Influence of a chemical reaction on viscous fingering: 
Effect of the injection flow rate

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Abstract

The hydrodynamic viscous fingering instability can be influenced by a simple viscosity changing chemical reaction of type $A + B \rightarrow C$, when a solution of reactant $A$ is injected into a solution of $B$ and a product $C$ of different viscosity is formed. We investigate here numerically such reactive viscous fingering in the case of a reaction decreasing the viscosity to define the optimal conditions on the chemical and hydrodynamic parameters for controlling fingering. In particular, we analyze the influence of the injection flow rate or equivalently of the Péclet number (Pe) of the problem on the efficiency of the chemical control of fingering. We show that the viscosity decreasing reaction has an increased stabilizing effect when Pe is decreased. On the contrary, fingering is more intense and the system more unstable when Pe is increased. The related reactive fingering patterns cover then respectively a smaller (larger) area than in the non-reactive equivalent. Depending on the value of the flow rate, a given chemical reaction may thus either enhance or suppress a fingering instability. This stabilization and destabilization at low and high Pe are shown to be related to the Pe-dependent characteristics of a minimum in the viscosity profile that develops around the miscible interface thanks to the effect of the chemical reaction.

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A hydrodynamic viscous fingering (VF) instability can deform the interface between two different fluids when a high mobility fluid of lower viscosity displaces a more viscous and hence less mobile one in a porous medium [1–5]. In numerous industrial and environmental problems such as enhanced oil recovery, CO$_2$ sequestration, combustion, hydrology, soil remediation, etc. [6–10], this fingering instability can interplay with chemical reactions. In the past few decades, viscous fingering has been analyzed in reactive systems on both miscible and immiscible interfaces [11–31]. If the reaction does not modify the viscosity in-situ, the chemical species are passively advected by the flow and the fingering properties of the interface remain similar to those of the nonreactive system [11–15]. The flow in the fingering patterns can on the other hand change the spatio-temporal distribution of the reactants and influence the yield of the reaction. Active influence of chemistry on fingering can be obtained as soon as the chemical reaction taking place around the interface between the two fluids modifies their physical properties and, in particular, their viscosity [32]. The reaction then influences the stability as well as the spatio-temporal dynamics of the flow. In turn, the hydrodynamic flow affects mixing and thus the amount and spatial distribution of chemical species and a highly nonlinear feedback is established between chemistry and hydrodynamics.

For cases where reactions actively change the viscosity in-situ, numerical simulations have first shown on the basis of a bistable chemical reaction scheme that the properties of miscible VF are modified when the reaction changes the viscosity across the reactive miscible interface [16, 17]. The bistable nature of chemical kinetics is then responsible for a new phenomenon of droplet formation isolating regions of high or low viscosity within connected domains of the other steady state. In other studies, the active influence of $A + B \rightarrow C$ types of chemical reaction on miscible viscous fingering has been studied both experimentally [19, 20, 22, 27–29] and theoretically [21, 23–26, 28, 30]. Podgorski et al. [19] have in particular studied experimentally chemically-driven fingering at the miscible reactive interface between two aqueous solutions of same viscosity when a reaction between a cationic surfactant and an organic salt produces an elastic more viscous worm-like micellar fluid. Various fingering regimes have been identified depending on concentrations, fluid characteristics and injection flow rate (or equivalently Péclet number, defined as the ratio
of the convective to diffusive transport rates).

In some experiments by Nagatsu et al., a less-viscous acidic or basic aqueous solution was injected into a more-viscous polymeric solution, the viscosity of which depends on pH \cite{20, 22, 27}. It is observed that, when the viscosity is increased (decreased) by the reaction, fingers are widened (narrowed), which is mainly due to suppressed (enhanced) shielding effects. Interestingly, opposite results have been observed at moderate reaction rates for systems with a viscosity decrease \cite{22} and increase \cite{27}. In the case where the non reactive displacement is stable (more viscous solution displacing a less viscous once), it has even been shown experimentally that the reaction is able to trigger VF \cite{28}. Depending whether the reaction increases or decreases viscosity, a different fingering pattern is then obtained.

