Influence of miscible viscous fingering on spreading of solutes in porous media

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Abstract

Viscous fingering (VF) between miscible fluids of different viscosities can affect the dispersion of localized fluid samples in porous media and the adsorption of solutes initially dissolved in this sample onto the porous matrix. Here, we investigate numerically the influence of VF due to the difference in sample solvent and displacing fluid viscosities by considering a sample solvent more or less viscous than the carrier fluid in a three component system. The model equations are Darcy's law for the fluid velocity coupled with a mass-balance equation for the sample solvent and solute concentrations. The influence of the retention parameter κ' controlling the adsorption of the solute on the porous matrix and various other parameters that control VF is studied.

1 Introduction

Miscible viscous fingering is an interfacial fluid flow instability that occurs when a less viscous fluid displaces another more viscous miscible one in a porous medium, leading to the formation of finger like patterns at the interface of both fluids [1]. VF impacts a variety of practical applications such as oil recovery, filtration and hydrology, liquid chromatography, and even medical applications. In aquifer contamination problems and in liquid chromatography (LC), used to separate the chemical components of a sample by passing it through a porous medium, VF can be of concern when the sample solvent has a viscosity different from that of the mobile phase [2, 3, 4, 5]. Depending whether the sample is more or less viscous than the carrier fluid, the log-mobility ratio R (defined as $R = ln(\mu_2/\mu_1)$ where μ_2 and μ_1 are the viscosities of the sample and of the carrier) is respectively positive or negative. In the case of a linear displacement of a finite slice of fluid, $R > 0$ leads to fingering of the rear interface of the sample where the less viscous carrier invades the more viscous sample. If $R < 0$, it is on the contrary the frontal interface of the sample that develops fingers where the less viscous sample displaces the more viscous bulk solution [5]. The possible adsorption of solutes can be affected by this VF leading to broader contamination zones in aquifers or anomalous peak shapes in reversed-phase liquid chromatography [4]. The objective of our work is to analyse theoretically the effects of miscible viscous fingering due to viscosity differences between the sample solvent and the carrier fluid on the adsorption of retained solutes. Moreover, we analyze the differences in fingering dynamics between the positive and negative log-mobility ratio cases to compare to LC experimental results [6].

2 Mathematical formulation and nonlinear simulation

We consider a two dimensional porous medium of length L_x and width L_y in which a sample of finite width is injected at an initial time $t = 0$. This sample contains a solute/analyte dissolved in a solvent of viscosity μ_2 different from the eluent (carrier fluid) viscosity μ_1. Once the sample is injected in the porous matrix the solute can adsorb onto the porous matrix following a reversible adsorption-desorption reaction. We assume

that the analyte concentration in the sample solvent is small, hence μ does only depend on the concentration of the sample solvent c which is equal to c_1 in the sample and to zero in the displacing fluid.

The fluid is assumed incompressible and the flow inside the porous medium is governed by Darcy's law. The governing equations of the system are the convection-diffusion evolution equation for the concentration c of the sample solvent and a mass balance equation for the solute concentration c_m in the mobile phase. We take a concentration dependent viscosity $\mu(c) = \mu_1 e^{R\frac{c}{c_1}}$, where $R = ln\frac{\mu_2}{\mu_1}$. If $R > 0$, then $\mu_1 < \mu_2$ and the rear interface of the sample is unstable due to viscous fingering while the frontal interface is stable. If $R < 0$, the rear interface is stable while viscous fingering operates at the frontal interface. A linear isotherm adsorption model is assumed between the stationary and mobile phase for the solute concentration. To non-dimensionalise the governing equations, we choose the velocity of displacing fluid U as the characteristic velocity, a length scale $L_c = D_x/U$, and a time scale $t_c = D_x/U^2$ [1, 2, 3]. Switching to a coordinate system moving with speed U, the non-dimensional governing equations in terms of the stream function $\psi(x, y)$ become :

