# Non-linear adsorption in a multidimensional and double-porosity model of fractured rock solute transport

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**Abstract** We define a model of solute transport in fractured rock as a combination of the standard concepts into a multidimensional double-porosity. As addition to the previous work, we formulate the model with non-linear equilibrium adsorption in each domain. The numerical code uses the finite volume space discretisation and the operator splitting method in time to separate the complex processes. Test problems in 1D and 2D demonstrate the effect of non-linearity to the concentration profile and breakthrough curve shapes.

**Keywords** discrete fracture network; dual-porosity; immobile zone; non-linear adsorption; solute retention

## INTRODUCTION

The paper deals with conceptual models and numerical algorithms for solute transport in the fractured rock. The known difficulties are related to geometric complexity of fracture networks and resulting computational cost and parameter uncertainty. The modelling is based on several approaches; the typical are equivalent continuum, discrete fracture networks, and double-porosity models (Bear et al., 1993).

Recently we introduced a concept of combining network of large discrete fractures and double-porosity continuum representation of the remaining fractures – multidimensional double-porosity model. In this paper we introduce the process of adsorption to solid phase, with general non-equilibrium isotherm. Comparing to Gallo et al. (2006), we formulate and solve the mobile-immobile model with non-linear adsorption isotherm instead of linear.

There are no special numerical methods derived in literature for combination of the non-equilibrium exchange between mobile and immobile zone and the equilibrium non-linear adsorption. Using the operator-splitting method, known e.g. as a tool for separation of advection and diffusion, we can extract all the additional transport processes to be solved separately with simple analytical formulas (Hokr et al., 2003).

In the test problems, we demonstrate the qualitative difference between solute retention and concentration front retardation as consequence of either the transfer from the discrete fracture into the rock matrix, or the transfer from mobile to immobile water in a geometrically same place, or the equilibrium adsorption.

## MODEL CONCEPT AND GOVERNING EQUATIONS

We consider a system of 3D continuum, 2D discrete fracture network and a network of 1D fracture intersection. Each part is further composed of a mobile and immobile part (double continuum). We denote the problem domain $\Omega = \Omega_1 \cup \Omega_2 \cup \Omega_3$, $\Omega_i \subset R_3$, $i = 1,2,3$, where $\Omega_3$ is polyhedron (3D continuum), $\Omega_2$ is a system of planar polygons (discrete fracture network) and $\Omega_1$ is a systems of lines (intersections of fractures = "pipes"). The governing equations of flow are ($i = 1,2,3$)

$$\vec{u}_i = K_i \nabla p_i \quad in\Omega_i \tag{1}$$

$$\kappa_i \frac{\partial p_i}{\partial t} - \nabla \cdot \vec{u}_i = q_i^+ + q_i^- + \sum_{j=1, j\neq i}^{3} \frac{\tilde{q}_{ij}}{\mu_i} \quad in\Omega_i \tag{2}$$

$$\tilde{q}_{ij} = \sigma_{ij}(p_i - p_j) \qquad i \neq j$$

where the unknowns are $u_i(\vec{x},t)$ the velocity and $p_i(\vec{x},t)$ the piezometric head and the parameters are $K_i$ the hydraulic conductivity, $\kappa_i$ the storativity, $q_i$ sources/sinks, $\sigma_{ij}$ transmissivity between domains, and the variable $\tilde{q}_{ij}$ (positive from $j$ to $i$) denotes the flux between domains of different dimensionality. The parameter $\mu_i$ is an additional dimension: $\mu_1$ is cross-sectional area of 1D domain, $\mu_2$ is thickness of 2D domain, and $\mu_3 = 1$ (dimensionless). The governing equations of the mass transport are

$$n_i^{(m)} \frac{\partial\left(c_i^{(m)} + f_i^{(m)}(c_i^{(m)})\right)}{\partial t} = -\nabla \cdot (c_i^{(m)} \vec{u}_i) + \nabla \cdot (\mathbf{D}_h^{(i)} \nabla c_i^{(m)}) +$$

$$q^+ c^{(m)*} + q^- c^{(m)} + \sum_{j=1, j\neq i}^{3} \frac{\tilde{q}_{ij}^{(c)}}{\mu_i} + r_i^{(m)} - \alpha_i (c_i^{(m)} - c_i^{(im)}) \tag{3}$$

$$n_i^{(im)} \frac{\partial\left(c_i^{(im)} + f_i^{(im)}(c_i^{(im)})\right)}{\partial t} = r_i^{(im)} + \alpha_i (c_i^{(m)} - c_i^{(im)}) \tag{4}$$

