Submitted to Physics of Fluids

A Numerical Study of Chemical Front Propagation in a Hele-Shaw Flow under the Buoyancy Effects

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Abstract

We consider the propagation of chemical fronts in a Hele-Shaw flow where the front is assumed to propagate with a curvature dependent velocity. The motivation is to model some recent experiments that employ aqueous autocatalytic chemical reactions in such a device. The density change across the front in such experiments is quite small so the Boussinesq approximation can be used, and the flow field generated is exclusively due to the buoyancy effects. We derive a free boundary formulation based on Darcy's law and a single layer potential, and describe the evolution in terms of the $\theta - L$ formulation, in which the tangent angle and the perimeter of the closed front are followed in time. Numerical solutions are obtained for this formulation with a rising and expanding bubble. As observed in the experiments, a fingering phenomenon which is different from the surface tension associated phenomenon appears in our calculations. The mechanisms that control the wavelength selection of the fingers are discussed.

1 Introduction

Buoyancy effects can be an important factor in the propagation rates and shapes of premixed gas flames, since the density of the mixture experiences a change when the reaction takes place. When the flame is propagating in a vertical direction, the velocity flow field can be significantly altered by a Rayleigh-Taylor type instability mechanism, and therefore it modifies the flame propagation. This effect is especially important for mixtures with low burning velocities. However, a detailed understanding of this interaction exists only for certain very simple systems. Reasons for this lack of understanding of more general systems include a large density ratio across the flame front for gas flames (typically 7) which greatly complicates the flame and flow interaction, and substantial changes in fluid properties across the front, such as the viscosity, thermal diffusivity and mass diffusivity of the fluids.

To isolate the interaction between the flame propagation and the buoyancy effect, it is very helpful to have a system where the aforementioned complications are absent. In some recent experiments conducted by Ronney and his coworkers[1−4], aqueous autocatalytic chemical reactions are employed to produce propagating chemical fronts which are analogous to premixed gas flame fronts. These fronts have very low density changes (typically 0.03%), which greatly simplify the reactive flow system. In a recent experiment of Ronney[5], the Hele-Shaw flow is chosen for the propagation of the chemical fronts because of its ability to be modeled by a two-dimensional system and the absence of any imposed flow. In this case, the flow is generated solely by the effects of the buoyancy associated with the density difference caused by the chemical reaction. We will be able to observe the effect of this very small density ratio on the front propagation, and indeed some fingering phenomenon is observed (Fig. 1) when the plates are placed vertically, which clearly suggests a finger wavelength selection mechanism. This particular finger wavelength is obviously very important to our understanding of the wrinkling fronts in the flow.

From the point of view of flame stability, Hele-Shaw flows were also considered by Joulin and Sivashinsky[6] to analyze the effects of momentum and heat losses on the flame stability, where buoyancy is also a crucial factor. In the case where there is no heat loss to the wall, they showed that the Landau-Darrieus instability is modified by the friction to adiabatic walls through a damping coefficient and a Saffman-Taylor type of instability[7]. The latter may dominate the stabilizing influence of gravity for downwardly propagating fronts in narrow channels, while for upward propagations the friction reinforces the Rayleigh-Taylor instability mechanism. In reality the wall is not adiabatic and heat generated from the reaction can be lost through the wall which is typically maintained at a constant temperature. The fluid density will therefore resume its value before the reaction and the buoyancy effect would be expected to be reduced. In the analysis of Joulin and Sivashinsky, this heat loss effect is indeed found to moderate the mechanisms shown in the adiabatic case, but the long wavelengths of wrinkling may still remain unstable.

In this paper, we derive a simplified free-boundary model for the front propagation system of the experiment mentioned above, and introduce a numerical method to solve this free-boundary problem. Boundary integral formulations have been very successful in numerical solutions for Hele-Shaw flows[8–11]. The mathematical model here can also be viewed as a modification to the system considered in the linear stability analysis of Joulin and Sivashinsky in that the heat loss effect is ignored while the effect of momentum loss to the walls is emphasized by using Darcy's law for Hele-Shaw flows in both fluid regions. The Saffman-Taylor instability is absent in our model since the fluid viscosity experiences only a very small change across the reaction front. The advantage of this simplified system is that the free boundary problem benefits from a boundary integral formulation where a single layer potential can be used to represent part of the solution. The consequence of the boundary integral formulation is that the original two-space-dimensional problem is reduced to a one-space-dimensional problem in a periodic domain where spectral accuracy can be achieved in a finite difference discretization, with the help of Discrete Fourier Transforms. The spatial accuracy of the approximation to the boundary integral is very important since the major difficulty in any numerical approach to this type of problems is the unstable nature of the physical flows. The time evolution of the chemical front is further complicated by the coupling between the fluid velocity and the chemical front propagation which involves a curvature dependent burning velocity. In a front tracking method that uses markers along the front, it is a well-known fact that constructing the normal unit vectors with finite difference approximations is numerically unstable. While in the G-equation formulation, a two-dimensional field is required and the constant burning velocity shows up in a nonlinear term. Our approach here is based on a boundary integral with the front position given by a set of markers on the boundary. The numerical difficulty to approximate the unit normal associated with a front tracking method mentioned above is eliminated when we choose to follow the evolutions of the tangent angle and the perimeter. This is so-called the $\theta - L$ formulation[12,13,11,14]. The advantage of this choice to describe a chemical front, as opposed to the more familiar G-equation formulation, is an improved accuracy and the reduction of the number of dimensions. The constant burning velocity can be described by a constant term in an ordinary different equation for the perimeter of the front, as opposed to the nonlinear term in the G-equation. There is no need to approximate the normal unit vector at a grid point on the front through the particle positions in the calculation, since the tangent angle at this point is provided from the formulation. Of course there is the disadvantage that topological changes of the front cannot be accommodated in this $\theta - L$ formulation. However, for the focus of this study, we are only interested in the growth of the unstable modes in a relatively early stage of the fingering development, rather than the overall topological structure of the product region. In this case, merging and splitting of the product region do not appear usually, as observed in the experiments. Another important advantage of this form is that the curvature is related to the tangent angle linearly, so the curvature dependence of the burning velocity can be implicitly treated in the time stepping procedure. This eliminates another potential numerical instability factor. The grid points along the front are arranged in such a way that they remain equally distributed in time, as long as they start to be so. Fast Fourier transforms are used throughout for differentiation, integration along the front, and the time evolution is also performed in the Fourier space. Spectral accuracy is therefore achieved in our numerical approximations to accommodate our needs for the best accuracy. As the perimeter of front grows in time due to the chemical reaction, and the structure becomes more and more complex, it is natural to add more points along the front. This can be conveniently achieved in the current formulation due to the fact that we we work in the Fourier space.

