Fingering instabilities in adhesive failure

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Abstract

We study the fingering instabilities that occur in the debonding of model adhesives. We perform a linear analysis of the instability in the lifting Hele-Shaw cell, relevant for the failure of soft adhesives. The analysis is done for a Newtonian fluid but can be extended to shear-thinning fluids without any problem. When compared to experiment, significant differences are found between the linear theory and the experimental results. We therefore, discuss three-dimensional effects, that are found to improve the agreement; nevertheless significant differences between theory and experiment persist.

Keywords: Adhesion; Instabilities; Viscous fingering

1. Introduction

Thin layers of adhesives are used in a large number of applications to bond surfaces together. Besides the adhesion of the fluid to the solid substrate itself, the bulk rheology of the adhesive is an important factor for instance for the dissipation in debonding processes. We focus here on the debonding process, in which air enters in the gap between two plates that are separated at constant speed, and at time \( t = 0 \) the gap is filled by the adhesive. Since the adhesive is much more viscous than air, the debonding leads automatically to the Saffman–Taylor instability: the boundary of the blob, assumed initially circular, may develop a large number of oscillations due to the occurrence of viscous fingering. As time goes on, their number decreases but the amplitude of the undulation grows, so that the oscillations of the interface transform into fingers. Here we propose a linear analysis of this instability for model “adhesives”. In order to avoid the discussion on the often complicated visco-elastic properties of adhesives, we study a model situation in which the adhesive is assumed either Newtonian or exhibits the simplest non-Newtonian rheology: a power-law viscosity. This is an oversimplification of the adhesion problem, since, most
adhesives are polymer-based, strongly non-Newtonian fluids. They exhibit several rather complicated properties together like yield stresses, normal stresses, non-local and time dependent viscosities, etc. However, it is a good starting point for the study of the fingering instabilities that may happen during debonding of adhesives.

The understanding of the dynamics of the air/fluid interface in the Newtonian case, which is not trivial, is a necessary first step to go further and understand the behaviour of adhesives. The experimental situation in debonding is similar to that of the classical fingering instability [5,6] which is rather well documented now (see for example [7–9]), in spite of the fact that the complete understanding of the dynamics in radial Hele-Shaw cell remains a challenge both analytically and numerically [10–12]. Specifically the understanding of the time-dependent patterns in this case remains a problem. Moreover, numerical treatments require complicated algorithms [13,14] and lengthy calculations. Most studies of radial fingering concern the divergent flow when air pushes a more viscous liquid from the center. However, convergent flows, similar to what happens in debonding, have also been studied a decade ago in the sector geometry [15,16]. For the lifting radial Hele-Shaw cell, the most important difference is that the flow is not Laplacian but satisfies a Poisson equation. This difference will appear within an analytical linear treatment which is explained hereafter. All studies so far ignore three-dimensional effects and assume validity of Darcy’s law combined with 2D boundary conditions [1]. Valid at short times, the predictions of the linear analysis, deduced from the model and compared to experimental data is a test of these assumptions for this experimental set-up. The comparison shows that the 2D model overestimates the number of fingers even at times $t \approx 0$. We therefore, perform a detailed treatment of the meniscus shape which suggests that strong 3D effect are expected. We consider these here also.

2. A viscous blob in contraction

We consider the adhesive as a viscous blob of viscosity $\mu_1$, surrounded by a less viscous fluid of viscosity $\mu_2$. The adhesive is confined between two rigid parallel walls separated by a small distance $B$ (see Fig. 1). We assume that one plate of this Hele-Shaw cell can be lifted so that the cell gap $B(t)$ is a function of time, in a manner that can be fixed by the experimentalist. Although $B(t)$ is an increasing function of time, the spacing is assumed to remain small at all times compared to the radius of the blob. As a first step, we consider two Newtonian fluids (adhesive and air); in a second analysis we will consider the problem when the adhesive is non-Newtonian. If the experiment starts with a large blob of fluid, we find that the number of unstable modes at $t = 0$ may be of the order of a hundred. This large number of modes subsequently decreases slowly in time while the, initially small, amplitude increases. This indicates that a linear treatment should be correct in principle for short times after the start of the experiment.

Fig. 1. Schematic of the experimental setup.
2.1. Mathematical formulation

The patterns occurring in debonding are free-boundary problems: the interface position is an unknown which is part of the dynamics. We are therefore, faced with a three-dimensional free boundary problem controlled by Stokes flow; this class of problems cannot be solved except in very peculiar cases and simple geometries, and without instabilities occurring. We will therefore, have to consider the simpler two-dimensional situation, and discuss corrections due to the three-dimensional nature of the problem later. We first recall the equations for the fluid flow, which are valid everywhere in the bulk and discuss the boundary conditions.

2.1.1. Hydrodynamic flow for a Newtonian fluid

Why is there an instability? The lifting of the upper plate results in a lateral stretching flow, sucking the interface inwards. The classical Saffman–Taylor instability [1] is expected to occur as soon as the blob is made of the more viscous fluid, i.e., \( \mu_1 > \mu_2 \), at least when both fluids are Newtonian. Let us write the governing equations for this modified Hele-Shaw problem, and subsequently introduce dimensionless units for the mathematical formulation of the instability problem. We consider that the viscous blob occupies a surface area \( \Omega(t) \), limited by the interface \( \partial \Omega(t) \), when \( \vec{r} \) is the two-dimensional coordinate \( \vec{r} = (x, y) \).

