EXPERIMENTAL AND NUMERICAL INVESTIGATIONS OF MATRIX DIFFUSION IN A FRACTURED SANDSTONE BLOC

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Abstract - Exploited coal seams are used to deposit residues from thermal energy and waste incineration plants at North Rhine Westphalia, Germany. Such an underground repository requires the proof that dissolved contaminants may not reach the biosphere. Numerical models may be used as tool to predict the local and temporal migration of pollutants. In order to investigate the migration of heavy metals in a porous sandstone bloc containing a single fracture two tracer experiments were performed under precisely defined hydraulic flow conditions. The tracer experiments were performed applying pyranine, cadmium and lead as solutes and the hydrochemical conditions correspond to a depth of 1000 m below ground surface. For the numerical model the fracture was considered as a two-dimensional parallel plate conduit and the rock matrix was treated as a three-dimensional porous medium. Together with rock properties determined in laboratory the model yielded a good agreement between measured and simulated breakthrough curves by calibrating the fracture aperture and longitudinal dispersion.

Keywords - underground repository, fractured sandstone, migration experiment, numerical model

INTRODUCTION

During the early 80's the problem of hazardous solutes, which might return from underground radioactive repositories back to the biosphere, attracted a lot of interest. Important results were gained at STRIPA in Sweden (Abelin *et al.*, 1991), FANAY-AUGÈRES in France (Cacas *et al.*, 1990) and at GRIMSEL in Switzerland (Lieb, 1993). In the early 90´s

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the concept of subsurface storage of industrial residues from electric power plants in coal mines at North-Rhine-Westphalia (Germany) became an alternative to surface waste dump sites (Jäger *et al.* 1990). The residues consist of filter ashes and desulphurization residues, which are injected as hydraulic suspension into exploited coal seams. Numerical

Simulations showed that dissolved contaminants might only return to the biosphere when the mines are closed and the pumping of groundwater is stopped (Himmelsbach & König, 1997; König & Himmelsbach, 1997).

Flow processes in fractured rocks were considered first to occur in a conduit of parallel plates separated by a constant aperture (Snow, 1969). Experimental (Witherspoon *et al.*, 1980) and theoretical investigations showed that this simplification might be inadequate for flow and transport processes in fractured rocks due to channeling processes (Gentier *et al.,* 1989; Neuzil & Tracy, 1981; Rasmuson & Neretnieks, 1986; Dykhuizen, 1992; Tsang & Tsang, 1987). The most studies, however, neglected the influence of matrix diffusion. A numerical model for transport in a one-dimensional fracture with diffusion into the matrix was presented first by Grisak & Pickens (1992), while the transport equation and analytical solution for this problem was published by Tang *et al.* (1981). This paper describes a bench scale experiment that addresses both effects, matrix diffusion and advective-dispersive transport in the fracture. In addition a 3D-model based on the finite element method can be verified using the experimental data from the experiment.

EXPERIMENTAL ARRAY

In order to investigate the migration behavior of heavy metals two migration experiments using cadmium and lead as solute and pyridine as tracer were performed in the laboratory under precisely defined hydraulic boundary conditions. The hydrochemical conditions of migrating pore fluid correspond to a depth of approximately 1,000 m below ground surface. The sandstone bloc has a length of 24 cm, a height of 24 cm, a width of 21 cm and the single fracture lies exactly in the middle of the bloc. A small plastic ribbon having a thickness of 350 µm separates the two halves of the bloc. By this method the fracture aperture could be fixed to a definite and constant value. The matrix porosity was determined applying the mercury porosimetry. Since the tracer experiments should investigate advective-dispersive fracture transport processes affected by matrix diffusion, any superposition of this process by advection due to an unsaturated matrix had to be avoided. The experimental setup was therefore saturated with artificial formation water that corresponds to the depth of the disposal site (Himmelsbach & Wendland, 1999). During the tracer experiments the synthetic formation water was pumped at a constant flux rate of Q=4.57 ml/h through the fracture plane using a peristaltic pump (Fig. 1). The injection lasted 1 min and was performed with a syringe through the septum at the lower injection point. Due to sealed areas an irregular flow pattern in the fracture plane was expected. The correct description of the transport process required therefore a 3D-model.