For an example of front propagation we want to consider that closely matches a particular experimental setup, a circular region is perturbed initially and ignited instantly. As chemical reaction takes place, this product region rises and expands in the Hele-Shaw cell. The expansion of the product region is a consequence of the chemical reaction, and the rise of the product region is due to the gravity effect. As we expect from the experimental observation and the linear stability analysis, the upper part of the circular front will be unstable to small perturbation due to the Rayleigh-Taylor type of instability, leading to some fingering phenomenon, and the bottom part of the front will remain smooth as long as the wavelength of the perturbation is not long enough. The questions are how the Rayleigh-Taylor instability will interact with the front propagation, and what wavelengths of wrinkling are most pronounced in this process. To address these issues, we study the effects of various parameters, such as the curvature dependence, cell width, and density ratio, in a sequence of calculations in a hope to determine the factors of the wavelength issue. Since the curvature dependence corresponds to smoothing of the front, it is not surprising to discover through numerical calculations that in this particular model, indeed the wavelength of the fingers is determined by the level of curvature dependence of the burning velocity.

The paper is organized as follows. In section 2, we formulate the free boundary problem and introduce a single layer potential to represent the pressure driven fluid velocity field in terms of the front positions. In section 3, we describe the $\theta - L$ formulation for a closed front and the evolution equations for θ and L. In section 4, the numerical methods are introduced, then we discuss our results in section 5. Finally, a conclusion and some discussions on the future work that includes the heat loss effects are given.

2 Formulation of the Problem

Here we consider the viscous and incompressible fluid in a Hele-Shaw cell between two parallel plates that is placed vertically, with a cell gap w. After averaging the flow quantities across the cell gap with the consideration of the no-slip boundary conditions on the plate walls, the averaged velocity $\mathbf{u}$ and the averaged pressure p satisfy the following Darcy's law in two space dimensions:

$$\mathbf{u} = -\frac{w^2}{12\mu}\nabla(p + \rho g y), \tag{1}$$

$$\nabla \cdot \mathbf{u} = 0. \tag{2}$$

Here μ is the fluid viscosity and g is the gravity acceleration. We use the coordinate y to denote the upward direction.

In the presense of a front separating two different fluids, the above equations are satisfied in both regions. In the situation discussed here, a chemical front separates the reactant and product regions, and propagates in the flow field with certain burning velocity. The effects of the front on the fluids are given through certain jump conditions prescribed along the front. If we take the divergence of Eq. (1) and observe the incompressibility condition (2), it is easy to see that the pressure p satisfies the Laplace equation in both regions. It is therefore natural to prescribe the jump conditions for the solution in terms of the jumps in p and its normal derivative $\partial p/\partial n$. The jump in p is usually associated with a force balance across the front and it is given by the Laplace-Young condition, which attributes the pressure jump to the surface tension effect. In our particular case, the fluids in the reactant region and the product region are *miscible,* with slight differences in fluid physical properties such as viscosity and density. It is therefore reasonable to assume that the surface tension effect can be ignored. As the consequence, the jump in pressure across the front

$$[p] = 0, \qquad x \in \Gamma, \tag{3}$$

where Γ is the curve representing the reacting front.

The jump in the normal derivative of p is caused by a density difference across the front and is therefore related to the gravity effect, which is the focus of this study. The density of the fluid in the reactant region (ρ_1) is reduced by a very small amount to ρ_2 when the reaction takes place and the fluid is converted to the product. Since we assume that the heat release is small and there is no volume expansion along the front, the normal velocity across the front is required to be continuous. Therefore, according to Eq. (1),

$$[\frac{\partial p}{\partial n}] = g\Delta\rho\cos\theta, \tag{4}$$

where θ is the tangent angle of the curve with respect to the horizontal line at the point $\mathbf{x}$ on Γ, as illustrated in Fig. 2, and the density difference $\Delta\rho = \rho_1 - \rho_2 > 0$ is assumed to be very small relative to ρ_1 and ρ_2.

We now try to find a representation for the pressure in terms of a single layer potential by considering a nonintersecting, closed curve Γ in the infinite space only. As we will show, finite boundary effects can be incorporated by adding a potential flow. First, we consider the far field flow condition. As $\mathbf{x} \to \infty$, $\mathbf{u} \to 0$. So the pressure should approach the hydrostatic pressure, that is,

$$p \to p_0(\mathbf{x}) = -\rho_1 g y. \tag{5}$$

A single layer potential can be introduced to provide a function that is harmonic in both regions, continuous across the front but with a required jump in the normal derivative[15]. Combining the far field pressure with such a boundary integral, we have

$$\begin{aligned} p(\mathbf{x}) &= \frac{1}{2\pi}\int_\Gamma \log|\mathbf{x}-\mathbf{y}|[\frac{\partial p}{\partial n}]dS_\mathbf{y} + p_0(\mathbf{x}) \\ &= \frac{g\Delta\rho}{2\pi}\int_\Gamma \log|\mathbf{x}-\mathbf{y}|\cos\theta(\mathbf{y})dS_\mathbf{y} - \rho_1 g y. \end{aligned} \tag{6}$$

Given the front position, we can therefore represent the fluid velocity away from the front by using Eq. (1) and (6). For $\mathbf{x} \notin \Gamma$,

$$\mathbf{u}(\mathbf{x}) = -\frac{w^2}{12\mu}\nabla\left[\frac{g\Delta\rho}{2\pi}\int_\Gamma \log|\mathbf{x}-\mathbf{y}|\cos\theta(\mathbf{y})dS_\mathbf{y} + (\rho_i - \rho_1)gy\right] \tag{7}$$

where ρ_i is the density for region i (1 for the reactant and 2 for the product).