Assuming that the Darcy’s law remains valid, as in the Saffman–Taylor fingering, the two-dimensional horizontal fluid velocities are given by:

\[
\vec{V}_{1,2}(\vec{r}, t) = -\frac{B(t)^2}{12 \mu_{1,2}} \nabla P_{1,2}(\vec{r}, t) \quad \text{in} \quad \Omega(t) \quad (1)
\]

(Index 1 or 2 are for different fluids).

The validity of Darcy’s law relies on a number of assumptions: the Stokes approximation for the flow (zero Reynolds number limit), the fact that the pressure does not depend on the vertical coordinate \( z \) and the small gap limit \( B(t) \ll A_0 \).

Due to mass conservation, the surface area \( \Omega(t) \) decreases. Because the gap grows, there exists a vertical component of the fluid velocity, contrary to what happens for the classical Saffman–Taylor viscous fingering experiment. The vertical velocity of the fluid \( U_{1,2} \) is simply given by:

\[
U_{1,2}(\vec{r}, z, t) = \frac{B(t)}{B(t)} \frac{\dot{B}(t)}{B(t)} z \quad \text{in} \quad \Omega(t) \quad (2)
\]

which yields the modified incompressibility equation:

\[
\nabla^2 \vec{V}_{1,2}(\vec{r}, t) = -\frac{\dot{B}(t)}{B(t)} (3)
\]

(\( \nabla_2 \) is the two-dimensional horizontal gradient, the dot signifies a derivative with respect to time.)

Combining Eqs. (1) and (3) gives a Poisson equation for the pressure field. Before discussing the boundary conditions at the interface, we consider the more complicated case where the blob is made of a power-law fluid, i.e., the fluid viscosity has a power-law dependence on the shear rate (the velocity gradient) in the fluid.

2.1.2. Hydrodynamic flow in a power-law fluid

Our aim is to establish the correct Darcy’s law for these fluids, taking into account the advantage of the lubrication approximation. Our derivation is exact in this context, which means we do not need to introduce new assumptions to describe the flow. In the limit \( \epsilon = B(t)/A_0 \ll 1 \) which pertains to our problem, the dominant shear rate is \( \dot{\gamma} = \frac{v_1}{A_0} \) being any component of the velocity in the blob. A naive estimation of the shear rate is given by the magnitude of the velocity divided by \( B(t) \). The shear rate evaluated with the horizontal velocity is then larger than the elongation rate, which is simply given by \( \dot{B}(t)/B(t) \) since the ratio between them is of order \( 1/\epsilon \), which is large in our problem.
If we define dimensionless variables and choose the initial radius of the blob \( A_0 \) as length units, the unit of time is given by the ratio \( B(t)/B(0) \) at time \( t = 0 \). These units are the same for the Newtonian and the non-Newtonian case. We assume a circular geometry: this is true in case where the contraction occurs without instabilities but remains a very reasonable assumption when the instabilities start to grow. Then, the viscosity of the power-law fluid is given by \( \mu_1 = \mu_0 |\varepsilon|^{\alpha_a} \), \( \nu_1 \) being the dimensionless radial velocity: \( \nu_1, \varepsilon = \frac{\tau}{\nu_1} \), the unit of shear being given by \( B_0/B_0 \); the exponent \( \alpha \) of shear thinning (or thickening, depending on the sign of \( a \)) for the power-fluid can be measured independently by rheometry. This model is of course not applicable for very small or very high shear [17].

Since the gap between the two plates is small and the flow rather slow, we use the Stokes approximation. Due to the power-law character of the fluid, the Stokes approximation in the lubrication limit does not guarantee that the pressure is \( z \)-independent as is the case for a Newtonian fluid. Comparing the different components of the Stokes equation, it becomes evident that the \( z \) component of the pressure gradient is smaller than the horizontal component by a factor \( \varepsilon \). Therefore, we neglect the \( z \) dependence of the pressure. We scale the resulting pressure by: \( 12|A_0|B_0|/B_0 \). Finally, to leading order in \( \varepsilon \) the Stokes equation gives for the radial component of the velocity:

\[
\frac{\partial}{\partial r} \left( \frac{\partial a_r^\prime}{\partial r} \right) - \frac{\partial a_r}{\partial r} = 12 \frac{\partial P(r,t)}{\partial r} \quad (4)
\]

We choose: \( u_r = f(r)g(z) \) for the horizontal velocity which greatly simplifies the previous PDE and we find for \( g(z) \):

\[
g(z) = \left| z - \frac{(b(t)/2)^{a}}{(b(t)/2)^{a}} \right|^2 \quad (5)
\]

with \( a = (2 + \alpha)/(a + 1) \) and \( b(t) \) the dimensionless thickness of the cell. Note that \( g(z) \) vanishes on both plates, a necessary boundary condition for viscous flow. Then, we derive for \( f(r) \)

\[
\frac{\partial}{\partial r} \left( \frac{\partial f(r)}{\partial r} \right) = \frac{12}{a^2+1} \frac{\partial P(r,t)}{\partial r} \quad (6)
\]