The experimental study of Nagatsu et al. \cite{20} showed that at ‘large’ injection rate, or equivalently high Péclet number (Pe), an instantaneous chemical reaction can have opposite effects on miscible VF when a less viscous (acidic or basic) solution is injected radially into a more viscous (e.g. polymeric solution) one in a Hele-Shaw cell depending whether the reaction locally increases or decreases the viscosity. In the viscosity increase case, the VF pattern is “denser” in the sense that it covers a more compact area in the Hele-Shaw cell than the non-reactive pattern. On the contrary, a VF pattern covering a smaller area (also qualified as “less dense pattern”) was reported in the viscosity decrease reactive case. Recently, new experiments have been carried out focusing on the influence of the injection rate on viscosity increasing and decreasing reactive systems \cite{29, 33}. Interestingly, it was found that, at lower Pe, the trends are opposite than at high Pe i.e. for viscosity decreasing reactions, the system can be stabilized at low injection flow rates. These experiments \cite{20, 29, 33} thus clearly show that, in the presence of a viscosity decreasing reaction, the reactive VF patterns can be controlled by varying the Péclet number. Moreover, when the reaction induced viscosity decrease is large enough, a suppression of the VF instability can be obtained at small Pe. In numerical studies, the explicit influence of the injection rate on reactive VF has however not been addressed explicitly.

In this context, our objective is here to analyze numerically the influence on the VF instability of changes in the injection flow rate i.e. changes in the Pe number of the problem when a simple $A + B \rightarrow C$ chemical reaction decreases the viscosity \textit{in situ}. To this end, we integrate numerically the reaction-diffusion-convection (RDC) equations of reactive VF in porous media and analyze the properties of the fingering patterns for different values of
We show that a viscosity-decreasing reaction enhances stabilization or destabilization of the interface at respectively low and high Pe, with regard to the non-reactive system. This is related to the possibility at low Pe for chemistry to build up a minimum in the viscosity profile that blocks the further progression of fingering and stabilizes the system. On the contrary, at high Pe, chemistry does not have time to act to decrease the viscosity and the classical enhanced destabilization when the flow rate is increased is then observed.

These results highlights the optimum conditions on flow conditions to obtain stabilization by reactions of VF. This is of practical importance as it paves the way to a possible chemical control of fingering instabilities appearing in many practical situations ranging from geophysical to environmental problems.

This paper is organized as follows. The problem description and the related RDC model are given in Sec. II. In Sec. II B, the numerical method used to integrate the model is discussed. The characteristics of VF patterns and in particular the influence of the Péclet number are studied in Sec. III. The non-reactive and reactive cases are given in III A and III B, respectively. A quantitative analysis and parametric study are carried out in Secs. III C and IV. At the end, conclusions and outlook are given in Sec. V.

II. PROBLEM DESCRIPTION AND GOVERNING EQUATIONS

Consider a homogeneous two-dimensional porous medium or horizontal thin Hele-Shaw cell of length $L_x$ and width $L_y$ with constant permeability $\kappa$ in which a miscible solution of reactant $A$ with viscosity $\mu_A$ is injected from left to right into a solution of reactant $B$ with viscosity $\mu_B$ at a constant speed $U$ along the $x$-direction (Fig.1). We assume that the initial concentrations of $A$ and $B$ are both equal to $a_0$. The initial position of the miscible interface is $x_0$. Upon contact between the two solutions, a simple $A + B \rightarrow C$ chemical reaction takes place in the miscible interface zone where $A$ and $B$ meet by diffusion, react, and yield the product $C$ of viscosity $\mu_C$. The objective is to analyze numerically how the dynamic decrease of viscosity driven by the reaction can influence the VF instability and in particular what is the influence of the injection speed $U$ on this effect.

To analyze the problem, the system is considered as incompressible and neutrally buoyant. The dynamics is modeled using Darcy’s law for the velocity field along with three reaction-
FIG. 1. Sketch of a two-dimensional porous medium of length $L_x$ and width $L_y$ with permeability $\kappa$ in which a solution of reactant $A$ with viscosity $\mu_A$ is displacing a solution of reactant $B$ of viscosity $\mu_B$ from left to right at a constant speed $U$. Here $x_0$ and $a_0$ are the initial contact position and initial concentration of reactants, respectively.