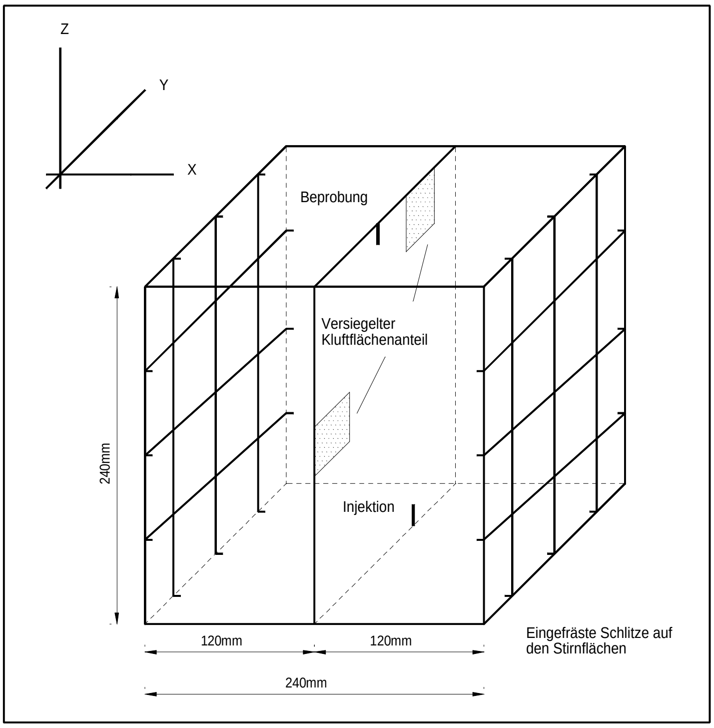


Figure 1: Schematic representation of the sandstone bloc

NUMERICAL MODEL

The model considers single-phase flow through a homogeneous, saturated, fractured porous medium under steady state conditions. For the coupled model including porous and fracture flow the mass balance is given as:

$$\Delta \boldsymbol{q} = Q \qquad (1)$$

where $\boldsymbol{q}$ is the specific discharge and Q represents the source terms (Bear, 1979). Assuming an isotropic and homogeneous medium, the specific discharge can be given by:

$$\boldsymbol{q} = -K \frac{rg}{h} \left(\frac{p}{rg} + z \right) \qquad (2)$$

in which K is the permeability, r is the fluid density, g is the gravity acceleration, h is the dynamic viscosity, p is the hydrostatic pressure and z is the vertical position. For porous media the permeability K is determined by percolation experiments. Assuming the fracture as a conduit defined by parallel plates, the permeability K for the fracture is given by (Berkowitz & Bear, 1987):

$$K = \frac{a^2}{12} \tag{3}$$

in which a is the aperture of the fracture.

The transient transport of dissolved solutes is governed by the advection-dispersion equation (Bear, 1979):

$$\frac{\partial c}{\partial t} + \frac{\boldsymbol{q}}{n}\nabla c - \nabla(\boldsymbol{D}\nabla c) = Q\left(c^* - c\right) \tag{4}$$

where c is the solute concentration, t is the time, $\boldsymbol{q}$ is the specific discharge, $\boldsymbol{D}$ is the hydrodynamic dispersion tensor, containing the mechanical dispersion and molecular diffusion, c^* is the solute concentration of the sources and n is the constant effective porosity in the blocs. For the fractures, n is set equal to 1.

