Nonlinear Mathematical Model of Contaminant Distribution in Unsaturated Catalytic Porous Media

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Abstract— **The nonlinear mathematical model of a contaminant distribution in unsaturated catalytic porous media to the filter-trap in isothermal conditions is presented. The mathematical model takes into account the micro and the meso/macro scale factors of the heat and mass transfer processes. The numerical solution of the respective boundary value problem was obtained by the method of finite differences. The analytical solution for mass transfer in nanoparticles was presented as well.**

Keywords— mathematical model; boundary-value problem; numerical method; refinement; nanoadsorbent

I. INTRODUCTION

Pesticide use and the disposal of radioactive, biological, and chemical wastes can lead to much higher but localized levels of soil contamination [1, 2].

At the same time, researchers actively investigate heat and mass transfer processes on the mezo- and micro levels. For example, modern industrial equipment allows the possibility to inject special nanoparticles into the soil with the purification purpose [3]. Therefore, a lot of scientists around the world are involved into creation, developing, verification and validation of corresponding mathematical models for the fundamental understanding of the various processes of chemical and physical migration behaviour taking into account the catalytic micro- and nanoporous particles (catalytic porous media) [4–6].

On the other hand, the Ukrainian scientific school of underground mass transfer processes modelling have presented a range of mathematical models for problems of filtration consolidation [7, 8], water cleaning in wetland [9], diffusion in a multiphase body [10], iron removal from underground water [11, 12], stresses-strained state of the earth damps [13, 14], flushing process for saline soils [18] etc. Consequently, they prepared a good basement for next level of mathematical models which may connect together macro- and micro-scaled processes of heat and mass transfer.

II. FORMULATION OF THE PHYSICAL PROBLEM

Let us consider the problem of vertical migration of contaminants (e.g., fertilizers, pesticides, radionuclides etc) in a layer of soil (Fig. 1). The layer of soil is fulfilled with colloid adsorbents (e.g. nanosapropel) for the purification process. That is why is called catalytic porous media.

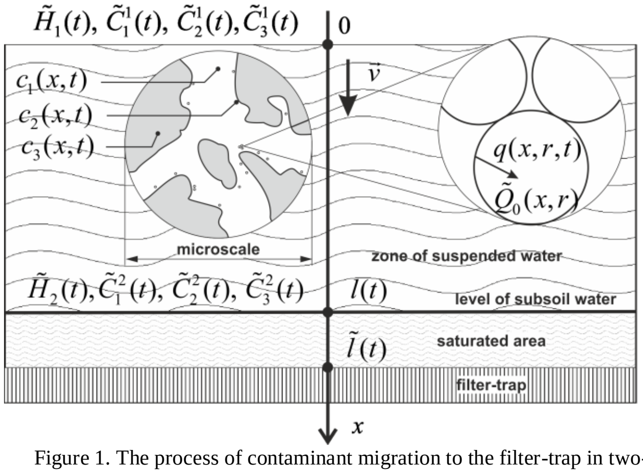

Figure 1. The process of contaminant migration to the filter-trap in two-dimentional case

The pore spaces between the soil grain particles are partially filled with water, partially with air (unsaturated zone or zone of suspended water).

At a depth $\tilde{l}$ in the ground is a filter-trap filled with a sorbent (such as vermiculite) is located. There is a piezometric pressure on the upper and lower surfaces of the unsaturated zone of a soil $\tilde{H}_1$ and $\tilde{H}_2$ ($\tilde{H}_1 > \tilde{H}_2$), respectively. The distribution of contaminant concentrations at the initial time $t=0$: $\tilde{C}_1^0(x)$, $\tilde{C}_2^0(x)$, $\tilde{C}_3^0(x)$, and $\tilde{Q}^0(x,r)$ are known. The contaminant concentrations $\tilde{C}_1^1(t)$, $\tilde{C}_2^1(t)$ and $\tilde{C}_3^1(t)$ on the upper surface and $\tilde{C}_1^2(t)$, $\tilde{C}_2^2(t)$, $\tilde{C}_3^2(t)$ on the level of subsoil water are also known.

It is necessary to build the adequate mathematical model, find numerical solution and develop software algorithm for further investigation of the $c_1(x,t)$, $c_2(x,t)$, $c_3(x,t)$ and $q(x,r,t)$ concentrations distribution on the large filtration area at a given time steps.

III. Mathematical Model

Transfer of salts dissolved in water and heat by filtration flow occurs under the influence of the pressure gradients and the concentration of salts. The filtration of salt solutions and the heat transfer proceed in accordance with the generalized Darcy's and Fick's laws.

