Transport of Active Solute in a Fractured Porous Medium with Nonequilibrium Adsorption

Bakhtiyor Khuzhayorov, Zhabbor Mustofoqulov

Professor, Department of Mathematical modelling, Samarkand State University, Samarkand, Uzbekistan

Assistant Professor, Department of Radio electronica, Jizzakh Polytechnic Institute, Jizzakh, Uzbekistan

ABSTRACT: In the paper an active solute transport problem in a fractured-porous medium is considered with taking into consideration linear and non-linear non-equilibrium adsorption, corresponding to Henry and Freundlich isotherms at equilibrium state. The problem is numerically solved and the influence of non-equilibrium adsorption on solute transport characteristics is established.

KEYWORDS: adsorption, adsorption kinetics, concentration, diffusion; fractured-porous media; mass transfer, relative flow rate, solute transport.

I. INTRODUCTION

Studies on fluid flow and solute transport in fractured porous medium (FPM) unambiguously show the effect of fractures on the effective permeability and the effective diffusion coefficient of FPM. For non-reactive (inert) solutes, the free space of porous blocks between fractures represents storage, where the solute is retained for a long time. This prolongs the time of release of these solutes to the surface, thereby increasing the technical and economic parameters of underground reservoirs as storage for various pollutants [1-3, 18].

In the case of radioactive substances, the decay time is prolonged because of the delay in the porous blocks before the substance exits to the surface [4, 5]. The porosity of blocks (matrix), that are in contact with fractures, allows the transition of substance from fractures to blocks (or vice versa), is called as diffusion porosity or porosity of the matrix [6, 8]. We note that this concept was introduced earlier in [16].

The recent studies show that the diffusion of pollutants into the matrix can lead to a significant retardation of its transport in the fracture. In [7], using the finite element method, the sensitivity of the model was analyzed from the parameters included in the model during the transport of substance in a fracture with diffusion into the matrix. These parameters are: fracture opening, fluid velocity in the fracture, diffusion porosity (matrix porosity), matrix distribution parameter, dispersivity.

Theoretical studies of the injection of radionuclides into the well in a fractured-porous medium are well described by authors of [10, 12, 13, 14]. In these work the models I and II were used. Each model is implemented for various boundary conditions at the location of the well, namely, for a constant concentration and an exponentially decaying concentration. In the model I radionuclide is transported in the fracture through radial convection (advection) and longitudinal dispersion, while in the model II only radial advection is taken into account. Both models take into account the delay effects associated with radioactive decay, as well as linear adsorption isotherms in both the fracture and the porous matrix. Solute transport from the fracture to the porous block
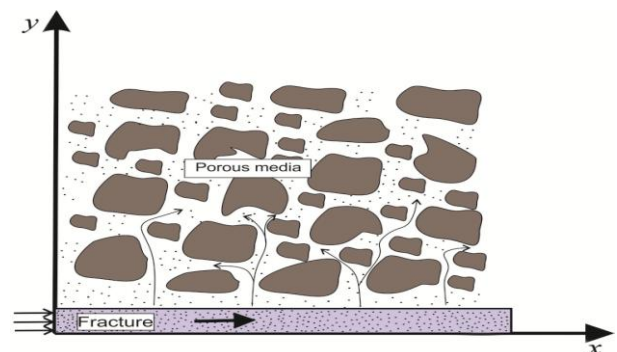

Fig.1. Schematic structure of the solute

transport of in FPM

is accounted for by molecular diffusion. It is established that the role of longitudinal dispersion is manifested for relatively large periods of radionuclide injection.

In [9] the solution of the solute transport problem in a fractured medium is presented, where the solute diffuses from the fracture into the porous matrix. The solute is inert and does not react with the matrix skeleton and fracture rock. Transport equation in the fracture is one-dimensional equation of convective (advective) diffusion, and in the porous blocks – a one-dimensional diffusion type equation. The analytical solution of the problem was used to interpret the laboratory data obtained in [7].

In this paper we consider a solute transport problem in a medium, consisting of a single fracture and adjusted porous block (Figure 1). Solute transport in the fracture governed by convection, diffusion (hydrodynamic dispersion), adsorption processes, while in the porous block only diffusion and adsorption processes occur. Two kinds of adsorption kinetics in both elements of the medium are considered. At the asymptotic stage (for large times) the dynamics of adsorption corresponds to Henry and Freundlich isotherms. A solute transport problem is posed and numerically solved. Solute concentration in mobile liquid and concentration of adsorbed mass are computed for different values of initial parameters. Relative solute mass transport in different form through common boundary of the fracture and the porous block is estimated.