The diffusion-convection (RDC) equations for the concentrations:

\[ \nabla \cdot \mathbf{u} = 0, \]
\[ \nabla p = -\frac{\mu(a, b, c)}{\kappa}\mathbf{u}, \]
\[ \frac{\partial a}{\partial t} + \mathbf{u} \cdot \nabla a = D_A \nabla^2 a - ka b, \]
\[ \frac{\partial b}{\partial t} + \mathbf{u} \cdot \nabla b = D_B \nabla^2 b - ka b, \]
\[ \frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = D_C \nabla^2 c + ka b, \]

where $a$, $b$, and $c$ denote the concentrations of the reactants $A$ and $B$ and of the product $C$, respectively, $k$ is the kinetic constant, $p$ is the pressure, $D_A$, $D_B$ and $D_C$ are the diffusivities of the reactants $A$ and $B$ and the product $C$, respectively, $\mathbf{u} = (u, v)$ is the two-dimensional flow velocity and $\kappa$ is the constant permeability. The viscosities of the solution when only one species is present in concentration $a_0$ are defined as $\mu_A$, $\mu_B$ and $\mu_C$, respectively in the presence of the reactants $A$, $B$ or of the product $C$. Following previous theoretical work on viscous fingering [3, 16, 17, 21, 23–26, 29, 30, 34], we assume the viscosity as an exponential function of the concentrations of $A$, $B$ and $C$ as

\[ \mu(a, b, c) = \mu_A e^{[R_b b + R_c c]/a_0}, \]
where $R_b$ and $R_c$ are the log-mobility ratios defined as
\[
R_b = \ln \left( \frac{\mu_B}{\mu_A} \right) \quad \text{and} \quad R_c = \ln \left( \frac{\mu_C}{\mu_A} \right),
\] (7)

For the non-reactive VF case or the equivalent specific reactive case when the product $C$ has the same viscosity as one of the reactant (i.e. $R_b = R_c$), the system is unstable when the lower viscosity solution of $A$ displaces the more viscous solution of $B$ i.e. when $\mu_A < \mu_B$ or $R_b > 0$. Let us analyze how this stability is changed when both $\mu_C$ and the injection speed $U$ are varied.

A. Non-dimensional Equations

To specifically let the injection speed appear in the dimensionless problem under the form of a Péclet number, the reference scales for length, velocity, time, concentration, viscosity, diffusivity and pressure are taken as $L_y, U, L_y/U, a_0, \mu_A, D_C$ and $\mu_A U L_y/\kappa$, respectively. For simplicity, equations are written in a reference frame moving with speed $U$ by transforming variables as $x \rightarrow x - U t e_x$ and $u \rightarrow u - U e_x$ with $e_x$ being the unit vector along $x$ direction. The dimensionless form of (1)–(6) can then be written as

\[
\nabla \cdot u = 0, \quad \nabla p = -\mu(a, b, c)(u + e_x), \quad \frac{\partial a}{\partial t} + u \cdot \nabla a = \delta_a \text{Pe}^{-1} \nabla^2 a - D_a a b, \quad \frac{\partial b}{\partial t} + u \cdot \nabla b = \delta_b \text{Pe}^{-1} \nabla^2 b - D_a a b, \quad \frac{\partial c}{\partial t} + u \cdot \nabla c = \text{Pe}^{-1} \nabla^2 c + D_a a b,
\] (8–12)

\[
\mu(a, b, c) = e^{\left(R_b b + R_c c\right)},
\] (13)

where $D_a = k a_0 L_y/U = \tau_h/\tau_c$ is the dimensionless Damköhler number defined as the ratio of the hydrodynamic time scale $\tau_h = L_y/U$ to the chemical time scale $\tau_c = 1/ka_0$. The Péclet number $\text{Pe} = U L_y/D_c = \tau_h/\tau_D$ is the ratio of the convective time $\tau_h$ to the diffusive time $\tau_D = D_c/U^2$ while $\delta_a = D_A/D_C$ and $\delta_b = D_B/D_C$ are the diffusion coefficient ratios. Taking the curl of the momentum equation and defining the stream function $\psi(x, y)$ as $u = \partial \psi/\partial y$
and \( v = -\partial \psi / \partial x \), we get

\[
\nabla^2 \psi = R_b (\psi_x b_x + \psi_y b_y + b_y) + R_c (\psi_x c_x + \psi_y c_y + c_y),
\]

\[
a_t + a_x \psi_y - a_y \psi_x = \delta_a Pe^{-1} \nabla^2 a - D_a a b,
\]

\[
b_t + b_x \psi_y - b_y \psi_x = \delta_b Pe^{-1} \nabla^2 b - D_a a b,
\]

\[
c_t + c_x \psi_y - c_y \psi_x = Pe^{-1} \nabla^2 c + D_a a b,
\]

where the subscripts \( x \) and \( t \) represent the respective derivatives. The last term in (15)–(17) corresponds to the reaction rate \( \mathcal{R} \):

\[
\mathcal{R}(x, y, t) = D_a a(x, y, t) b(x, y, t).
\]

