MODELING AN ELASTIC FINGERING INSTABILITY IN A REACTIVE HELE-SHAW FLOW∗

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Abstract. In this paper we develop a mathematical model of Hele-Shaw flows in which two immiscible fluids react and form a gel-like phase at the interface, a phenomenon which has been observed in recent experiments. This phase significantly stiffens the interface. The interface is modeled as an elastic membrane which has a bending rigidity dependent on the local curvature. A dispersion relation is derived using an energy variational method. Several types of instabilities are categorized, and how various physical parameters affect the instability is investigated. Our model is able to account for some of the anomalous fingering instabilities observed in experiments by Podgorski et al. [Phys. Rev. E, 76 (2007), 016202]. Consistent with the experimental observations, the fingering instability in a reactive system is shown to be greater than in a nonreactive system.

Key words. interfacial instabilities, Hele-Shaw flow, elastic membrane, bending rigidity

AMS subject classifications. 76E17, 76D27, 74B05

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1. Introduction. In the classical setting of viscous fingering, two fluids meet in a Hele-Shaw cell at an interface where the Young–Laplace condition is satisfied: the pressure difference between two sides is equal to the product of the surface tension and the mean curvature. If the displacing fluid has a smaller viscosity than the displaced fluid, the interface is unstable, and ramified fingering patterns will emerge. The purely hydrodynamic instability resulting from viscosity contrast has been extensively studied; a small selection of published works includes [1, 14, 15, 17, 22, 26]. Many additional efforts have been put into more complicated systems involving Hele-Shaw flows. For instance, the inertia [4, 6, 12] and the rarefaction [11] of the fluids have been shown to have stabilizing effects on the fingering instability. Effects due to the Coriolis force have been studied in a rotating system [3, 8, 27].

When complex fluids are involved, Darcy’s law should be modified to incorporate the non-Newtonian effects. The mathematical challenge that the pressure field is no longer harmonic makes analytic and numerical studies difficult. The resultant fingering patterns are significantly different from those in classical Newtonian fluids. For example, Kondic, Shelley, and Palffy-Muhoray [16] showed numerically using a modified Darcy’s law that shear-thinning fluids can suppress the tip-splitting instability. Lindner, Coussot, and Bonn [18] found that the fingering patterns in a yield stress fluid are more ramified with lower injection rates, which is opposite to classical viscous fingering. While these anomalous phenomena are due to the property changes in the bulk fluids, complexities could also arise from the modified boundary conditions.

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Podgorski et al. [24] performed experiments using two miscible fluids which react at the interface, producing a gel-like phase at the interface which significantly increases its stiffness. Away from the interface the two fluids remain Newtonian. Although each fluid has the same density and viscosity, instabilities are always observed no matter which fluid displaces which.

In this study we construct a mathematical model for two immiscible chemically reactive fluids displacing each other in a Hele-Shaw cell. To understand the role of the gel-like phase produced by reaction, we consider the interface to be an elastic membrane with a bending rigidity which is a function of the local curvature. This provides a simple model of the constitutive weakening of the gel-like phase as a geometric property of the interface. In particular, interfacial deformation typically breaks the homogenous molecular arrangement and results in local weakening of the gel-like phase. It is assumed that the characteristic chemical reaction time is much smaller than the characteristic flow time, so that the Damköhler number is large [7, 19]. The two fluids are assumed to be Newtonian, and thus with no reaction the problem is identical to the classical Hele-Shaw flow problem. This scenario is not unlike the one in which the presence and redistribution of surfactants brings about variations in the surface tension [2, 28, 31]. Our theory is compatible with the experimental work of Podgorski et al. [24], except that in their case the fluids are miscible. Numerical studies on miscible fluid displacement can be found in [9, 13]. In our case, we assume that the gel-like phase produced at the interface prohibits further mixing of the fluids, and therefore miscibility plays an insignificant role in the fingering instability. The model we build explains why more ramified fingering patterns are observed in a reactive system [24].

The present paper is organized as follows. In section 2 we derive the dispersion relation of an infinitesimal perturbation to a circular interface, leaving the boundary condition in its most general form. In section 3 an energy variational method is implemented to obtain an explicit form for the boundary condition. In section 4 we define a curvature-dependent rigidity function and motivate such a choice. Section 5 is dedicated to categorizing different types of instabilities and analyzing how they are determined by the physical parameters of the system. In section 6 and section 7 we examine, respectively, the maximum growth rate of the perturbation and conditions for self-similar fingering patterns. They both indicate a greater instability for reactive systems compared to nonreactive ones.

