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Effect of solute immobilization on the stability problem within the fractional model in the solute analog of the Horton-Rogers-Lapwood problem*

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Abstract. The paper is devoted to the linear stability analysis within the solute analogue of the Horton-Rogers-Lapwood (HRL) problem. The solid nanoparticles are treated as solute within the continuous approach. Therefore, we consider the infinite horizontal porous layer saturated with a mixture (carrier fluid and solute). Solute transport in porous media is very often complicated by solute immobilization on a solid matrix of porous media. Solute immobilization (solute sorption) is taken into account within the fractal model of the MIM approach. According to this model a solute in porous media immobilizes within random time intervals and the distribution of such random variable does not have a finite mean value, which has a good agreement with some experiments. The solute concentration difference between the layer boundaries is assumed as constant. We consider two cases of horizontal external filtration flux: constant and time-modulated. For the constant flux the system of equations that determines 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. We have 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. We have 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 wave number. For moderate time-dependent filtration flux the differential equation with Caputo fractional derivatives has been obtained for the description of the behavior near the convection instability threshold. This equation is analyzed numerically by the Floquet method; the parametric excitation of convection is observed.

1 Introduction

The investigation of the passive transport of a solute through a porous media is of interest not only for the numerous practical applications, but also from the theoretical point of view because of its deviation from the linear Fick's law [1]. As has been demonstrated in experiments (for example, [2,3]), due to the rather complex spatial structure of porous media, the diffusion process is slower than predicted by Fick's law. Many studies have been devoted to the non-Fickian effect on processes in porous media (see, for example, [4–6]). Frequently the deceleration is usually explained by the adsorption of the solute particle by the porous matrix, in other words, particles can stick to the solid matrix and do not move for a sufficient time. Such behaviour is called the immobilization and is often modelled by the MIM (mobile/immobile media) approach [7]. According to this approach the solute concentration can be divided into two phases: mobile and immobile, which are connected with each other through the kinetic law.

The MIM approach was first suggested in [8] with the linear kinetics law for the concentrations of immobile and mobile solutes. Although this model predicts the retardation of the diffusion process, it gives a poor fit to the experimental data. The improvement of this model was suggested in [9] and developed in [7] by the introducing of the first-order kinetics model. The developed model adequately describes the diffusion of solutes at low concentrations (lower than the concentration of saturation of a solid porous matrix). A model of this type is often called the linear sorption model or standard MIM model. The results obtained by this model have good agreement with

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some experiments [10,11], but do not explain the problem of the localized concentration peak dissolution. For this problem at large times the standard MIM model predicts an exponential decline of concentration with respect to time, meanwhile, experimental studies demonstrated that it should be the power law. To describe the corresponding behaviour at large time the fractional mobile/immobile model (fMIM) was developed [12]. According to this model a solute in porous media immobilizes within random time intervals and the distribution of such random variable does not have a mean value. The kinetic law is the linear relation between the influx to the immobile phase and the fractional Caputo derivative of mobile phase concentration with respect to time. In paper [13] it was demonstrated that this model gives a right behaviour at long time for low solute concentrations. At high concentration the author of [14] has developed the non-linear fractional MIM model which describes a long-time limit.

The present work is devoted to the study of solutal convection in a horizontal layer of a porous medium under an imposed horizontal filtration flux. The problem in a similar configuration, without immobilization and external filtration flux, was considered in [15]. It was obtained that the convective regime has a form of a set of convective cells, with a width equal to the layer thickness. The convection under an external horizontal flux was studied in [16]. It was shown that the influence of a steady external flux leads to the excitation of an oscillatory mode with the same wavelength of critical perturbations and the same critical value of the Rayleigh-Darcy number, which was obtained without external flux. The effect of the immobilization of the solute particles on the convection was investigated in [17] using the standard MIM model [7]. It was numerically obtained that due to immobilization the critical values of the parameters become dependent on the external flux intensity. In the present paper the immobilization was considered in the framework of the fractal linear model of the mobile/immobile medium (fMIM) approach.

The mechanism of instability in the series of systems with fractional derivatives was investigated in [18,19]. The analysis of pattern formation and stability in the fractional diffusion-reaction system was presented in [20,21]. The authors found new oscillatory-type instabilities and investigated their spatial spectrum.

The paper is organized in the following way. Section 2 is devoted to the problem statement, where governing equations and boundary conditions are described. In sect. 3 the case of the steady filtration flux is investigated analytically using the Laplace-Fourier transformation method. The case of time-dependent filtration flux is considered numerically in sect. 4. Section 5 provides some conclusions.