Therefore, the boundary value problem of the contaminant migration in a catalytic porous medium in one-dimentional nonlinear case was solved using a mathematical model with the following equations [15–17]:

the equation of contaminant migration with concentration c_1 in a convectively mobile pore solution

$$\sigma_1\frac{\partial c_1}{\partial t}=\frac{\partial}{\partial x}\left(D_1(c_1)\frac{\partial c_1}{\partial x}\right)+\frac{\partial}{\partial x}\left(D_{T_1}\frac{\partial T}{\partial x}\right)-\\ -\upsilon(c_1)\frac{\partial c_1}{\partial x}-\gamma_1c_1+\gamma_2c_2, \tag{1}$$

the equation of contaminant migration with concentration c_2 located in the water bound with the soil skeleton with account for the intraparticle transfer

$$\frac{\partial c_2}{\partial t}=\frac{\partial}{\partial x}\left(D_2(c_2)\frac{\partial c_2}{\partial x}\right)+\frac{\partial}{\partial x}\left(D_{T_2}\frac{\partial T}{\partial x}\right)+\\ +\gamma_1c_1-\gamma_2c_2+\gamma_3c_3-\theta\left.\frac{\partial q}{\partial r}\right|_{r=R}, \tag{2}$$

the equation of contaminant migration with concentration c_3 located in the soil skeleton

$$\frac{\partial c_3}{\partial t}=\frac{\partial}{\partial x}\left(D_3(c_3)\frac{\partial c_3}{\partial x}\right)+\frac{\partial}{\partial x}\left(D_{T_3}\frac{\partial T}{\partial x}\right)+\\ +\gamma_2c_2-\gamma_3c_3=\sigma_1\frac{\partial c_3}{\partial t}, \tag{3}$$

the equation of intraparticle mass transfer of contaminant with current concentration q

$$D_0(q)\left(\frac{\partial^2 q}{\partial r^2}+\frac{2}{r}\frac{\partial^2 q}{\partial r}\right)+D_{T_0}\left(\frac{\partial^2 T}{\partial r^2}+\frac{2}{r}\frac{\partial T}{\partial r}\right)=\frac{\partial q}{\partial t}, \tag{4}$$

the equation of convective heat transfer

$$\frac{\partial}{\partial x}\left(\lambda_T\frac{\partial T}{\partial x}\right)-\rho c_\rho\upsilon\frac{\partial T}{\partial x}=c_T\frac{\partial T}{\partial t}, \tag{5}$$

the equation of moisture transfer

$$\mu(h)\frac{\partial h}{\partial t}=\frac{\partial}{\partial x}\left(K\left(h,c_1,T\right)\frac{\partial h}{\partial x}\right)-\\ -\frac{\partial}{\partial x}\left(\nu\frac{\partial c_1}{\partial x}\right)-\frac{\partial}{\partial x}\left(\nu^T\frac{\partial T}{\partial x}\right)+f, \tag{6}$$

the generalized equation of the Darcy law in nonisothermal conditions for filtration of the salt solutions

$$\upsilon=-k(c_1,T)\frac{\partial h}{\partial x}+\nu_c(c_1)\frac{\partial c_1}{\partial x}+\nu_T\frac{\partial T}{\partial x}, \tag{7}$$

the adsorption isotherm which at η =0 becomes the traditional Freundlich isotherm and at β=1 - the Langmuir isotherm

$$q(x,r,t)\big|_{r=R}=\frac{k_f\cdot c_2^\beta(x,t)}{1+\eta\cdot c_2^\beta(x,t)}, \tag{8}$$

boundary conditions for concentrations c_1, c_2, c_3, q and piezometric head h

$$l_1c_1(0,t)=\tilde{C}_1^1(t),\quad l_2c_1(l,t)=\tilde{C}_2^1(t), \tag{9}$$

$$l_3c_2(0,t)=\tilde{C}_1^2(t),\quad l_4c_2(l,t)=\tilde{C}_2^2(t), \tag{10}$$

$$l_5c_3(0,t)=\tilde{C}_1^3(t),\ l_6c_3(l,t)=\tilde{C}_2^3(t), \tag{11}$$

$$l_5T(0,t)=\tilde{T}_1(t),\quad l_6T(l,t)=\tilde{T}_2(t), \tag{12}$$

$$h(x,0)=\tilde{H}_0(x),\ h(0,t)=\tilde{H}_1,\ \ h(l,t)=\tilde{H}_2, \tag{13}$$

$$T(x,0)=\tilde{T}_0(x),\ \ c_1(x,0)=\tilde{C}_0^1(x), \tag{14}$$

$$c_2(x,0)=\tilde{C}_0^2(x),\ c_3(x,0)=\tilde{C}_0^3(x), \tag{15}$$

$$q\big|_{t=0}=\tilde{Q}^0(x,y,r),\ \left.\frac{\partial q(x,y,r,t)}{\partial r}\right|_{r=0}=0, \tag{16}$$