The derivative of the velocity is not continuous at the center-line of the cell but the relevant physical quantity is: the viscous shear stress is continuous and can be derived indefinitely. Focusing on the averaged value of the radial velocity, as for the standard Darcy’s law: \( V_{r}(r,t) = \frac{1}{b(t)} \int_0^{b(t)} u_r \, d\zeta \), we get a modified Darcy’s law:

\[
V_{r}(r,t) = -\frac{N b(t)^{\alpha}}{b(t) P(r,t)^{\alpha+1}} \frac{\partial P(r,t)}{\partial r} \quad (7)
\]

with \( N = (12)^{\frac{1}{\alpha+1}} (2a+1) \). We recover the Newtonian case with \( \alpha = 0 \) and \( \alpha = 2 \). Rigorously speaking, this law applies only in the radial direction. We can extend this relation when the radial symmetry is broken. If the orthogonal velocity remains small compared to the radial velocity, a calculation without difficulty shows that the orthogonal velocity satisfies a similar relationship so that finally, within the lubrication approximation we obtain the vential Darcy’s law:

\[
\tilde{V}_{r}(r,t) = -\frac{N b(t)^{\alpha}}{b(t) P(r,t)^{\alpha+1}} \nabla P(r,t) \sim -\frac{N b(t)^{\alpha}}{[\nabla P(r,t)]^{\alpha+1}} \nabla P(r,t) \quad (8)
\]

but with the restriction \( |V_{\theta}| \ll |V_{r}| \) or \( |\partial P(r,\theta,t)/\partial \theta| \ll |\partial P(r,\theta,t)/\partial r| \). This restriction is satisfied without any problem at the beginning of the lifing.

The modified Darcy’s law obtained above has been tested experimentally in some detail for Saffman–Taylor fingering, in a linear Hele–Shaw cell with various power-law fluids ([17]). A rather good agreement was found between the theoretical predictions deduced from this law [18,19] and the experimental results. A much more sophisticated treatment has been achieved in [20,21] which recovers this power law behaviour in a specific limit. Then, combining the modified Darcy’s law with the conservation of mass given by Eq. (3), we derive a p-Poisson
type equation for the pressure field which simplifies into a Poisson equation for a Newtonian fluid. Therefore, we have:

\[
\nabla \frac{1}{|\mathbf{y}|} \nabla P(r, t) = \frac{\dot{b}(t)}{Nb(t)^3(1+\alpha)} \nabla P(r, t)
\]

(9)

Although the remaining part of the paper is mostly devoted to the Newtonian case, we stress here that the more realistic non-Newtonian analysis can be obtained using the exact same strategy.

2.2. Interfacial relations

2.2.1. Interfacial laws assuming cylindrical symmetry

To discuss the interfacial relations, three-dimensional effects are neglected and we use the interfacial laws established by Saffman and Taylor [1] for viscous fingering: continuity of the normal averaged horizontal velocity and a two-dimensional Laplace equation for the pressure. These interfacial conditions have been successfully tested experimentally in the Hele-Shaw cell with a constant gap

\[
B(t) = B_0 .
\]

A numerical treatment for the lifting Hele-Shaw cell, using the classical boundary conditions has been performed in the case of a liquid/gas interface by Shelley et al. [13]. In the linear Hele-Shaw cell, departure from the continuity of velocity and Laplace’s equation have been analyzed theoretically in the small capillary number limit, yielding corrections in

\[
C_2/3 \quad [22,23] .
\]

These corrections are due to the existence of a thin film of variable thickness which separates the viscous finger from the plates. In adhesion, the geometry is different and the existence of such films has not been demonstrated; therefore, it is likely that the full three-dimensional corrections have to be taken into account. The three-dimensional effects are due to the quasi “catenoid” shape of the blob with a radius at \( z = \frac{b}{2} \) smaller than that at \( z = 0 \) or \( z = b \). This will greatly affect the two-dimensional Laplace law for the pressure since the transverse curvature has a sign opposite to the two-dimensional curvature.

The classical boundary conditions read:

\[
\begin{align*}
\vec{n} \cdot \dot{\mathbf{V}}_1 &= \Omega \mathbf{n} = -N \frac{b(t)^2}{|V_1|^2} \frac{\partial P_1}{\partial n} \quad \text{for } r = R(\theta, t) \\
\vec{n} \cdot \dot{\mathbf{V}}_2 &= -\frac{b(t)^2}{\eta} \frac{\partial P_2}{\partial n} \quad \text{for } r = R(\theta, t)
\end{align*}
\]

(10)

\( \eta = \mu_2/\mu_0 \) being the ratio between viscosity of the outer fluid (assumed viscous here) and the inner fluid whose viscosity \( \mu_0 \) has been chosen as unit. \( \dot{\mathbf{V}}_1 \) is the normal velocity of the boundary.

The Young–Laplace law gives the jump of the pressure:

\[
P_1 - P_2 = T \kappa \quad \text{for } r = R(\theta, t)
\]

(11)

with \( T \) the control parameter, proportional to the surface tension:

\[
T = \frac{B_0 \gamma}{(12A_0\mu[\partial R_0])}
\]

and \( \kappa \) denotes the local 2D-curvature. The curvature is chosen positive for convex curves, by convention. So, for a circular blob of radius \( R(t) \), \( \kappa \) is \( 1/R(t) \). Since Laplace’s law in 3D is related to the mean curvature, the 2D approximation over-estimates the pressure jump at the interface.

2.2.2. Shape of the viscous blob, a model to improve the boundary conditions

To improve these boundary conditions, one has to take into account the shape of the blob, deduce the curvature of the interface and properly average it in order to maintain a 2D flow approach. As already mentioned, it is impossible to solve the full 3D free boundary problem of an interface for Stokes flow; consequently we do not know the exact shape of the interface. The usual assumption in hydrodynamics is to employ a no-slip boundary condition on the
plates: $z = b(t)$ or $z = 0$, the tangential component of the flow vanishes so in principle the border cannot move. Of course, the dynamics of the border also depends on the wetting properties of the fluid on the substrate. This complicates the problem tremendously. We will assume here that the viscous blob has the property to completely wet the plates so that a film remains behind as the boundary moves; we assume that this follows Tanner’s law. Nevertheless, the problem remains complicated because it involves the moving contact line: we call its radius $R_c(t)$.