2 Problem statement

Let us consider the flow of a mixture through the horizontal layer of a porous medium (the problem configuration is sketched in fig. 1). The mixture consists of solid nanoparticles and ambient fluid; the solid nanoparticles are considered as a solute within the continuous approach. The flow inside the layer is induced by an external horizontal filtration with velocity $\mathbf{V}$. 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 some sufficient time, is taken into account. We model the immobilization process using the MIM (mobile/immobile media) approach. According to [7] the total solute concentration is assumed to have two phases: the mobile phase (with volume concentration C) and the immobile phase (with volume concentration Q). Solute concentrations at the upper and lower boundaries of the layer are considered to be constant. The solute is heavier than the carrier fluid, which can lead to a solutal convection.

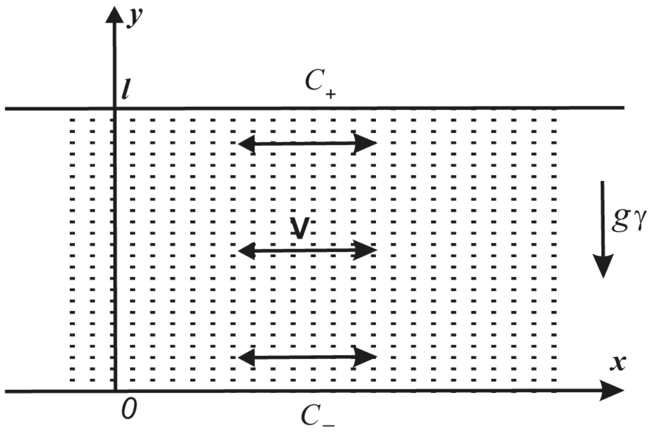

Fig. 1. Sketch of the problem configuration.

It is convenient to write the equations of the solutal convection in porous media within the framework of the Bousinesq approximation [1]:

$$\begin{gathered}\frac{\eta}{\kappa}\phi\mathbf{V} = -\nabla p + \rho\beta_c g C\gamma,\\ \nabla\cdot\mathbf{V} = 0,\\ \partial_t C + \partial_t Q = D\nabla^2 C - \mathbf{V}\cdot\nabla C.\end{gathered} \tag{1}$$

We use following notation ∂_t for the time derivative, $\mathbf{V}$ for two-dimensional flow velocity, γ for a unit vector along gravity, g is the gravity acceleration, p for a deviation of the pressure from the hydrostatic one. Here ρ and η are the density and dynamic viscosity of the ambient liquid and D, ϕ, κ are the effective diffusivity, porosity and permeability of porous media.

This system represents the Darcy law with the buoyancy force [1], the incompressibility condition and the advective-diffusion equation with additional influx into the immobile phase ($\partial_t Q$). To close the system of equations one needs to add 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. We used the fractional MIM model with the following kinetic equation [12]:

$$Q = \frac{\lambda}{\Gamma(1-\alpha)}\int_0^t \frac{C(r,t')}{(t-t')^\alpha}\mathrm{d}t', \tag{2}$$

where λ is the mobility parameter and α is the exponent of the Levy stable law. The mobility parameter characterizes the portion of the immobile concentration in the total concentration. The exponent of the Levy stable law varies from 0 to 1 and describes the relaxation of the system to the dynamical equilibrium state between mobile and immobile phases. For $\alpha = 0$ the relaxation time is infinite, the process of mobilization is blocked and equilibrium cannot be reached; whereas in the opposite case $\alpha = 1$ corresponds to the instantaneous relaxation ($Q = \lambda C$). Meanwhile from the theoretical point of view the range is $0 < \alpha < 1$, in practical application for usual porous materials $\alpha > 0.5$.

In order to obtain a dimensionless equation system the following scales have been chosen: l^2/D as the scale for time, l for length, $C_0 = C_+ - C_-$ for concentration, D/l for velocity and $D\eta\phi/\kappa$ for pressure. In dimensionless form the governing equation system eqs. (1), (2) reads

$$\begin{gathered}\mathbf{V} = -\nabla p + RpC\gamma,\\ \nabla \cdot \mathbf{V} = 0,\\ \partial_t C + \partial_t Q = \nabla^2 C - \mathbf{V} \cdot \nabla C,\\ Q = \frac{\lambda}{\Gamma(1-\alpha)} \int_0^t \frac{C(r,t')}{(t-t')^\alpha} \mathrm{d}t',\end{gathered} \tag{3}$$

with boundary conditions

$$\begin{aligned} y = 0: \quad & C = 0, \quad V_y = 0,\\ y = 1: \quad & C = 1, \quad V_y = 0,\\ x = \pm\infty: \quad & V_x = Pef(t). \end{aligned} \tag{4}$$

