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Influence of the solute immobilization on linear stability within the solute analogy of Horton–Rogers–Lapwood problem

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Abstract. Linear stability analysis within the solute analogy of Horton–Rogers–Lapwood (HRL) problem has been investigated. Solute immobilization (solute sorption) of nanoparticles by the porous medium is taken into account within the fractal model of MIM approach. The solute concentration difference between the layer boundaries and the horizontal external filtration flux are assumed as constant. The system of equations that determine the frequency of neutral oscillations and the critical value of the Rayleigh-Darcy number is derived. Neutral curves of the critical parameters on the governing parameters are plotted. Stability maps are obtained numerically in a wide range of parameters of the system. It was found that taking immobilization into account leads to an increase in the critical value of the Rayleigh-Darcy number with an increase in the intensity of the external filtration flux. The case of weak time-dependent external flux is investigated analytically. It was shown that the modulated external flux leads to an increase in the critical value of the Rayleigh–Darcy number and a decrease in the critical wavenumber.

1. Introduction
The present work is devoted to the study of the linear stability problem for solutal convection in a horizontal layer of a porous medium at a given vertical concentration gradient with an imposed external horizontal filtration flux, taking into account the immobilization of the solute.

The problem in a similar formulation, but without immobilization and in the absence of an external flux, was solved in [1] for the first time. It was shown that the convective regime has a form of a set of convective cells, which width is equal to the thickness of the layer. The influence of an external horizontal flux on convective regimes was investigated in [2]. It was shown that, although the presence of external flux leads to generation of an oscillatory mode of the instability, but the intensity of the external flux affects only the frequency of the oscillations. The wavelength of critical perturbations and the critical value of the Rayleigh-Darcy number do not changed. The effect immobilization on the problem under consideration was investigated in [3]. The map of the observed convection regimes was obtained numerically. It was found that taking into account the immobilization the critical values of the parameters become dependent on the external flux intensity. In [3] the immobilization was considered in the framework of the MIM (mobile-immobile medium) model with linear sorption kinetics [4]. However, as shown in [5], the modeling of the immobilization will be more realistic if we use the fractal linear model of the mobile/non-mobile medium (fMIM) [6]. The present paper is devoted to such modeling.
2. Problem statement

We study the solutal convection of a mixture of solid nanoparticles and ambient fluid in a horizontal layer of a porous medium under external filtration flux (Figure 1). The system is subjected to the gravity field. The solid nanoparticles are considered as a solute within the continuous approach. We took into account the effect of solute immobilization when the solute particles can stick to the solid matrix of the porous medium and do not move for a sufficient time. According to MIM approach [4], we consider that the total solute concentration is divided into two phases: the mobile phase (with volume concentration \( C \)) and the immobile phase (with \( Q \)). Solute concentrations at the upper and lower boundaries of the layer are kept constant. The solute is assumed to be heavier than the carrier fluid, thus the difference of densities leads to a solutal convection.

![Figure 1. Problem configuration. The layer of thickness \( l \) is infinite in the \( x \)-direction. The flow through the layer with velocity \( \mathbf{V} \) is imposed. The unit vector \( \mathbf{\gamma} \) shows the direction of gravity field.](image)

The equation of the solutal convection in porous media within the framework of the Boussinesq approximation in dimensionless form can be written as

\[
\mathbf{V} = -\nabla p + R_p C \mathbf{\gamma},
\]

\[
\nabla \cdot \mathbf{V} = 0,
\]

\[
\partial_t (C + Q) = \nabla^2 C - \mathbf{V} \cdot \nabla C,
\]

\[
Q = \frac{\lambda}{\Gamma(1-\alpha)} \int_0^t C(r,t') \, dt'.
\]