Comparing the present RDC model (14)–(17) with those previously studied in the literature [5, 21, 23, 26, 35], we note that: (i) when \( D_a = 0 \) we recover the classical model for non-reactive viscous fingering similar to the one studied by Tan and Homsy [5, 35]; (ii) when \( D_a \neq 0, Pe = 1 \) and \( R_b = 0 \) we obtain the model of reactive VF for solutions of \( A \) and \( B \) of same viscosity as analyzed numerically by Gérard and De Wit [21]; (iii) when \( D_a \neq 0, Pe = \delta_a = \delta_c = 1 \) we get back to the reactive VF model with \( A, B \) and \( C \) of different viscosity but species diffusing all at the same rate as studied by Hejazi et al. [23] and Nagatsu and De Wit [26].

As the dynamics of the reactive zone is independent of boundary conditions as long as the unstable fingered front does not confront its periodic extension [5], we use periodic boundary conditions in both directions. The initial conditions for the stream function and product concentration \( c \) are taken as \( \psi(x, y) = 0 \) and \( c(x, y) = 0 \), for all \((x, y)\), respectively. For the initial concentrations of the reactant \( A \) and \( B \) solutions, we use a step front between \( A = 1, B = 0 \) on the left and \( B = 1, A = 0 \) on the right of \( x = x_0 \) with a random noise of amplitude of order \( 10^{-2} \) added in the front to trigger the instability. The dimensionless system size is \( \mathcal{A} \times 1 \), where \( \mathcal{A} = L_x/L_y \) is the aspect ratio. Equations (14)–(17) together with the initial and boundary conditions form an initial-boundary value problem with six dimensionless control parameters—namely, \( R_b, R_c, D_a, \delta_a, \delta_b \) and \( Pe \). To decrease the wide range of possibilities, we fix here \( \delta_a = \delta_b = 1 \) to focus on the effect of the reaction (variable \( Da \) and \( Rc \) for a given \( R_b \)) and flow speed (variable \( Pe \)) on the fingering instability.
B. Numerical Method

To solve (14)–(17), we use a pseudo-spectral numerical scheme based on the discrete Fourier transform library FFTW 3.3.4 [5, 16, 17, 21, 36]. In order to avoid any interaction between the unstable fingered front and its periodic extension, we choose a domain with a large aspect ratio. The physical and computational domain size \((L_x \times L_y)\) are \(32 \times 1\) and \(4098 \times 128\), respectively. The time step of numerical integration is chosen as \(dt = 10^{-4}\). To validate our code, we have successfully reproduced previous nonlinear simulation results of non-reactive [5, 35] and reactive [21, 23, 26] systems.

III. RESULTS

A. Non-reactive system

It is already known that, in absence of any reaction effect \((Da = 0 \text{ or } R_b = R_c \text{ [23, 26]})\), increasing the injection speed (i.e. increasing Pe in our dimensionless formulation of the problem) increases the destabilization of the interface by VF when \(R_b > 0 \text{ [4, 5, 35]})\). As a reference case, this observation is shown in Fig. 2 which illustrates the concentration of reactants \((A \text{ and } B)\) for \(Pe = 100\) and \(Pe = 1000\), respectively, at four different times. As Pe increases, fingering becomes more intense and the wavelength of the pattern decreases as the interface becomes more unstable. It is also observed that, at low Pe, the deformed interface tends to flatten as time evolves thanks to transverse diffusion.

Figure 3 compares the one dimensional profiles of the concentrations of \(A\) and \(B\), and the logarithm of viscosity \(\ln(\mu)\) at a fixed transverse location \(y = L_y/2\) for \(Pe = 100\) at time \(t = 30\) and \(Pe = 1000\) at \(t = 5\), respectively. As a reference, the white line \(y = L_y/2\) is shown in the corresponding two-dimensional map of \(\ln(\mu)\) on the top of the panels. While the concentrations and viscosity profiles at large Pe show bumps characteristic of the fingering instability, these profiles are quasi linear between the end-point values at small Pe indicating a more stable interface. This stabilizing effect at low Pe is in agreement with previous results [3, 5, 35].
FIG. 2. Equivalent non-reactive ($R_b = R_c = 2$) system: Concentrations of $A$ and $B$ for $Pe = 100$ [left] and 1000 [right] at four different times (from top to bottom). Concentration fields are scaled between zero (blue) and one (red). The viscosity (not shown) varies in a similar way as the concentration of $B$.