2. Derivation of the dispersion relation. We consider that in a Hele-Shaw cell with gap thickness $h$ a fluid is injected at the origin, displacing another fluid which prefills the cell. Both fluid flows obey Darcy’s law [22],

$$u_k = -M_k \nabla P_k,$$

where $M_k = \frac{h^2}{12 \mu_k}$ is the mobility and $\mu_k$ is the viscosity of fluid $k$ (the subscripts $k = 1$ and 2 refer respectively to the interior and exterior fluid, as in Figure 1). We assume that an expanding circular interface $\Gamma_0$ with a radius $a$ is perturbed to $\Gamma_t$ with radius

$$r(\theta, t) = a(t) + \epsilon b(t) \cos(n \theta),$$

where $\epsilon \ll 1$, $n \geq 2$ is an integer, $\theta$ is the polar angle, and $b(t)$ represents the time-dependent amplitude of the perturbation. Denote by $\Omega_t$ the region enclosed by $\Gamma_t$. 

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Fig. 1. A circular interface with a mode-6 perturbation.

The local curvature of $\Gamma_t$ is

$$H(\theta) = \frac{1}{a} + \epsilon \frac{(n^2 - 1)b \cos(n\theta)}{a^2} + O(\epsilon^2).$$

As to the dynamic boundary condition on $\Gamma_t$, we denote the local pressure jump by

$$[P]_t \equiv P_1 - P_2,$$

and that on the unperturbed interface $\Gamma_0$ is denoted by $[P]_0$. In the case of two immiscible nonreactive Newtonian fluids, the pressure difference is given by

$$[P]_t = \sigma H,$$

where $\sigma > 0$ is the surface tension. This Young–Laplace equation is the most common boundary condition for Newtonian fluids in equilibrium, and it continues to hold for many non-Newtonian fluids [5, 16]. More generally, however, $[P]_t$ may contain $H$ and its derivatives. In section 3 we will derive the dynamic boundary condition for the reactive system by using an energy variational method.

We first solve for the Hele-Shaw base flow. Let $\phi_k (= -M_k P_k)$ be the velocity potential of the fluids, which satisfies Laplace’s equation in polar coordinates,

$$\frac{\partial^2 \phi_k}{\partial r^2} + \frac{1}{r} \frac{\partial \phi_k}{\partial r} + \frac{1}{r^2} \frac{\partial^2 \phi_k}{\partial \theta^2} = 0.$$

The solution to (2.6) with the unperturbed interface $\Gamma_0$ is

$$\phi_1^{(0)} = J \left( \log \frac{r}{a} + \frac{M_1}{M_2} \right), \quad \phi_2^{(0)} = J \left( \log \frac{r}{a} + 1 \right) + [P]_0 M_2,$$

where $J = \frac{1}{2\pi} \frac{d}{dt} \left( \pi a^2 \right)$ is the constant rate of injection. The required solution of (2.6) in the region bounded by $\Gamma_t$ is

$$\phi_k = \phi_k^{(0)} + (-1)^k \beta \left( \frac{r}{a^n} \right)^{(-1)^{k+1}} \cos(n\theta).$$
Here $\beta$ can be determined by the continuity boundary condition ($\frac{\partial \phi_1}{\partial r} = \frac{\partial \phi_2}{\partial r}$ on $\Gamma_t$) as

$$\beta = - \frac{\epsilon}{n} \left( \frac{Jb}{a} + a\hat{b} \right),$$

where $\hat{b} = \frac{d}{dt}b(t)$. Substituting (2.7)–(2.9) into the boundary condition (2.4), we have

$$[P]_t = \left[ \left( \frac{\phi_2}{M_2} - \frac{\phi_1}{M_1} \right) \right]_{\Gamma_t} = J \frac{M_2 - M_1}{M_1 M_2} \frac{eb\cos(n\theta)}{a} M_1 + M_2 \frac{Qb}{2\pi a} \frac{M_2}{M_1 M_2} (\frac{Qb}{2\pi a} + ab) + [P]_0.$$

The above expression can be rewritten as

$$\left( \frac{b}{a} \right)^{-1} \frac{d}{dt} \left( \frac{b}{a} \right) = \frac{J}{2\pi a^2} (n\Psi - 2) - \frac{M_1 M_2}{M_1 + M_2} ([P]_t - [P]_0) \frac{n}{\epsilon ab \cos(n\theta)},$$

where $\Psi = \frac{M_1 - M_2}{M_1 + M_2}$ is the Atwood number of mobility. The left-hand side of (2.11) has the relative amplitude of the disturbance $b/a$. Equation (2.11) is the general dynamic boundary condition which contains the pressure condition in its most general form.