The boundary conditions on horizontal and vertical plane read

\[
y = 0: \quad C = 0, \quad V_y = 0,
\]

\[
y = 1: \quad C = 1, \quad V_y = 0,
\]

\[
x = \pm \infty: \quad V_x = \text{Pe} \, f(t),
\]

where \( \partial_t \) denotes the time derivative, \( \mathbf{V} \) is the two-dimensional flow velocity, \( p \) is pressure, \( \mathbf{\gamma} \) is the unit vertical vector, \( \Gamma(x) \) is the Euler gamma function. The kinetics of immobilization (equation (1d)) is described by the fractal MIM model [6] with two parameters: \( 0 < \alpha \leq 1 \) is the exponent of the Levy stable law and \( \lambda \) is the mobility parameter. The mobility parameter denotes the portion of the immobile concentration in the total concentration, meanwhile the exponent of the Levy stable law describes the relaxation of the system to the state of dynamical equilibrium between phases. For \( \alpha = 0 \) the relaxation time is infinite, the process of mobilization is blocked and equilibrium cannot be achieved; whereas the opposite case \( \alpha = 1 \) corresponds to the instantaneous relaxation (\( Q = \lambda C \)).

We choose \( l^2/D, l, C_0 = C_c - C_s, D/l, D \eta \phi / \kappa \) as the scales for time, length, concentration, velocity and pressure respectively. Here \( l \) is the layer thickness, \( \eta \) is dynamic viscosity and \( D, \phi, \kappa \) are the effective diffusivity, porosity and permeability of porous media. In chosen scales the
problem (equations (1a)-(2c)) is characterized by two governing parameters Rayleigh–Darcy number and Péclet number

\[ R_p = \frac{\beta C_g g l k \rho_f}{\phi \eta D}, \quad Pe = \frac{Vl}{D}. \]

The problem (equations (1a)-(2c)) admitted uniform seepage solution, namely

\[ C_h = y, \quad V_h = \left( Pe f(t), 0 \right), \quad \text{ (3)} \]

Let us consider the perturbation of the solution (equation (3)) in the following form:

\[ c = C - y, \quad \mathbf{v} = (u, w) = \left( V_x - Pe f(t), V_y \right), \quad p = \bar{p} - p_h. \]

Thus, in terms of the stream function \( u = -\partial_y \psi, \quad w = \partial_x \psi \) the governing system (equations (1a)-(1d)) can be rewritten in linear form as following

\[ \partial_x (c + q) + Pe f(t) \partial_c c + \partial_y \psi = \nabla^2 c, \quad \text{(4a)} \]

\[ \nabla^2 \psi = -R_p \partial_x c, \quad \text{(4b)} \]

\[ q = \frac{\lambda}{\Gamma(1 - \alpha)} \int_0 t - t' \partial_x \partial_y \partial_c c \partial_t', \quad \text{ (4c)} \]

where \( \partial_x \) and \( \partial_y \) denote corresponding derivatives, while \( \nabla^2 = \partial_x^2 + \partial_y^2 \) is the two-dimensional Laplace operator. With boundary condition

\[ c, q, \psi \big|_{\gamma = 0} = 0 \quad \text{ (5)} \]

the system (equations (4),(5)) represents the linear stability problem.

3. Steady external flux

Let us consider the time-independent external filtration flux or \( f(t) = 1 \). In order to find the solution we applied the Laplace-Fourier transformation method [7] to the governing equation system (equations (4a)-(4c)). Let \( \tilde{F}(r, s) = \int_0^\infty \int F(r, t) \exp(-st) dt \) is the Laplace transform of some function \( F(r, t) \) varying in time and \( \tilde{F}(k, s) = \int \int F(r, t) \exp(-i k \cdot r) d k \) is the Fourier transform of \( F(r, t) \) varying in space. Thus in Laplace-Fourier space the system (Equations 4a-c) has the form

\[ \hat{c}(k, s)(s + \lambda s^\alpha) - \hat{c}(k, 0) = -k^2 \hat{c} - Pe i k \hat{c} - i k \hat{\psi}, \quad \text{(6a)} \]

\[ -k^2 \hat{\psi} = -i k R_p \hat{c}, \quad \text{(6b)} \]

Substitution Equation 6b into Equation 6a defines the expression for mobile volume concentration in Laplace-Fourier space via

\[ \hat{c}(k, s) = \hat{c}(k, 0)(s + \lambda s^\alpha + k^2 + ik Pe - k^2 R_p k^{-2})^{-1}, \quad \text{(7)} \]

which coincide with [5].