In principle, we have to compare the velocity of the contact line with the velocity of lifting. To simplify the problem, it seems reasonable to assume that the mobility of the interface is controlled mostly by the radial component of the velocity which gives for any $z$ inside the cell for: $0 < z < b(t)$:

$$R(\theta, z, t) = R_c(t) + (R_c(t) - R(\theta, t)) \left[ \frac{2z}{R(t)} - 1 \right]$$

where $R(\theta, t)$ is the smallest radius, at the middle of the patch. It is related to the average radius of the blob by the relation: $R(\theta, t) = R[1 + (R_c - R)/R]$ which ensures mass conservation. This relation is suggested by the existence of a 3D Poiseuille flow, at the origin of the Darcy’s approximation for $\alpha = 2$. It turns out that simple analytical results can be derived only for the case $\alpha = 0$, which is the Newtonian case, so we restrict ourselves to Newtonian fluids, hereafter.

First we derive the complete expression for the mean curvature when we use the cylindrical coordinates for the writing of the surface equation: $R(\theta; z, t)$ (hereafter, to simplify the notations, we will drop the arguments of $\tilde{R}$):

$$K^{1/2} = -\frac{\partial^2 \tilde{R}}{\partial \theta^2} \left[ R^2 + \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2 \right] - 2 \frac{\partial \tilde{R}}{\partial \theta} \frac{\partial^2 \tilde{R}}{\partial \theta \partial \theta} \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2$$

$$+ \left( R^2 + 2 \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2 - \frac{\partial^2 \tilde{R}}{\partial \theta^2} \right) \left[ 1 + \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2 \right]$$

with

$$D = R^2 \left[ 1 + \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2 \right] + \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2$$

We drop all quadratic terms which are derivatives with respect of the angle $\theta$ (in $(\partial \tilde{R}/\partial \theta)^2$) because we restrict ourselves to small modulations of the boundary. In this case, Eq. (13) simplifies to:

$$\kappa = -\frac{\partial^2 \tilde{R}}{\partial \theta^2} \left[ R^2 + \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2 \right] + \left[ 1 - \frac{1}{R} \frac{\partial^2 \tilde{R}}{\partial \theta^2} \right] \left[ 1 + \left( \frac{\partial \tilde{R}}{\partial \theta} \right)^2 \right]^{1/2}$$

We consider the average of $\tilde{K}$: $\kappa = 1/b(t)$ \(\tilde{K}\). We need to discuss the mobility of the contact line which in any case is much smaller that the center of the blob. Either the contact line is completely pinned, so that it does not exhibit viscous instability or the contact line has the same instabilities as the free surface. The result between these two cases is slightly different, and it turns out that the comparison with the experiment is in favour of the second hypothesis. Let us first calculate the case where the contact line remains circular, whatever the amplitude of the instabilities.

We define intermediate quantities in order to simplify the notations: $\rho = 4(R_c - R)/b(t)$, $\rho_1 = R_c/R - 1 - \rho^2$, $u_1 = \rho/\sqrt{1 + \rho^2}$. If $\rho_1$ is positive, the average is given by

$$\kappa(t) = -\frac{2}{b(t)} u_1 + \frac{1}{\sqrt{\rho_1}} \sqrt{\pi} \tanh \sqrt{\pi} \sqrt{1 + \rho^2} \left[ 1 - \frac{1}{2} \frac{\partial^2 \tilde{R}}{\partial \theta^2} \left( 1 + \frac{\rho^2}{\rho_1} \right) \right]$$

$$- \frac{1}{2} \frac{\partial^2 \tilde{R}}{\partial \theta^2} \sqrt{1 + \rho^2} \frac{\rho^2}{\rho_1}$$
If $\rho_1$ is negative:

$$\kappa = -\frac{2}{b(t)}u_1 + \frac{1}{R\sqrt{\rho_1}} \text{Atanh}\left(\frac{\sqrt{-\rho_1}}{\sqrt{1 + \rho^2}}\right) \left[ 1 - \frac{1}{2K} \frac{\partial^2 \hat{K}}{\partial \theta^2} \left( 1 - \frac{1 + \rho^2}{\rho_1} \right) \right] - \frac{1}{2K} \frac{\partial^2 \hat{K}}{\partial \theta^2} \frac{1 + \rho^2}{\rho_1}$$

(17)

Depending on the experimental conditions, all values for $\rho_1$ and therefore $\rho_1$ are possible and need to be controlled if quantitative comparisons have to be made. Even at short times in the experiment $\rho_1$ can be small, finite or large: $(R_c - R(t))$ which is always smaller than $(1 - R(t))$ is small, but also $b(t)$ varies. We consider two different limits here: small and large $\rho_1$.

If $\rho$ is small, taking into account Eq. (16), we obtain, to leading order:

$$\kappa \sim -\frac{8(R_c - R(t))}{b(t)^2} + \frac{1}{R} \left[ 1 - \frac{1}{2K} \frac{\partial^2 \hat{K}}{\partial \theta^2} \right]$$

(18)

If $\rho$ is large on the contrary, we get:

$$\kappa \sim -\frac{2}{b(t)} \left( 1 - \frac{1}{\rho_1^2} \right) + \frac{1}{\rho_1} \log(\rho) \left[ 1 - \frac{1}{2K} \frac{\partial^2 \hat{K}}{\partial \theta^2} \right]$$

(19)