The equations describing flow and transport in the fractures and in the matrix are coupled by the continuity law and the flow equation (1) is solved using the finite element method. For the transport equation (4) the **S**ymmetrical **S**treamline **S**tabilization scheme is used (Wendland & Schmid, 1996). A 3D-model combines 2-D elements for the fracture and 3-D elements for the porous matrix (Fig. 2).

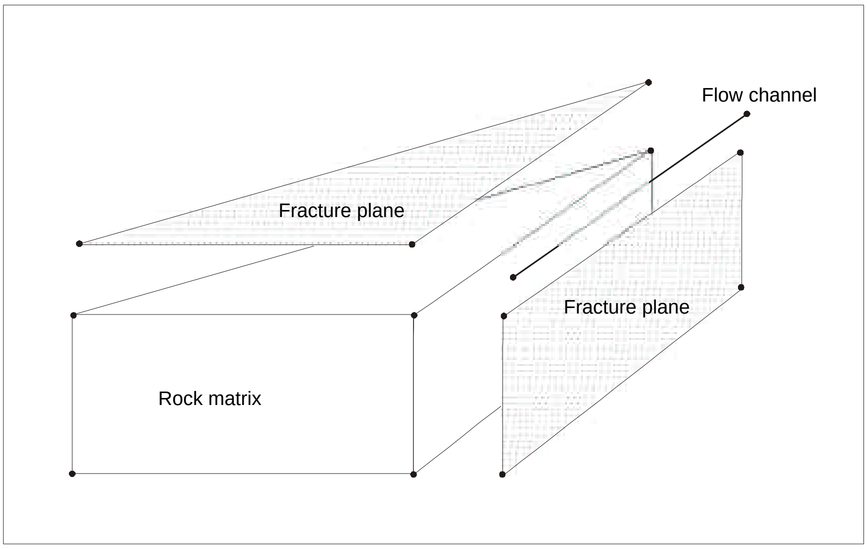


Figure 2: Combination of different element types in a 3D-model

PHYSICAL PARAMETERS

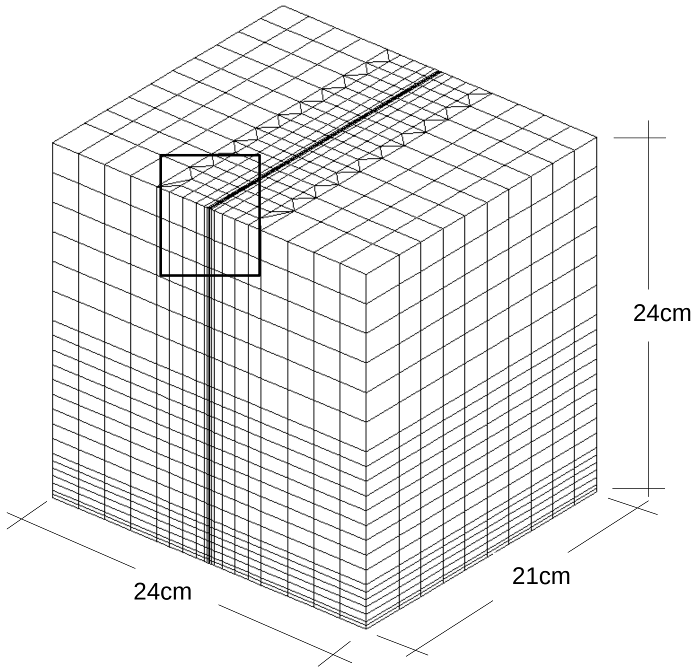
The defined fracture opening was achieved by a thin plastic film between both bloc halves. Considering the roughness of the bloc surfaces the effective hydraulic aperture was committed to 507 µm for the numerical model of the sandstone bloc. From diffusion cell experiments using similar rock samples the pore diffusion coefficient for pyranine was determined to D_p=2*10^{-7} cm^2/s (Harnischmacher, 1996). Since the model of the sandstone bloc considers only a smooth opening between parallel plates such a simulation would underestimate the effect of matrix diffusion, because the fracture roughness causes an increased exchange surface. The diffusion coefficient used in numerical simulation was therefore increased to D_p=5*10^{-7} cm^2/s. The matrix porosity of the sandstone bloc was determined to an average of 7.2% with a maximum amounting to 8,7 %. Due to the fact that the rock porosity increases in the direct vicinity of fractures (Frick, 1993) an effective porosity of 8.5% was assumed. With regard to the sharp breakthrough curve the longitudinal dispersivity was set to α_L= 0.6 cm which resembles in sense of numerical stability a lower limit. The injection occurs as Dirac pulse directly into the fracture. In order to avoid numerical oscillation an input function was generated which elongates the injection to a time interval of 6 min. Compared to the duration of the experiment of 60 h it can still be regarded as Dirac pulse and the numerically generated mass of 32.2 µg equals the injected tracer mass. The physical parameters are summarized in Table 1.