The inverse Laplace-Fourier transformation can be expressed as a sum of residues: \( 2 \pi i \sum \text{Res} \left( \exp(st) f(s) \right) \). According to Cauchy’s residue theorem we can define the contour integral in such a way that \( \exp(st) \to 0 \). Thereby in order to analyze the stability of the solution of equation (7) its denominator has to be considered.

Let us find the neutral perturbation (for which \( s = -i \omega \)). Substitution it into denominator of equation (7) and extraction of real and imaginary parts gives the system of equation, which define the frequency of critical perturbations and the critical value of the Rayleigh–Darcy number as

\[ \omega - \lambda \text{ Im} (i^{\alpha}) \omega^\alpha - k Pe = 0, \quad \text{ (8a)} \]
\[ R_p = \frac{\left( \pi^2 n^2 + k_z^2 \right)^2}{k_z^2} + \frac{\left( \pi^2 n^2 + k_z^2 \right)}{k_z^2} \lambda \Re(i^{3\alpha})\omega^\alpha, \]  

(8b)

It should be noted that without immobilization (\( \lambda = 0 \)) these expressions (equation (8)) match the well-known solution, which has been first obtained in [2] for the case when the external flux has no influence on the convection threshold. Indeed substituting \( \lambda = 0 \) one can get

\[ \omega = k_z \text{Pe}, \]  

(9a)

\[ R_p = \frac{\left( \pi^2 n^2 + k_z^2 \right)^2}{k_z^2}. \]  

(9b)

This convection threshold (equation (9b)) is the same for the case without external filtration flux. As it can be seen from equation (8a), if external flux is absent (\( \text{Pe} = 0 \)) the oscillatory convection does not occur and \( \omega = 0 \). Thus the critical value of the Rayleigh–Darcy number is the same as described by equation (9b).

Figure 2. Dependences of the critical wave number (a), the Rayleigh–Darcy number (b) and the oscillation frequency (c) on the Péclet number for \( \alpha = 0.8 \) (solid line), \( \alpha = 0.85 \) (dash line) and \( \alpha = 0.9 \) (dash dot line). \( \lambda = 2 \).

3.1. Limiting case of instantaneous relaxation. Natural results can be obtained easily for limiting case \( \alpha = 1 \). From equation (8a) it can be seen that oscillation frequency has the form
\[ \omega = \frac{k_P e}{1 + \lambda} . \]  

Meanwhile equation (8b) for \( \alpha = 1 \) will give the same result for the critical Rayleigh–Darcy number as equation (9b). These results are reasonable and proof that equations (8a), (8b) are consistent. Indeed, this limiting case corresponds to the linear relation between immobile and mobile concentrations; \( Q = \lambda C \), which means that instead of Equation 1c1 one can solve the ordinary diffusion equation with additional factor for time derivative \((1 + \lambda)\). Thus, the frequency has to be reduced by this factor and convection threshold does not change.

3.2. General case. The numerical computations of equations (8a), (8b) are presented on Figure 2, for the values of the exponent of the Levy stable law widely used in experiments.

As it is shown on Figure 2, the dependencies become nonlinear with sufficient values of Peclet number. Hence, in case of steady external filtration flux the account of immobilization leads to increase of convection threshold with growth of filtration intensity.

4. Weak non-steady external flux

Let us consider the influence of time-dependent filtration flux on the convection threshold, so in this case \( f = A \cos \Omega t \). We assume that the flux is weak, it allows to find the solution as a series of \( \varepsilon = k_P e \ll 1 \):

\[ c = c^{(0)} + \varepsilon c^{(1)} + \varepsilon^2 c^{(2)} + \ldots \]
\[ q = q^{(0)} + \varepsilon q^{(1)} + \varepsilon^2 q^{(2)} + \ldots \]
\[ \gamma = \varepsilon \gamma^{(1)} + \varepsilon^2 \gamma^{(2)} + \ldots \]

where \( \gamma = k_P^2 R_J k^{-2} - k^2 \) is the critical Rayleigh–Darcy number without immobilization (see equation (9b)).