II.   FORMULATION OF THE PROBLEM

We consider the case where the solute has chemical-biological or radiation activity. Chemical-biological activity means the interaction of the solute with the rock skeleton or solute degradation under the influence of chemical or biological processes [15, 16]. In radioactive disposals due to the natural decay solute concentration gradually decreases. It should be noted that due to the decay or degradation for a certain time, the concentration of the substance can reach zero values completely.

Since here the adsorption of solute on the rock surface is considered, we are to consider decay or degradation of the adsorbed solute also. We assume that the decay or degradation of the solute both in the free volume and in the adsorbed state occurs according to the same law, but in the general case, with different parameters. At these assumptions mass transport equations in the fracture and the porous block have the form [8, 17]:

$$b\left(\frac{\partial c_f}{\partial t} + \rho\frac{\partial s_f}{\partial t} + V\frac{\partial c_f}{\partial x}\right) = bD_f^*\frac{\partial^2 c_f}{\partial x^2} + \theta_m D_m^*\frac{\partial c_m}{\partial y}\bigg|_{y=0} - b\lambda_f c_f - b\rho\lambda_{sf} s_f, \qquad 0 \le x < \infty, \tag{1}$$

$$\frac{\partial c_m}{\partial t} + \frac{\rho}{\theta_m}\frac{\partial s_m}{\partial t} = D_m^*\frac{\partial^2 c_m}{\partial y^2} - \lambda_m c_m - \frac{\rho}{\theta_m}\lambda_{sm}s_m, \qquad 0 \le y < \infty \tag{2}$$

where $c_f = c_f(t,x)$ - the concentration of the solute in the fracture, m^3m^{-3}; $c_m = c_m(t,x,y)$ - the concentration in the matrix, m^3m^{-3}; $s_f = s_f(t,x)$ - the concentration of the adsorbed solute in the fracture, m^3kg^{-1}; $s_m = s_m(t,x,y)$ - the concentration of adsorbed solute in the matrix, m^3kg^{-1}; D_f^*, D_m^* - effective diffusion coefficients in the fracture and the matrix, respectively, m^2s^{-1}; ρ - density of saturated medium, kgm^{-3}; V - average flow velocity in the fracture, ms^{-1}; b - fracture width, m; θ_m - the matrix porosity coefficient, t - time, s; λ_f, λ_m are coefficients of chemical-biological degradation of solute in the fracture and in the matrix, respectively, or radioactive decay coefficients $(\lambda = \lambda_f = \lambda_m)$, s^{-1}; λ_{sf}, λ_{sm} are the coefficients of chemical-biological degradation of the adsorbed solute in the fracture and in the matrix, respectively. In the case of radioactive solute we can put $\lambda = \lambda_{sf} = \lambda_{sm}$.

We consider two models of adsorption.

Model I. Here we assume that the adsorption of solute in FPM occurs according to linear equilibrium kinetics [17]

$$\frac{\partial s_f}{\partial t} = \alpha_f\left(k_f c_f - s_f\right), \tag{3}$$

$$\frac{\partial s_m}{\partial t} = \alpha_m\left(k_m c_m - s_m\right), \tag{4}$$

where α_f, α_m are the coefficients characterizing the intensity of the adsorption processes in the fracture and in the matrix, respectively, s^{-1}, k_f, k_m are the adsorption coefficients in the fracture and in the matrix, m^{3}kg^{-1}.

From equations (3) and (4) at $t \to \infty$ we obtain linear equilibrium adsorption $s_f = k_f c_f$ and $s_m = k_m c_m$, respectively (Henry isotherms).

Model II. Here, instead of the linear kinetics of adsorption (3) - (4), we consider the non-linear kinetics [19]

$$\frac{\partial s_f}{\partial t} = \alpha_f \left(k_f c_f^N - s_f \right), \tag{5}$$

$$\frac{\partial s_m}{\partial t} = \alpha_m \left(k_m c_m^N - s_m \right), \tag{6}$$

where N is a constant. From equations (5) and (6) at $t \to \infty$ we obtain the non-equilibrium equations $s_f = k_f c_f^N$, $s_m = k_m c_m^N$, respectively (Freundlich isotherm).