$$\bigtriangledown^2\psi \;\; = \;\; -R\left(\frac{\partial\psi}{\partial x}\frac{\partial c}{\partial x} + \frac{\partial\psi}{\partial y}\frac{\partial c}{\partial y} + \frac{\partial c}{\partial y}\right), \tag{1}$$

$$\frac{\partial c}{\partial t} + \frac{\partial\psi}{\partial y}\frac{\partial c}{\partial x} - \frac{\partial\psi}{\partial x}\frac{\partial c}{\partial y} \;\; = \;\; \frac{\partial^2 c}{\partial x^2} + \epsilon\frac{\partial^2 c}{\partial y^2}, \tag{2}$$

$$(1 + \kappa')\frac{\partial c_m}{\partial t} + \left(\frac{\partial\psi}{\partial y} - \kappa'\right)\frac{\partial c_m}{\partial x} - \frac{\partial\psi}{\partial x}\frac{\partial c_m}{\partial y} \;\; = \;\; \delta\left[\frac{\partial^2 c_m}{\partial x^2} + \epsilon_m\frac{\partial^2 c_m}{\partial y^2}\right]. \tag{3}$$

where $\epsilon = D_y/D_x$, $\epsilon_m = D_{m_y}/D_{m_x}$, $\delta = D_{m_x}/D_x$. Here D_x, D_y and D_{m_x}, D_{m_y} are the axial and transverse dispersion coefficients of the sample solvent and solute, respectively. κ' is the retention parameter defined as $\kappa' = (1 - \epsilon_{tot})K/\epsilon_{tot}$, ϵ_{tot} is the total porosity and K is the adsorption-desorption equilibrium constant.

A pseudo-spectral method [2] is used to solve equations (1-3) numerically. The boundary conditions are periodic in both axial and transverse directions. The width corresponds to a Peclet number $Pe = UL_y/D_x$ while $L = UL_x/D_x$ is the dimensionless length of the two-dimensional domain. The initial conditions for both solvent concentration and mobile phase concentration of analyte correspond to a convectionless fluid embedding a rectangular sample of concentration $c = 1, c_m = 1$ of size $Pe \times l$ in a $c = 0, c_m = 0$ background, l being the dimensionless length of the sample. A random noise is added to the initial condition corresponding to two back to back step functions between $c = 0, c = 1$ and $c_m = 0, c_m = 1$ to trigger the fingering instability on a pertinent computing time [2].