The boundary value problem, eqs. (3), (4) is characterized by two governing parameters: the Rayleigh-Darcy number and the Péclet number

$$Rp = \frac{\beta_c C_0 g l \kappa \rho}{\phi \eta D}, \qquad Pe = \frac{Vl}{D}.$$

The problem (eqs. (3), (4)) admits the uniform solution defined by $C = y$ and $\mathbf{V} = (Pef(t), 0)$. Our interest is on the linear analysis of the perturbations of this solution. To find perturbations let us consider $c = C - y$, $\mathbf{v} = (u, w) = (V_x - Pef(t), V_y)$ and introduce the stream function ψ as $u = -\partial_y \psi$ and $w = \partial_x \psi$. Thus, neglecting the non-linear terms in eq. (3), it can be derived that

$$\begin{gathered}\nabla^2 \psi = -Rp \partial_x c,\\ \partial_t c + \partial_t q + Pef(t)\partial_x c + \partial_x \psi = \nabla^2 c,\\ q = \frac{\lambda}{\Gamma(1-\alpha)} \int_0^t \frac{c(r,t')}{(t-t')^\alpha} \mathrm{d}t',\end{gathered} \tag{5}$$

with boundary conditions

$$c, q, \psi|_{y=0,1} = 0. \tag{6}$$

In order to find the solution of eqs. (5), (6) it is convenient to apply the Laplace-Fourier transformation method [22]. If $\mathcal{L}[F](\mathbf{r}, s) = \int_0^\infty F(\mathbf{r}, t) \exp\{-st\}\mathrm{d}t$ is the Laplace transform of some function $F(\mathbf{r}, t)$ varying in time and $\hat{F}(\mathbf{k}, t) = \iiint_{-\infty}^{\infty} F(\mathbf{r}, t) \exp\{-i\mathbf{k} \cdot \mathbf{k}\}\mathrm{d}\mathbf{k}$ is the Fourier transform of $F(\mathbf{r}, t)$ varying in space, then in the Laplace-Fourier space the system eq. (5) can be rewritten as a single equation for the Laplace-Fourier transform of perturbation of mobile concentration c:

$$\begin{aligned}&\mathcal{L}[\hat{c}](\mathbf{k}, s)(s + \lambda s^\alpha) - \hat{c}(\mathbf{k}, 0) =\\ &-\mathcal{L}[\hat{c}](\mathbf{k}, s)\left(k^2 - k_x^2 Rp k^{-2}\right)\\ &-ik_x Pe \mathcal{L}[f(t)\hat{c}(\mathbf{k}, t)],\end{aligned} \tag{7}$$

where $\mathbf{k}$ is a wave vector and k_x its x-component, $k^2 = k_x^2 + \pi^2 n^2$. To solve this equation the external filtration flux $f(t)$ has to be specified.

3 Stationary flux

Let us start by considering the time-independent external filtration flux ($f(t) = 1$). The solution of eq. (7) in the Laplace-Fourier space is

$$\begin{aligned}&\mathcal{L}[\hat{c}](\mathbf{k}, s) =\\ &\hat{c}(\mathbf{k}, 0)\left(s + \lambda s^\alpha + k^2 + ik_x Pe - k_x^2 Rp k^{-2}\right)^{-1}.\end{aligned} \tag{8}$$

The inverse Laplace-Fourier transformation of eq. (8) gives the system of equations, which defines the frequency of critical perturbations and the critical value of the Rayleigh-Darcy number, which, for the critical oscillatory perturbation with $s = -i\omega$, reads

$$\begin{gathered}\omega - \lambda \operatorname{Im}\left(i^{3\alpha}\right)\omega^\alpha - k_x Pe = 0,\\ Rp = \frac{(\pi^2 n^2 + k_x^2)^2}{k_x^2} + \frac{(\pi^2 n^2 + k_x^2)}{k_x^2} \lambda \operatorname{Re}\left(i^{3\alpha}\right)\omega^\alpha.\end{gathered} \tag{9}$$

To check that our solution is consistent, let us consider some limiting cases. It is natural that without immobilization ($\lambda = 0$) eq. (9) gives

$$\begin{gathered}\omega = k_x Pe,\\ Rp = \frac{(\pi^2 n^2 + k_x^2)^2}{k_x^2},\end{gathered} \tag{10}$$

which matches the well-known solution, when the external flux has no influence on the convection threshold [16]. It should be noted that the critical value of the Rayleigh-Darcy number is the same, if the external flux is absent ($Pe = 0$). In this case the oscillatory convection does not occur: $\omega = 0$.