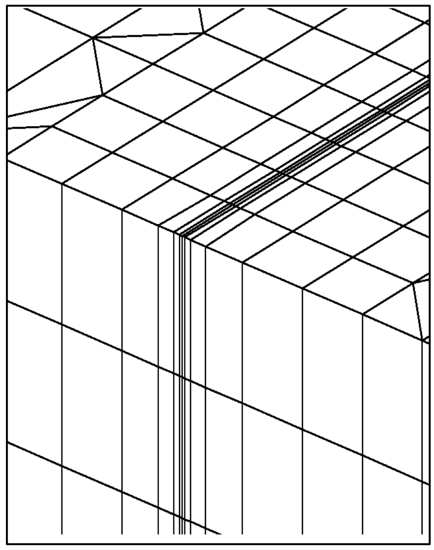
Table 1: Parameters used for the numerical simulation

Parameter	Value	Unit	Source
Flow-Simulation			
Flux rate	4.57	ml/h	measured
Matrix conductivity	$1*10^{-9}$	m/s	measured
Fracture opening	507	µm	calibrated (>350µm)
Transport-Simulation			
Tracer mass	32.2	µg	measured
Injection volume	0.307	ml	measured
Matrix porosity	8.5	%	measured (7.2%)
Diffusion coefficient in the matrix	$5*10^{-7}$	cm^2/s	measured ($2*10^{-7}$ cm^2/s)
Long. dispersivity in the fracture	0.6	cm	assumed

DISCRETIZATION

Since the flow velocity in the fracture is high and the concentration profiles along the fracture are sharp, the model needs a fine grid spacing to avoid numerical oscillation (Fig. 3).





Enlarged detail view

Figure 3: Spatial discretization of the sandstone bloc

The experimental array is vertically discretized into 22 layers. Near the injection point with the highest velocity and sharpest solute concentration front, the element spacing amounts therefore to only 0.25 cm. In direction of flow (from the bottom to the top) the spacing of the elements is increasing and the uppermost layer has a thickness of 4.00 cm. Compared to the fast transport in the fracture, the diffusion processes in the sandstone matrix occur considerably slower. In order to address this context numerically, the spatial discretization of the rock matrix normal to the fracture was chosen to follow a logarithmic increasing grid spacing. Hence, the first nodal row is located at merely 0.002 cm parallel to the fracture interface and the subsequent nodes are at distances of 0.005 cm, 0.14 cm and 0.40 cm respectively. Due to the slow propagation of the diffusion front into the rock matrix all further nodes parallel to the fracture are located at constant distances of 1 cm. The resulting mesh used for the simulation has 9108 nodes, which are interlaced to 8668 3D-elements describing the matrix and 435 2D-elements for the fracture. In order to account for the sealed fracture areas, the corresponding 2D-elements were deleted from the mesh.

The time discretization for this transport simulation was chosen according to the COURANT criterion. Considering a maximal flow velocity of approximately 2.5*10^{-4} m/s at the injection point and the local spatial discretization of 0.25 cm a maximal time step length of Δt=20s was applied.