FIG. 3. Spatial profiles of concentrations of $A$ (dashed blue line), $B$ (dash-dotted red line), and of $\ln(\mu)$ (solid magenta line) along the injection direction at $y = L_y/2$ for $R_b = R_c = 2$ and (a) Pe = 100 at $t = 30$ or (b) Pe = 1000 at $t = 5$, see Fig. 2. The top figures represent the corresponding two-dimensional map of $\ln(\mu)$ through which the one dimensional sections are shown.

B. Reactive system

Let us now analyze the effect of $Pe$ on reactive VF when an $A + B \rightarrow C$ reaction produces the product $C$ of lower viscosity (negative value of $R_c$) such that the viscosity of the system develops in time a minimum around the reactive front.
FIG. 4. Reactive VF at $R_b = 2$, $R_c = -2$ and $D_a = 1$. The first and second columns represent the concentrations of $A$, $B$, $C$, viscosity in log-scale ($\ln(\mu)$) and $R$ for $Pe = 100$ and 1000 at various times, respectively. $A$ and $B$ are scaled between zero (blue) and one (red), $C$ is scaled between zero (blue) and 0.5 (red), and $\ln(\mu)$ and $R$ are shown in their absolute values.

Figure 4 shows the concentrations, $\ln(\mu)$, and reaction rate $R$ at $D_a = 1$, $R_b = 2$ and $R_c = -2$ for two values of Péclet numbers $Pe = 100$ (first column) and 1000 (second column). Two opposite behaviours are obtained at low and high $Pe$: at $Pe = 1000$, fingering is more intense than in the non reactive case with coarsening, and more repetitive shielding and tip splitting [26]. The fingered zone extends on a larger spatial extent than in the non reactive case (Fig.2) suggesting that the reaction has here a destabilizing effect. A comparison of the transverse averaged viscosity profile in the non reactive (Fig.3b) and reactive (Fig.5b) cases shows that, at $Pe = 1000$, the decrease in viscosity induced by the reaction leads to a sharper viscosity jump which can explain the increased destabilisation. As a consequence,
fingering extends both in the $A$- and $B$-rich regions with the reaction rate being localised at the fingered frontier between the two reactants. On the contrary, at $Pe = 100$ (Fig.4, first column), a minimum in viscosity develops in the course of time where the less viscous $C$ separates the two reactants $A$ and $B$ (Fig.5a). The reaction rate correspondingly decreases in time and remains strongly localised at a given location. The time scales are also longer as more time is needed to cover the same distance. Interestingly, fingering is weak and remains longer in the boundary zone where the less viscous $C$ displaces the more viscous $B$ then in the stable part of the non-monotonic profile where A pushes the less viscous $C$. This means that, in experiments where often a dye is used to visualize the fingering pattern, the instability would quickly become unnoticeable if the dye is diluted in the injected A reactant [29].

A comparison of the spatio-temporal distribution of $A$ in Fig.2 (non reactive) and 4 (reactive) leads thus to the conclusion that, at high $Pe$, reactive fingering is more intense with more ramified fingers that cover a larger area in the presence of reaction. On the contrary, at low $Pe$, fingering is stabilized by the reaction. The effect of the reaction decreasing the viscosity has thus an opposite effect on the flow at high and low $Pe$, as observed experimentally [20, 29].

![FIG. 5. Same as Fig. 3 but for the reactive case at $D_a = 1$, see Fig. 4.](image-url)
C. Quantitative analysis

In order to understand the opposite dynamics at low and high Pe, and to quantify the influence of varying Pe on reactive VF, we compute the one-dimensional transversely averaged profiles of given quantities, $\zeta(x, y, t)$ as

$$\langle \zeta(x, t) \rangle = \frac{1}{L_y} \int_0^{L_y} \zeta(x, y, t) \, dy,$$

where $\zeta$ can be, for instance, concentration, viscosity, etc. In absence of fingering ($R_b = R_c = 0$), these profiles are equivalent to the one-dimensional reaction diffusion profiles. For the simulations of Fig. 4, the temporal evolution of some of these transversely averaged profiles is shown in Fig. 6.