Another limiting case for which the convection threshold does not change is $\alpha = 1$. Indeed, substituting it into eq. (9) gives the same value of the critical Rayleigh-Darcy number, but the value of the frequency will decrease, namely

$$\omega = \frac{k_x Pe}{1 + \lambda}.$$

Indeed, this limiting case corresponds to the linear relation between immobile and mobile concentrations; $Q = \lambda C$. Thus the advective diffusion equation (3) is

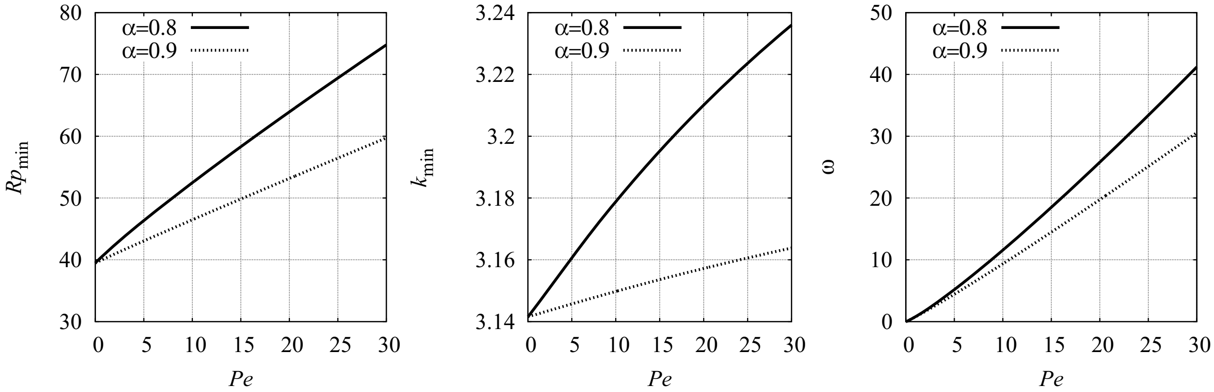

Fig. 2. Dependences of the critical Rayleigh-Darcy number (left panel), the critical wave number (middle panel) and the frequency of critical perturbation oscillation (right panel) on the Péclet number for $\alpha = 0.8$ (solid line) and $\alpha = 0.9$ (dashed line); $\lambda = 2$.

transformed to the ordinary diffusion equation with additional factor for the time derivative $1 + \lambda$.

In the general case the dependence of critical perturbations parameters on the problem parameters is investigated. The critical value of Rayleigh-Darcy number is defined as the minimal possible value (Rp_{min}) the corresponding values of the horizontal wave number (k_{min}) and the frequency of critical perturbations (ω_{min}) are obtained from eqs. (9).

The dependences of critical parameters on the Péclet number are presented in fig. 2. It is seen that the increase of the external flow intensity leads to the growth of all the parameters but the growth of the wave number is very tiny. This result corresponds to the findings of [17]. Also in the case of $\alpha = 0.8$ the effect is greater than for $\alpha = 0.9$ because the immobilization dynamics is more intensive for $\alpha = 0.8$. We remember here that in the case $\alpha = 1$ we find an instateneous relaxation without any dynamics, but the mechanism of stabilisation is a dynamical transition of concentration between mobile and immobile phases.

Figure 3 shows the dependences of the critical parameters on the mobility parameter (λ). The most interesting effect is observed for moderate values of λ. The small values corresponds to the weak immobilization and the obtained results are consistent to the findings of [16]. In the case of great λ values ($\lambda > 16$) the most part of the solute is kept in the immobile phase and the dynamics of phase transition slows down. Due to this fact, the critical value of the Rayleigh-Darcy number increases but also turns to the constant value, for $\lambda > 16$ all other parameters turn to the values which correspond to the steady state [15] (case without external flow). The same effects are obtained from the analysis of α variation (see fig. 4). The immobilization dynamics is the most intensive for $\alpha = 0.5$ and as α is increasing, the effect of immobilization decays.