In Laplace-Fourier space the system (equations (4a)-(4c)) yields

\[ \hat{c}(k, s)(s + \lambda s^\alpha) - \hat{c}(k, 0) = \gamma \hat{c}(k, s) - \varepsilon i A \cos \Omega t c(r, t) . \]  

In the first-order the mobile volume concentration can be written as

\[ \hat{c}^{(1)}(k, s) = -\hat{c}(k, 0) \left( \frac{\gamma^1}{(s + \lambda s^\alpha)} \right) + i A f(k, s) , \]  

where

\[ f = \frac{1}{2\pi i} \int_0^\infty \cos \Omega t e^{-i \omega t} \int_{-i\infty}^{i\infty} (p + \lambda p^\alpha)^{-1} e^{i\omega t} dp = \frac{1}{2} \left( \frac{1}{s + i\Omega + \lambda (s + i\Omega)^\alpha} + \frac{1}{s - i\Omega + \lambda (s - i\Omega)^\alpha} \right) . \]

The inverse transformations of equation (12) give a linear growth due the term with \( \gamma^1 \), which corresponds to the pole of order 2. Thus, in the first order the perturbations of the Rayleigh–Darcy number vanishes: \( \gamma^1 = 0 \); meanwhile the solution for mobile concentration becomes

\[ \hat{c}^{(1)}(k, s) = \hat{c}(k, 0) \left( \frac{i A}{2} \left( \frac{1}{s + i\Omega + \lambda (s + i\Omega)^\alpha} + \frac{1}{s - i\Omega + \lambda (s - i\Omega)^\alpha} \right) \right) , \]  

In the second-order by analogy the solution of equation (11) can be obtained as

\[ \hat{c}^{(2)}(k, s) = \hat{c}(k, 0) \left( \frac{A^2}{4(s + \lambda s^\alpha)} \left( \frac{1}{s + i\Omega + \lambda (s + i\Omega)^\alpha} + \frac{1}{s - i\Omega + \lambda (s - i\Omega)^\alpha} \right) \right) + \frac{\gamma^2}{s + \lambda s^\alpha} \ldots . \]

By vanishing the terms corresponding to the pole of order 2 the rectification of \( \gamma \) gives
Thus, the expected correction to the critical Rayleigh–Darcy number is

\[
R_p = \left( \frac{\pi^2 n^2 + k_f^2}{k_f^2} \right)^2 + \left( \frac{\pi^2 n^2 + k_f^2}{k_f^2} \right)^2 \frac{\lambda \Omega^\alpha \cos \left( \frac{\pi \alpha}{2} \right)}{4 \Omega^2 + \lambda^2 + \lambda \Omega^2 + \lambda \Omega^\alpha \cos \left( \frac{\pi \alpha}{2} \right)}. \tag{15}
\]

Note, that in the absence of immobilization (\( \lambda = 0 \)) or in limiting case with \( \alpha = 1 \), the correction (equation (14)) vanishes, which coincide with known results [8]. The dependence of the correction on external flux frequency is plotted on Figure 3. As it is expected the correction decrease dramatically with growth of external frequency.

5. Conclusion
The linear stability problem for solutal convection in a horizontal layer of a porous medium at a given vertical concentration gradient with imposed external horizontal filtration flux is studied. The effect of solute immobilization when the solute particles can stick to the solid matrix of the porous medium is taken into account in the framework of fractal linear MIM model. The solute concentration difference between the layer boundaries and the horizontal external filtration flux are assumed as constant. Two cases are considered: steady and weak time-dependent external filtration flux.

In the case of steady flux the system of equations that determine the frequency of neutral oscillations and the critical value of the Rayleigh-Darcy number is derived (equations (8a),(8b)). Neutral curves of the critical parameters on the governing parameters are calculated. Stability maps are obtained numerically in a wide range of parameters of the system. It was found that taking immobilization into account leads to an increase in the critical value of the Rayleigh-Darcy number with an increase in the intensity of the external filtration flux.

The case of weak time-dependent external flux is investigated analytically. The results show that at low flow velocity the modulation makes additional positive contribution to the critical value of the Rayleigh–Darcy number. As a result, the formula, which determines the threshold value, consists of two positive terms: the first term is the main critical value without external flow, and the second term is related to the effect of the external flow modulation (equation (15)). Thus the introduction of modulation leads to stabilization.

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