where $c_1(x,t)$, D_1, D_{T_1} – concentration, coefficients of convective diffusion of contaminant and thermodiffision in the filtration flow; $c_2(x,t)$, D_2, D_{T_2} – concentration, coefficient of molecular diffusion of contaminant and thermodiffision coefficient in water connected with soil skeleton; $c_3(x,t)$, D_3, D_{T_2} – concentration, coefficient of diffusion of contaminant in soil skeleton and thermodiffision coefficient; $q(x,r,t)$, D_0, D_{T_0} – concentration, diffusion coefficient of contaminant and and thermodiffision coefficient in particles with radius R, which located in soil skeleton; c_T; c_ρ – specific heats of solid and liquid phases; λ_T – thermal conductivity; $K\left(h,c_1,T\right)$ – coefficient of moisure expansion; $\mu(h)$ – coefficient of moisture capacity; k_f,β,η – adsorption isotherm coefficients; θ – coefficient of micro- or nanoparticle mass transfer influence on mass transfer near the ground skeleton; υ – filtration velocity; K – filtration coefficient; γ_1, γ_2, γ_3 – mass transfer coefficients; ν_c, ν_T - coefficients of chemical and thermal osmosis; σ_1 – porosity of soil; x – vertical coordinate; l_i, $i=\overline{1,6}$ – differential operators for boundary conditions; t – time, $0<t<t_1$, r - radius (radial variable) $0<r<R$.

IV. Numerical Solution

The complicated boundary-value problem (1)-(16) have been solved with different numerical approaches. The finite difference method was used in general. Therefore, the difference grid was introduced for the variables *x, r, t* with the steps h_1, h_2, and τ accordingly:

$$\omega_{h_1h_2\tau}=\left\{\left(x_i,r_j,t_k\right)\middle|\begin{array}{l}x_i=ih_1,\ r_j=jh_2,\ t_k=k\tau,\\ i=\overline{0,n_1},\ j=\overline{0,n_2},\ k=\overline{0,n_3},\\ h_1n_1=l,\ rn_2=R,\ \tau n_3=T,\end{array}\right\} \quad (17)$$

Equations (1), (3), (5), (6), (7) have been discretized with the Samarskii monotonic difference scheme, and equation (2) with an implicit difference scheme [19]. Let us show the mathematical manipulation for equation (1). At the first, we have been wrote the finite-difference analogue of the corresponding differential equation (1):

$$\sigma_1\frac{c_{1,i}^{(k+1)}-c_{1,i}^{(k)}}{\tau}=\frac{\chi_i^{(k)}}{h_1}\left(\begin{array}{l}d_{1,i+1}^{(k)}\dfrac{c_{1,i+1}^{(k+1)}-c_{1,i}^{(k+1)}}{h_1}-\\ -d_{1,i}^{(k)}\dfrac{c_{1,i}^{(k+1)}-c_{1,i-1}^{(k+1)}}{h_1}\end{array}\right)+$$
$$+\frac{(r^+)_i^{(k)}}{D_{1,i}^{(k)}}d_{1,i+1}^{(k)}\frac{c_{1,i+1}^{(k+1)}-c_{1,i}^{(k+1)}}{h_1}+$$
$$+\frac{(r^-)_i^{(k)}}{D_{1,i}^{(k)}}d_{1,i}^{(k)}\frac{c_{1,i}^{(k+1)}-c_{1,i-1}^{(k+1)}}{h_1}- \quad (18)$$
$$-\gamma_1c_{1,i}^{(k+1)}+\gamma_2c_{2,i}^{(k+1)}+\frac{1}{h_1}\left(\begin{array}{l}(d_{T_1})_{i+1}^{(k)}\dfrac{T_{i+1}^{(k+1)}-T_i^{(k+1)}}{h_1}-\\ (d_{T_1})_i^{(k)}\dfrac{T_i^{(k+1)}-T_{i-1}^{(k+1)}}{h_1}\end{array}\right),$$

$$c_{1,i}^{(0)}=\tilde{C}_1^0(x_i),c_{1,0}^{(k)}=\tilde{C}_1^1(t_k),c_{1,n_1}^{(k)}=\tilde{C}_1^2(t_k),\quad i=\overline{1,n_1-1},\ k=\overline{0,n_3}. \quad (19)$$