As we move across Γ, the pressure gradient experiences a jump, so is the term $(\rho_i - \rho_0)$, in such a way that the normal fluid velocity is continuous. However, the tangential velocity across the front is not continuous. Therefore, the front here can be viewed as a vortex sheet with the strength depending on the orientation of the local front. This is similar to the vortex sheet formulation[16,17,11] except that the strength is given by a different mechanism here. To evaluate this normal velocity, we can take the limit of Eq. (7) with $\mathbf{x}$ approaching Γ from either side, observing the special properties of the normal derivative of a single layer potential near the boundary[15]. As the consequence, we obtain the following normal velocity of the fluid at a point $\mathbf{x}$ on Γ,

$$\begin{aligned} u_n(\mathbf{x}) &= \mathbf{u}(\mathbf{x})\cdot\mathbf{n} \\ &= -\frac{w^2 g\delta}{12\nu}\left[\frac{1}{2\pi}\int_\Gamma \frac{\partial}{\partial n_\mathbf{x}}\log|\mathbf{x}-\mathbf{y}|\cos\theta(\mathbf{y})dS_\mathbf{y} + \frac{1}{2}\cos\theta(\mathbf{x})\right], \end{aligned} \tag{8}$$

where $\mathbf{n} = (\sin\theta, -\cos\theta)$ is the unit normal vector, pointing from the product region to the reactant region, $\nu = \mu/\rho_1$ is the kinematic viscosity of the fluid, and $\delta = \Delta\rho/\rho_1$ is the relative change of the density. Note that this boundary integral is different from the one in Eq. (7) in that the integral is evaluated on Γ so the integrand is not well defined when $\mathbf{y} = \mathbf{x}$. However, it can be shown that this singularity is actually removable.

This velocity field is supposed to vanish as $\mathbf{x} \to \infty$. If we want to solve the flow in a bounded region Ω, we would need to modify the pressure so the velocity satisfies the corresponding boundary condition. This can be achieved by adding a suitable harmonic function to the existing pressure for correction of the boundary conditions. It is known that Hele-Shaw flows are analogous to potential flows in that we can only impose the normal velocity on the boundary. To impose this boundary condition for the pressure correction, the velocity field shown above can be evaluated on the boundary, assuming that the product region will not spread to the boundary,

$$\mathbf{u}(\mathbf{x}) = -\frac{w^2 g\delta}{24\pi\nu} \int_\Gamma \frac{\mathbf{x}-\mathbf{y}}{|\mathbf{x}-\mathbf{y}|^2} \cos\theta(\mathbf{y}) dS_{\mathbf{y}}, \quad \mathbf{x} \in \partial\Omega. \tag{9}$$

Now we require the pressure correction $\tilde{p}(\mathbf{x})$ to satisfy the following equation

$$\nabla^2 \tilde{p} = 0, \tag{10}$$

and the boundary condition

$$\frac{\partial \tilde{p}}{\partial n} = -\frac{g\Delta\rho}{2\pi} \int_\Gamma \frac{(\mathbf{x}-\mathbf{y}) \cdot \mathbf{n}}{|\mathbf{x}-\mathbf{y}|^2} \cos\theta(\mathbf{y}) dS_{\mathbf{y}}, \quad \mathbf{x} \in \partial\Omega. \tag{11}$$

The total pressure, now including the hydrostatic and boundary corrections, is

$$p(\mathbf{x}) = \frac{g\Delta\rho}{2\pi} \int_\Gamma \log|\mathbf{x}-\mathbf{y}| \cos\theta(\mathbf{y}) dS_{\mathbf{y}} + p_0(\mathbf{x}) + \tilde{p}(\mathbf{x}), \tag{12}$$

and it will generate a velocity field that satisfies the zero normal velocity condition on the boundary.

3 Evolution of the Front

The dynamics of the chemical front is determined by the fluid flow and the self propagation. Namely, the chemical front is convected by the flow field and propagates along its normal direction with a burning velocity. To track the front as an evolving curve $\Gamma(t)$, it is sufficient to specify the normal velocity of the front, which is the combination of the local fluid velocity in the normal direction, and a front propagating velocity, the burning speed S. We can choose a parameter α for the closed curve $\Gamma(t)$, then Γ can be represented by a periodic function $\mathbf{x}(\alpha, t), 0 \leq \alpha \leq 2\pi$, for each $t > 0$. The evolution equations for $\mathbf{x}(\alpha, t)$ can be derived. As we pointed out in Section 1, computing the local normal vectors on the curve based on a discrete set of markers is a numerically difficult task. We now describe the alternative approach we use in this work that avoids this particular numerical difficulty.

