according to known/estimated reaction rate laws. The rates for primary minerals are based on experimentally determined rates published in the literature. Mineral surface areas (ranges of values) are calculated from specific surface areas (BET) measured on the fine-grained fault gouge filling the fractures and geometric considerations. Fast rates have been used for the secondary minerals so the results resemble local equilibrium for these phases.

The interaction between the injected hyperalkaline solution and the rock (fault gouge filling the fracture) causes a series of mineral reactions. The main processes are the dissolution of albite and precipitation of tobermorite (CSH phase), prehnite (CASH phase) and mesolite (Na–Ca zeolite) close to the injection borehole, and the precipitation of natrolite and analcime (Na-zeolites) farther away down the flow field. Fig. 4 shows the pH distribution in the fracture after 1 year since the start of injection of the high pH solution (the planned duration of the experiment) for the hydraulic dipole set-up used for dipole experiment 4. The two plots correspond to two different values of the initial surface areas of the primary minerals. The flow field was taken from an MCOTAC calibration of dipole experiment 4, but it was not updated during the calculations (Darcy velocities were assumed to be constant with time). Even with these simplifications, the results show that a large degree of spatial heterogeneity is expected, given the heterogeneous nature of the flow field. Also, the amount of reaction (reflected in the figure by changes in pH) is greatly dependent on the available reactive surface area.

6. Conclusions

Single and double-porosity models have been applied to fit the tracer breakthrough in order to extract values for some transport parameters for two different model approaches, optimise for the long-term experiment and discriminate for certain processes. The one-dimensional dual porous medium approach and the two-dimensional heterogeneous porous medium approach succeeded to fit dipole experiment 4, the dual porous medium approach using multiple flow paths. Although, the distance between injection and extraction boreholes was only about 1 m, measured breakthrough could only be fitted by the two-dimensional heterogeneous porous medium approach with a complex flow field geometry assuming heterogeneous porosity and hydraulic conductivity distributions within the HPF shear zone. Attempts to model all dipole tests with a similar model parameter set, except for dipole geometry and applied pumping rates, failed. The reason seems to be the heterogeneity of the shear zone, even in the small volume of the shear zone where all the dipole experiments have been performed. This indicates that for both model concepts the heterogeneity has to be taken into account, at least to fit all individual dipole experiments. In addition, both model concepts have been applied to predict the breakthrough curve for a sorbing tracer. Using a non-sorbing and a sorbing tracer in the same experiment could allow for the discrimination of transport processes in such an extended dipole flow field because the modelling showed quite different breakthrough curves when calculated by both model concepts.

The two-dimensional heterogeneous porous medium approach was applied to predict the reactive transport of a high pH solution. The spreading of the pH solution was calculated according to the heterogeneous flow field fitted to a dipole experiment. It is strongly influenced by the available reactive surface area. Similar patterns are possible for the foreseen injection of radionuclides together with the high pH solution.

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