Let initially the medium is filled with pure (without solute) liquid. From the inlet section of the fracture ($x = 0$) liquid with solute concentration c_0 is injected under constant average flow velocity V. At $x \to \infty$ in the fracture and at $y \to \infty$ in the porous block no solute transport occurs. On the common boundary of the fracture and the porous block we adopt the continuousity of the concentration fields. Then the initial and boundary conditions we can take in the following form

$$c_f(0, x) = c_m(0, x, y) = 0, \tag{7}$$

$$c_f(t, 0) = c_0, \tag{8}$$

$$c_f(t, x) = c_m(t, x, 0), \tag{9}$$

$$\frac{\partial c_f(t, \infty)}{\partial x} = 0, \tag{10}$$

$$\frac{\partial c_m(t, x, \infty)}{\partial y} = 0, \tag{11}$$

$$s_f(0, x) = s_m(0, x, y) = 0. \tag{12}$$

So, we are to solve (1), (2) and (3), (4) (or (5), (6)) with conditions (7) - (12).

III. NUMERICAL SOLUTION OF THE PROBLEM

We solve the system of equations (1) - (2) with (3) - (4) under the conditions (7) - (12) by the method of finite differences [20].

In the porous block domain we introduce the following net domain $\omega_{\tau h_1 h_2} = \left\{ (t_k, x_i, y_j); \, t_k = k\tau, \, x_i = ih_1, \, y_j = jh_2, \, i = \overline{0, I}, \, j = \overline{0, J}, \, k = \overline{0, K} \right\}$, where τ - step of the grid with respect to time, h_1, h_2 - step of the grid with respect to x and y coordinates, respectively, I, J - enough large integers that are to be chosen such to cover concentration change area. The time step τ we choose as $\tau = \frac{t_{\max}}{K}$, where $t_{\max}$ is maximal time, we study the problem in the time segment $t \in [0, t_{\max}]$.

Equations (1) - (2) and (3) - (4) are approximated as follows:

$$\frac{cf_i^{k+1} - cf_i^k}{\tau} + \rho\frac{sf_i^{k+1} - sf_i^k}{\tau} + V\frac{cf_i^{k+1} - cf_{i-1}^{k+1}}{h_1} = D_f^*\frac{cf_{i+1}^{k+1} - 2cf_i^{k+1} + cf_{i-1}^{k+1}}{h_1^2} +$$

$$+\frac{\theta_m D_m^*}{b}\frac{cm_{i,1}^k - cm_{i,0}^k}{h_2} - b\lambda_f cf_i^{k+1} - b\rho\lambda_{sf} sf_i^{k+1}, \quad i = \overline{1, I-1},\ k = \overline{0, K-1},$$

$$(13) \quad \frac{cm_{i,j}^{k+1} - cm_{i,j}^k}{\tau} + \frac{\rho}{\theta_m}\frac{sm_{i,j}^{k+1} - sm_{i,j}^k}{\tau} = D_m^*\frac{cm_{i,j+1}^{k+1} - 2cm_{i,j}^{k+1} + cm_{i,j-1}^{k+1}}{h_2^2} - \lambda_m cm_{i,j}^{k+1} - \frac{\rho}{\theta_m}\lambda_{sm} sm_{i,j}^{k+1},$$

$$i = \overline{0, I},\ j = \overline{1, J-1},\ k = \overline{0, K-1},$$

(14)

$$\frac{sf_i^{k+1} - sf_i^k}{\tau} = \alpha_f k_f cf_i^k - \alpha_f sf_i^{k+1},\ i = \overline{0, I},\quad k = \overline{0, K-1}, \tag{15}$$

$$\frac{sm_{i,j}^{k+1} - sm_{i,j}^k}{\tau} = \alpha_m k_m cm_{i,j}^k - \alpha_m sm_{i,j}^{k+1},\ i = \overline{0, I},\ j = \overline{0, J},\ k = \overline{0, K-1},$$

(16)

where $cm_{i,j}^k$, cf_i^k, $sm_{i,j}^k$, sf_i^k are the grid functions corresponding to c_m, c_f, s_m, s_f, respectively.

Equations (13) - (16) are implicit finite difference schemes with respect to grid functions $cm_{i,j}^k$, cf_i^k, $sm_{i,j}^k$, sf_i^k.