3.1 The $\theta - L$ Formulation

The alternative is to follow the evolution of the tangent angle of the front at $\mathbf{x}(\alpha)$, as a function $\theta(\alpha)$, and the total arclength of the closed front $L(t)$. Under certain conditions, namely that α is an equal arclength parameter and a reference point is given, θ and L completely determines the curve Γ. To begin with, we can easily establish a transformation between $\mathbf{x}$ and the tangent angle θ, and the arclength derivative s_α. Given a parametrization $\mathbf{x}(\alpha) = (x(\alpha), y(\alpha))$, the transformation between $\mathbf{x}$ and θ, s_α is quite simple:

$$\theta(\alpha) = \tan^{-1}\left(\frac{y_\alpha}{x_\alpha}\right), \tag{13}$$

$$s_\alpha = \sqrt{x_\alpha^2 + y_\alpha^2}, \tag{14}$$

for θ and s_α in terms of $\mathbf{x}$; or

$$\begin{cases} x(\alpha, t) = x(0, t) + \int_0^\alpha \cos(\theta(\alpha', t)) s_\alpha d\alpha', \\ y(\alpha, t) = y(0, t) + \int_0^\alpha \sin(\theta(\alpha', t)) s_\alpha d\alpha', \end{cases} \tag{15}$$

for $\mathbf{x} = (x(\alpha), y(\alpha))$ in terms of θ and s_α.

Although the motion of a curve in a two-dimensional space is determined completely by its normal velocity, adding a tangential velocity to the motion will not change the motion of the curve. However, a tangential velocity along the curve can certainly change the parameterization along the curve. We want to explore this additional freedom we have to provide a better environment for a numerical spatial discretization, namely, a parameter α that is an equal arclength parameter for Γ. In our problem, the normal velocity of the curve at $\mathbf{x}(\alpha)$, $V(\alpha)$, which is taken positive for inward motion (pointing from the reactant to the product), comes from the fluid velocity in the normal direction $V_f(\alpha)$ evaluated at $\mathbf{x}(\alpha)$, and a curvature dependent burning velocity. Here we assume that the laminar burning velocity has the following form

$$S = S_L \exp(-b\kappa), \tag{16}$$

where S_L is the laminar burning velocity of the chemical reaction for a front that has a flat projection onto the plates, and it is assumed to be a constant, measured from the experiment; κ is the curvature of the curve Γ; and b is taken to be another parameter measured from the experiment. It should be pointed out that the chemical reaction surface in the Hele-Shaw cell is curved in a plane that is normal to the plates, with a typical radius of curvature proportional to w[6]. The laminar burning velocity S_L in our case has taken into consideration of this factor and it is different from the laminar burning velocity for a planar front. The reason to choose this form of the burning velocity, rather than the linear form, is that it is strictly positive. While the linear form ($S = S_L(1 - b\kappa)$) can be considered the first order approximation of the current form we use here. It is also noted that a more general flame stretch dependence of the burning velocity would include a fluid velocity strain rate effect[18]. At this stage, we will consider the curvature effects only.

Combining these two velocities,

$$V(\alpha) = V_f(\alpha) - S_L \exp(-b\kappa(\alpha)). \tag{17}$$

If we impose an artificial tangential velocity based on the normal velocity[11],

$$U(\alpha) = \int_0^\alpha \theta_\alpha V d\alpha - \frac{\alpha}{2\pi} \int_0^{2\pi} \theta_\alpha V d\alpha, \tag{18}$$

then the parameter α will remain an equal arclength parameter as long as it starts to be one, and the evolution equations for θ and L are given by

$$\theta_t = \frac{1}{s_\alpha}(V_\alpha + \theta_\alpha U) \tag{19}$$

$$L_t = -\int_0^{2\pi} \theta_\alpha V d\alpha \tag{20}$$

where $s_\alpha = L/2\pi$ is independent of the parameter α. The two equations, together with an additional evolution equation for the reference point $\mathbf{x}(0) = \mathbf{x}^0$,

$$\mathbf{x}_t^0 = -V(0)\mathbf{n}(0) = -V(0)(\sin\theta(0), -\cos\theta(0)), \tag{21}$$

completely characterized the dynamics of the front. Once θ, L and $\mathbf{x}^0$ are found, the front positions $\mathbf{x}(\alpha)$ can be easily obtained by using Eq. (15).

There are several advantages to this formulation. There will be no need to construct the local normal vector $\mathbf{n}$ at each marker point by differencing the positions of neighboring points, since it is given explicitly by the tangent angle θ. In the context of front propagation, it is particularly attractive to see that a constant term in V becomes extremely simple in this evolution system. It contributes some terms that are explicitly integrable. The additional tangential velocity makes sure that the parameter α stays an equal arclength parameter, that is, all markers are uniformly distributed along the front which makes it possible to use Discrete Fourier Transforms, and s_α equals to the mean of itself. The curvature becomes simply θ_α/s_α and the dependence of the normal velocity on the curvature can be naturally treated in the Fourier space. The usual numerical instability caused by the curvature dependence can be avoided by a simple implicit scheme. We will discuss this in the next subsection. Finally, all the derivatives and integrals can be obtained by FFT and spectral convergence will be achieved.

In analyzing the front propagation of the product region, it is often needed to calculate some geometric quantities of the region, such as the total area and the center of the mass. With the $\theta - L$ formulation, these quantities are naturally given, in terms of the solution $\mathbf{x}(\alpha, t)$, via the use of the divergence theorem. For example, given the product region Ω_0, its area can be obtained by

$$A_0 = \int_{\Omega_0} \nabla \cdot (x, 0) d\mathbf{x} = \int_{\Gamma} (x, 0) \cdot \mathbf{n} ds = s_\alpha \int_0^{2\pi} x(\alpha) \sin\theta(\alpha) d\alpha. \tag{22}$$

For the center of mass (x_c, y_c), a similar expression can be obtained. For example,

$$y_c = \frac{1}{A_0} \int_{\Omega_0} y d\mathbf{x} = \frac{1}{A_0} \int_{\Omega_0} \nabla \cdot (0, \frac{1}{2} y^2) d\mathbf{x} = -\frac{s_\alpha}{2A_0} \int_0^{2\pi} y^2(\alpha) \cos(\theta(\alpha)) d\alpha. \tag{23}$$

These integrals can be all calculated by FFT with spectral accuracies, which makes the analysis of the product region much easier.