4 Non-stationary flux

Let us consider the time-dependent external filtration flux as a harmonic function with an amplitude A and a frequency Ω: $f = A\cos \Omega t$. In this case eq. (7) in the Laplace-Fourier space has the form

$$\begin{aligned}&\mathcal{L}\left[\hat{c}\right](\mathbf{k},s)\left(s+\lambda s^{\alpha}\right)-\hat{c}\left(\mathbf{k},0\right)=\\&-\mathcal{L}\left[\hat{c}\right](\mathbf{k},s)\left(k^{2}-k_{x}^{2}Rpk^{-2}\right)\\&-ik_{x}PeA\mathcal{L}\left[\cos\Omega t\hat{c}\left(k,t\right)\right].\end{aligned}\tag{11}$$

Equation (7) contains only the multiplication of amlitude A and Péclet number Pe, so we will assume that $A = 1$ and Pe plays the role of external flux amlitude. Despite the fact that a full solution of this system can be found only numerically, we provided an analytic investigation for the case of weak flux, when $\varepsilon = k_x Pe \ll 1$. Hence we take ε as a small parameter and seek the solution in the form of expansions with respect to ε

$$h_j = h_j^{(0)} + \varepsilon h_j^{(1)} + \varepsilon^2 h_j^{(2)} + \dots,$$

where the symbol h_j denotes the field functions c, q and β, where $\beta = {k_x}^2 Rpk^{-2} - k^2$. The value $\beta = 0$ corresponds to the critical form of perturbations in the case without immobilization (see eq. (10)).

In the first order with respect to ε we obtain the concentration of the mobile phase as

$$\begin{aligned}&\mathcal{L}\left[\hat{c}^{(1)}\right](\mathbf{k},s)=\\&-\frac{\hat{c}(\mathbf{k},0)}{(s+\lambda s^{\alpha})}\left(\frac{\beta^{(1)}}{(s+\lambda s^{\alpha})}+\frac{i}{2}G\left(\mathbf{k},s\right)\right),\end{aligned}\tag{12}$$

where

$$G = \frac{1}{s + i\Omega + \lambda(s + i\Omega)^{\alpha}} + \frac{1}{s - i\Omega + \lambda(s - i\Omega)^{\alpha}}.$$

It can be seen from eq. (12), that the term, which is proportional to $\beta^{(1)}$, corresponds to a pole of second order. Hence the inverse Laplace transformation of eq. (12) leads us to calculate the residue for the pole of second order which is proportional to the linear function of time. As a result this term will increase with time. But physically there is no reasons for such growth, thus we should

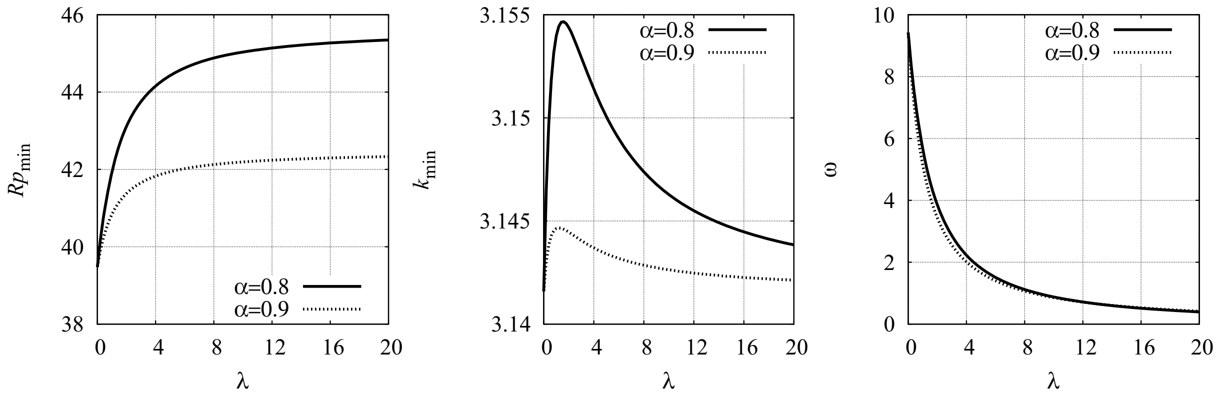

Fig. 3. Dependences of the critical Rayleigh-Darcy number (left panel), the critical wave number (middle panel) and the frequency of critical perturbation oscillation (right panel) on the mobility parameter for $\alpha = 0.8$ (solid line) and $\alpha = 0.9$ (dashed line); $Pe = 3$.