In the convective flow regime, the fingering pattern starts to develop around the reactive interface as soon as solutions $A$ and $B$ react and produce a less-viscous product $C$, see Figs. 6(a) and 6(b). As the system evolves in time, we see that increasing amounts of $A$ and $B$ are consumed and that the total production of the product $\langle c(x, t) \rangle$ increases. The corresponding reaction rate $\langle R(x, t) \rangle$, shown in Fig. 6(e), decreases in time when $A$ and $B$ are consumed and are progressively separated by $C$. Fig. 6(c) shows the development of viscosity as time evolves. At low Pe, a viscosity minimum develops in time at the back of the reaction front where the product concentration is maximum which can also be seen from Figs. 4(c) and 4(d). Owing to the viscosity minimum, the interface between $A$ and $C$ is stabilized, which can clearly be observed in Fig. 4(a) as the interface tends to flatten. On the contrary, the interface between $B$ and $C$ where the less viscous $C$ pushes the more viscous $B$ indicates the presence of VF. Nevertheless, transverse diffusion finally dominates VF, and the interface between $B$ and $C$ eventually stabilizes again [see Fig. 4(a–e)].

Let us now analyze quantitatively fingering patterns at larger Pe. We have noticed in Fig. 4(f–j) that reactive VF is destabilizing at high Pe in contrast to a stabilizing trend at low Pe. Figures 6(b,d,f) show that, at high Pe, when VF is present, the transversely averaged concentration profiles feature bumps indicating the presence of forward and backward fingering. In contrast to fingering at the back, forward fingering shows merging and tip-splitting, see Fig. 4(f–j). Similar to the concentration, the log-viscosity, Fig. 6(d), and reaction rate profiles, Fig. 6(f), show similar features. The center of mass of these profiles is shifted towards the right of the reaction front indicating the presence of more elongated
FIG. 6. Transversely averaged concentration (top row), viscosity (middle row), and reaction rate (bottom row) profiles corresponding to simulations of Figs. 4 for Pe = 100 (left column) and Pe = 1000 (right column). The dashed, dash-dotted and solid lines in panels (a,b) depict concentrations of $A$, $B$ and $C$, respectively. The black, red, green, blue and magenta colors in panels (a,c,e) correspond to $t = 0, 10, 20, 40$ and 50, respectively, while those in panels (b,d,f) correspond to $t = 0, 1, 3, 5$ and 10, respectively.

fingerling in the $B$-rich region. While, at low Pe, the viscosity minimum formed at the back (or left) of the reaction front gives rise to stabilization, it is completely absent at high Pe
causing VF to expand significantly around the reaction zone.

IV. PARAMETRIC STUDY

We have seen that fingering is stabilized at lower Pe when the viscosity decreases thanks to a chemical reaction. To gain more insight into this stabilization effect, a parametric study is next carried out at several low Pe values to understand the effect of varying the Damköhler number $D_a$ and the viscosity of the product by changing the log-mobility ratio $R_c$.

A. Effect of mobility ratio $R_c$ at $R_b > 0$

FIG. 7. From top to bottom in each column: concentrations of $A$, $B$ and $C$, ln($\mu$) and $\mathcal{R}$ for various time steps ($t = 10$, $20$, $30$ and $40$) at Pe = 100 with $R_c = -2$ (first column), $R_c = 0$ (second column) and $R_c = 2$ (equivalent of the non-reactive case, third column). Other parameter values are as in Fig. 4.

The effect of changing the log-mobility ratio $R_c$ is shown on Fig. 7. We consider the three values $R_c = -2$, $0$, $2$. We remind that, when $R_b = R_c$ (=2 here), the consumption of $B$ is balanced by the production of $C$, hence the dynamics of the reactive case is equivalent to that of the non-reactive system. When $0 < R_C < R_b$, the viscosity decreases by the reaction
but the viscosity profile remains monotonic in space. On the contrary, if \( \text{Re}_c < 0 \), a minimum in viscosity develops in time. For \( \text{Re}_b = 2 \), the cases \( \text{Re}_c = 2 \), \( \text{Re}_c = 0 \) and \( \text{Re}_c = -2 \) represent thus the (i) non-reactive VF, (ii) reactive VF with monotonic viscosity, and (iii) reactive VF with a viscosity minimum, respectively.