The next notation was used here:

$$d_{1,i}^{(k)}=\frac{D_{1,i}^{(k)}+D_{1,i-1}^{(k)}}{2},\qquad D_{1,i}^{(k)}=D_1(c_{1,i}^{(k)},T_i^{(k)}),$$
$$(d_{T_1})_i^{(k)}=\frac{(D_{T_1})_i^{(k)}+(D_{T_1})_{i-1}^{(k)}}{2},\qquad r_i^{(k)}=(r^+)_i^{(k)}+(r^-)_i^{(k)},$$
$$\chi_i^{(k)}=\frac{1}{1+\dfrac{h_1\left|r_i^{(k)}\right|}{2D_{1,i}^{(k)}}}=1-\frac{h_1\left|r_i^{(k)}\right|}{2D_{1,i}^{(k)}}+O(h_1^2),$$
$$(r^+)_i^{(k)}=\frac{-\upsilon_i^{(k)}+\left|\upsilon_i^{(k)}\right|}{2}\ge 0,\ (r^-)_i^{(k)}=\frac{-\upsilon_i^{(k)}-\left|\upsilon_i^{(k)}\right|}{2}\le 0\,.$$

Thomas algorithm was used for calculating distribution of the $c_1(x,t)$ salt concentration. And the difference scheme (18), (19) consequently was presented in next form:

$$\begin{cases}a_i^1c_{1,i-1}^{(k+1)}-\overline{c}_i^1c_{1,i}^{(k+1)}+b_i^1c_{1,i+1}^{(k+1)}=-c_{1,i}^{(k)}-f_i^{1,(k+1)},\\ c_{1,0}^{(k+1)}=\mu_1^1c_{1,1}^{(k+1)}+\mu_2^1,\\ c_{1,n_1}^{(k+1)}=\mu_3^1c_{1,n_1-1}^{(k+1)}+\mu_4^1,\end{cases} \quad (20)$$

where

$$a_i^1=\frac{\tau}{\sigma_1}\frac{d_{1,i}^{(k)}}{h_1}\left(\frac{\chi_i^{(k)}}{h_1}-\frac{(r^-)_i^{(k)}}{D_{1,i}^{(k)}}\right),\ b_i^1=\frac{\tau}{\sigma_1}\frac{d_{1,i+1}^{(k)}}{h_1}\left(\frac{\chi_i^{(k)}}{h_1}+\frac{(r^+)_i^{(k)}}{D_{1,i}^{(k)}}\right),$$

$$\overline{c}_i^1=1+\frac{\tau}{\sigma_1}\left(\begin{array}{l}\dfrac{\chi_i^{(k)}(d_{1,i+1}^{(k)}+d_{1,i}^{(k)})}{h_1^2}+\\ +\dfrac{1}{h_1D_{1,i}^{(k)}}((r^+)_i^{(k)}d_{1,i+1}^{(k)}-(r^-)_i^{(k)}d_{1,i}^{(k)})+\gamma_1\end{array}\right),$$

$$f_i^{1,(k+1)}=\frac{\tau}{\sigma_1}\left(\gamma_2c_{2,i}^{(k+1)}+\frac{1}{h_1}\left(\begin{array}{l}(d_{T_1})_{i+1}^{(k)}\dfrac{T_{i+1}^{(k+1)}-T_i^{(k+1)}}{h_1}-\\ -(d_{T_1})_i^{(k)}\dfrac{T_i^{(k+1)}-T_{i-1}^{(k+1)}}{h_1}\end{array}\right)\right),$$

$$\mu_1^1=0\,,\ \mu_2^1=\tilde{C}_1^1\,,\ \mu_3^1=0\,,\ \mu_4^1=\tilde{C}_1^2\,.$$

Finally, the $c_1(x,t)$ salt concentration distribution at time level *(k+1)* may be presented with the following relation:

$$c_{1,i}^{(k+1)}=\alpha_{i+1}^1c_{1,i+1}^{(k+1)}+\beta_{i+1}^1 \quad (21)$$

where $\alpha_{i+1}^1=\dfrac{b_i^1}{\overline{c}_i^1-\alpha_i^1a_i^1}$, $\beta_{i+1}^1=\dfrac{a_i^1\beta_i^1+c_{1,i}^{(k)}+f_i^{1,(k+1)}}{\overline{c}_i^1-\alpha_i^1a_i^1}$,

$i=\overline{1,n_1-1},\ k=\overline{1,n_3},\ \alpha_1^1=\mu_1^1\equiv 0,\ \beta_1^1=\mu_2^1\equiv\tilde{C}_1^1.$

Thus, the Thomas algorithm was used to solve such kind of tridiagonal system of equations [20].