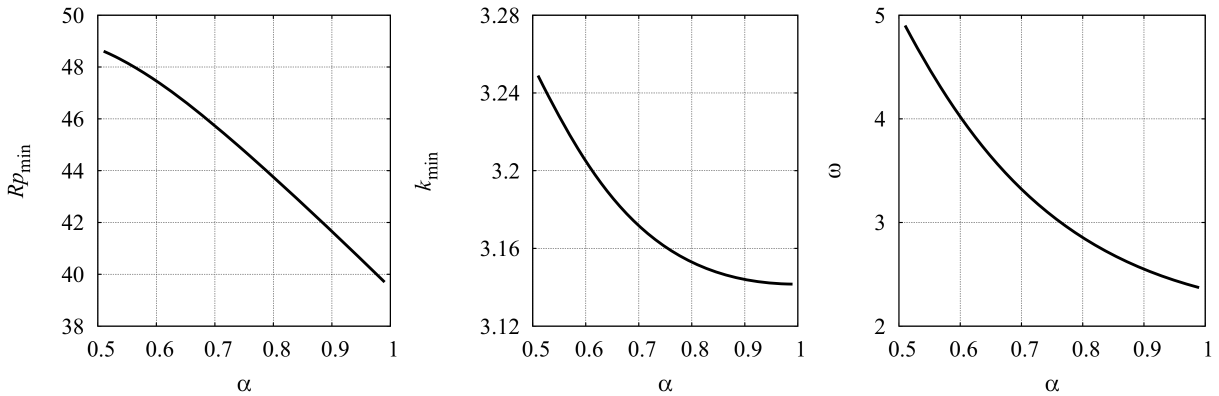

Fig. 4. Dependences of the critical Rayleigh-Darcy number (left panel), the critical wave number (middle panel) and the frequency of critical perturbation oscillation (right panel) on the exponent of the Levy stable law for $Pe = 3$, $\lambda = 2$.

state that $\beta^{(1)} = 0$ (see [23]). In order to obtain the perturbation of the critical Rayleigh-Darcy number we have to consider the second order with respect to ε.

Indeed in the second order the solution of eq. (11) reads

$$\mathcal{L}\left[\hat{c}^{(2)}\right](\mathbf{k}, s) = \frac{\hat{c}(\mathbf{k}, 0)}{s + \lambda s^\alpha}\left(-\frac{G}{4(s + \lambda s^\alpha)} + \frac{\beta^{(2)}}{s + \lambda s^\alpha} + \dots\right).$$

from which the second oder for β series can be obtained as

$$\beta^{(2)} = \frac{1}{4}\,\frac{2\lambda(\Omega)^\alpha\cos(\alpha\pi/2)}{\Omega^2 + \lambda^2\Omega^{2\alpha} + 2\lambda(\Omega)^\alpha\cos(\alpha\pi/2)}.$$

In this way, the corresponding correction to the critical Rayleigh-Darcy number is

$$Rp = \frac{(\pi^2 n^2 + k_x^2)^2}{k_x^2} + Rp^* = \frac{(\pi^2 n^2 + k_x^2)^2}{k_x^2} + \left(\pi^2 n^2 + k_x^2\right)^2 \frac{Pe^2}{4} \times \frac{\lambda\Omega^\alpha\cos(\pi\alpha/2)}{\Omega^2 + \lambda^2\Omega^{2\alpha} + \lambda\Omega^\alpha\cos(\pi\alpha/2)}. \qquad (13)$$

The result of the tabulation of the expression of the correction to the critical Rayleigh-Darcy number is presented in fig. 5. It is shown that the correction decreases dramatically with the increasing of the external flux frequency value Ω. it should be noted that the correction (eq. (13)) vanishes in the case without immobilization ($\lambda = 0$) or either when $\alpha = 1$, these results have a good agreement with the results obtained in [24] and with the findings of sect. 3.

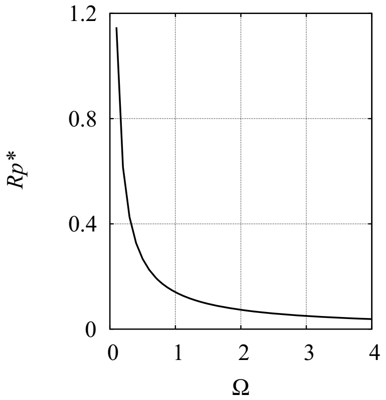

Fig. 5. Dependence of the correction to the critical Rayleigh-Darcy number (Rp^* from eq. (13)) on the external frequency Ω for $A = 2$, $\alpha = 0.8$ and $\lambda = 2$.