By comparing concentrations of \( A \), \( B \) and \( C \) for various time steps in these three cases, we see that, when \( \text{Re}_c < 0 \), the viscosity minimum has the following effects: (i) The interface between \( A \) and \( C \) stabilizes rapidly and the mixing of reactant \( A \) decreases as compared to the other two cases, (ii) as time evolves, the mixing region between \( C \) and \( B \) increases and stops fingering in time, displacing more \( B \) by the product \( C \). The reactive VF is stabilized at low Pe by the viscosity minimum compared to the reactive VF case with monotonic viscosity or the non-reactive VF.

![FIG. 8. (a) RD profiles for ln(\( \mu \)), and (b) temporal variation of the mixing length for A (main panel), B (inset), and C (inset) for \( \text{Re}_c = -2 \) (solid line), 0 (dashed line), and 2 (dash-dotted line). Other parameter values are as in Fig. 7.](image)

The origin of this stabilization can be explained through the long time asymptotic one-dimensional reaction diffusion (RD) profiles of ln(\( \mu \)), as shown in Fig. 8(a). If \( \text{Re}_c = -2 \), the reaction diffusion viscosity front moves in time from the higher viscosity region of \( B \) to the lower viscosity region of \( A \), see Fig. 6(a). Due to the presence of lower viscosity region containing \( C \), the profile of ln(\( \mu \)) develops a minimum in the \( A \)-rich region. While
the gradients \( \frac{d(\ln \mu)}{dx} \) are decreasing with \( R_c \) on the left of the reaction front \( [x - x_0 < 0] \), those on the right \( [x - x_0 > 0] \) are increasing. Owing to this, when \( R_c = -2 \), the miscible interface between \( A \) and \( C \) is more stable as is the case when a higher viscosity fluid displaces a lower viscosity one. As a consequence, the mixing length \( L_a \) decreases rapidly as time evolves and finally reaches a steady value which is the lowest among all cases, as shown in the main panel of Fig. 8(b).

In contrast to the interface between \( A \) and \( C \), the interface between \( C \) and \( B \) is more unstable when \( R_c = -2 \) because \( \frac{d(\ln \mu)}{dx} \) is then the steepest, see Fig. 8(a). This can also be noticed in the evolution of the mixing length of \( B \) and \( C \) in Fig. 8(b). The instability at the interface between \( B \) and \( C \) starts earlier and the mixing length \( L_b \) and \( L_c \) increase more in time as \( R_c \) decreases. As time evolves (far from the onset), due to transverse diffusion, \( L_b \) and \( L_c \) reach a steady value which increases with decreasing \( R_c \). The displacement of \( B \) is thus larger when \( R_c = -2 \) in comparison to \( R_c = 0 \) and 2 (non-reactive). From Figs. 4–8, we can thus conclude that, when \( R_c = -2 \), the front between \( A \) and \( C \) stabilizes, the mixing between \( B \) and \( C \) is increased and the displacement of \( B \) is larger.

**B. Effect of Péclet number \( Pe \)**

![FIG. 9. Same as Fig. 7 but for \( R_c = -2 \) and different values of \( Pe \): 100 (first column), 150 (second column) and 300 (third column).](image)
FIG. 10. (a) RD profiles of $\ln(\mu)$ for $Pe = 100$ (blue), 150 (red), 300 (black) and 1000 (magenta) where the solid and dashed lines represent reactive and non-reactive systems, respectively. (b) Temporal evolution of the mixing length for $A$ (main panel), $B$ (upper inset) and $C$ (lower inset) at $Pe = 100$ (blue solid line), 150 (red dashed line), 300 (black dash-dotted line) and 1000 (magenta dotted line). Other parameter values are as in Fig. 9.