Note that $[P]_t - [P]_0$ contains a $\cos(n\theta)$ term, as we will see in section 4, so that the right-hand side of (2.11) is independent of $\theta$. We remark that if the absolute amplitude $b$ is to be considered we will have

$$b^{-1} \frac{d}{dt} (b) = \frac{J}{2\pi a^2} (n\Psi - 1) - \frac{M_1 M_2}{M_1 + M_2} ([P]_t - [P]_0) \frac{n}{\epsilon ab \cos(n\theta)}.$$

### 3. Elastic boundary condition

In order to obtain an expression for $[P]_t$ that suitably represents the force exerted on the gel-like interface, we define the elastic energy of the interface as

$$E_b = \frac{1}{2} \int_{\Gamma_t} \omega ds - \int_{\Omega_t} \int dV,$$

where $\omega$ is the elastic energy density per unit length and $s$ is the arc length. The first term is the elastic energy of the membrane; the second term takes account of the volume constraint due to the incompressibility of the flow. Note that we impose no constraints on the circumference of the membrane. If the circumference is fixed, then another term $\alpha \int_{\Gamma_t} ds$ should be added to the right-hand side of (3.1), where the tensile stress $\alpha$ serves as the Lagrangian multiplier. The equation of mechanical equilibrium satisfied on $\Gamma_t$ is the vanishing of the first variation,

$$\delta E_b = 0,$$

under the condition of rotational symmetry [20, 21].

There are two typical choices of $\omega$ in (3.1): if $\omega = \sigma(constant)$, then $E_b$ is the surface energy of a fluid-fluid interface, and from (3.2) we obtain the Young–Laplace equation (2.5); if $\omega = \nu_0 H^2$, then (3.1) gives rise to the elastic energy of a membrane with constant bending rigidity $\nu_0$ [20]. In the latter situation, the first variation of $E_b$ in three dimensions can be found in [21] or [25]. In this paper, we treat the interface as
an elastic membrane with bending rigidity changing according to the local curvature; that is,
\begin{equation}
\omega = \nu(H)H^2.
\end{equation}

Using (3.3), we substitute (3.1) into (3.2) to get
\begin{equation}
[P]_t = -\frac{1}{2} \nu'' H^2 H_s^2 - \nu' \left( \frac{1}{2} H^4 + 3H_s^2 + 2HH_s + \nu' \left( \frac{1}{2} H^3 + H_s \right) \right).
\end{equation}

The details of the computation can be found in [10]. This dynamic boundary condition on the perturbed interface and its counterpart on the unperturbed interface are to be used to derive the dispersion relation. Note that variable bending rigidity has been investigated previously in the context of multicomponent vesicles in a viscous fluid [29, 30]. But the rigidity function therein depends on the surface phase, which is different from the case here.

4. Bending rigidity function. The bending rigidity of an elastic membrane may change as a result of various deformations of the membrane such as stretching, bending, and twisting. Here we treat the interface as a one-dimensional curve with zero width, and assume that the only change in the rigidity is dependent on the mean curvature \(H\). This approach overlooks the fact that the membrane may get stretched or compressed during the motion, thus neglecting certain mathematical complexities arising from the microstructure of the elastic interface. However, as mentioned earlier, deviations of any kind of the interface from a straight line should always decrease the rigidity, because such deviations break the homogeneity of the surface molecular arrangement, and thus the membrane can be deformed more easily. We refer to this as “curvature weakening.” At the extremity \(H \to \infty\), we set \(\nu\) to converge to a small positive value. Following this motivation, we define the bending rigidity function as
\begin{equation}
\nu(H) = \nu_0 (Ce^{-\lambda H^2} + 1 - C).
\end{equation}