The initial and boundary conditions (7) - (12) are approximated as:

$$cf_i^0 = cm_{i,j}^0 = 0, \tag{17}$$

$$cf_0^{k+1} = c_0, \tag{18}$$

$$cf_i^{k+1} = cm_{i,0}^{k+1}, \tag{19}$$

$$cf_I^{k+1} = cf_{I-1}^{k+1}, \tag{20}$$

$$cm_{i,J}^{k+1} = cm_{i,J-1}^{k+1}, \tag{21}$$

$$sf_i^0 = sm_{i,j}^0 = 0,\quad i = \overline{0, I},\ j = \overline{0, J},\ k = \overline{0, K-1} \tag{22}$$

Equations (13) - (16) are reduced to the following form

$$A_1 cf_{i-1}^{k+1} - B_1 cf_i^{k+1} + C_1 cf_{i+1}^{k+1} = -F_i^{(1)},\ i = \overline{1, I-1},\quad k = \overline{0, K-1}, \tag{23}$$

$$A_2 cm_{i,j-1}^{k+1} - B_2 cm_{i,j}^{k+1} + C_2 cm_{i,j+1}^{k+1} = -F_{i,j}^{(2)},\ i = \overline{0, I},\ J = \overline{1, J-1},\ k = \overline{0, K-1}, \tag{24}$$

$$sf_i^{k+1} = R_1 cf_i^k + E_1 sf_i^k,\quad\quad i = \overline{0, I},\quad k = \overline{0, K-1}, \tag{25}$$

$$sm_{i,j}^{k+1} = R_2 cm_{i,j}^k + E_2 sm_{i,j}^k,\quad\quad i = \overline{0, I},\ J = \overline{0, J},\quad k = \overline{0, K-1}. \tag{26}$$

where $A_1 = \dfrac{D_f^*}{h_1^2} + \dfrac{V}{h_1}$, $B_1 = \dfrac{1}{\tau} + 2\dfrac{D_f^*}{h_1^2} + \dfrac{V}{h_1} + \lambda_f$, $C_1 = \dfrac{D_f^*}{h_1^2}$, $A_2 = \dfrac{\tau D_m^*}{h_2^2}$, $B_2 = 1 + 2\dfrac{\tau D_m^*}{h_2^2} + \tau\lambda_m$, $C_2 = \dfrac{\tau D_m^*}{h_2^2}$, $R_1 = \dfrac{\tau\alpha_f k_f}{1 + \tau\alpha_f}$,

$E_1 = \dfrac{1}{1 + \tau\alpha_f}$, $R_2 = \dfrac{\tau\alpha_m k_m}{1 + \tau\alpha_m}$, $E_2 = \dfrac{1}{1 + \tau\alpha_m}$, $F_{ij}^{(1)} = \dfrac{1}{\tau}c_i^k + \dfrac{\theta_m D_m^*}{bh_2}\left(cm_{i,1}^k - cm_{i,0}^k\right) - \left(\dfrac{\rho}{\tau} + \rho\lambda_{sf}\right)sf_i^{k+1} + \dfrac{\rho}{\tau}sf_i^k$,

$F_{ij}^{(2)} = cm_{ij}^k - \dfrac{\rho}{\theta_m}\left(1 + \tau\lambda_{sm}\right)sm_{i,j}^{k+1} + \dfrac{\rho}{\theta_m}sm_{i,j}^k$.

The systems of linear algebraic equations (23) - (24) are solved by the Tomas' method [11]. After determining the concentration fields, it is possible to determine the adsorption fields sf_i^k, $sm_{i,j}^k$ from (25), (26).

IV. DISCUSSION OF RESULTS

Using the numerical results, profiles of concentrations c_f and c_m, as well as adsorbed mass concentrations s_f and s_m, both in the fracture and in the matrix are drawn.

In calculations the following initial values of parameters are used: $c_0 = 0.01\,\text{m}^3\text{m}^{-3}$, $\theta_m = 0.2$, $D_m^* = 1\cdot10^{-6}\,\text{m}^2\text{s}^{-1}$, $D_f^* = 6\cdot10^{-5}\,\text{m}^2\text{s}^{-1}$, $V = 5\cdot10^{-4}\,\text{ms}^{-1}$, $b = 4\cdot10^{-4}$ m, $\rho = 2.5\cdot10^3\,\text{kgm}^{-3}$, $\alpha_m = \alpha_m = 4\cdot10^{-4}\,\text{s}^{-1}$ and the various λ.