In the previous section, we have seen that the onset time decreases i.e. the system is initially more unstable as $R_c$ decreases. We now fix $R_c = -2$ and analyze the effect on fingering of changing $Pe$ keeping it nevertheless at small values. Specifically, concentrations, $\ln(\mu)$ and the reaction rate are shown for $Pe : 100$ (first column), 150 (second column) and 300 (third column) in Fig. 9. We see that the system becomes more unstable when increasing $Pe$. This can be understood by inspecting the one-dimensional RD profiles shown in Fig. 10(a). The non-reactive displacement (dashed lines) is more unstable at higher $Pe$ because the gradient of viscosity $d(\ln\mu)/dx$ is correspondingly sharper. Similarly, viscosity gradients in the reactive RD systems (solid lines) are larger when $Pe$ increases as diffusion is then less efficient to smooth the viscosity profile. Consequently, the RDC system also becomes more unstable with increasing $Pe$, as shown in Fig. 9 and on the evolution of the mixing lengths, see Fig. 10(b) where we see that the onset time of the fingering instability decreases with increasing $Pe$. The smaller $Pe$, the quicker the mixing lengths tend to a steady state value at low $Pe$ whereas at large $Pe$ the mixing lengths are increasing instantaneously irrespective of the viscosity minimum at the interface.
C. Effect of Damköhler number, $Da$

To study the effect of varying the Damköhler number on the stabilization of fingering instability thanks to reactions decreasing the viscosity, Figure 11 depicts the concentrations, $\ln(\mu)$ and the reaction rate $\mathcal{R}$ at successive times for three values of $D_a$. We see that, when increasing $D_a$ (i.e. the reaction occurs faster), the viscosity minimum develops more quickly (see also Fig. 12(a)), the amount of product $C$ formed at a given time increases, and the reaction rate $\mathcal{R}$ decays faster because the reactants $A$ and $B$ are increasingly separated by the product $C$. As a consequence, when $D_a$ increases, the miscible interface between $A$ and $C$ stabilizes faster and the steady value of $L_a$ decreases. In parallel, the interface between $B$ and $C$ becomes uniform in time, and the corresponding values of $L_b$ and $L_c$ saturates (see Fig. 12(b)). The system is thus globally more stable when $D_a$ is larger.

We conclude thus from this parametric study that the displacement tends to stabilize (destabilize) at lower Pe (high Pe) for $R_c < 0$ ($R_c \geq 0$), and larger $D_a$ (smaller $D_a$). The optimal conditions to avoid fingering can thus be achieved when the rate of injection of displacing fluid is kept as low as possible to allow the viscosity minimum to build up.
FIG. 12. Same as Fig. 10 for variable $D_a$: 0.5 (thick dashed line), 1.0 (thick solid line) and 5.0 (thick dot-dashed line). The thin dashed line in panel (a) represents the non-reactive case.

V. CONCLUSION AND OUTLOOK

We have here analysed the influence of the injection flow rate on reactive VF driven by a simple $A + B \rightarrow C$ type chemical reaction decreasing the viscosity in situ. To do so, we have numerically integrated Darcy’s law for the evolution of the flow velocity and RDC equations for the concentrations coupled by a viscosity profile depending dynamically on the concentration of the chemical species. The injection flow rate has been varied by changing the values of the dimensionless parameter $Pe$. Nonlinear simulations have been performed to characterise the properties of reactive VF when a solution of a reactant $A$ displaces a solution of $B$ to produce the less viscous product $C$ at the miscible reactive interface. At lower $Pe$, the VF instability is less intense in both reactive and non-reactive cases because the viscosity gradients are smoothed out by diffusion. The reactive VF pattern covers nevertheless a larger area i.e. is spatially denser than the non-reactive pattern. These observations are in good agreement with experiments [20, 29]. Similarly to the non-reactive case, at higher $Pe$, VF is enhanced in reactive systems when the viscosity minimum does not have time to build up. Less-dense fingering patterns and more mixing are then observed. In other words, the fingering patterns at high $Pe$ cover a smaller area than at low $Pe$. In terms of displacement efficiency, the presence of a viscosity minimum at lower $Pe$ is found to optimize a homogeneous and regular displacement with less convective mixing.
Our study provides a mathematical framework to control VF in many geophysical processes e.g. reactive pollutant displacement, CO$_2$ sequestration and EOR. Recently, it has been shown that fingering instabilities in the application of EOR can be controlled by introducing a viscosity minimum in the zone of contact between the two fluids via the formation of foam between the injected gas and displaced oil [9]. In this context, the present study (i) provides a convection between viscosity minimum and stabilization, (ii) introduces a way to control VF by controlling the injection rate, (iii) shows that, at low injection rate, the reactive VF improves the sweep efficiency in comparison to the non reactive conditions.

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