Here the maximum rigidity \(\nu_0\) is the largest resistance of the membrane to disturbances and corresponds to the tightest arrangement of the surface molecules. The value of \(\nu(H)\) decreases from \(\nu_0\) to \((1 - C)\nu_0\) as \(H\) increases from 0 to infinity. So \(C\) measures the fraction of intermolecular bonds broken through surface deformation. We call \(C\) the rigidity fraction. The quantity \(1/\lambda\) is the characteristic curvature beyond which the \(\nu(H)\) has a substantial decrease (Figure 2). As seen from experiments by Podgorski et al. [24], there appears to be a typical magnitude of radius of the finger tips over which the finger will split. This indicates a possible choice of \(\lambda\).

We can evaluate \(\nu(H)\) at the perturbed circular interface \(\Gamma_t\) and substitute it into (3.4) to get, up to \(O(\epsilon)\),
\begin{equation}
[P]_t = \frac{\delta E_h}{\delta n} = \nu_0 C e^{\frac{-\lambda^2}{2a^2}} \frac{2\lambda^2 - a^2}{2a^5} - \frac{\nu_0}{2a^3}
\end{equation}

\begin{equation}
+ \epsilon \nu_0 \cos(\theta) \left( Ce^{\frac{-\lambda^2}{2a^2}} B_1(\lambda, a, n) + (1 - C) B_2(a, n) \right),
\end{equation}

where \(B_1(\lambda, a, n)\) and \(B_2(a, n)\) are given by
\begin{equation}
B_1(\lambda, a, n) = \frac{1}{2a^5} \left( (4\lambda^4 - 10\lambda^2 a^2 + 2a^4)n^4 - (12\lambda^4 - 24\lambda^2 a^2 + 5a^4)n^2 + (8\lambda^4 - 14\lambda^2 a^2 + 3a^4) \right),
\end{equation}
\begin{equation}
B_2(a, n) = \frac{2n^4 - 5n^2 + 3}{2a^4}.
\end{equation}
Since \([P]_0\) is simply the \(O(1)\) term in (4.2), we have

\[
[P]_t - [P]_0 = \epsilon \nu_0 b \cos(n \theta) \left( C e^{-\frac{\eta^2}{2}} B_1(\lambda, a, n) + (1 - C) B_2(a, n) \right).
\]

Substituting (4.4) into the boundary condition (2.11), we have

\[
\left( \frac{b}{a} \right)^{-1} \frac{d}{dt} \left( \frac{b}{a} \right) = \frac{\tilde{\gamma} \eta}{\lambda^3} S_n(C, \eta, \tilde{J}, \Psi, \lambda),
\]

where

\[
S_n(C, \eta, \tilde{J}, \Psi, \lambda) = \frac{\eta^{5/2}}{2n(n^2 - 1)}(A_1(n^2 + 1) + A_2),
\]

\[
\tilde{\gamma} = \frac{\nu_0 M_1 M_2}{M_1 + M_2}, \quad \tilde{J} = \frac{\lambda^3 J}{\eta^2},
\]

and

\[
A_1(C, \eta) = C e^{-\eta}(-4\eta^2 + 10\eta - 2) - 2(1 - C),
\]

\[
A_2(C, \eta) = C e^{-\eta}(12\eta^2 - 24\eta + 5) + 5(1 - C).
\]

The interface is linearly unstable to the \(n\)th mode perturbation if and only if

\[
S_n(C, \eta, \tilde{J}, \Psi, \lambda) > 0.
\]

The first term on the right-hand side of (4.6) signifies the effect of injection, which is negative if \(\Psi < \frac{2}{n}\); the second term signifies the effect of bending. When \(\nu(H) \equiv \nu_0\) (i.e., \(C = 0\)) we have \(A_1 = -2\) and \(A_2 = 5\), meaning that bending forces always stabilize the interface. But we shall see that choosing a curvature-dependent bending rigidity (e.g., (4.1)) may lead to a destabilizing effect for the system.

5. **Parametric study.** Here we survey the ways in which a fingering instability can occur for our elastic interface model. Our focus is (4.5), aided by (4.6) and (4.7). Before proceeding, we summarize the *five* dimensionless parameters that are
Table 5.1
Signs of some linear combinations of $A_1(C, \eta)$ and $A_2(C, \eta)$.