3   Results

We are interested to investigate the disengagements of the retained solute zone with respect to the sample solvent zone. Density plots of concentration fields of c_m for different values of the retention parameters κ' are plotted at successive times in Fig.1. VF at the rear interface occurs because the displacing fluid has a lower viscosity than the sample solvent $(R > 0)$. If $\kappa' = 0$, the VF pattern of the unretained solute is the same as that of the solvent (Fig.1a). In the presence of adsorption the retained analyte develops fingering of its distribution zone at either the rear or frontal interface or both when in contact with the unstable interfaces of the sample solvent zone (Figs.1b-c). This retained analyte zone disengages rapidly from the sample solvent zone for large values of κ' giving then bearely no distortion at both interfaces (see Fig.1d). However for low viscous sample solvent case $(R < 0)$, if $\kappa' = 0$, VF occurs at the frontal interface (Fig.1e). The disengagement of the analyte zone from the fingering sample solvent zone with unperturbed interfaces occur for lower retention parameter κ' than the case of corresponding positive log-mobility ratio $R = 2$ (see Fig. 1f-h). These results can be explained quantitatively through transverse average profiles of c_m defined by $\bar{c}_m(x, t) = \frac{1}{Pe}\int_0^{Pe} c_m(x, y, t)dy$ and its variance due to the effects of VF, σ_f, defined in [1, 2, 3]. It is observed that the larger κ', the quicker the separation of both analyte and sample solvent plugs. The separation are much more quicker for $R < 0$ than $R > 0$. This model explains clearly the effects of sample solvent on the peak shape of analytes eluted in LC experiment as discussed in [6].
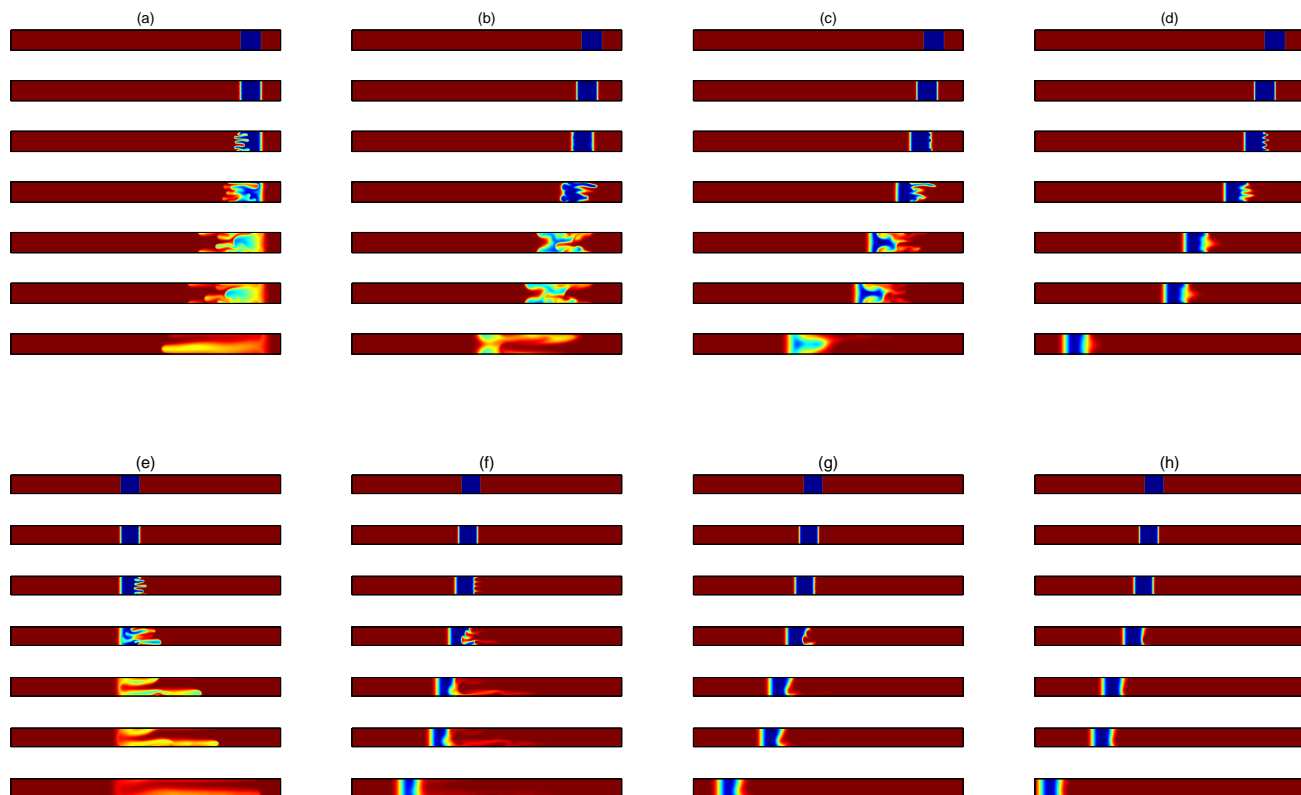
Figure 1: Density plots of the mobile phase concentration of the solute c_m at successive times in a frame moving at the velocity of eluent; First row for $R = 2$: (a) $\kappa' = 0$ (VF analogous to that of the sample solvent), (b) $\kappa' = 0.3$, (c) $\kappa' = 0.5$, (d) $\kappa' = 1$ and second row for $R = -2$: (e) $\kappa' = 0$, (f) $\kappa' = 0.2$, (g) $\kappa' = 0.3$, (h) $\kappa' = 0.4$ From top to bottom : $t = 0, 500, 1000, 2000, 4000, 5000, 10000$.

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