Analogical mathematical manipulations were provided for equations (3), (5), (6), (7) and (2). Some of them desribed in details in following papers [17]. For intraparticle contaminant concentration analysis (4) may be used as numerical finite-difference analysis as well as analytical. Let us show the steps for situation when $D_0(q)=D_0=const$ and $D_{T_3}=0$. Thus we can obtain:

$$\frac{\partial q}{\partial t}=D_0\left(\frac{\partial^2q}{\partial r^2}+\frac{2}{r}\frac{\partial^2q}{\partial r}\right) \quad (22)$$

The solution of (22) with appropriate boundary conditions $q(x,r,0)=\tilde{Q}_0(r)$ and $q(x,R,t)=0$ can be found in analytical way:

$$q(x,r,t)=\frac{1}{r}\sum_{n=1}^{\infty}\beta_ne^{-\frac{n^2\pi^2D_0t}{R^2}}\sin\frac{n\pi r}{R}, \quad (23)$$

where $\beta_n=\dfrac{2}{R}\displaystyle\int_0^R r\tilde{Q}_0(r)\sin\frac{n\pi r}{R}dr$.

Let us find the analytical solution of (22) for the case where the doundary condition is not homogeneus:

$$q(x,R,t)=\tilde{Q}_1, \quad (24)$$

where $\tilde{Q}_1 = const$.

To reduce the nonhomogeneous boundary condition to homogeneous boundary condition, let us use the following substitution

$$q(x,r,t) = u(x,r,t) + \tilde{Q}_1, \quad (25)$$

where $u(x,r,t)$ – is the unknown function. Then

$$\frac{\partial q}{\partial t} = \frac{\partial u}{\partial t}, \ \frac{\partial^2 q}{\partial r^2} = \frac{\partial^2 u}{\partial r^2}, \ \frac{\partial q}{\partial r} = \frac{\partial u}{\partial r},$$

$$q(x,R,t) = u(x,R,t) + \tilde{Q}_1 = \tilde{Q}_1,$$

$$q(x,r,0) = u(x,r,0) + \tilde{Q}_1 = \tilde{Q}_0(r).$$

And hence

$$u(x,R,t) = 0,$$

$$u(x,r,0) = \tilde{Q}_0(r) - \tilde{Q}_1.$$

Thus, we have the following boundary-value problem for the function $u(x,r,t)$:

$$\frac{\partial u}{\partial t} = D_0\left(\frac{\partial^2 u}{\partial r^2} + \frac{2}{r}\frac{\partial u}{\partial r}\right), \ \mathbf{X} \in \Omega, \ r \in (0, R), \ t > 0, \quad (26)$$

$$u(x,r,0) = \tilde{Q}_0(r) - \tilde{Q}_1, \ \mathbf{X} \in \Omega, \ r \in (0, R), \quad (27)$$

$$u(x,R,t) = 0, \ \mathbf{X} \in \Omega, \ t > 0. \quad (28)$$

And the solution of (26)-(28) is the following:

$$u(x,r,t) = \frac{1}{r}\sum_{n=1}^{\infty}\beta_n e^{-\frac{n^2\pi^2 D_0 t}{R^2}} \sin\frac{n\pi r}{R}, \quad (29)$$

where $\beta_n = \frac{2}{R}\int_0^R r(\tilde{Q}_0(r) - \tilde{Q}_1)\sin\frac{n\pi r}{R}dr$.

Returning to the replacement (7), we obtain the analytical solution (22) with the appropriate boundary conditions in the form of the following function:

$$q(x,r,t) = \tilde{Q}_1 + \frac{1}{r}\sum_{n=1}^{\infty}\beta_n e^{-\frac{n^2\pi^2 D_0 t}{R^2}} \sin\frac{n\pi r}{R}, \quad (30)$$

where $\beta_n = \frac{2}{R}\int_0^R r(\tilde{Q}_0(r) - \tilde{Q}_1)\sin\frac{n\pi r}{R}dr$.

Conclusion

The physical problem of soil purification was formulated according to agroindustry requirements and critical analysis of a number of scientific papers. The nonlinear mathematical model of the contaminant vertical migration in unsaturated catalytic porous media to the filter-trap in isothermal conditions was defined. Catalytic porous media were presented with colloid nanoadsorbents (e.g. nanosapropel). The mathematical model took into account the micro and the meso/macro scale factors of the heat and mass transfer processes. The numerical and analytical solutions of the complicated boundary-value problem have been proposed.

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