3.2 Decomposition of the Burning Velocity

Since the variable $\theta(\alpha)$ gains a value 2π each time α is incremented by 2π, it is better to work on another variable that is a periodic function, so FFT can be conveniently used. In this case, we write

$$\theta(\alpha) = \alpha + \phi(\alpha), \quad 0 \leq \alpha \leq 2\pi, \tag{24}$$

then ϕ is a 2π-periodic function of α. In our numerical implementation that follows, we will solve for ϕ, instead of θ, which is readily available from the definition of ϕ above.

When we consider a front evolution problem involving a mean curvature flow, there is a natural numerical stability consideration. If the dependence of the burning velocity is linear, the $\theta - L$ formulation leads to a very straightforward implementation of an implicit scheme. In our current formulation, the dependence of S on κ is nonlinear. Nevertheless we can write

$$\exp(-b\kappa) = 1 - b\kappa + R(b\kappa), \tag{25}$$

where R denotes the remainder of the expansion for small values of b.

It is obvious that the linear term can cause a stability problem if it is treated by an explicit method, but the remaider can be expected to be quite small if the front is sufficiently smooth. Since $\kappa = \theta_\alpha/s_\alpha = (1+\phi_\alpha)/s_\alpha$, we have

$$V_\alpha = \frac{S_L b}{s_\alpha}\phi_{\alpha\alpha} + \frac{\partial V_f}{\partial \alpha} - S_L \frac{\partial R}{\partial \alpha}. \tag{26}$$

In this form, the first term is linear and is expected to be treated implicitly in the next section for numerical discretization.

Now the evolution equations for ϕ and s_α based on Eq. (19) and (20) become

$$\phi_t = \frac{1}{s_\alpha}\left(\frac{S_L b}{s_\alpha}\phi_{\alpha\alpha} + \frac{\partial V_f}{\partial \alpha} - S_L \frac{\partial R}{\partial \alpha} + (\phi_\alpha + 1)U\right) \tag{27}$$

$$\frac{\partial s_\alpha}{\partial t} = -\frac{1}{2\pi}\int_0^{2\pi} (\phi_\alpha + 1)\left(V_f - S_L \exp(-b\kappa)\right) d\alpha \tag{28}$$

4 Numerical Discretization

4.1 Discretization of the Boundary Integral

We discretize the closed curve $\Gamma(t)$ by a collection of markers $\{\mathbf{x}_i(t), i = 1, ..., N\}$ but do not follow the motion of these markers explicitly. Instead, the update of the marker positions is obtained by the update of θ and L via the transformation (15). For a given time, we denote $\alpha_i = 2\pi i/N$, $\theta_i = \alpha_i + \phi(\alpha_i)$, and the positions $\{\mathbf{x}_i, i = 1, ..., N\}$ are used to construct the boundary integral and evaluate the fluid velocity induced by the single layer potential. It is well known that the trapezoidal rule for the discretization of a closed boundary integral has a spectral rate of convergence as $N \to \infty$. Therefore we use it here to discretize the integral. However, the kernel is singular when $\mathbf{x} \to \mathbf{y}$ and it requires special care. Since we are taking the normal derivatives, it is a well known fact that this singularity is actually removable[19] and we can show that

$$\lim_{\mathbf{y}\to\mathbf{x}} \frac{\partial}{\partial n_{\mathbf{x}}} \log|\mathbf{x} - \mathbf{y}| = \frac{\kappa}{2} \tag{29}$$

where κ is the local curvature. Therefore, we can approximate the integral in Eq.(8) by

$$V_f(\alpha_i) = A\left[\frac{s_\alpha}{N}\sum_{j\neq i} \frac{(\mathbf{x}_i - \mathbf{x}_j)\cdot \mathbf{n}_{\mathbf{x}_i}}{|\mathbf{x}_i - \mathbf{x}_j|^2}\cos\theta_j + \frac{1}{2}\left(\frac{s_\alpha}{N}\kappa_i + 1\right)\cos\theta_i\right] \tag{30}$$

where $A = w^2 g\delta/12\nu$.

Alternatively, we can use the alternate-point quadrature method which also gives spectral accuracy in calculating the principal value integrals[20,10]. In this quadrature method, we discretize the integral for each particular $\mathbf{x}_i$ by taking the summation over alternating points, so that the singular term is skipped. There will be no need to use the limit (29) if we use the alternate-point quadrature method. For the discretization with this method, we have

$$V_f(\alpha_i) = A\left[\frac{2s_\alpha}{N}\sum_{\substack{j=0\\ j+i\ odd}}^{N-1}\frac{(\mathbf{x}_i-\mathbf{x}_j)\cdot\mathbf{n}_{\mathbf{x}_i}}{|\mathbf{x}_i-\mathbf{x}_j|^2}\cos\theta_j+\frac{1}{2}\cos\theta_i\right] \tag{31}$$

In this work, we use this alternate-point quadrature method since it appears to have a better numerical stability property in our calculations.