Some results of calculations for two models of kinetic adsorption are shown in Fig.2-4. In Figures 2-3 solute concentration surface and the adsorbed mass surface for the two adsorption kinetics (Model I, II) are presented. In Fig.2 surfaces of c_m/c_0 and s_m for $k_m = k_f = 3\cdot10^{-5}\,\text{m}^3\text{kg}^{-1}$ and at the case $\lambda = \lambda_f = \lambda_m = \lambda_{sf} = \lambda_{sm}$ are shown.

Comparing the presented data, it can be seen that the solute decay leads to a reduction of concentration distribution area width both for the solute and the adsorption fields.

The comparison of Fig.2a and Fig.2b shows that the solute decay in the fracture and the porous block leads to a narrowing of the solute distribution zone. The solute concentration due to decay quickly fades in the fracture, which affects on the concentration distribution in the porous matrix. However, the maximum value of the adsorbed mass does not change when the solute decays.

In Fig.3 similar surfaces are given at $k_m = k_f = 3\cdot10^{-5}\,\text{m}^3\text{kg}^{-1}$, $N = 0.83$ for the non-linear non-equilibrium adsorption kinetics (Model II) at the steady state corresponding to the Freundlich isotherm. In this case, the maximum value of the adsorbed masses at the point $(0,\,0)$ was equal to $0.6\cdot10^{-6}\,\text{m}^3\text{kg}^{-1}$, while for the equilibrium case of the adsorption kinetics corresponding to the Henry isotherm, this value is equal to $0.28\cdot10^{-6}\,\text{m}^3\text{kg}^{-1}$.

As can be seen from the presented data, the concentration fields, as the intensity of adsorption increases, are of limited distribution - the range of concentration changes will narrow. In Fig. 2a, the propagation of the concentration field in the y direction is extended from 0.75 m at $t = 10000$ s, and in Fig. 3a one can see, the concentration distribution area does not reach 0.75 m. From the presented data we see that at the same other parameters, the non-linear law of adsorption leads to the narrowing of the solute concentration distribution in comparison with the linear law.

