Miscible Viscous Fingering: Application in Chromatographic Columns and Aquifers

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Abstract: Miscible fluid flows occur in a wide range of industrial and biological processes in which viscous fingering is observed. Here our main focus is to model miscible viscous fingering (VF) using COMSOL Multiphysics. We study the effect of the positive and negative log-mobility ratio on the fingering phenomenon that appears in chromatographic columns and aquifers. Study has been conducted in 2D Eulerian frame for different injection speed and different log-mobility ratio. The dependence of mixing length and number of the fingers on various governing parameters in applications to chromatography and aquifers are presented. We also tried to examine the performance of our model by computing execution times for different meshes.

Keywords: viscous fingering, chromatographic columns, Hele-Shaw cell.

1. Introduction

When a less viscous fluid displaces a more viscous one in a porous medium or Hele-Shaw cell, the interface between the two miscible fluids does not remain flat and deforms into fingers growing in time [1]. It occurs due to the faster movement of less viscous fluid than the more viscous one, for a given pressure gradient. Fingering affects in aquifers, in packed bed reactors, and detrimental to chromatographic separation, groundwater contamination and many other systems. In these cases the two fluids under consideration do not extend over semi-infinite regions. Rather the displaced fluid is extended over a finite region and bounded with two semi-infinite displacing fluid region. This situation is drastically different form the situation when one fluid is displaced by another fluid both extended over semi-infinite domains, which appears in petroleum recovery from underground reservoir.

Viscous fingering is also of much concern in the dispersion of finite polluted viscous zones inside aquifers and in chromatography columns [2-5]. Liquid chromatography process separates the chemical components of a given sample by passing it through a porous medium. Displacement of the sample by a carrying fluid the eluent of different viscosity may lead to viscous fingering of either the front or the rear interface of the sample slice, leading to deformation of the initial planar interface. In these two applications, the viscous sample is generally of finite length. Viscous fingering occurs then at the interface where the less viscous fluid displaces the more viscous one, the other interface being stable. Numerical studies of the influence of fingering on dispersion of finite viscous samples using COMSOL Multiphysics has allowed the understanding of the fingering dynamics of such localized viscous zones where the instability is then only a transient phenomenon contributing to widening of the peak and comparison with other classical semi-infinite domain model [6] has been obtained.

2. Problem formulation

A fluid of viscosity μ_{01}, injected at a uniform velocity $u = U_0$ from the left of the domain, displaces another fluid of viscosity μ_{02} in a homogeneous porous medium. Both the fluids are considered to be incompressible and neutrally buoyant to neglect the density driven fingering. The problem has been modeled by coupling evolution equation for sample solvent concentration with a fluid flow equation in two-dimensional porous media or Hele-Shaw cell and they are:

$$\nabla.\vec{u} = 0 \tag{1}$$

$$\nabla p = -\frac{\mu}{k}\vec{u} \tag{2}$$

$$\frac{\partial c}{\partial t} + \vec{u}.\nabla c = D\nabla^2 c \tag{3}$$

The first equation expresses the equation of continuity, which represents the conservation of mass for incompressible fluids. Eq. (2) corresponds to the momentum equation in the form of a Darcy's law for the flow in porous media. The transport of solvent concentration is characterized by the convection-diffusion equation (Eq. (3)). For the flow in porous media $\vec{u}$ represents the Darcy-velocity, k is the permeability constant of the medium, p dynamic pressure and μ the kinematic viscosity of the fluid. For fluid motion in Hele-Shaw cell the above formulation is valid if the small gap d between the two parallel plates is such that the permeability of the medium is $k = d^2/12$.

In our model the viscosity of the fluids depends on the concentration of the solvent. The kinematic viscosity has been considered as an exponential function of the concentration [1]:

$$\mu(c) = \mu_{01} e^{Rc} \qquad (4)$$

where R is the log-mobility ratio given by

$$R = \ln(\mu_{02}/\mu_{01}) \qquad (5)$$

μ_{10} and μ_{02} are the kinematic viscosities of the displacing and displaced fluids, respectively.