<table>
<thead>
<tr>
<th></th>
<th>$A_1 &gt; 0$</th>
<th>$A_1 \leq 0$</th>
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<tbody>
<tr>
<td>$A_2$</td>
<td>Undetermined</td>
<td>$&gt; 0$</td>
</tr>
<tr>
<td>$A_1 + A_2$</td>
<td>Undetermined</td>
<td>$&gt; 0$</td>
</tr>
<tr>
<td>$5A_1 + A_2$</td>
<td>$&gt; 0$</td>
<td>Undetermined</td>
</tr>
<tr>
<td>$5A_1 + 2A_2$</td>
<td>$&gt; 0$</td>
<td>$\geq 0$</td>
</tr>
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</table>

important in the system: the rigidity fraction $C$, the injection rate $\hat{J}$, the Atwood number of mobility $\Psi$, the characteristic radius $\lambda$, and $\eta = (\frac{\lambda}{a})^2$, which is the inverse square of the nondimensional radius. We can see from (4.6) that the $n = 1$ mode is linearly stable, which corresponds to a shift of the center of the circular interface. Instabilities may arise for $n > 1$. By assuming that $n = 2$ is the smallest wavenumber for any given set $(C, \eta, \hat{J}, \Psi, \lambda)$, we categorize four different types of linear stabilities as follows:

1. **stable** if the perturbation of any wavenumber diminishes; i.e., $S_n \leq 0$ for all $n \geq 2$;
2. **unstable** if the perturbation of at least one wavenumber grows; i.e., $S_n > 0$ for some $n \geq 2$;
3. **intrinsically unstable** (I-unstable) if the perturbation of large-enough wavenumbers grows; i.e., there exists an integer $n_0 \geq 2$ such that $S_n > 0$ for all $n > n_0$;
4. **thoroughly unstable** (T-unstable) if the perturbation of any wavenumber grows; i.e., $S_n > 0$ for all $n \geq 2$.

It follows immediately from the above definition that the T-instability is a subset of the I-instability, and the I-instability is a subset of the instability.

**5.1. I-instability.** Since $S_n$ is a fifth-order polynomial in $n$ with the highest coefficient $\frac{1}{2} \eta^{3/2} A_1$ and $A_1$ is a function of $C$ and $\eta$, I-instability is a property dependent only on $C$ and $\eta$ but not on $\hat{J}$, $\Psi$, or $\lambda$. Moreover, positivity of $A_1$ guarantees the positivity of $S_n$ for large enough $n$. This proves the following result.

**Theorem 5.1.** The system is I-unstable if and only if $A_1 > 0$.

For a system to be I-stable for all radii $a$, $A_1$ must be negative for all $\eta > 0$. It can be shown that this yields the condition

$$C \leq \min_{\eta > 0} \frac{e^\eta}{\eta^3 - 2\eta^2 + 5\eta - 1} \approx 0.56,$$

and the minimum is attained at $\eta \approx 0.81$.

**5.2. Zero flux: Instability and T-instability.** In general, both instability and T-instability depend on the parameters $\Psi$ and $\hat{J}$. When $\hat{J} = 0$ we can prove that I-instability implies T-instability. To see this, let us assume that the system is I-unstable (so $A_1 > 0$); then it follows from Table 5.1 that $A_1(n^2 + 1) + A_2 > 5A_1 + A_2 > 0$ for all $n \geq 2$. Therefore we have the next result.

**Theorem 5.2.** If $\hat{J} = 0$, then I-instability is equivalent to T-instability.

The different stability regions when $\hat{J} = 0$ are plotted in Figure 3. As more fluid is injected, $\eta$ decreases. We can see that the regions of these instabilities are bounded by a finite strip in $\eta$ in the $C$-$\eta$ plane. Note that if $C$ is smaller than some critical value around 0.5, the interface is stable for all $\eta$. We have the following criterion for stability when $\hat{J} = 0$. 

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Theorem 5.3. If \( \hat{J} = 0 \), the system is stable if and only if \( 5A_1 + A_2 \leq 0 \).

