Influence of Double Diffusive Effects on Miscible Viscous Fingering

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Miscible viscous fingering (VF) occurs classically in porous media when a fluid of a given viscosity displaces another miscible more viscous fluid leading to a deformation of the corresponding interface into fingerlike patterns [1]. This paradigmatic pattern-forming instability has been the focus of numerous works devoted to study petroleum recovery [1], polymer processing [2], pollution spreading in soils [3] or efficiency of engineering separation techniques [3,4] to name a few. Except for some peculi...
The same liquid solvent where the scalars are present with a value $S_1$ and $F_1$, giving a viscosity $\mu_1$ is injected uniformly at an average velocity $U$ in the longitudinal direction $x$. Following earlier studies [21], the viscosity is assumed to depend exponentially on the values $S$ and $F$ as $\mu = \mu_1 \exp[R_s(S-S_1)/(S_2-S_1)] + R_f(F-F_1)/(F_2-F_1)],$ where $\mu_1$ is the viscosity of the injected solution and $R_s = (S_2-S_1)\text{dln}(\mu)/\text{dS}$ and $R_f = (F_2-F_1)\text{dln}(\mu)/\text{dF}$. If $R_s > 0$, then for $R_f = 0$, we have $\mu_1 < \mu_2$ i.e. a less viscous solution of $S$ displaces a more viscous one which is an unstable situation. If $S$ stands for temperature, this would be the case of a hot fluid displacing a cold fluid for instance while if $S$ stands for a viscosity increasing solute, this would correspond to a less concentrated solution of $S$ displacing a more concentrated one. Similarly, $R_f > 0$ for $R_s = 0$ leads to instability with regard to the $F$ component. When both $S$ and $F$ related effects are coupled and $\delta = 1$, the system is classically VF unstable when $\mu_1 < \mu_2$, i.e., $R_s + R_f > 0$. We seek here to obtain destabilization conditions for $R_s + R_f < 0$ when $\delta > 1$.

The evolution equations are Darcy’s law and transport equations for $S$ and $F$ in a coordinate system moving at the injection speed $U$. Considering the characteristic length $L_c = D_s/U$ and time $t_c = D_s/U^2$, the scalar values $S$ and $F$, pressure, velocity and viscosity are nondimensionalized as $\bar{S} = (S-S_1)/(S_2-S_1)$, $\tilde{F} = (F-F_1)/(F_2-F_1)$, $\tilde{\mu} = K(p-p_0)/\mu_1D_s$, $\tilde{\mu}_f = u_0/U$ and $\mu = \tilde{\mu}/\mu_1$, respectively, where $p_0$ is an arbitrary reference pressure. $K$ is the permeability and $\hat{\imath}$ is the unit vector along $x$. Dropping hats for convenience, the dimensionless equations with $\nabla \cdot \mathbf{u} = 0$ are

$$
\nabla p = -\mu(u + \hat{\imath}),
$$

$$
\mu = e^{R_s x + R_f f},
$$

$$
s_x + \bar{S} \cdot \nabla S = \nabla^2 S,
$$

$$
f_x + \bar{F} \cdot \nabla F = \delta \nabla^2 F.
$$

In the absence of any transverse instabilities $u = 0$, and the one-dimensional $s$, $f$, and viscosity base states are given analytically by $\bar{S} = \text{erfc}(-x/2\sqrt{\delta})/2$, $\tilde{F} = \text{erfc}(-x/2\sqrt{\delta})/2$, and $\tilde{\mu} = \exp[R_s \bar{S} + R_f \tilde{F}]$, where $x = 0$ is the initial interface position.

