Solutal convection in horizontal porous layer with immobilization and clogging

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Abstract. The paper is devoted to the investigation of the solutal convection in a horizontal layer of porous media with horizontal seepage of the mixture and solute immobilization. The flow through porous media is modelled within the standard Darcy–Bousinesq model and immobilization is described by the mobile/immobile media (MIM) approach. The high concentrations of solute are considered which lead to taking into account the dependence of media permeability on porosity. The variation of porosity is modelled as a linear function of solute concentration in the immobile phase. It is shown that immobilisation leads to the stabilization of homogeneous filtration regime and to slow down of the perturbation dynamics. The increasing of critical parameters with the intensification of sorption is not monotonous due to saturation of porous matrix with the adsorbed solute.

1. Introduction

The paper continues a series of study of solutal convection in a horizontal layer of a porous medium with imposed horizontal pumping. For the first time, the investigation of the similar problem configuration, but without solute immobilization and without external filtration flux, was made in [1]. A set of convective cells, with a width equal to the layer thickness, were obtained analytically. The influence of an external horizontal flux on such convective regime was studied in [2]. The authors showed that the steady external flux leads to excitation of an oscillatory mode, however, the wavelength of critical perturbations and the critical value of the Rayleigh–Darcy number do not change. Later on, the problem was complicated by the account of solute particle immobilization [3, 4]. The authors used a widely applied MIM approach [5, 6], according to which the presence of two solute phases (mobile and immobile) can be considered. The interphase exchange by solute between phases is described by a kinetic equation. The investigation was carried out numerically in the framework of linear MIM model [3] and fractal one [4]. It was demonstrated that the particle immobilization leads to the dependence of the critical parameters on the external flux intensity.

In the present paper, we focus on the solute mixture with high concentration. To accurate modelling of such phenomenon, the effect of saturation of the immobile phase (the concentration of immobile solute should not exceed some value) is taken into account. As it was shown in [7] the second-order kinetic model, in which the adsorption rate linearly depends on the difference between the limiting saturation concentration of porous medium and the concentration of...
immobile solute, is a promising candidate. Moreover, to describe the effect of porosity decreases due to occupation of the pore space by immobile solute particles, we use the most simple and universal Carman–Kozeny equation \[8\]), which links the permeability and porosity of the media.

The paper contains six sections. Section 1 is an introduction where the previous study and motivation is discussed. Section 2 is devoted to problem statement and derivation of governing equations. In Section 3 the base solution as a regime of the homogeneous horizontal equation is obtained. The Section 4 contains the derivation of equations for small perturbations and its reduction to ODE for normal perturbations. The Section 5 is devoted to discussion about neutral curves which are obtained from the numerical solution of equations for normal perturbations. Section 6 is the conclusion where the main findings of the present paper were summarized.

2. Problem statement

We consider a flow of a mixture through a horizontal layer of a porous medium. Configuration of the problem is sketched on Figure 1. The mixture consists of solid nanoparticles and ambient fluid; the solid nanoparticles are considered as a solute within continuous approach. The filtration velocity on the horizontal boundaries is prescribed \( V = (V(y), 0) \).

To describe nanoparticle transport of solute through a porous medium we use the mobile-immobile media (MIM) model (or the two-region solute transport model). Within such a model it is assumed that the solute can be partitioned into distinct mobile (or flowing, with volume concentration \( c \)) and immobile (stagnant, with volume concentration \( q \)) phases. The solute exchange between these two phases can be modelled by the kinetic equation, which determines the dependence of the solute influx \( (\partial_t q) \) on the solute concentrations in both phases, or it also can be interpreted as phase transition kinetics. The equation system for solute concentration reads

\[
\begin{align*}
\partial_t c + \partial_t q &= D\nabla^2 c - \nabla \cdot c, \\
\partial_t q &= R(q, c).
\end{align*}
\]

(1)

Here the coupling function \( R(q, c) \) can be defined by the specific type of the MIM model.

The simplest and widely used model is the standard or linear MIM model with the following kinetic equation \[6\]

\[
\partial_t q = \alpha (c - K_d q),
\]

(2)

where \( \alpha \) is the mass transport coefficient and \( K_d \) is the solute distribution coefficient.