Proof. If \( \hat{J} = 0 \), then the sign of \( S_n \) is dictated by the sign of \( A_1(n^2 + 1) + A_2 \). If the system is stable, it must be stable for \( n = 2 \), so \( 5A_1 + A_2 \leq 0 \). On the other hand, if \( 5A_1 + A_2 \leq 0 \), then \( A_1 \leq 0 \) (Table 5.1). So \( A_1(n^2 + 1) + A_2 \leq 5A_1 + A_2 \leq 0 \) for all \( n \geq 2 \).

It is noteworthy that our model allows an instability to occur even if \( \hat{J} = 0 \): no injection takes place. The origin of the instability is the curvature weakening effect, which is signified by the positivity of the quantity \( A_1(n^2 + 1) + A_2 \).

5.3. Nonzero flux: The role of the Atwood number. We can see from (4.6) that if \( \hat{J} \neq 0 \), the Atwood number \( \Psi \) plays a crucial role in determining the stability of the system. It will suffice to consider only \( \hat{J} \geq 0 \), so long as we allow \( \Psi \) to be in the interval \([-1, 1]\). We remark that in a linear Hele-Shaw cell if \( \Psi = 0 \), the injection does not affect the stability result at all. Nonetheless, it is conceivable that the speed at which one fluid intrudes into the other does influence the stability behavior even if they have the same mobility. In this case, the inertia of the fluids, which is neglected in Darcy’s law, must be taken into consideration [12].

5.3.1. Positive Atwood number. For positive \( \Psi \), the injection always stabilizes perturbations with wavenumber \( n \leq \frac{3}{\Psi} \) but destabilizes those with wavenumber \( n > \frac{3}{\Psi} \). As \( a \to \infty \) (i.e., \( \eta \to 0 \)), the second term in (4.6) decreases, implying that modes larger than \( \frac{3}{\Psi} \) will become eventually unstable. In the \( C-\eta \) plane this is illustrated by the existence of a tail region near the \( C \)-axis for any mode larger than \( \frac{3}{\Psi} \) (Figure 4).

The following is a result of the ordering of different modes.

Theorem 5.4. Suppose that \( \Psi \in (0, 1] \) and \( S_{n_0} < 0 \), where \( n_0 = \lceil \frac{3}{\Psi} \rceil \) (the smallest integer larger than or equal to \( \frac{3}{\Psi} \)); then \( S_n < S_{n_0} < 0 \) for all \( n > n_0 \).

Proof. Since \( n_0 \) is chosen so that \( n_0 \Psi - 2 > 0 \) and

\[
S_{n_0} = (n_0 \Psi - 2)\hat{J} + \frac{1}{2a^3}n_0(n_0^2 - 1)(A_1(n_0^2 + 1) + A_2) < 0,
\]
we must have
\[(5.2) \quad (5A_1 + A_2) + (n_0^2 - 4)A_1 = A_1(n_0^2 + 1) + A_2 < 0.\]
From (5.2) it follows that $A_1 < 0$, because otherwise both $A_1$ and $5A_1 + A_2$ are positive (Table 5.1).
Define
\[f(x) = \frac{x(x^2 - 1)(A_1(x^2 + 1) + A_2)}{2a^3(\Psi x - 2)};\]
then
\[S_{n_0} = (n_0\Psi - 2)(\hat{J} + f(n_0)).\]
We will show that $f'(x) < 0$ for all $x \geq n_0$. Indeed, computing $f'(x)$, we find that
\[(5.3) \quad f'(x) = \frac{1}{a^3(\Psi x - 2)^2}[(\Psi x^3 - 3x^2 + 1)(A_1(n_0^2 + 1) + A_2)
+ (2\Psi x^5 - 5x^4 - (n_0^2 + 1)\Psi x^3 + 3(n_0^2 + 1)x^2 - n_0^2)A_1].\]
Since for $x \geq n_0$, $\Psi x^3 - 3x^2 + 1 = x^2(\Psi x - 3) + 1 > 0$ and
\[2\Psi x^5 - 5x^4 - (n_0^2 + 1)\Psi x^3 + 3(n_0^2 + 1)x^2 - n_0^2
= (\Psi x - 3)x^2(2x^2 - n_0^2 - 1) + (x^4 - n_0^2) > 0,\]
we can directly compute that, for $n > n_0$,
\[S_n - S_{n_0} = (n\Psi - 2)(\hat{J} + f(n)) - (n_0\Psi - 2)(\hat{J} + f(n_0))
\leq (n\Psi - 2)(\hat{J} + f(n_0)) - (n_0\Psi - 2)(\hat{J} + f(n_0))
= \frac{(n - n_0)\Psi S_{n_0}}{n_0\Psi - 2} < 0.\]
As $\hat{J}$ increases, the instability is triggered at an earlier stage (i.e., larger radius); however, the number of incipient fingers does not depend on $\hat{J}$ (Figures 5 and 6).
5.3.2. Special case $\Psi = 1$. When the viscosity of the injected fluid is negligible compared to that of the displaced fluid, the Atwood number $\Psi$ is 1. This can be approximated by injecting a gas (e.g., air) into a viscous fluid, and is the most common experimental configuration \[22, 26\]. From (4.6) we can see that $\dot{J}$ is always destabilizing, and instabilities occur if $\dot{J}$ is greater than the critical value