3. Model in COMSOL Multiphysics

Here, the classical model of miscible viscous fingering (VF) has been investigated, using the COMSOL Multiphysics CFD modules. The implementation has been done using two-phase Darcy's law model, which couples the steady Darcy flow equation with the time dependent convection-diffusion equation for the concentration. This model considers two phases of fluids having different saturations s_1 and s_2. The concentration of the fluid c is the product of the saturation s_1 and the density ρ_1. With the assumptions of constant values of density $\rho = \rho_1 = \rho_2$, porosity ε_p, permeability κ, relative permeability $\kappa_{r1} = \kappa_{r2} = 1$, viscosity $\mu = \mu_1 = \mu_2$, in the model equations of two-phase Darcy's law (tpdl) of COMSOL Multiphysics reduce to the governing equations (1)-(3). The dependence of the viscosity on the concentration is maintained in this model through the saturation s_1, can be given as

$$\mu = \mu_{01} e^{R\, s_1} \qquad (6)$$

In this model a rectangular domain of length L_x and width L_y is considered. A fluid of viscosity μ_{01} is entering the domain from the left of it with a uniform velocity U_0, which displaces another fluid of viscosity μ_{02}.

The above-mentioned situation is implemented as follows: The inlet boundary condition at $x = 0$ is specified as the Dirichlet conditions for the velocity as well as the saturation of the displacing fluid, which mentions the injection speed of the displacing fluid $u = U_0$ and saturation $s_1 = 0$. At the outlet boundary $x = L_x$ the concentration of the displacing fluid is specified as Neumann condition $\partial c / \partial x = 0$, whereas the pressure as Dirichlet condition $p = 0$. Neumann type boundary conditions are specified at the boundaries $y = 0$ & $y = L_y$ for the velocity, i.e. the fluid does not leak out of the domain through these boundaries.

As initial condition a linear velocity profile has been chosen which satisfy the boundary conditions. The saturation of the displacing fluid is taken to be

$$s_1(t=0) = \begin{cases} 0, & x < L_x/8 \\ 0.5(1+\zeta f(x,y)), & x = L_x/8 \\ 1, & L_x/8 < x < L_x/8 + W \\ 0.5(1+\zeta f(x,y)), & x = L_x/8 + W \\ 0, & x > L_x/8 + W \end{cases}$$

Here the function $f(x,y)$ represents a two dimensional random function having mean 0.5 and ranging from 0 to 1. This describes a disturbance to the initial condition to trigger the instability. ζ denotes the amplitude of the disturbance. The initial state of the saturation of the displacing fluid s_1 is implemented using a combination of three analytic functions and a random function. These settings have been used for our simulation with their values listed in the Table 1. To obtain the fingering instability the initial disturbances are given at the interface of both miscible fluids only, unlike Holzbecher [6], in which a similar model of VF has been obtained using the initial disturbances over the whole region of less viscous displacing fluid.

The model equations are discretized according to the finite element method discretization. The coupled system is a time dependent problem as the concentration of the fluid goes through temporal evolution. The resultant non-linear equations are solved using the non-linear solver MUMPS while for the temporal evolution we use the backward Euler method. The numerical solution is obtained in a Eulerian system.

Table 1: List of parameters

Parameters	**Symbols**	**Value & Unit**
Length of the domain	L_x	0.08 mm 0.128 mm
Width of the domain	L_y	0.02 mm 0.032 mm
Amplitude of the disturbance	ζ	0.01
Log-mobility ratio	R	-2, 2, 3
Injection speed	U_0	1 mm/s 0.5 mm/s
Viscosity of the displacing fluid	μ_1	10^{-3} Pa-s
Aspect ratio	A	4
Length of the finite sample	$W = L_x/8$	0.01 mm 0.016 mm

Initially we did a test run of our simulation with three different types of meshes over a rectangular domain of size 0.08×0.02 mm^2. The injection speed for these test runs is $U_0 = 1$ mm/s, the log-mobility ratio is taken to be $R = 2$ and the length of the finite sample is chosen $W = L_x/8 = 0.02$ mm. The number of elements, degrees of freedom for which the problem is solved for and the simulation taken time for those three meshes is listed in the Table 2. It has been observed that second meshing (extra fine) gives quite good result and takes moderate computational time. So, “extra fine” meshing has been used for further simulations.