None of the two equations Eqs. (18) and (19) recover the 2D case. The contribution of the instability $(\frac{\partial^2 \hat{K}}{\partial \theta^2})$ seems to be reduced by a factor 2 if we compare Eq. (18) to Eq. (15) with $\frac{\partial \hat{K}}{\partial z} = 0$. This is due to the approximation of a completely pinned contact line $R_c = 1$. At $z = 0$, $b(t)$. The averaging reduces this effect. On the contrary, if one assumes that the contact line evolves in the same way as the middle line and destabilises also in the same way, so $\frac{\partial^2 \hat{K}}{\partial \theta^2} / \hat{R} = \frac{\partial^2 \hat{K}}{\partial \theta^2} / R$, the average of the curvature is simplified and gives for $\rho_1$ positive:

$$\kappa(t) = -\frac{2}{b(t)}u_1 + \frac{1}{R\sqrt{\rho_1}} \text{Atanh}\left(\frac{\sqrt{-\rho_1}}{\sqrt{1 + \rho^2}}\right) \left[ 1 - \frac{1}{2K} \frac{\partial^2 \hat{K}}{\partial \theta^2} \left( 1 - \frac{1 + \rho^2}{\rho_1} \right) \right]$$

(20)

and for negative $\rho_1$:

$$\kappa = -\frac{2}{b(t)}u_1 + \frac{1}{R\sqrt{\rho_1}} \text{Atanh}\left(\frac{\sqrt{-\rho_1}}{\sqrt{1 + \rho^2}}\right) \left[ 1 - \frac{1}{2K} \frac{\partial^2 \hat{K}}{\partial \theta^2} \left( 1 - \frac{1 + \rho^2}{\rho_1} \right) \right]$$

(21)

It is not sure that this description is better than that for the quasi-static contact line. It seems likely that the truth is between the two solutions. We will therefore use these different results for $\kappa(t)$ in our comparison with the experiment.

2.3. Self-similar regular collapsing solution

Contrary to what happens for the classical radial Hele-Shaw flow which has been examined in the same context by [5,6], the pressure here is no longer Laplacian since it satisfies a Poisson equation. For both Newtonian ($a=0$) and non-Newtonian fluids, we have to solve Eq. (9). The outer fluid (2) being assumed Newtonian, Eq. (9) simplifies to:

$$\tau_{D,t} = \frac{\eta b(t)}{2} \frac{\partial^2 \hat{K}}{\partial \theta^2}$$

(22)

The base state $\hat{P}^1_0$, $\hat{P}^2_0$, $\hat{R}(t)$ is the hydrodynamic state of reference with a boundary: $\hat{R}(t)$ that contracts while always remaining circular: $r = \hat{R}(t)$. At time $t = 0$, it coincides with the situation we examine. An obvious time-dependent solution of the system of equations: Eqs. (9)–(11) and (22) is easily found for a blob which remains
circular for all times:

\[
P_{01}(r, t) = \frac{1}{2} \left[ \Lambda(t)^{1+\alpha} (r^2 + y^2)^{1+\alpha/2} - R_0(t)^{2+\alpha} \right] + \frac{T}{4-q} \kappa(t) \quad \text{for} \quad r < R_0(t)
\]

with \( \Lambda(t) \) being given by: \( \dot{b}(t)/[2N(\dot{b})(t^{1+2\alpha/(1+\alpha)})] \) and \( \kappa(t) \) the curvature of the blob.

\[
P_{02}(r, t) = \frac{\eta}{4} \frac{b(t)}{b^2(t)} (r^2 + y^2) - R_0(t)^2 + \frac{\eta T}{4-q} \kappa(t) \quad \text{for} \quad r > R_0(t)
\]

with:

\[
R_0(t) = \sqrt{b_0 b(t)}
\]

This self-similar collapsing solution of the hydrodynamic flow is simple but may not be observable in the experiment, since it may be unstable as soon as \( \eta < 1 \) as follows from the linear stability analysis presented below.

2.4. Predictions of the linear analysis

2.4.1. The Newtonian case

As shown in Fig. 2, a large number of small oscillations can be observed at very short times in the experiment. The number of oscillations being large and the amplitude small suggests that a linear treatment can yield useful information. Although the calculation is straightforward \([5,6]\), we recall it to extend the treatment to the more complicated case of the power-law fluid. We assume that at time \( t = 0 \), a small perturbation is present at the boundary. As time goes on, it becomes:

\[
R_0(\theta, t) = R_0(t) + \epsilon C_n(t) \sin(n\theta), \quad 0 < \epsilon \ll 1
\]

\( C_n(t) \) being the growth rate of the mode \( n \). To linear order, the pressure fields are perturbed linearly (\( \epsilon \) is small):

\[
P_{1,2}(r, \theta, t) = P_{01,2}(t) + \epsilon p_{1,2}(r, \theta, t)
\]

We can write down the \( O(\epsilon) \) problem as:

\[
\nabla^2 p_{1,2}(r, \theta, t) = 0
\]

Fig. 2. Observation using a CCD camera of the fingering instabilities at different stages of the experiment. Shown here is a typical evolution for a speed of 20 \( \mu \text{m/s} \) and initial spacing of 0.5 mm. Diameter shown is 40 mm.
On the boundary, for \( r = R(\theta, t) \), the continuity Eq. (10) gives:

\[
b(t)^2 \frac{\partial p_1}{\partial r} + \frac{1}{2} \frac{b(t)}{b(t)} C_n(t) \sin n \theta + \frac{C_n(t)}{b(t)} \sin n \theta = 0
\]

(29)

and

\[
\frac{\partial p_2}{\partial r} = \eta \frac{\partial p_1}{\partial r} \quad \text{for} \quad r = R(\theta, t)
\]

(30)

From the Young–Laplace equation, we derive:

\[
p_1 - p_2 + \frac{1}{2} \frac{b(t)}{b(t)} R_0(t) C_n(t) \sin n \theta = \frac{T F(n, t)}{R_0(t)} C_n(t) \sin n \theta
\]