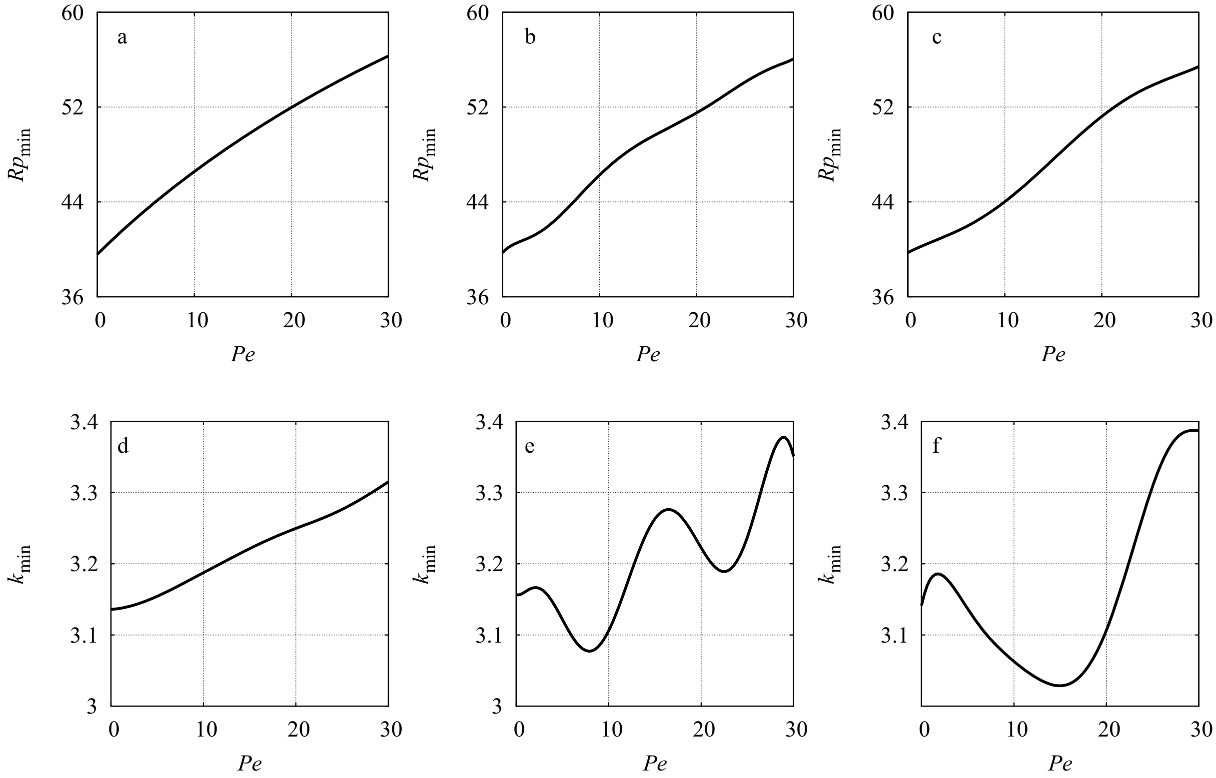

Fig. 6. Dependences of the Rayleigh-Darcy number (a)–(c) and the critical wave number (d)–(f) on the Péclet number for different frequencies of the external flux: $\Omega = 1$ ((a), (d)), $\Omega = 8$ ((b), (e)), $\Omega = 16$ ((c), (f)). $A = 1$, $\alpha = 0.8$ and $\lambda = 2$.