where the unknowns are $c^{(m)}$ the concentration in the mobile zone and $c^{(im)}$ the concentration in the immobile zone, the parameters are $D_h$ the tensor of hydrodynamic dispersion, $c^*$ the injected concentration, $\tilde{q}_{ij}^{(c)}$ the auxiliary solute source/sink from the interaction between domains of different dimension, $\alpha$ is the rate of mobile-immobile exchange, $r_i^{(m)}$, $r_i^{(im)}$ the chemical reaction production terms,. The parameters $n^{(m)}$ and $n^{(im)}$ (porosities) represent the relative volume of mobile and immobile zone of the media, the subscripts $i, j$ denote the part of the domain with respective dimension. The adsorption term $f_i^{(m)}(c_i^{(m)})$ (and corresponding for the immobile zone) is arbitrary continuous non-decreasing function, e.g. the Freundlich isotherm can be written as

$$f_i^{(m)}(c_i^{(m)}) = \rho_i^s K_F c_i^{(m)a_F} \frac{\phi(1 - n_i^{(m)} - n_i^{(im)})}{n_i^{(m)}} \tag{5}$$

$$f_i^{(im)}(c_i^{(im)}) = \rho_i^s K_F c_i^{(im)a_F} \frac{(1-\phi)(1 - n_i^{(m)} - n_i^{(im)})}{n_i^{(im)}} \tag{6}$$

where $\phi$ is fraction of sorption surface between the mobile zone and the immobile zone and $\rho_s$ is the solid density (in the test problems below included in the $K_F$ value).

## NUMERICAL SOLUTION

The model is implemented in our simulation code FLOW123D with batch processing. The discretisation is realized with tetrahedrons in 3D and triangles in 2D. The fluid flow is solved using mixed-hybrid formulation of finite element method with pressure and velocity results, see also Královcová *et al.* (2006). The transport problem is solved with the finite volume method. We use operator splitting method for single transport processes separation (advection and mobile-immobile exchange), whose technical realisation employing an analytical solution of mobile-immobile exchange is described in Hokr *et al.*(2003).

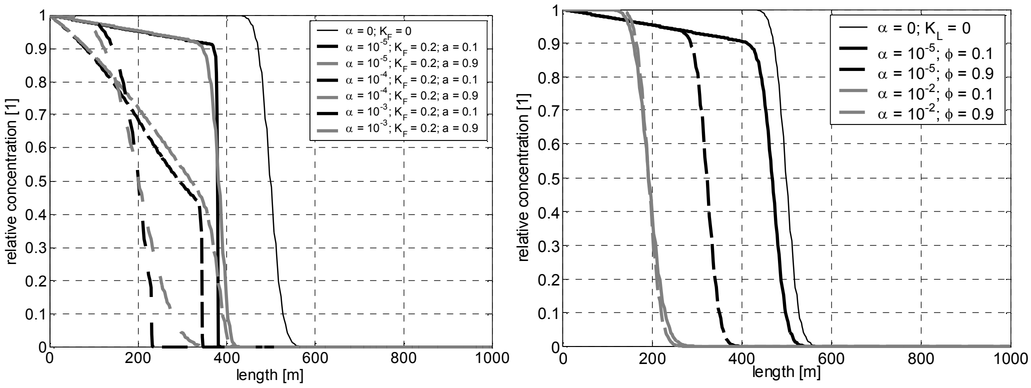


**Fig. 1** Profile of the concentration in the final time of simulation for the 1D test problem – effect of mobile-immobile exchange (rate $\alpha$), Freundlich sorption isotherm (exponent *a*), and the sorption fractioning $\phi$.

# MODEL PROBLEMS

## One-dimensional test problem

We consider the constant-concentration inflow problem to test the numerical algorithm. The problem is defined as 1000m long line, with constant flow $1\,\mathrm{m\,d^{-1}}$, diffusion coefficient $D=0.5\mathrm{m^2\,d^{-1}}$, zero initial concentration and given inflow concentration $1\mathrm{kg\,m^{-3}}$. We consider the mobile-immobile exchange with the rate $\alpha$ (several variants) and linear and Freundlich adsorption isotherm. The volume of the mobile zone and the immobile zone are the same ($n_m$=0.2, $n_{im}$=0.2), as well as the sorption fractioning ($\phi$=0.5).

The results for selected parameters are displayed in Fig. 1. In the first graph, the case with exponent close to 1 (linear case) is compared to the small exponent (concave sorption curve), leading to larger retardation for smaller concentration and producing steepening of the concentration front. The second graph shows that for small transfer to the immobile zone, only the sorption in the mobile zone contributes to retardation, while for the almost equilibrium case, sorption effect is in both and does not depend on fractioning.

## Combination of continuum and fracture network

The problem is defined as a 2D continuum rectangle 12.75 × 8 m, with discrete fracture network composed of lines (section of fractured media). The pressure head is prescribed on the left and right boundaries with gradient 0.001 (steady flow). Prescribed concentration $c_{IN}$= 1g $\mathrm{l^{-1}}$ is on the inflow boundary (Fig.2).The initial state is zero concentration. The problem is solved in the time interval 1000years. The material parameters for the fracture and the continuum are given in Tab. 1. The three variants of mobile-immobile transfer rate represent the cases of very small mass exchange, medium influence, and almost equilibrium. The three variants of Freundlich exponent represent the effect of non-linearity with respect to the quantitatively similar linear case ($a_F$=1).