4.2 Front Evolution

First we take the Fourier transform of Eq.(27) to obtain

$$\widehat{\phi}_t = \frac{1}{s_\alpha}\left[-\frac{S_L b}{s_\alpha}k^2\widehat{\phi}+ik(\widehat{V_f}-S_L\widehat{R})+\widehat{\theta_\alpha U}\right], \tag{32}$$

where k is the wave number. In a time discretization, we denote $u^n = u(n\Delta t)$ to be update of u at n-th time step. The above equation can be discretized by the following first order scheme, with the leading curvature term treated implicitly,

$$\frac{\widehat{\phi}^{n+1}-\widehat{\phi}^n}{\Delta t} = -\frac{S_L b}{s_\alpha^2}k^2\widehat{\phi}^{n+1}+\frac{ik}{s_\alpha}(\widehat{V_f}^n-S_L\widehat{R}^n)+\frac{1}{s_\alpha}(\widehat{\theta_\alpha U})^n \tag{33}$$

We can solve this directly to obtain

$$\phi^{n+1} = \frac{1}{1+\Delta t\sigma(k)}\left[\widehat{\phi}^n+\frac{i\Delta tk}{s_\alpha}(\widehat{V_f}^n-S_L\widehat{R}^n)+\frac{\Delta t}{s_\alpha}(\widehat{\theta_\alpha U})^n\right], \tag{34}$$

where

$$\sigma(k) = \frac{S_L b}{s_\alpha^2}k^2. \tag{35}$$

The update of s_α is obtained by solving the ODE explicitly using the first-order Euler's method,

$$s_\alpha^{n+1} = s_\alpha^n + \Delta t\Phi^n, \tag{36}$$

where

$$\Phi^n = -\frac{1}{2\pi}\int_0^{2\pi}(\theta_\alpha V)^n d\alpha. \tag{37}$$

Here the integral is calculated by the FFT.

The time step Δt is controlled by a CFL condition. There are at least two relevant factors here: a conventional constraint that the distance a marker moves in a single time step should be less than the mesh size along the curve; and another constraint that two neighboring markers should not collapse into one after one time step, which is mostly a condition governed by the local curvature. Here we enforce the following CFL by combining these two considerations:

$$\Delta t \leq \frac{C}{V_{\max}} \min\left(\frac{2\pi s_\alpha}{N}, \frac{1}{\kappa_{\max}}\right), \tag{38}$$

where $V_{\max}$, $\kappa_{\max}$ are the maximal velocity, which is the sum of the fluid velocity and the burning velocity, and the maximal curvature along the front. A positive constant C between 0 and 1 is chosen for each calculation.

5 Results

5.1 Choices of parameters

First of all, the system can be nondimensionalized by introducing a characteristic velocity U and a characteristic length W for the cell size in the two-dimensional plane. The other physical parameters are naturally given by the problem. For Eq. (8), which contains the most of the dynamics in this problem, the terms inside the bracket remain the same forms for any scaling of the length variable, such as $\mathbf{x} = W\mathbf{x}'$, therefore the dimension of the right-hand-side of Eq. (8) is determined by the factor $A = w^2 g\delta/12\nu$. On the other hand, the characteristic velocity U on the left-hand-side is not obvious. In this case, we choose the velocity scale based on the viscosity and pick $U = 12\nu/w$. The burning velocity shall be scaled with this particular characteristic velocity too, and a characteristic time will be obtained by W/U. When we rewrite Eq. (8) as

$$u_n = -B\left[\frac{1}{2\pi}\int_\Gamma \frac{\partial}{\partial n_{\mathbf{x}}} \log|\mathbf{x}-\mathbf{y}|\cos\theta(\mathbf{y})dS_{\mathbf{y}} + \frac{1}{2}\cos\theta(\mathbf{x})\right]. \tag{39}$$

Here $\mathbf{x}$ and u_n are both nondimensionalized length and velocity variables, and $B = w^3 g\delta/(12\nu)^2$ is a dimensionless constant. In this work, we choose the physical parameters $\rho, \Delta\rho, \mu, w$ to reflect the experiment setup as closely as possible. In the experiment that we try to model here, the box size is $20 \times 30 \times 0.2$ cm^3 and the length scales are naturally chosen such that $W = 20$ cm and $w = 0.2$ cm. With these choices of the physical parameters, combined with the physical properties of the water for the fluid considered, such as the fluid density and viscosity, we arrive at the nondimensional number $B \approx 0.11$.

5.2 The Exact Solution for an Expanding Circle

Here we show that an analytic solution exists for the initially circular front. As expected, this circular front will expand while it remains circular, and the center of the circle shall move up. Motivated by this speculation, we look for a solution in this form:

$$\mathbf{x}(\alpha,t) = (0, y_c(t)) + r(t)(\cos(\alpha), \sin(\alpha)), \quad 0 \le \alpha \le 2\pi, \ t \ge 0, \tag{40}$$

with the initial condition $y_c(0) = y_c^0$ and $r(0) = r_0$. For the fluid velocity in the normal direction on the front, we can find from Eq. (39),

$$u_n(\mathbf{x}(\alpha,t)) = \frac{B}{2}\sin\alpha, \quad 0 \le \alpha \le 2\pi, \tag{41}$$

since the boundary integral vanishes identically for a circle. This normal velocity is obviously the projection of a constant upward velocity $B/2$ onto the normal directions. Therefore, in the case that there is no front propagation, the fluid motion corresponds to a rigid body translation in the upward direction with a velocity $B/2$. It is important to note that this velocity is independent of the size of the front, as long as the front remains circular. This suggests that the front propagation part can be decoupled from the translation due to the gravity effects. So we conclude

$$y_c(t) = y_c^0 + \frac{B}{2}t. \tag{42}$$

If we add the front propagation part with the burning velocity $S = S_L \exp(-b\kappa)$ to the system, we can obtain $r(t)$ independently by solving the following ordinary differential equation,

$$\frac{dr(t)}{dt} = S_L \exp(-\frac{b}{r(t)}), \quad r(0) = r_0 > 0. \tag{43}$$

A numerical solution for this equation can be easily obtained.