This model provides the correct description of solute transport, especially for low initial concentration, which was confirmed theoretically (for example, \[9\]) and experimentally \[10\]. However to investigate solute transport of moderate and even high concentration, which is the aim of present article, we apply nonlinear kinetic function with Langmure saturation of porous matrix \( (q_0) \) in the form

\[
\partial_t q = \alpha (c(q_0 - q) - K_d q),
\]

(3)

Thus, the problem can be described within the Darcy–Boussinesq model with account of nonlinear MIM model for solute immobilization as \[6, 11\]

\[
\begin{align*}
\frac{\partial}{\partial t} (c + q) &= -\nabla \cdot c + \nabla^2 c, \\
-\nabla p &= \frac{\eta \phi_0}{\kappa(\phi)} V + \rho_0 c g j, \\
\nabla \cdot V &= 0, \quad p = p' + \rho g z, \\
\frac{\partial q}{\partial t} &= \alpha (c(q_0 - q) - K_d q), \\
\kappa(\phi) &= \kappa_0 \phi^3/(1 + \phi)^2, \quad \phi = \phi_0 - q.
\end{align*}
\]

(4)
Here $D$ is the effective diffusivity, $\alpha$ is the mass transport coefficient, $K_d$ is solute distribution coefficient, $V$ is the vector of filtration velocity, $\kappa$ is the permeability of the porous medium, $\kappa_0$ is Kozeny–Carman constant, $\eta$ is the dynamic viscosity, $\rho$ is the fluid density, $\beta_c$ is the coefficient of concentration expansion, $p$ is the deviation of pressure from the hydrostatic one, $g$ is the gravity acceleration, $j$ is the unit vertical vector and $\phi$ is the porosity of the medium with $\phi_0$ is the porosity of the clean medium.

Boundaries of the layer are impermeable for the fluid. Solute concentrations at the upper and lower boundaries are kept constant and denoted by $C_+$ and $C_-$, respectively, the gradient of pressure along the boundaries is constant, thus boundary conditions read

$$
\begin{align*}
V \cdot j |_{y=0,1} &= 0, \\
-c |_{y=0} &= C_+, \\ c |_{y=1} &= C_-, \\
\frac{\partial p}{\partial x} |_{y=0,1} &= -A.
\end{align*}
$$

(5)

We chose $h$, $h^2/D$, $\phi D/h$, $\phi D\eta/\kappa$, $C_0 = C_+ - C_-$ as the scales for the distance, time, velocity, pressure and concentration, respectively.

In the dimensionless form the problem (4) can be rewritten as follows

$$
\begin{align*}
\frac{\partial}{\partial t} (c + q) &= -V \cdot \nabla c + \nabla^2 c, \\
\nabla p + \frac{1}{\kappa} V + Rp c j &= 0, \\
\frac{\partial q}{\partial \phi} &= ac (q_0 - q) - bq, \\
\nabla \cdot V &= 0, \\
\kappa(\phi) &= \phi^3/(1 + \phi)^2, \quad \phi = \phi_0 - C_0 q, \\
V |_{y=0,1} &= 0, \\
c |_{y=0} &= 1, \quad c |_{y=1} = 0.
\end{align*}
$$

(6)

The boundary value problem (6) is characterized by governing parameters: $Rp = C_0 g L \kappa_0 \rho \beta_c / (D \eta \phi_0)$ is the solutal Rayleigh–Darcy number, $Pe = A \kappa_0 h / (D \eta \phi_0)$ is the Pécel number, $a = \alpha K_d C_0 h^2 / D$ and $b = \alpha h^2 / D$ are dimensionless adsorption and desorption rates respectively.
3. Homogeneous horizontal filtration

Let us find stationary solution of the problem (6), which does not depend on horizontal coordinate, in the form

\[ c = c^0(y), \quad q = q^0(y), \quad (V) = (V)^0 = (u^0(y), 0), \quad \phi = \phi^0(y), \quad p = p^0. \]

The equations describing this regime read

\[
\begin{aligned}
&\frac{\partial^2 c^0}{\partial y^2} = 0, \\
&0 = a(q_0 - q^0) - b q^0, \\
&\nabla p^0 + \frac{1}{\kappa} V^0 + Rp c^0 j = 0,
\end{aligned}
\]  \tag{7}

where symbol \( \partial^2 \) denotes the second derivative with respect to vertical coordinate \( y \).