RESULTS

Two multi-tracer experiments were performed. During the first one pyranine and cadmium were injected simultaneously and the second one was performed with pyranine and lead. The breakthrough curve of cadmium was nearly the same as for pyranine. Also the time needed for first detection at the sampling port as well as the time elapsed to reach the maximum concentration were nearly the same (Fig. 4). Hence, under high saline pore water conditions the heavy metal cadmium seems to be highly mobile and shows no retardation effect. Under the same hydrochemical conditions, which are characterized by a salt content that is three times higher than seawater, the heavy metal lead was absorbed to such an extent that no breakthrough curve could be measured.

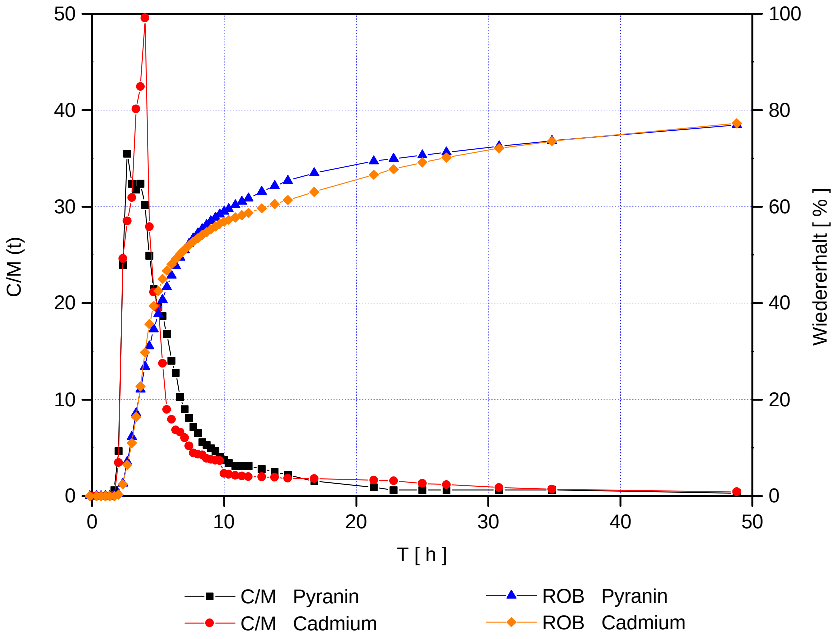


Figure 4: Measured breakthrough curves and recovery rate at the outflow point of the sandstone bloc for cadmium and pyranine

The numerical simulation was done with the computer code SICK100 (Schmid *et al.*, 1991) and covers a time period of 11h divided into 2000 time steps. Due to the fine spatial discretization it required approximately 1.6 CPU hours on a DEC 500/333 workstation. The breakthrough curves for some selected nodes along the flow direction in the fracture are shown in Figure 5. The sharpest concentration profile occurs at a distance of 0.4 cm from the injection point (node C) and becomes more elongated for other nodes lying at increasing distances of 0.9 cm to 16.0 cm (nodes E and F respectively). The maximum distance of 24.0 cm corresponds to the sampling point (node B). The influence of dispersion and matrix diffusion becomes evident.