The decay of the solute in this case also leads to a reduction in the solute concentration distribution and the adsorption mass concentration in the fracture and porous block (Fig. 3a, b).
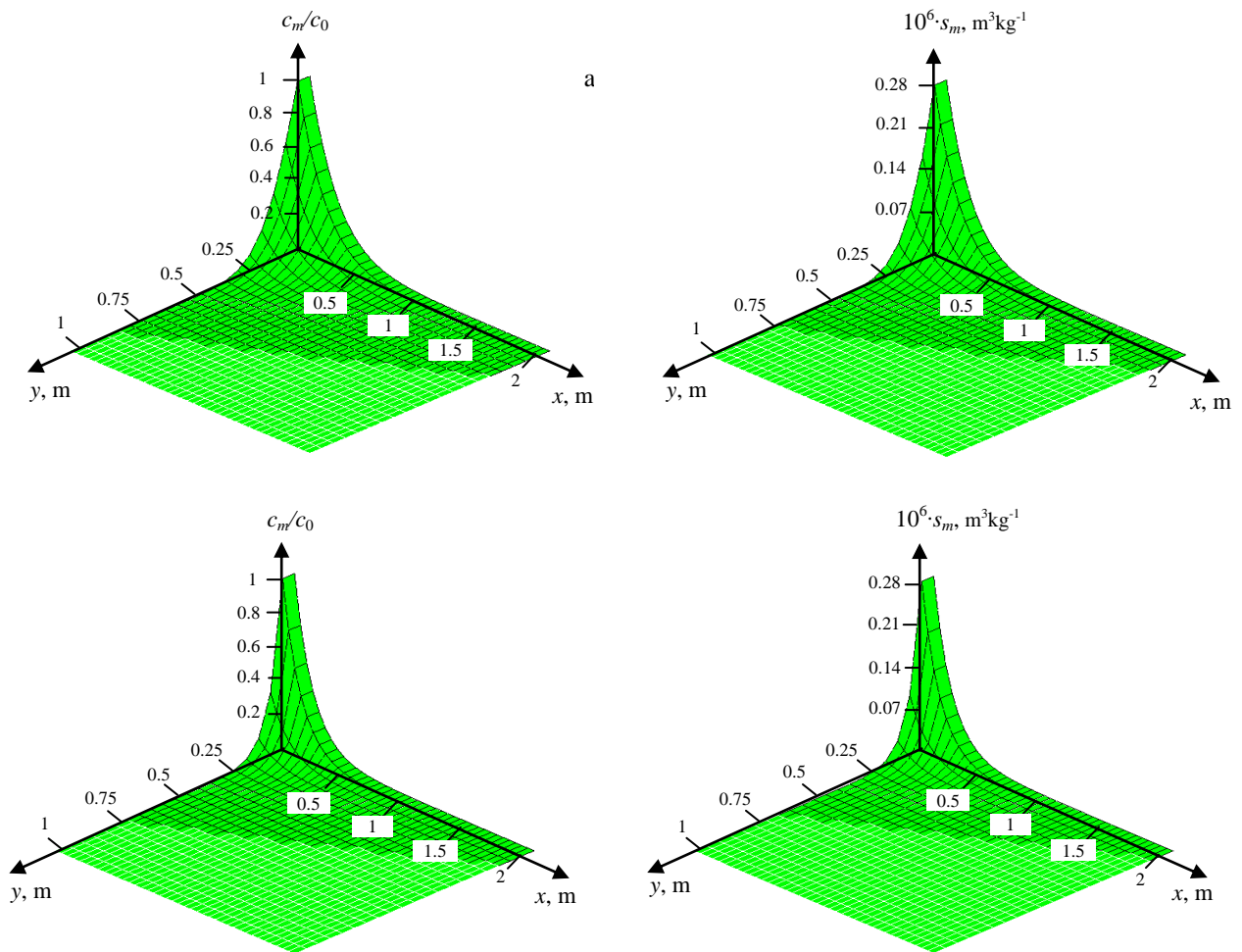
Fig.2. Surface c_m/c_0 and s_m for the linear kinetic adsorption according to Henry isotherm with $\lambda = 0$ (a), $\lambda = 3 \cdot 10^{-4}$ s^{-1} (b), $t = 10000$ s.
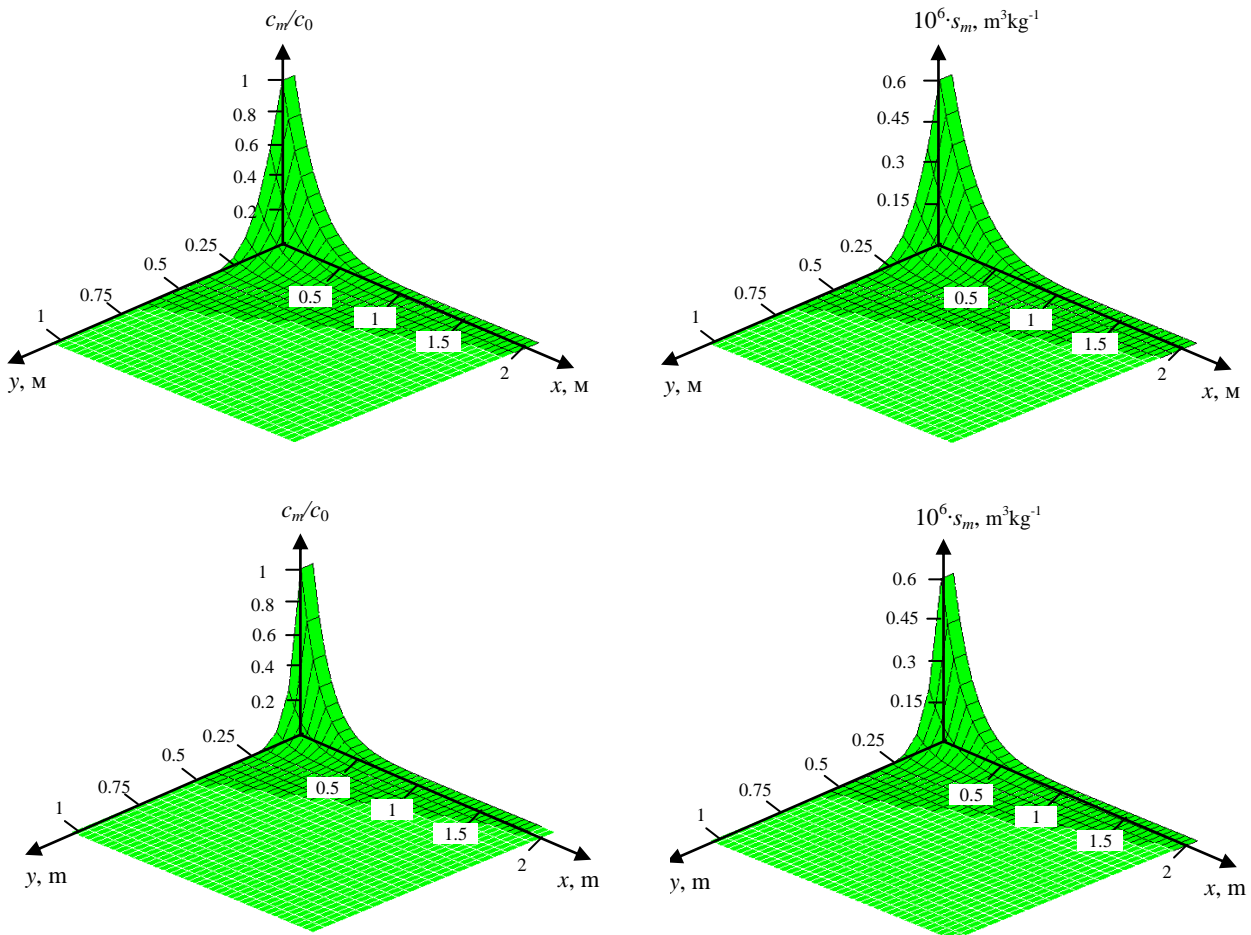
Fig. 3. Surface c_m/c_0 and s_m for the non-linear kinetic adsorption according to Freundlich isotherm with $\lambda = 0$ (a), $\lambda = 3 \cdot 10^{-4}$ s^{-1} (b), $t = 10000$s, $N = 0.83$