Taking into account boundary conditions (5), the solution of (7) can be written in the form

\[
\begin{aligned}
p^0 &= -Pex - \frac{Rp}{2} y^2, \\
\phi^0 &= \phi_0 + C_0 q^0, \\
u^0 &= \kappa(\phi^0) Pe = \frac{(\phi_0 - C_0 q^0)^3}{(1 - \phi_0 + C_0 q^0)^2} Pe, \\
q^0 &= \frac{aq_0}{ay + b y}, \\
c^0 &= y.
\end{aligned}
\]  \tag{8}

As it can be seen, the stationary solution of the problem (6) depends on the sorption parameters as well as on filtration rate (Peclet number). The linear stability analysis of this regime with respect to small perturbations is performed in the next Section.

4. Small perturbations

Let us proceed to the stability analysis of the homogeneous filtration. To this end, we represent the fields of velocity, pressure and solute concentrations in mobile and immobile phases as the sums of basic state fields and small perturbations:

\[ q = q^0 + Q, \quad c = c^0 + C, \quad V = V^0 + W, \quad p = p^0 + P, \quad W = (u, v). \]

The system of linearized equations for small perturbations reads

\[
\begin{aligned}
\partial_t (C + Q) &= -u^0 \partial_x C - v + \nabla^2 C, \\
W &= \kappa^0 (-\nabla P + Rp C j) + \kappa (-\nabla p^0 + Rp c^0 j), \\
\partial_t Q &= a C (q_0 - q^0) - (b + ac^0) Q, \\
\nabla \cdot W &= 0, \\
\kappa^0 &= \kappa(\phi^0) = \frac{(\phi_0 - C_0 q^0)^3}{(1 - \phi_0 + C_0 q^0)^2}, \\
W \cdot j|_{y=0,1} &= 0, \\
C, Q|_{y=0,1} &= 0, \\
\partial_x P|_{y=0,1} &= 0,
\end{aligned}
\]  \tag{9}

here symbols \( \partial_t, \partial_x \) and \( \partial_y \) denote the derivatives with respect to \( t, x \) and \( y \).

It is convenient to rewrite this equation system in terms of stream function \( \psi \) related to the
velocity components as \( u = -\partial_y \psi, \ v = \partial_x \psi \):

\[
\begin{align*}
\partial_t (C + Q) &= \partial^2_x C + \partial^2_y C - \kappa^0 Pe \partial_x C - \partial_x \psi, \\
\partial_t Q &= aC \left( q_0 - q^0 \right) - \left( b + ac^0 \right) Q, \\
\partial^2_x \psi + \partial^2_y \psi &= -RP0 \partial_x C + \frac{\kappa'}{k^0} \partial_y q^0 \left( \kappa' PeQ - \partial_y \psi \right) + Pe \left( \kappa' \partial_y Q - \kappa'' Q \partial_y q^0 \right), \\
\kappa' &= \frac{\partial \kappa}{\partial \phi} (\phi^0) = \frac{(\phi^0)^2 (3 - \phi^0)}{(1 - \phi^0)^3}, \quad \kappa'' = \frac{\partial^2 \kappa}{\partial \phi^2} (\phi^0) = \frac{6\phi^0}{(1 - \phi^0)^4}, \\
\psi |_{y=0,1} &= 0, \quad C, Q |_{y=0,1} = 0.
\end{align*}
\]

A solution to problem (10) can be found in the form of normal perturbations, which are periodic in the time and along horizontal direction: \( Q, C, \psi \sim \exp(kx - \omega t) \), where \( k \) is horizontal wave number and \( \omega \) is the frequency of neutral perturbations. In the terms of normal neutral perturbations the problem (10) becomes

\[
\begin{align*}
-i \omega (C + Q) &= (\partial^2_y - k^2 - ik\kappa^0 Pe)C - ik\psi, \\
(\partial^2_y - k^2)\psi &= -iRPe0C - \frac{\kappa'}{k^0} \partial_y q^0 \partial_y \psi + Pe \left( \frac{\kappa'^2 \partial_y q^0}{k^0} + \kappa' \partial_y - \kappa'' q^0 \right) Q, \\
-i \omega Q &= aC \left( q_0 - q^0 \right) - \left( b + ac^0 \right) Q, \\
\psi |_{y=0,1} &= 0, \quad C, Q |_{y=0,1} = 0.
\end{align*}
\]