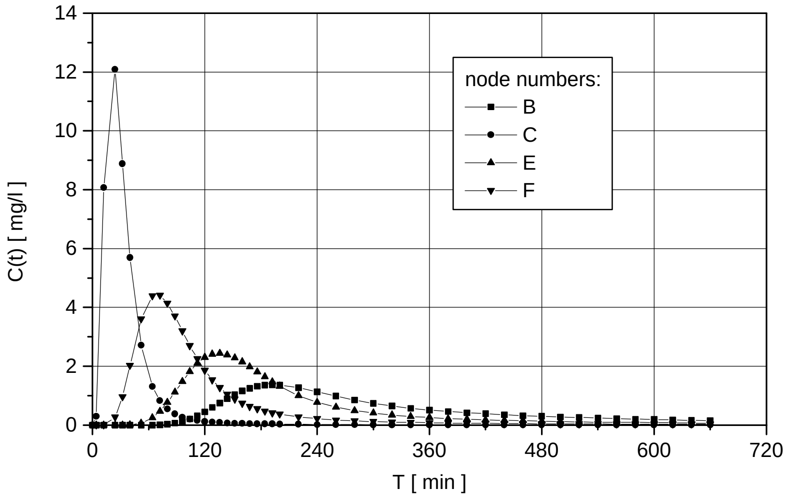


Figure 5: Breakthrough curves at different nodes along the flow path

In order to assess the quality of the numerical simulation it is necessary to compare the simulated breakthrough curve with the one determined experimentally (Fig. 6). The point of maximal concentration is not exactly reproduced but the simulation resembles the experimental breakthrough curve in its whole and in particular the long tailing effect due to matrix diffusion. To the end of the experiment, the injected tracer mass was not discarded completely reaching a recovery rate of only 80%.

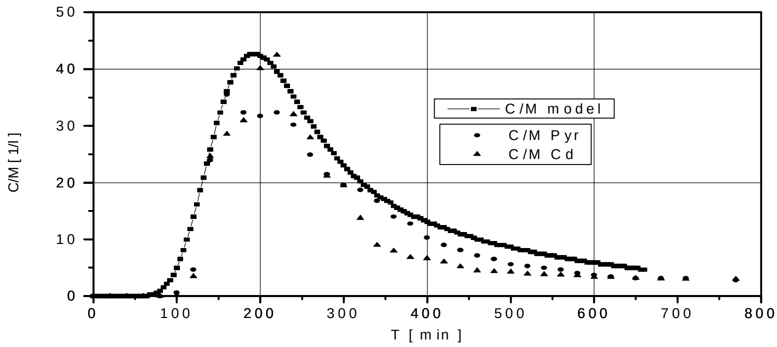


Figure 6: Simulated breakthrough curves compared to measured values
at the outflow point of the sandstone bloc

The remaining concentrations in the fracture are shown in Figure 7. The sealed fracture areas (see also Fig. 1) act as a barrier forming a pool of stagnant solutes. Behind this impermeable barrier the tracer concentrations are affected only by matrix diffusion. A second tracer experiment (Himmelsbach & Wendland, 1999) carried out with the same bloc produced a breakthrough curve with two maxims. This behavior occurred probably due to the remaining tracer mass from the first experiment. The good agreement between numerical and experimental curves could be achieved only by the appropriate description of the flow field with a three-dimensional model. Since the remaining residual deviations are due to the unknown texture of the fracture surface, a subsequent measuring of the fracture aperture and surface roughness might yield an even better match between experiment and simulation.