Table 2: Comparison of three different meshes

No. of elements	**Degrees of freedom**	**Computation time (s)**
12684	32249	250
63930	160982	1475
383814	961778	6111

4. Results and Discussion

Here we will discuss various properties of VF at the frontal (R<0) and rear interface (R>0) of a finite sample for the same viscosity ratio between the sample and the displacing fluid. Fingering dynamics of such finite samples have been shown on Fig. 1 and Fig. 2 for R = 2, -2 respectively, which give a visualization of the difference between the two cases. The evolution of the dynamics is shown in a stationary frame for different time. Rear interface of the sample becomes unstable due to unfavorable viscosity contrast in the case of the positive log-mobility ratio (R>0). The frontal interface remains stable due to the favorable viscosity gradient. On the other hand, opposite phenomenon is observed for the case R<0, where the frontal interface becomes unstable whereas the rear interface remains stable. Extensive qualitative numerical studies have been done for such dynamics with positive and negative log-mobility [2, 3]. The numerical results for both values of R are compared with the simulation results obtained by Mishra et al. [3], using a Fourier-spectral method. As anticipated the similar VF pattern at the early times until t = 20 seconds is seen out of the same noise and is seen through a visual inspection of the Fig. 1 and Fig. 2. Our simulation domain in these cases is a rectangle of size 0.08×0.02 mm^2. Fluid is injected with speed 1 mm/s.

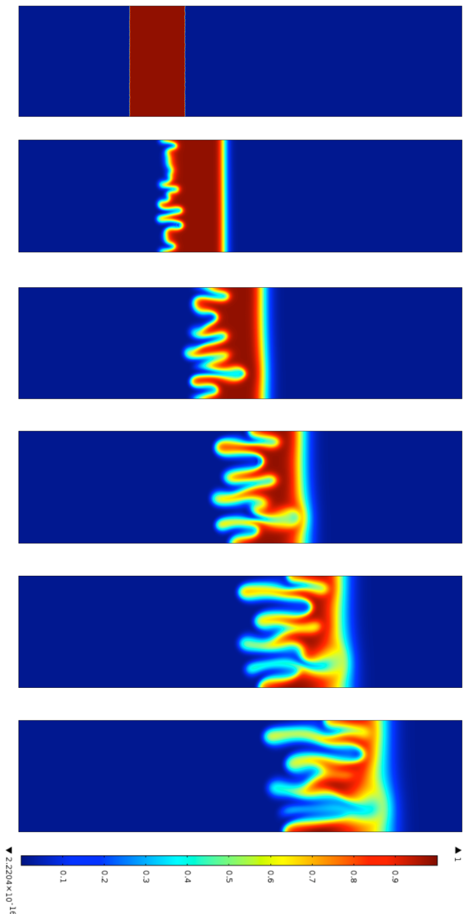

Figure 1. Surface plots of saturation S_1 at successive times with U_0 =1 × 10^{-3} *m/s, R*=2. From top to bottom *t*=0, 5, 10, 15, 20, 25 seconds.

The forward finger moves faster than the backward finger similar to the observation of spectral methods [3]. Mixing length becomes larger in *R*=-2 than the case of *R*=2.

Fig. 3 depicts the dynamics when the displacing fluid is injected with a speed U_0 = 0.5 mm/s. It is to be noticed that the unstable rear interface becomes less severe as the injection speed reduces here. The forward fingers [3] do not touch the frontal interface even at time t = 40 seconds, while it reaches at time t = 15 seconds with U_0 = 1 mm/s (see Fig. 1). As the displacing fluid invades slowly the onset time of fingering delayed than the larger U_0 = 1 mm/s and mixing becomes slower, which has also been observed in a recent experimental paper [4].

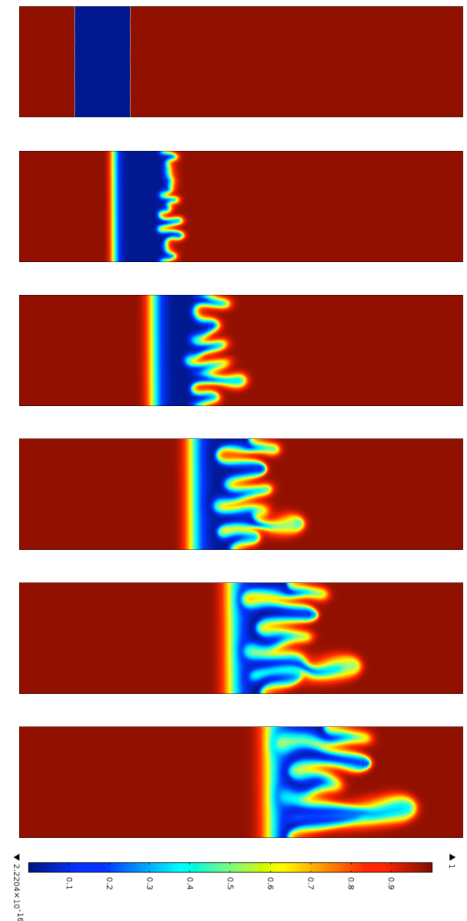

Figure 2. Surface plots of saturation S_1 at successive times with U_0 =1 × 10^{-3} *m/s*, *R*= -2. From top to bottom *t*=0, 5, 10, 15, 20, 25 seconds.