(31)

where \( F(n, t) \) has to be determined in three different limits, using the results of the previous paragraph. If we assume a pure cylindrical geometry for the interface, we obtain:

\[F(n, t) = n^2 - 1.\]

The solutions to this set of equations that have bounded pressure at infinity are:

\[p_2 = A_2(t) \left( \frac{R_0(t)}{r} \right)^n \sin n \theta \]

(32)

and is finite for \( r = 0 \):

\[p_1 = A_1(t) \left( \frac{r}{R_0(t)} \right)^n \sin n \theta \]

(33)

From the continuity equation at the boundary it follows that: \( A_2 = -\eta A_1 \). Substitution into the boundary conditions reveals that \( A \) and \( C \) satisfy the coupled equations and the boundary conditions:

\[
\frac{dC_n}{dt} + n \frac{A_1}{R_0(t)} b(t)^2 + \frac{b(t)}{2b(t)} C_n(t) = 0
\]

(34)

\[A_1(1 + \eta) + \frac{(1 - \eta)}{2} \frac{b(t)}{b(t)} R_0(t) C_n(t) = \frac{T}{R_0(t)^2} C_n(t) F(n, t)
\]

(35)

\[C_n(0) = 1
\]

(36)

Eliminating \( A(t) \) gives a unique equation for \( C_n(t) \):

\[
\frac{dC_n(t)}{dt} + \frac{1}{2} \frac{b(t)}{b(t)} \left( \frac{b(t)}{R_0(t)} \left( 1 - \eta \right) \right) \frac{A_1}{R_0(t)} + \frac{2nF(n, t) T}{R_0(t)} b(t)^2 = 0
\]

(37)

We shall use this result first to examine the classical 2D case.

2.5. Growth rate of the contour undulation in 2D

For the 2D case: \( F(n, t) = n^2 - 1 \), and we can integrate \( C_n(t) \) explicitly to obtain:

\[
C_n(t) = h(t)^{\delta - 1/2} \exp \left[ -\beta \int_0^t (h(t))^{1/2} \, dt \right] \quad \text{for} \quad 0 < t < 1
\]

(38)

with \( \delta \) the viscosity contrast \( \delta = (1 - \eta)/(1 + \eta) \) and \( \beta = Tn(n^2 - 1)b_0^{1/2}(1 + \eta) \). This gives the dynamics of each mode under the condition that they do not couple to each other, i.e., at short times. As was expected, the circular blob is unstable for \( \eta < 1 \) but is stable for \( \eta > 1 \). From Eq. (38), we deduce two characteristic values for the wavenumber:
(i) the critical wave-number \( n_c \) which corresponds to \( \frac{dC_n}{dt} = 0 \), (ii) the wavenumber \( n_m \) which corresponds to the fastest growing mode and the condition:

\[
\frac{\partial}{\partial n} \left( \frac{dC}{dt} \right) = 0
\]

\( n_c \) is solution of a cubic equation and can be evaluated easily for two limits:

\( \delta \to 1 \) which corresponds to the classical limit of a viscous blob in air and \( \delta \to 0 \) which means that the viscosity contrast is weak. In between these two limiting cases, the calculation must be done numerically. When we consider that \( \eta = 0 \) so \( \mu_2 = 0 \) (air/liquid interface), we get:

\[
n_c = \left( \frac{bb_0^{3/2}}{2B(t)^{3/2}} + \frac{1}{4} \right)^{1/2} - \frac{1}{2}
\]

(39)

Perhaps the most interesting result from this simple linear stability analysis is \( n_m \) which represents the number of penetrating fingers if one naively anticipates that the fastest growing fingers win the competition compared to the others. This rather naive physical intuition needs to be compared to the weakly nonlinear treatment since our analysis is only valid at short times when the lifting process occurs.

From Eq. (38), we deduce that:

\[
n_m^2 = \frac{1 - \eta B(t)}{6T} \left( \frac{B(0)^{3/2} A_0^2}{B(t)^{3/2}} \right) + \frac{1}{3}
\]

(40)

We give here the result in physical units. Since, in the experiment, the initial number of modes is rather large, one can neglect the number \( \frac{1}{3} \) and finds that the number of modes decreases as \( B(t)^{-3/4} \), if the velocity of the lifting is constant, as in the experiments [3]. A possible other comparison with the experimental result then consists in the measure of number of fingers at time \( t = 0 \) and the time evolution of this number as time goes on.

### 2.5.1. Results with an averaged meniscus

As mentioned above, two limits exist that are easy to handle, although in a real experiment, it is not sure that we are concerned with either limit. This has to be checked carefully. The dimensionless number which selects the regime is:

\[
\rho = \frac{4(1 - R(t)/b(t))}{b(t)}
\]

When \( \rho \) is small, the expected number of fingers is given by:

\[
n_m^2 \approx \frac{16 A_0^2 B(t)}{3 B(t)^{3/2}} = \frac{1 - \eta B(t)}{3T} \left( \frac{B(0)^{3/2} A_0^2}{B(t)^{3/2}} \right) + \frac{2}{3}
\]

(41)

while in the limit of large \( \rho \), we get:

\[
n_m^2 \approx \frac{8}{3} \rho^2 \log(\rho) \frac{A_0^2 B(t)}{B(t)^{3/2}} = \frac{1 - \eta \rho B(t)}{6T} \log \rho \left( \frac{B(0)^{3/2} A_0^2}{B(t)^{3/2}} \right) + \frac{1}{3}
\]

(42)