If $\delta = 1$, $\bar{S} = \tilde{F}$, and $R_s + R_f > 0$ corresponds to a monotonically increasing $\tilde{\mu}$, i.e., to the unstable situation, the reverse case $R_s + R_f < 0$ being stable. If $\delta > 1$, depending on the relative values of $R_s$ and $R_f$, six different $\tilde{\mu}$ profiles can be constructed (Fig. 1) [19,22] with differential diffusion effects triggering nonmonotonic viscosity profiles [23] in time in zones II, III, V, and VI. The boundaries of these various zones [13] can be found by noting that $\tilde{\mu}$, the derivative of $\tilde{\mu}$ with regard to $x$, changes sign at the two points satisfying $x^2 = \frac{4\delta}{\delta - 1} \ln(-\sqrt{\delta R_s}/R_f)$ which exist when $\sqrt{\delta R_s}/R_f < -1$. Further, the viscosity gradient at $x = 0$ is locally positive if and only if $\sqrt{\delta R_s}/R_f > 0$. Thus, a monotonically decreasing viscosity profile occurs when $R_f < \min(0, -\sqrt{\delta R_s})$ which is region IV in Fig. 1. Although regions III and V are within the domain $R_s + R_f < 0$, they both have nonmonotonic viscosity profiles which could trigger VF instabilities locally in the zones where $\tilde{\mu}_s > 0$.

To understand how these nonmonotonic effects develop in time and more strikingly how instabilities can also be obtained in regions IV$_{c,d}$ where the viscosity decreases monotonically, a time-dependent LSA must be performed. At $t = 0$, the stability problem can be solved analytically to yield $R_s(1 - k/\sqrt{2 + \sigma}) + R_f(1 - k/\sqrt{2 + (\sigma/\delta)}) = 2\sigma/k$ where $\sigma$ is the instantaneous growth rate for a disturbance of wave number $k$. This reveals that the system is stable at $t_0 = 0$ in regions III, IV$_{a,b,c}$ in Fig. 1 which is defined by $R_f < \min(-R_s - \delta R_s)$ while it is right away unstable in IV$_d$ and V. For $t_0 > 0$, the LSA is performed numerically using a QSSA [19,21]. As $t_0$ increases, the unstable regions expand across the $(R_s, R_f)$ plane until in the large time asymptotic limit the only stable regions are IV$_{a,b}$ whose combined regions are defined by $R_f > \min(0, -\delta^{3/2} R_s)$. This condition predicts that region IV$_c$ eventually becomes unstable in time while region IV$_d$ remains stable. This suggests that, beyond destabilization due to the build-up of nonmonotonic viscosity profiles in time, differential diffusion is able to destabilize a monotonic increasing viscosity profile as is the case in zones IV$_{c,d}$.

To test these predictions and analyze the influence of differential diffusion on nonlinear fingering, we performed numerical simulations of the full nonlinear problem. A
stream function–vorticity formulation of Eq. (1) is solved using a pseudospectral method [24] on a 2D finite domain of size $L_x \times L_y$.

Figure 2 shows various VF cases for $R_s + R_f > 0$ and various $\delta$ i.e. for a globally less viscous fluid displacing a more viscous one. Figure 2(a) depicts classical VF for $\delta = 1$ occurring in quadrant $I_a$ of Fig. 1 and characterized by tip splitting [1,24,25]. In this case, for a given constant $\lambda$, the resulting fingering pattern is the same for each point in the parameter space on the line $R_s + R_f = \lambda$. If now $\delta > 1$ in zone $I_a$, differential diffusion leads to a mushroom structured head as seen at the tip of the fingers in Fig. 2(b). The tip splitting mechanism is not observed any longer as the steep viscosity gradient at their origin is faded away by the faster diffusion of $F$. Clear mushroom structures in both forward and reverse fingers are observed in region $I_b$ of Fig. 1, as shown in Figs. 2(c) and 2(d). For $R_f > 0$, an increase of $\delta$ at fixed $(R_s, R_f)$ amplifies the extrema of the nonmonotonic change of $\mu$, transversely across the head of a finger [Figs. 2(e) and 2(f)] favoring the mushroom shape.