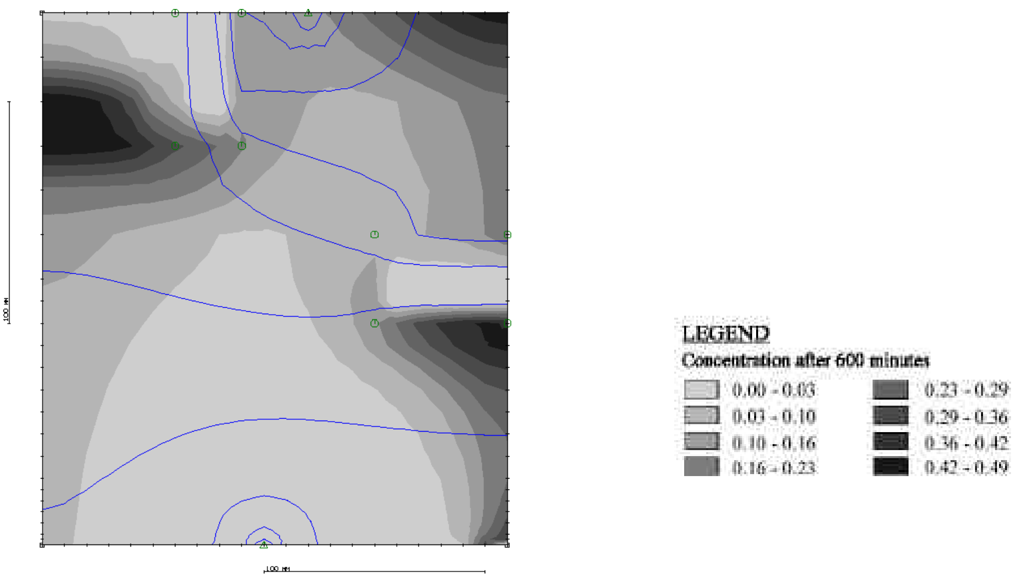


Figure 7: Concentration distribution within the fracture plane at the end of the tracer experiment

CONCLUSIONS

A laboratory experiment carried out under defined hydrochemical and flow conditions demonstrates that cadmium behaves as a conservative tracer under high saline water and neutral pH conditions. The observed breakthrough curve for cadmium in the single fracture experiment corresponds to the one obtained for pyranine leading to the conclusion that cadmium presents low sorption tendency in the considered formation water. Therefore numerical simulations viewing the risk analysis of an underground repository in a natural fracture system should not consider any effects of sorption and retardation in case of residues contaminated by cadmium (HIMMELSBACH & KÖNIG 1997, KÖNIG & HIMMELSBACH

1998). The adsorptive behavior of lead observed in this migration experiment agrees with results obtained by PAAS (1997) with batch experiments for typical sediments from the Ruhr Carbon area. Viewing the long time safety of underground repositories a strong lead adsorption as observed in this migration experiment can be considered a positive effect. For other heavy metals with adsorption disposition between cadmium and lead new migration experiments in laboratory or field scale are necessary in order to predict their transport behavior in a fractured host rock.

The detailed three-dimensional discretization of the sandstone bloc ensured a satisfactory simulation of the migration experiment. Previous simulations with a two-dimensional model did not lead to adequate results due to the complex flow field in the fracture plane. The closed experiment design with no flow over the boundaries leads to a variable flow field in the fracture. The velocity near the boundaries is very low leading to an increased tailing effect. In this case a one-dimensional analytical model can not describe the transport process correctly. Empirical approximations would be necessary. Due to the sealed areas in the fracture plane the real two-dimensional flow field could only be approximated by a three-dimensional model. A good agreement between measured and calculated breakthrough curves was obtained because the essential diffusion, sorption and porosity coefficients for the porous matrix had been previously determined in laboratory. An inverse simulation with these unknowns would not allow a unique solution. The comparison of measured and calculated breakthrough curves provides the verification of the numerical model assuring his applicability for real large-scale problems.

In a future work the authors will present results of numerical test simulations carried out with the same model for a coarse fracture surface. Stochastic generated fracture apertures approximate the natural roughness leading to a more realistic description of the physical problem.

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LITERATURE

Abelin, H., Birgersson, L., Moreno, L., Widén, H., Ågren, T. & Neretnieks, I. (1991): A Large-Scale Flow & Tracer Experiment in Granite: 2. Results and Interpretation. *Water Resour. Res.*, **27(12)**, 3119-3135.

Berkowitz, B. & Bear, J. (1987): Groundwater Flow and Pollution in Fractured Rock Aquifers. In: Novak, P. (1987): [Ed.]: Developments in Hydraulic Engineering. - *Elsevier Appl. Science Publ.*, **4**:175-238.

Bear, J. (1979): Hydraulics of Groundwater. New York, Mc-Graw-Hill Inc.