It is worthwhile to remember that \( T = B_0/\{12A_0^2 b(t) [B(t)] \} \) and \( \rho \approx \{1 - R(t)/A_0 B(t), R(t) \) being the ratio between the present radius and the radius at \( t = 0 \), and that this calculation assumes that the contact line does not move.
It turns out that in the experiment, the parameter \( \rho \) can be of order unity and \( \rho_1 \sim -\rho^2 \). A calculation that is tedious but without difficulty gives:

\[
\frac{1}{\rho} \left[ \frac{1}{\rho} \left( 1 + \frac{1}{\rho^2} \right) \right] \sqrt{1 + \rho^2} + \frac{16}{3} \left( 1 + \rho^2 \right)^{1/2} \frac{A^2_2 R_0}{B(t) - 3B(0)} - \frac{A^2_2 R_0}{6A_0(1 - R^2)} \frac{B(t)(1 - 2R)}{\sqrt{1 + \rho^2}}
\]

The limit \( \rho \) small or \( \rho \) large allows to recover the results given by Eq. (41) or Eq. (42). If we allow an oscillation of the contact line, in the same limit \( \rho_1 = -\rho^2 \) we obtain:

\[
\frac{1}{\rho} \left[ \frac{1}{\rho} \left( 1 + \frac{1}{\rho^2} \right) \right] \sqrt{1 + \rho^2} + \frac{2}{3} \left( 1 + \rho^2 \right)^{1/2} \frac{A^2_2 R_0}{B(t)} + \frac{R - 1}{3\rho} \left[ \frac{1}{\rho} \left( 1 + \frac{1}{\rho^2} \right) \right] \sqrt{1 + \rho^2}
\]

which recovers the 2D case in the small \( \rho \) limit exactly.

2.5.2. Fingering with a power-law fluid

If the fluid 1 has a power-law viscosity, the perturbation of the continuity equation (equivalent to Eq. (29)) is then:

\[
\frac{\partial \rho \phi}{(1 + \alpha)\sqrt{g^2 R_0(t)^2}} + \frac{1}{2 \lambda(t)} C_\phi(t) \sin n\phi + \frac{dC_\phi(t)}{dt} \sin n\phi = 0
\]

For simplicity, we neglect the viscosity of the outer fluid. Also, the Laplace equation is modified to:

\[
p_{\phi} + \Lambda(t)^{1/2} R_0(t)^{1/2} C_\phi(t) \sin n\phi = \frac{T(\alpha^2 - 1)}{R_0(t)^{1/2}} C_\phi(t) \sin n\phi
\]

The perturbed flow is given by:

\[
p_{\phi} = A(t)^{\phi}(t) \frac{\sin(n\phi)}{R_0(t)^{1/2}}
\]

\( \delta(n) \) being solution of \( \delta(\alpha - a) = n^2(1 + a) \). By elimination, one obtains the dynamical equation for \( C_\phi \):

\[
\frac{dC_\phi(t)}{dt} + \frac{1}{2} C_\phi(t) \Omega(t) \left( \frac{b(t)}{b(t)} \right) = 0
\]

with:

\[
\Omega(t) = 1 - \frac{\delta(n)}{(1 + \alpha)} + \frac{\delta(n)(\alpha^2 - 1)T}{\Lambda(t)^{1/2} R_0(t)^{1/2}(1 + a)}
\]

In the case of shear-thinning, the viscosity decreases with the shear rate, so that \( a < 0 \). If we calculate \( \Omega(0) \) for this situation, we obtain:

\[
\Omega(0) = 1 - \frac{\delta(n)}{(1 + \alpha)} + \frac{\delta(n)(\alpha^2 - 1)T}{(1 + a) - \frac{1}{2} \alpha^2 T}
\]
where, to simplify the problem, we use the cylindrical curvature. The stability of the initial contour is given by the sign of $\Omega(0)$. Then a blob of power-law fluid is unstable when values of $\delta(n)$ exist that make $\Omega(0)$ negative. We recall that, in an ordinary experiment, the number $n$ of oscillations is of order several dozens. We can therefore consider two limits: $a$ very close to zero (weakly non-Newtonian fluid) and $a$ close to the threshold value $-1$. Here also, we assume that the observed pattern corresponds to the fastest fingers which implies $\partial \Omega/\partial n = 0$. In this case, taking into account Eq. (49), we derive $n_m$:

$$n_m^2(3 + 2a) + a/2(u^2 + 4(1 + a^n_m)^{1/2} + u^2/2) - 1 = \frac{1 + a}{F} \frac{h^{1+3+nu/2}}{(2N)^{1+nu/2}}$$

If the number of fingers is large, we obtain for $n_m$:

$$n_m^2 = \frac{1 + a}{(3 + 2a)F} \frac{h^{1+3+nu/2}}{(2N)^{1+nu/2}}$$

In the simplest model, this predicts a decrease of the number of fingers as $e^{-(u+3a)/4}$, which depends on the properties of the complex fluid through $a$.

3. Experiment

3.1. Set up and model system

The experimental setup and protocol have already been described in detail by Derks et al. [3]. In brief, we study the stretch flow of a thin adhesive layer that is confined between two parallel circular plates (see Fig. 1) a geometry commonly used for probe tests. We use a Reologica StressTech rheometer equipped with a normal force transducer for the stretch flow experiments, using a plate-plate geometry. The upper plate is lifted at a constant driving velocity $V$ (typically 20 or 50 mm/s), leading to a debonding of the two surfaces. The stainless steel upper plate has a radius $A_0$ of 20 mm; the bottom plate is made of glass, which allows the observation in real time of the shape of the air–adhesive interface by filming with a CCD camera. The initial thickness of the adhesive layer $B_0$ is varied between 0.1 and 1.1 mm. As a Newtonian test fluid, we use a silicon oil of viscosity 30 Pa s.