The dynamics of the relative solute flow rate from the fracture to the matrix $Q = -\theta_m D_m^* \dfrac{\partial c_m}{\partial y}\bigg|_{y=0}$ is estimated and the graphs of its change are plotted. On the basis of Q the common relative mass transport through $y = 0$ is calculated as $Q_{com} = \int_0^\infty Q\,dx$ for each time t. In addition, the total relative solute mass transport through $y = 0$ is determined as

$$Q_{tot} = \int_0^t Q_{com}dt = \int_0^t\int_0^\infty Q\,dxdt \cdot$$

Some graphs for Q, Q_{com}, Q_{tot} are presented in Fig. 4. The distribution of Q at $t = 7200$s for two laws of adsorption is presented in Fig. 4a. As we see, for non-linear adsorption, up to certain distances in x values of Q are greater than in linear case at all other identical parameters. From the certain distance x the character of dependences is

changed and for non-linear adsorption the values of Q is smaller than in linear adsorption law. These phenomena can be explained by forming greater concentration gradients for nonlinear adsorption law up to certain x and its smaller

values for greater x. If can be seen also, decay of solute leads to the increasing of Q up to some x after that values Q are smaller than at the case of non-decay solute.
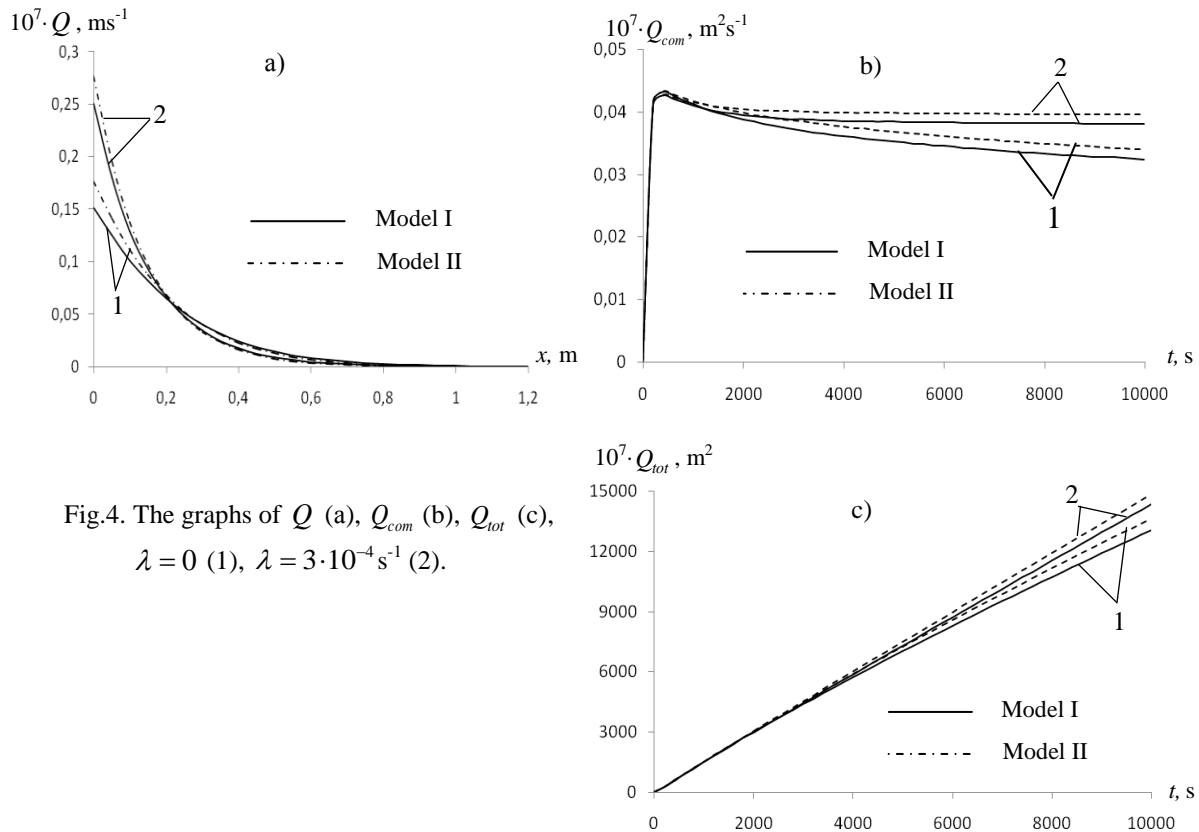