$$\dot{J}_c \equiv \frac{n(n^2 - 1)(A_1(n^2 + 1) + A_2)}{2a^3(n - 2)}.$$ (5.4)

Since $A_1 > 0$ implies $A_1(n^2 + 1) + A_2 \geq 5A_1 + A_2 > 0$ for all $n \geq 2$ (the same argument made prior to Theorem 5.2), the following result holds.
Theorem 5.5. If $\Psi = 1$, then $I$-instability is equivalent to $T$-instability.

As a consequence of Theorem 5.4, when $\Psi = 1$ and $S_3 < 0$, we have $S_k < S_3$ for all $k > 3$ (Figure 5). But for $\Psi \neq 1$ this is generally not true (Figure 6). We summarize this as follows.

Corollary 5.6. Let $\Psi = 1$ and $A_1 < 0$. If the system is unstable, then $n = 3$ always becomes the first unstable mode.

5.3.3. Nonpositive Atwood number. It follows from (4.6) that injection always has a stabilizing effect when $\Psi \leq 0$. Moreover, as $a \to \infty (\eta \to 0)$, $S_n$ will eventually be negative, and thus there is no trailing region near $\eta = 0$ (Figure 7). For a classical Hele-Shaw flow or if the bending rigidity is constant, the interface remains stable throughout the whole injection process. But if the chemical reaction is present, instability may be triggered by the destabilizing effect of curvature weakening, which is signified by the positivity of $A_1(n^2 + 1) + A_2$. This curvature weakening mechanism which promotes surface creation has also been hypothesized as the cause of other elastic instabilities in fluid dynamics [23].

Fig. 7. Contour plots of $S_n (n = 2, 3, 5)$ for $\Psi = -1, J = 1$. The dotted line encloses the $I$-stability region. For $C$ smaller than $\approx 0.56$, the system is stable for all $\eta$.

5.3.4. Special case $\Psi = 0$. When $\Psi = 0$, the interior and exterior fluids are indistinguishable in their influence on the fingering instability. Consequently the instabilities are indistinguishable. This is consistent with the experiments in Podgorski et al. [24]. Even though the fingering develops into very different patterns at a later stage, the shapes of the incipient fingers are remarkably similar (see Figures 1 and 2 in [24]). The stable region in the $C$-$\eta$ plane for $\Psi = 0$ is plotted in Figure 8.

6. Maximum growth rate. For a given $\hat{J}$, the most unstable mode $n_{max}$ can be obtained by setting $\frac{dS_n}{dn} = 0$. The maximum growth rate $G_{max}$ is defined to be the right-hand side of (4.5) when $n = n_{max}$. Figures 9 and 10 show plots of $n_{max}$ and $G_{max}$, respectively, as functions of $\eta$ for given $\hat{J}$, $\Psi$, and $C$. We can see that both $n_{max}$ and $G_{max}$ increase as $C$ increases, which implies that the reactive system is more unstable than the nonreactive one, and thus the reaction has a destabilizing effect. We also notice that for $C$ larger than a critical value (approximately 0.5), $n_{max}$ is no longer a monotonic function of $\eta$ but remains single-valued.
7. Self-similarity of the fingering pattern. In their recent work Li et al. [17] have obtained a way of suppressing the interfacial instabilities of a Hele-Shaw flow by controlling the time-dependent injection rate. Using Darcy’s law (2.1) and dynamic boundary condition (2.5), they found that if the injection rate is chosen to be 
\[ \frac{K_n(n^2-1)}{(n-2)m} \]
when the \( n \)-mode always has the largest growth rate. This means that the injection rate should be proportional to \( t^{1/3} \). Furthermore, they verified numerically and experimentally that the \( n \)-mode fingering pattern will persist, and a self-similar fingering pattern is maintained even when the system is far out of the linear region, which can be partially explained by the fact that both the number of unstable modes and the fastest growing mode do not change in time.
Here we extend the analysis of Li et al. [17] to the reactive system in which the boundary condition is taken to be (4.2). We can rewrite the function $S_n$ in (4.6) as