The results are presented by means of concentration contour in the selected time (Fig 2) and breakthrough curves in the outflow point of the fracture (Fig. 3). We can see the front sharpening with decreasing Freundlich exponent in the left and several steps of retardation (rock matrix, immobile zone, adsorbed) in Fig. 3 right. Presence of sorption also changes the breakthrough curve shape besides the retardation (steps).

**Table 1** Material parameters used for the test problem of the multidimensional (fracture-continuum) model, with mobile and immobile parts and non-linear Freundlich adsorption. The coefficients are defined within the equations (3) and (4), the subscript at $\alpha$ and $a_F$ denotes a problem variant.

| | $K$ [m s$^{-1}$] | $n_i$ [1] | $n_m$ [1] | $\phi$ [1] | $\mu_1$ [m$^2$] | $\mu_2$ [m] | $K_F$ [m$^{3aF}$ kg$^{-aF}$] | $\alpha_1$ [year$^{-1}$] | $\alpha_2$ [year$^{-1}$] | $\alpha_3$ [year$^{-1}$] | $a_{F1}$ [1] | $a_{F2}$ [1] | $a_{F3}$ [1] |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Rock (2D) | $3.2 \times 10^{-10}$ | 0.2 | 0.4 | 0.9 | - | 1 | 0.002 | 0.0001 | 0.01 | 1 | 0.1 | 0.5 | 1 |
| Fracture (1D) | $3.2 \times 10^{-5}$ | 0.2 | 0.4 | 0.9 | 1 | - | 0.002 | 0.0001 | 0.01 | 1 | 0.1 | 0.5 | 1 |

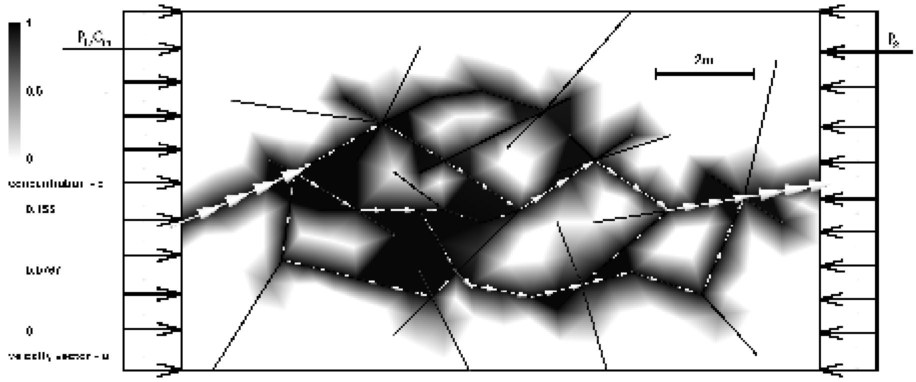


**Fig. 2** Illustration of fracture network and boundary condition, together with the results of the scalar field of concentrations in the time t=1000 years for the task with Freundlich sorption ($a_{F3}$) and no immobile zone. The velocity value is displayed by the arrow size.

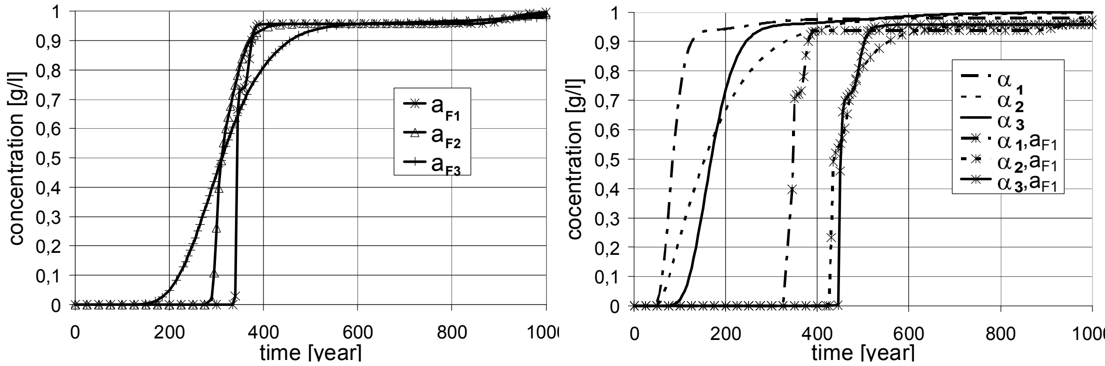


**Fig. 3** Breakthrough curves for the various exponents in the Freundlich isotherm (left) and various rates of mobile-immobile exchange (right) with or without sorption.

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