5.3 Small Perturbation of the Circle

When the circular front is initially perturbed, a natural question to ask is whether the circular front is stable subject to such a perturbation. In a physically two-dimensional flow, the buoyancy effect is responsible for the Rayleigh-Taylor instability such that a horizontal fluid interface with heavier fluid on the top will be unstable. However, the density difference in our case is so small, and there is an additional front propagation effect coupled to the fluid motion. It is not so clear as what to expect from such a perturbation of the circular front. The competing mechanisms in this model are the density difference, laminar burning velocity and the curvature dependence of the burning velocity. Our numerical studies will be a useful tool to explore the relationships among these parameters, and to analyze the wrinkling process of almost circular fronts.

First, we demonstrate by a numerical convergence study the fingering phenomenon arising from this model system. Since many Hele-Shaw flows are ill-posed, it is very important in a numerical calculation to distinguish the physical instability of the model from possible numerical instabilities. In our first example, we choose the initial front to be a small circle, perturbed by some small disturbance. To be more specific, we choose the initial front to have the form

$$r(\alpha) = r_0 + \epsilon(\cos(2\alpha) + \sin(3\alpha)) \quad 0 \le \alpha \le 2\pi, \tag{44}$$

and the center of the unperturbed circle is at $(0.5, 0.5)$. This parametrization does not have the equal arclength property. We use the procedure developed by Baker and Shelley[21] here to rearrange the points so that the new parameter is an equal arclength parameter.

To test the numerical convergence for this case, we fix the parameters $r_0 = 0.05$ and $\epsilon = 0.001$, and run the calculations starting with 128, 256, 512 and 1024 points along the front. We double the number of points once the perimeter is doubled. This is achieved easily by expanding the number of Fourier modes, setting those high frequency coefficients to be zero when we add points and let them grow in time. The role of the curvature smoothing is very critical to the fingering phenomenon. If there is no curvature dependence of S, it is well known that concave propagating fronts will lead to cusps[22]. In the experiments we try to model here, cusps are not observed and the curvature dependence of S is always present. However, for a certain numerical discretization, we can only resolve the dependence of S on the curvature above certain level. If the dependence is too weak, it will result in a numerical singularity which manifests as the appearance of an infinite local curvature somewhere along the front. On the other hand, we can show later that if the curvature dependence is too strong, the fronts are stable with respect to small perturbation and the fingering phenomenon is not observed. For the purpose of numerical convergence, we fix the parameter $S_L = 0.01$ and $b = 0.005$ in this particular case. Note that this particular dimensionless burning velocity roughly corresponds to a burning velocity 0.005 cm/sec in an actual experiment, which is the laminar burning velocity measured in a particular experiment[5] that we try to model here. In Fig. 3, the fronts at $t = 5$ from four different calculations with different numbers of grid points are shown. It is obvious that the results do converge to certain wrinkled fronts as N increases. This convergence study confirms that the fingering phenomena we observe are indeed physical, rather than numerical artifacts.

Given that the fingering phenomenon we observed is truly a physical phenomenon, we proceed to analyze the front dynamics for this particular case. In Fig. 4, we show the evolving fronts from our calculation for the flow with the above parameters and starting with $N = 512$ points. Notice that by $t = 5$, the fingering phenomenon along the upper part of the front is obvious, while the lower part remains smooth. This preference can be interpreted as an analogue of the Rayleigh-Taylor instability, where the upper part corresponds to the unstable mode, since the heavy (unreacted) fluid is above the light (reacted) fluid. The lower part corresponds to the stable mode, therefore it remains smooth. This phenomenon is persistent throughout our numerical investigation and is also consistently observed in the experiments. In Fig. 4(b), the detailed finger development is shown for the snapshots between $t = 5$ and $t = 5.5$. The development here is very similar to other Hele-Shaw flow calculations[10,11] and our numerical grid refinement has confirmed these fronts. It is noted that the "valley" part does not merge quickly, as expected by the burning process. This is because that the heavy fluid is "trapped" in the valley and the fluid velocity is quite large there, compared with the small burning velocity S_L.

One of the consequences of the wrinkling front is that the effective propagation rate would be increased, compared to the propagation rate of the circular front. To show this effect, we calculate the areas of the product region at $t = 5.5$ for both the case of the initially perturbed front (the above case) A_1, and the case of the exact circular front A_2. Here the solution for A_2, which involves the curvature effect, is obtained by solving the ordinary differential equation (43) numerically. In Fig. 5(a), we show the difference $(A_1 - A_2)$ between these two areas as a function of time t. It is very interesting to see that the difference takes off as the fingers develop. This shows that the buoyancy effects indeed is an important factor in the estimate of the effective propagation rate. Another quantity of interest is the center of mass of the product region. In Fig. 5(b), we show the difference between the center of mass of the wrinkled product region (y_c) and that of the circular region ($y_c^0 + Bt/2$). As it shows, the center of mass moves up faster in the wrinkled case. In Fig. 6(a), we show the curvature distribution of the front at $t = 0, 2, 4, 5$ and 5.5. It appears that the curvature stays bounded before a certain time, then suddenly it develops a structure with large magnitudes which corresponds to the fingers we observe. In Fig. 6(b), we plot the magnitudes of the first 50 Fourier coefficients of the function ϕ in complex form at different times. Given

$$\phi(\alpha) = \sum_{k=0}^{N} \left(a_k \cos(k\alpha) + b_k \sin(k\alpha)\right),$$

we plot $A(k) = \sqrt{a_k + b_k}$ for each $k \leq 50$. We see that initially the disturbance is concentrated on the lowest mode and then develops to a few higher modes. At $t = 5.5$, the majority of the disturbance is still spread over about 20 different frequency modes. This suggests that our calculations in the Fourier space is quite effective.

To convince ourselves that the particular fingering shape is not caused by our particular choice of the initial data, we turned the initial front upside down. The results are show in Fig. 7, with the detailed part of the fingering development shown in Fig. 7(b). It is striking that the fingers here are so similar to those in Fig. 4, except that they are developed in an earlier time (around $t = 4$) than in the previous case (around $t = 5$). Again, the fingering happens only at the upper part, with the lowering part remaining smooth. In other calculations not shown here, we introduce some perturbations that cover a larger frequency range, with amplitudes for different modes selected randomly. The resulting finger patterns remain similar to the above two cases.