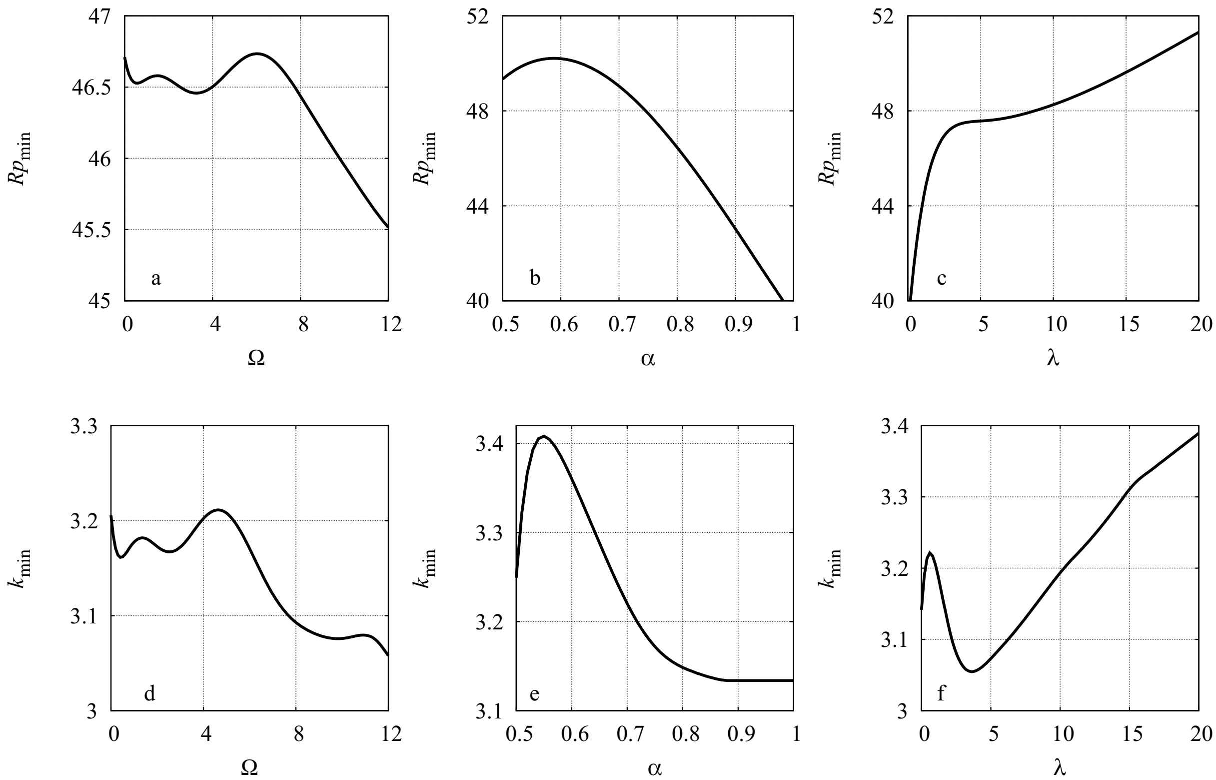

Fig. 7. Dependences of the Rayleigh-Darcy number (a)–(c) and the critical wave number (d)–(f) on the frequency of modulation ((a), (d)), on the exponent of the Levy stable law ((b), (e)) and on the mobility parameter ((c), (f)). The figures are plotted for the following values of parameters: $\alpha = 0.8$, $\lambda = 2$ ((a), (d)), $\Omega = 6$, $\lambda = 2$ ((b), (e)), $\Omega = 6$, $\alpha = 0.8$ ((c), (f)), $A = 1$ and $Pe = 10$.

The solution of eq. (11) under moderate time-dependent filtration flux was found numerically using the finite difference method. The expression for immobile concentration was modelled by the Riemann-Liouville fractional derivative according to [13]. The expected critical values of the parameters were obtained by the Floquet method [23]. The dependence of the critical values of parameters $Rp = Rp_{min}$ and $k_x = k_{min}$ on the Péclet number for different external flux frequency is plotted in fig. 6. It can be seen that when Ω is small, the critical Rayleigh-Darcy number value differs insignificantly from the value obtained for the steady flux case. With increasing external filtration frequency, the oscillations of Rp became significant, thus one can observe the parametric excitation of convection. A strong dependence of the critical wave number on the external flux parameters (*i.e.*, Péclet number and frequency Ω) allow to control the structure of convective cells.

In fig. 7 the dependences of critical parameters on frequency and immobilization parameters are presented. The observed effect is the same as previosly discussed: the increasing of frequency leads to the growth of variation in critical parameters. The effect of immobilization parameters is described in the standard way —the intensification of immobilization (increasing of λ and decreasing of α in the interval $0.5 < \alpha < 1$) leads to the growth of variation of critical parameters.

5 Conclusion

We have considered the linear stability problem for solutial convection. The investigation was carried out for a horizontal layer of a porous medium at a given vertical concentration gradient with imposed external horizontal filtration flux. The effect of solute immobilization when the solute particles can stick to the solid matrix of the porous medium is taken into account using fractal linear mobile/immobile media model. The solute concentration difference between the layer's boundaries is maintained constant. The investigation is limited within two cases: steady and unsteady external filtration flux.

The system of equations that determines 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 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 of the critical value of the Rayleigh-Darcy number with growth of 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 is the main critical value without external flow, and the second is related to the effect of the external flow modulation. Thus the introduction of modulation leads to stabilization.

The case of moderate values of time-dependent external flux is studied by numerical methods. The parametrical excitation of convection is observed and the possibility of controll for convective flow structure by the variation of external flux parameters is shown.

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Author contribution statement

BM, LK formulated the problem. BM derived eqs. (5)–(7). BM performed the analytical analysis of the weakness of the non-stationary flux. LK performed numerical simulations. BM and LK wrote the main manuscript text. All authors reviewed the manuscript.

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