The most striking effect of differential diffusion is however to be able to destabilize the situation of a more viscous fluid pushing a less viscous one in the zone $R_s + R_f < 0$ (Figs. 3 and 4). Let us first examine the case when the contribution of the slow and fast diffusing scalars is, respectively, destabilizing and stabilizing, i.e., when $R_s > 0$ while $R_f < 0$. This corresponds for instance to cold water invading a hot enough viscous glycerol solution such that the global viscosity decreases along $x$. In region V of Fig. 1, the overall viscosity gradient is stable, however, a locally unstable zone develops in time across the interface as the fast diffusing component moves quickly towards the more viscous zone. Fingers grow around the interface in the zone where locally $\bar{\mu}_s > 0$; however, they eventually fade away far from it when they encounter the two stable zones where $\bar{\mu}_s < 0$ (Fig. 3, left). Fingers elongate more along the flow than against it [25] leading to an asymmetry in the pattern with regard to the initial position of the interface. This kind of destabilization, occurring in region V of Fig. 1, and due to a differential diffusion induced nonmonotonic viscosity profile when the slow component is destabilizing, will be referred to as DNS VF. However, such nonmonotonicity is not necessary for an instability as seen in Fig. 3 (right) where the associated base state viscosity profile is strictly monotonically decreasing (zone IV$_d$ of Fig. 1). Such a destabilization referred to as double diffusive VF (DD VF) is due to a pure diffusive effect like in buoyancy-driven double-diffusion where fingers develop around a stable density profile when the slow (fast) diffusing species is destabilizing (stabilizing) [20].

We next examine the case when the fast diffusing scalar is the destabilizing component, the slow one being stabilizing, i.e., when $R_f > 0$ while $R_s < 0$ as is the case when a hot solution of glycerol invades cold water for instance. If $R_s + R_f < 0$ as in region III of Fig. 1, the base state viscosity profile consists of two locally unstable regions (one with a viscosity maximum, the other one with a minimum) developing in time at a symmetric increasing distance from the interface. This kind of destabilization, illustrated in Fig. 4, will be referred to as DNF-VF as here we have a differential diffusion induced nonmonotonic viscosity profile with the fast component being destabilizing. The interface where $\bar{\mu}_s < 0$ remains stable while VF

![FIG. 2 (color online). Fingering dynamics of $s$ for $L_y = 4096$ and $(R_s, R_f, \delta) = (a) (1,1,1), (b) (1,1,10), (c) (1, 3, 4), (d) (1, 1, 10), (e) and (f) are the viscosity fields corresponding to (c) and (d), respectively. The frame number is $t/10^3$.](image)

![FIG. 3 (color online). Dynamics of $s$ for (left) DNS VF, $R_s = 3, R_f = -3.6, \delta = 10, L_y = 4096$, (right) DD VF obtained for $R_s = 1, R_f = -3.2, \delta = 10, L_y = 2048$.](image)
occurs in the two zones where $\mu_r > 0$ at equal distances on either side of the interface. Forward fingers extend further than the reverse fingers as they develop along the flow while reverse fingers have to fight against it [25]. Here again, fingers have a mushroom type structure as $R_f > 0$. Such an instability is reminiscent of diffusive layer convection induced by buoyancy effects when a solution of the fast diffusing component overlies a denser solution of the slow species [26,27].

Although the mechanisms of DD, DNS, and DNF-VF triggered by differential diffusion bear similarities with those in buoyancy problems [20], there is a fundamental difference between them: buoyancy-driven patterns develop symmetrically around the unstable interface while in VF, triggered by a flow, asymmetric effects exist because fingers develop more easily along the flow than against it [25]. Analogous asymmetric effects can also be triggered by chemical reactions [27].

We have here studied how differential diffusion of two components each contributing to the viscosity of a solvent can profoundly affect the stability and nonlinear dynamics of viscous fingering. In the situation of a less viscous fluid pushing a more viscous one (i.e., when $R_v + R_f > 0$), double diffusion modifies the nonlinear dynamics and favors mushroom shaped fingers when the fast diffusing species is destabilizing. The most striking effects appear however when destabilization by differential diffusion of a more viscous fluid pushing a less viscous one is obtained. This occurs either due to the build-up in time of nonmonotonic viscosity profiles like in DNS (region V) or DNF (zone III) VF where locally unstable zones develop leading to spatially constrained nonlinear fingering. More interestingly, this also happens on a strictly monotonically decreasing viscosity profile (zone IV) because of a DD VF mechanism leading to smooth fingers. These various instability scenarios can be obtained by identifying the slow and fast component as mass and heat typically. Another way to experimentally look for the new instability scenarios predicted here is to use two nonreacting chemical species both influencing the viscosity of the solution but having sufficiently different diffusion coefficients. The wealth of available long chain molecules of variable length and thus tunable molecular weight and diffusion coefficients offers naturally to the experimentalist a tool kit to test the various proposed VF instability scenarios.

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