$$S_n(C, \eta, \hat{J}, \Psi, \lambda) \equiv (n\Psi - 2) \left( \hat{J} - J_R \right),$$

where $J_R = -\frac{n(n^2-1)(A_1(n^2+1)+A_2)}{2(n\Psi-2)}\eta^{3/2}$. If $\hat{J} = J_R$, the growth rate of perturbation is zero. We denote by $J_n^+$ and $J_n^-$, respectively, the upper and lower bounds of the injection rate such that the $n$-mode perturbation always has the fastest growth rate. This happens if

$$S_n \geq S_{n+1} > 0 \quad \text{and} \quad S_n \geq S_{n-1} > 0.$$

From (4.6) and (7.2) we obtain

$$J_n^+ = -\frac{n(n+1)\eta^{3/2}}{2\Psi}((5n^2 + 5n + 5)A_1 + 3A_2)$$

and

$$J_n^- = -\frac{n(n-1)\eta^{3/2}}{2\Psi}((5n^2 - 5n + 5)A_1 - 3A_2).$$

Plots of $J_n^+$ and $J_n^-$ for different values of $C$ are given in Figure 11. Note that a larger radius corresponds to a smaller $\eta$. When $C = 0$, $A_1$ and $A_2$ are constant, so from (7.3) we can see that $J_n^+$ and $J_n^-$ scale as $\eta^{3/2}$. This is because as the radius gets larger, the local flattening of the interface lowers the injection rate required to maintain a specific mode. For a moderate range of $C$, the flattening brings about a second and opposite effect, which is the increment in rigidity due to the change of curvature (or curvature-hardening; see Figure 2). In particular, when $C$ is sufficiently large (e.g., $C \approx 0.5$), then $J_n^+$ and $J_n^-$ become nonmonotonic, achieving local minima near $\eta \approx e^{-1/4}$. Therefore, for $\eta$ in this interval, the flux needs to be increased to maintain self-similar evolution. At certain values of the radius this increment in
rigidity becomes appreciable when $C$ is large enough. In spite of this, the overall $J_n^+$ and $J_n^-$ are smaller for larger $C$ (Figure 11). This again shows that the fingering instability is more pronounced in a reactive system than in a nonreactive system.

8. Conclusion. In this paper we have developed a mathematical model of a reactive circular Hele-Shaw flow, in which the chemical reaction between two fluids dominates the condition at the interface. The interface is treated as an elastic membrane with a bending rigidity $\nu(H)$ which is a function of the local curvature $H$. The specific choice of $\nu(H)$ in (4.1) is meant to be illustrative, and it captures the essence of curvature weakening. We have performed a linear stability analysis and investigated the effects of various parameters on the stability. This model successfully reproduces the destabilizing effects observed in a reactive system, where a gel-like phase is produced at the interface between two miscible fluids [24]. In particular, this model explains the occurrence of instability when the injection is zero and when two fluids have the same viscosity. In both cases, the interfacial instability is triggered by the curvature weakening effect of the elastic interface, as the classical Hele-Shaw flow predicts stable evolution in these regimes. Therefore our model may be useful in understanding the spontaneous blebbing motion in an oil-water-surfactant system, in which the curvature of the interface plays an important role [31]. More generally, we find that interfaces in the reactive system are more unstable than those in their nonreactive counterparts.

As a large system with five parameters, the reactive Hele-Shaw flow we consider here exhibits several interesting phenomena such as the crossing of different modes (Figures 4, 7, and 8), and nonmonotonicity of the most unstable mode (Figure 9) and of the injection rate (Figure 11). They are outcomes of the complex competition between the injection flux (which can be stabilizing or destabilizing), the flattening of the surface due to domain growth (stabilizing or destabilizing), and curvature weakening (destabilizing).

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REFERENCES