In the above calculations, boundary effects are ignored. When we apply the boundary correction, we are expected to add more perturbations to the front and the fingering phenomenon is expected to appear earlier. Our calculations confirm this prediction. In Fig. 8(a), we show the fronts at $t = 0, 1, 2$ and 2.3. By the time $t = 2$, we see that the fingers are already so pronounced and the detailed fronts in Fig. 8(b) show the similar Hele-Shaw fronts. It seems that the boundary effect does not add much to the scales of the developing fingers and can be considered as another perturbation effect to the evolving fronts.

The main question we ask here is the dominant factor that determines the scales of the developing fingers observed. In this model where the heat loss is not considered, it seems that the curvature dependence is one factor here. In all the calculations shown above, we have kept $b = 0.005$ and we have consistently observed four fingers in these calculations. Now we reduce the curvature dependence by letting $b = 0.0025$ and keeping all the other parameters. The results are shown in Fig. 9. It is not surprising that the scales of the fingers are reduced. In this case, doubling the number of points does not change the results and we are convinced that a weaker curvature dependence does result in a smaller scale of the fingers. Conversely, if we increase the curvature dependence to be strong enough, results show that the fingers will never be developed.

The competing mechanism between the fluid motion and the front propagation is probably the most important factor in the finger wavelength selection. The fluid motion is scaled by the parameter A and the propagation is controlled by the dimensional S_L. The relative competition is therefore determined by the ratio S_L/A. To study the competing mechanism, it is sufficient to fix A and vary the parameter S_L. In the calculations shown above, a dimensionless (scaled with respect to U) burning velocity $S_L = 0.01$ is used. In the next calculation, we increase S_L to 0.02 and keep all the other parameters as in the calculation of Fig. 4. It turns out that this leads to a stable situation where no fingers are found by the time $t = 5.5$ and the front has become almost circular. It would be natural to expect that reducing S_L would make it easier for fingers to develop. In Fig. 10, we show the evolving front for $S_L = 0.005$ at different times. It is noted that the fingers appear earlier than that of the case in Fig. 4, and the tips are not as smooth as those in Fig. 4. However, the wavelength of the fingers are comparable with Fig. 4. This suggests that the laminar burning velocity S_L has an effect that smooths out the Rayleigh-Taylor instability due to its curvature dependence. However, the fingering wavelength selection is determined by the curvature dependence only, at least in this model.

6 Conclusions

We have derived a simplified system for the chemical front propagation in a Hele-Shaw flow. This model uses Darcy's law and takes advantage of the small density change across the front that is observed in the experiments. This leads to a free boundary problem that can be represented by a boundary integral formulation. We use the $\theta - L$ formulation to evolve the front which gives high spatial accuracy in numerical solutions. In this model, we are able to emphasize the buoyancy effects related to a very small density change and explain the fingering phenomenon observed in experiments. The competing effects of the fluid motion, burning velocity and the curvature dependence of this burning velocity are explored with our numerical solutions. It is found that the level of curvature dependence of the burning velocity determines the wavelength of the fingers, and the laminar burning velocity S_L itself has an effect that controls the growth of the fingers.

A very important factor ignored in this system is the heat loss effect of the chemical front, where the density of the product will resume its previous value in some time after the reaction. In an ongoing project, we are studying these effects by solving the Euler-Darcy equations for the fluid and a heat loss equation for the temperature. With this model, we hope that we have a system that models the experiments quite accurately, and at the same time reasonable to solve numerically. This will allow us an opportunity to deepen our understandings of the chemical front propagation.

ACKNOWLEDGEMENTS

This work is motivated by Prof. P. Ronney's recent experiment and the author is extremely grateful to his suggestions, explanations and many discussions we had. The author also wish to thank T.Y. Hou for helpful discussions, and N. Beebe and D. Eyre for their help with the graphics. The use of FFT, redistribution codes developed by T. Y. Hou, J. Lowengrub and M. J. Shelley is also acknowledged. The calculations were performed at the center for scientific computing at the Department of Mathematics, University of Utah.

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Figure Captions

1. A snapshot of a chemical front in the experiment (photo courtesy of M. Abid and P. Ronney, University of Southern California).

2. An illustration showing the reactant and product regions and the $\theta - L$ formulation for the front.

3. A numerical convergence study: comparisons of the fronts at $t = 5.5$ with different numbers of grid points. From left to right, top to bottom: starting numbers are $N = 128, 256, 512$ and 1024.

4. Time evolution of a chemical front with an initially perturbed circular product region. (a). from $t = 0$ to $t = 5.5$; (b) from $t = 5$ to $t = 5.5$.

5. Comparisons of the area and center of mass with the exact solution for the unperturbed circular front.

6. (a). The curvature distribution of the chemical front and (b). the frequency distribution of the tangent angle θ.

7. Time evolution of a chemical front with the initial front from Fig. 4 turned upside down. (a). from $t = 0$ to $t = 4.5$; (b). from $t = 4$ to $t = 4.5$.

8. Time evolution of a chemical front with the same initial condition as in Fig. 4 but with a boundary correction. (a). from $t = 0$ to $t = 2.3$; (b). from $t = 1.9$ to $t = 2.3$.

9. Time evolution of a chemical front, with a weaker curvature dependence of the burning velocity ($b = 0.0025$). (a). from $t = 0$ to $t = 3.7$; (b). from $t = 3.3$ to $t = 3.7$.

10. Time evolution of a chemical front, with a smaller laminar burning velocity $S_L = 0.005$. (a). from $t = 0$ to $t = 3.3$; (b). from $t = 3$ to $t = 3.3$.