Cacas, M. C., Ledoux, E., de Marsily, G., Tillie, B., Barbreau, A., Durand, E., Feuga, B. and Peaudecerf, P. (1990): Modeling Fracture Flow With a Stochastic Discrete Fracture Network: Calibration and Validation 1. The Flow Model. *Water Resour. Res.*, **26(3)**, 479-489.

Dykhuizen, R. C. (1992): Diffusive Matrix Fracture Coupling Including the Effects of Flow Channeling. *Water Resour. Res.*, **28(9)**, 2447-2450.

Frick, U. (1993): Beurteilung der Diffusion im Grundwasser von Kristallingesteinen. Ein Beitrag zur Kristallinstudie 1993. Interner Ber., *NAGRA*, **92-92**: 84 p. NAGRA, Switzerland.

Gentier, S., Billaux, D. & van Vliet, L. (1989): Laboratory Testing of the Voids of a Fracture, Rock *Mechanics and Rock Engineering*, **22**, 149-157.

Grisak, G. E. & Pickens, J. F. (1992): Solute Transport Through Fractured Media 1. The Effect of Matrix Diffusion. *Water Resour. Res.*, **16(4)**, 719-730.

Harnischmacher, S. (1996): Ausbreitung von Stoffen in Kluft-Grundwasserleitern vor dem Hintergrund einer untertägigen Abfalldeponierung im Steinkohlengebirge. Diploma Thesis, University Bochum, Germany.

Himmelsbach, T. & König, Ch. (1997): Risk assessment of subsurface waste disposal in the near field of coal mines. - In: CHILTON, J. et al.: *Groundwater in the Urban Environment, Problems, Processes and Management*, **1**: 421 – 426. Netherlands.

Himmelsbach, T. & Wendland, E. (1999): Schwermetalltransport in Sandsteinen unter Bedingungen einer hochsalinaren Porenwasserlösung - Laborversuch und Modellierung . *Grundwasser*, 3(4), 103-112.

König, C. & Himmelsbach, T. (1997): Three-dimensional fracture analysis for risk assessment of subsurface disposal of industrial residues in coal mines. - 12th Intern. Conference on Computational Methods in Water Ressources; Kreta (Greece)

Lieb, R. W. (1993): Untersuchungen der Nagra für die Entsorgung radioaktiver Abfälle in der Schweiz. *Felsbau*, **11**, 13-16, 1993.

Paas, N. (1997): Untersuchungen zur Ermittlung der geochemischen Barriere von Gesteinen aus dem Umfeld untertägiger Versatzräume im Steinkohlenbergbau des Ruhrkarbons. – DMT-Ber. aus Forschung und Entwicklung 54, 234 S., Essen, Germany.

Rasmuson, A. & Neretnieks, I. (1986): Radionuclide Transport in Fast Channels in Crystalline Rock, *Water Resour. Res.*, **22(8)**, 1247-1256.

Schmid, G., Obermann, P., Braess, D., et al. (1991): SICK 1OO Berechnung von stationären und instationären Grundwasserströmungen und Stofftransport, Programmbeschreibung, Arbeitsgruppe Grundwassermodelle, Ruhr-Universität Bochum, Germany.

Snow, D. T. (1969): A Parallel Platte Model of Permeable Fractured Media, (Ph. D. Thesis), University of California, 1965.

Tang, D. H., Frind, E. 0. & Sudicky, E. A. (1981): Contaminant Transport in Fractured Porous Media: Analytical Solution for a Single Fracture. *Water Resour. Res.*, **17(3)**, 555-564.

Tsang, Y. W. & Tsang, C. F. (1987): Channel Model of Flow Through Fractured Media, *Water Resour. Res.*, **23(3)**, 467-479.

Wendland, E. & Schmid, G. (1996): Symmetrical Streamline Stabilization (S^3) for high advective transport processes, XI International Conference on Computational Methods in Water Resources, Cancun, Mexico.