\( k_{crit} = \pi \) (minima of the dependence \( RP^*(k) \)). It is seen that the critical value of wave number slightly changes with variation of sorption rates but this difference is not dramatically. We use the \( RP^* \) for obtaining the neutral curve because usually the full value of permeability is incorporated into Rayleigh–Darcy number and it is more convenient for comparison with previous investigations. It is known that without immobilization we have \( RP_{crit}^* = 4\pi^2 \) and \( \omega = kPe^* \) (see [2]). The immobilisation stabilises the homogeneous filtration because some part of solute transits to immobile phase as a result the critical value of Rayleigh–Darcy number increases. However, this increase is not monotonous because when the solute concentration in the immobile phase reached the value \( q_0 \) the adsorption process stops. In this case, the immobile phase becomes the source of solute due to desorption. The dependence of neutral perturbations frequency on wave number is close to linear as it is noted in [3], but the frequency decreases with the intensification of sorption processes. This effect can be explained by general slow down of dynamics due to immobilisation (see [3, 6]).

5. Results
The most interesting effects are in variation of the sorption parameters \( a \) and \( b \). The dependences of critical Raleigh–Darcy number \( RP \) value and the frequency of neutral perturbations \( \omega \) on wave number \( k \) for various values of sorption rates are presented in figure 2 and figure 3.

The neutral curves (see figure 2 and figure 3) is plotted for interval of wave numbers \( k \in [2, 4] \) because for classical Horton–Rogers–Lapwood problem [1] the critical value of wavenumber is \( k_{crit} = \pi \) (minima of the dependence \( RP^*(k) \)). It is seen that the critical value of wave number slightly changes with variation of sorption rates but this difference is not dramatically. We use the \( RP^* \) for obtaining the neutral curve because usually the full value of permeability is incorporated into Rayleigh–Darcy number and it is more convenient for comparison with previous investigations. It is known that without immobilization we have \( RP_{crit}^* = 4\pi^2 \) and \( \omega = kPe^* \) (see [2]). The immobilisation stabilises the homogeneous filtration because some part of solute transits to immobile phase as a result the critical value of Rayleigh–Darcy number increases. However, this increase is not monotonous because when the solute concentration in the immobile phase reached the value \( q_0 \) the adsorption process stops. In this case, the immobile phase becomes the source of solute due to desorption. The dependence of neutral perturbations frequency on wave number is close to linear as it is noted in [3], but the frequency decreases with the intensification of sorption processes. This effect can be explained by general slow down of dynamics due to immobilisation (see [3, 6]).

6. Conclusion
The problem of exciting of convective motion into the horizontal porous layer under horizontal seepage of mixture taking into account immobilization and clogging is investigated. The solution
Figure 2. The neutral curves into the parameter plane $R_p^*$, $k$ (left panel) and $\omega$, $k$ (right panel), where $R_p^* = R_p \kappa(\phi_0)$ and $\kappa(\phi_0) \approx 0.055$. The curves obtained for fixed adsorption rate value $a = 8$ and various values of desorption rate which are indicated in legend.

Figure 3. The neutral curves into the parameter plane $R_p^*$, $k$ (left panel) and $\omega$, $k$ (right panel), where $R_p^* = R_p \kappa(\phi_0)$ and $\kappa(\phi_0) \approx 0.055$. The curves obtained for fixed desorption rate value $b = 8$ and various values of adsorption rate which are indicated in legend.

of the problem in the regime of homogeneous horizontal filtration is obtained. The equations for small perturbation of obtained regime are derived and discussed. The stability problem is presented as a system of ODE and solved by standard differential sweep method. As a result, the neutral curves in parameter space are plotted. It is shown that immobilisation leads to stabilisation of considered regime and to slow down the perturbation dynamics. However, it is found that the increase of critical parameters with the intensification of sorption is not
monotonous due to saturation of porous matrix with the adsorbed solute.

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