Fig.4. The graphs of Q (a), Q_{com} (b), Q_{tot} (c), $\lambda = 0$ (1), $\lambda = 3 \cdot 10^{-4} \text{s}^{-1}$ (2).

The above discussed character of Q reflects on Q_{com} and Q_{tot} behavior. As in Q, the solute decay leads to greater values of Q_{tot} (Fig. 4 b, c). In the dynamics of Q_{com} on can observe non-monotonous dependence. In the case of nonlinear adsorption at all other constant parameters Q_{com} and Q_{tot} increase. At first sight can seem, that the decay should lead to the reduction of Q and, as a consequence, Q_{com}, Q_{tot}. However, here we deal not with the real solute mass flow through common boundary of two zones but with the relative mass flow. The fact, that non-linear adsorption leads to the increasing of Q_{com} and Q_{tot}, is the direct consequence of the same behavior of Q. Unlike Q_{com}, in Q_{tot} we have monotonous behavior. It is consequences of that, Q_{tot} represents the integrated characteristic of the solute mass transport.

IV. CONCLUSION

Solute transport in fractured porous media (FPM) have so far been studied relatively little. This is due to the complex geometrical configuration of such media and, consequently, the complexity of mathematical modeling of the transfer processes in them. In this paper a solute transport problem in a medium, consisting of a single fracture and adjusted porous block is considered. This schematization of FPM allows us analytically investigating characteristic features of the solute transfer process, in particular, internal mass transfer between the system of fractures and porous blocks.

The solute is treated as an active substance and its degradation and decay, as well as non-equilibrium adsorption both in the fracture and in the porous block are taken into account. The degradation coefficients in the transport equations are assumed to be equal in the case of a radioactive solute. Two models of kinetic adsorption are considered: linear and nonlinear. A solute transport problem for the one-dimensional case is numerically solved using the finite difference method. Based on numerical calculations, it was found that the solute decay slows down its spread in the fracture, and

as a result, in the porous block. In turn, this leads to a corresponding slowdown in the propagation of the adsorption field. It was established that, at all other parameters being equal, nonlinearity in kinetics leads to an intensification of the adsorption process. Consequently, with nonlinear kinetic adsorption, under stationary conditions corresponding to the Freundlich isotherm, a more accelerated slowdown in the solute propagation in the fracture and the porous block is observed. The relative current (Q), common (Q_{com}) and total (Q_{tot}) mass transfer from the fracture to the porous block are estimated. It is shown that up to some distances x for a decaying solute Q is greater than for a non-decaying one. For more x the nature of the change is reversed. For nonlinear adsorption kinetics, somewhat overestimated values were obtained. In Q_{com} a non-monotonic dynamics is appeared, which is explained by the occurrence of large concentration gradients at the initial moments of time. In Q_{tot} such non-monotonicity is not observed, which is explained by the integral behavior of this parameter. In all cases, the solute decay leads to an increase in the values of these relative solute fluxes. At first glance, this is paradoxical. However, it should be noted that these fluxes are relative; they do not characterize the total solute mass transported through the common boundary of the fracture and the porous block.

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