Finally on Fig. 4 we see the fingering dynamics at a higher log-mobility ratio (R = 3). We observe that the fingers grow faster and the forward finger reaches the stable interface earlier compared to the case with R = 2 (Fig. 3). This happens due to the larger viscosity gradient between both miscible fluids, similar to the results in [2-5].

5. Conclusions

We obtained the miscible VF patterns of a finite sample using COMSOL Multiphysics, which can be observed in chromatographic column and aquifers. The fingering instability in such cases is a transient phenomenon and broadens the length of the sample peaks. Fingering instability increases with increasing log-mobility ratio R, and the injection velocity U_0. The classical VF phenomenon like, merging and tip splitting are observed through this COMSOL Multiphysics simulation model. The results are very much comparable to the existing experiments [4] as well as other numerical simulations using spectral codes [2,3,5]. All these simulation performed here using two-phase Darcy's law (tpdl) of COMSOL Multiphysics 4.3a, are reproducible. In future, more detail simulations in this kind of pattern formations will be performed with different parameters of physical interest.

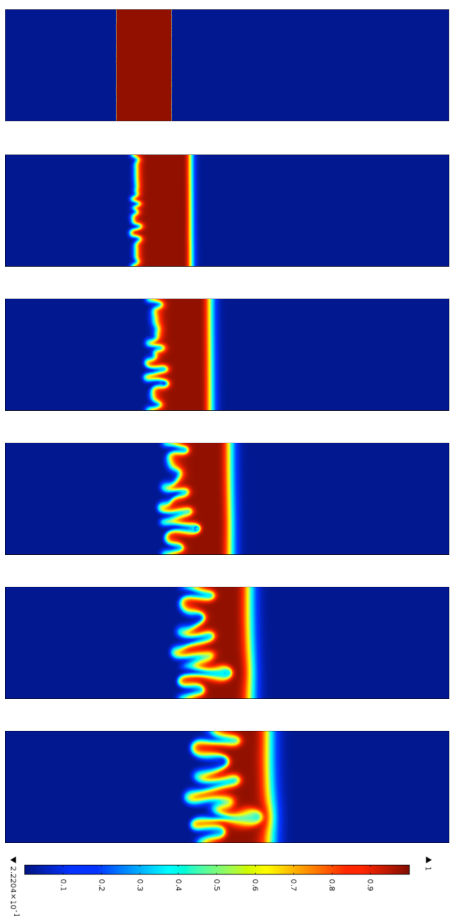

Figure 3. Surface plots of saturation s_1 at successive times with U_0 =5 × 10^{-4} *m/s*, R=2. From top to bottom t=0, 10, 20, 30, 40, 50 seconds.

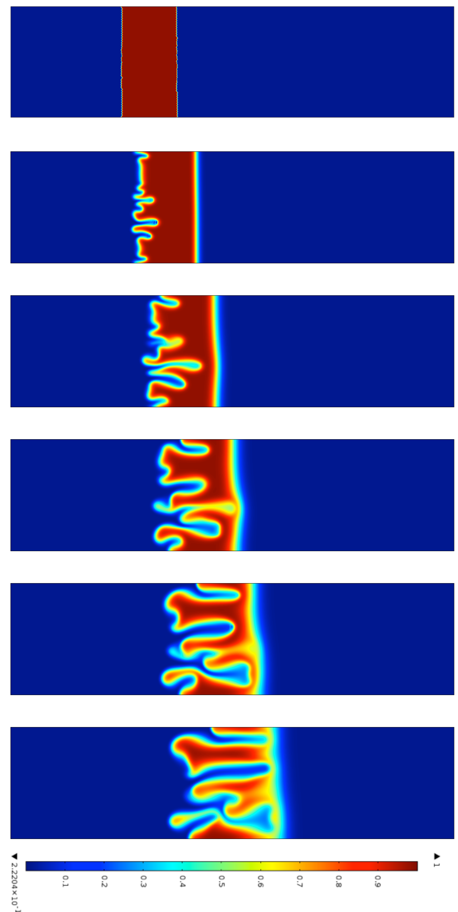

Figure 4. Surface plots of saturation s_1 at successive times with U_0 =5 × 10^{-4} *m/s, R*=3. From top to bottom t=0, 10, 20, 30, 40, 50 seconds.

6. References

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7. Acknowledgement

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