3.2. Fingering instabilities

Provided the typical wavelength of the fingering instabilities is smaller than the perimeter of the initially circular fluid blob, we observe the formation of fingers at the air-fluid interface during the debonding. We will investigate a set of experiments changing the initial plate spacing going from an experimental situation where little fingering is observed (large plate spacing) to a situation where strong fingering is observed (small plate spacing). We will limit ourselves here to experiments on the silicone oil; non-Newtonian fluids will be discussed elsewhere.

A typical sequence of images is shown in Fig. 2; we observe a large number of fingers in the beginning of the experiment, their number decreasing in time as the plate separation grows. To quantify our observations, we consider the wavelength of the fingering instabilities at the start of the experiments, where the linear stability analysis will most likely apply. We measure the finger width $\lambda$ simply by counting the number of fingers observed on an image, and define $\lambda$ as the perimeter of the interface divided by the number of fingers. Fig. 4 presents a typical result for the number of fingers as a function of time. As can be observed from the figure, the decrease of the number of fingers is exponential in time. This is in clear contradiction with the theoretical result from the 2D linear analysis, which predicts a power-law decrease (Eq. (40)). The result is however quite general; different silicon oils of widely differing viscosity (from 1 Pa s up to 500 Pa s) show similar exponential behaviour, the regression coefficient always being larger than 0.98.
Fig. 3. Number of fingers at early times during the experiment, for different initial plate spacings. The linear theory predicts values that are an order of magnitude larger than those observed experimentally; taking three-dimensional effects into account improves the agreement with theory especially for large values of $B$.

At this point, there are two possible explanations for the discrepancy between theory and experiment. The first option is that non-linear effects [24,25] are important, so that one has to go beyond linear instability to account for the experimental results. The second option is that three-dimensional effects, which were neglected in the theoretical treatment leading to Eq. (40), are important.

In order to distinguish between these two, we can compare the finger width as a function of the initial distance between the plates at early times. At early times, the instability has just set in, and therefore the linear stability analysis should apply. Fig. 3 depicts the initial number of fingers, counted as soon as they are clearly distinguishable, for different spacings between the plates. The comparison with the linear analysis (Eq. (40)) is telling: the stability analysis predicts a number of fingers that is roughly an order of magnitude larger than the experimental values. This implies that the two-dimensional approximation that led to Eq. (40) needs to be reconsidered. This was done (for exactly this reason) in the theoretical part of the paper, leading to Eqs. (41) and (44).

The key parameter characterising the amplitude of the 3D effects is $\rho$, which gives the deviation from a purely cylindrical fluid blob $\rho = 4(R(z = b/2) - R(z = 0))/B$ ($R$ and $B$ being defined here in physical units). Unfortunately, this quantity evolves during the experiment since it depends on the mobility of the contact line itself so on the wetting properties of the fluid/substrate. This is the reason why we focus on the early times, so that a direct comparison can be made with the data of Fig. 3. A direct measurement using high-resolution photography gives $\rho = 0.142 \pm 0.04$. Therefore, we should compare the experimentally observed number of fingers with the theoretical prediction for small $\rho$. Eq. (41). This is done in Fig. 3, from which we can observe that the agreement is somewhat improved by taking the 3D effects into account, especially for large $B$. In any case, the overall form of the dependence of the number of fingers as a function of $B$ is similar between experiments and the 3D theory. There still remains a discrepancy for small $B$, that we do not fully understand for the moment.

Theoretical results from the three-dimensional analysis, also predict a power-law like decrease of the number of fingers in time, while the decrease of the number of fingers in the experiment is exponential. The comparison is not completely straightforward however since the dimensionless parameter $\rho$ which measures the lateral retraction of the meniscus compared to the cell thickness varies as time goes on. We conclude that in any case the comparison with the linear theory is not very favourable. Therefore, the mechanism for this exponential behaviour is probably a non-linear one: if pairs of fingers coalesce together during the debonding (and this is the only possible mechanism for the decrease of the number of fingers) and if one assumes that the time $t_0$ required
to divide the number of fingers by 2 is always the same, we automatically get a coalescence cascade. This leads naturally to an exponential decrease of the number of fingers, as observed in the experiments. The experimental determination of the characteristic time $\tau_0$ (see Figs. 4 and 5) shows that it is a linear function of the spacing. We have no explanation for this dependence at the moment.

4. Discussion and conclusions

We studied the fingering instabilities that occur in debonding of model adhesives. We perform a linear analysis of the viscous instability in the lifting Hele-Shaw cell, and compare with experiments. In the experiment we observe
the formation of fingers at the air-fluid interface during the debonding. We observe a large number of fingers in the beginning of the experiment, their number decreasing in time, as the plate separation grows. The decrease of the number of fingers turns out to be exponential in time, in clear contradiction with the theoretical result from the 2D linear analysis, which predicts a power-law decrease (Eq. (40)). We suggest that there may be two explanations possible for the discrepancy. First, non-linear effects [24,25] may be important. The second option is that three-dimensional effects, which were neglected in the theoretical treatment leading to Eq. (40), are important.

In order to distinguish between these two, we compare theory and experiment at early times, when the instability has just set in. This is the first time that a detailed and quantitative comparison between theory and experiment is performed. The linear stability analysis should apply here, because we are at the onset of instability. Unfortunately, it predicts a number of fingers that is roughly an order of magnitude larger than the experimental values. The sophisticated numerical analysis performed in [13,14] and more recently in [25] is restricted to the 2D model with the standard boundary conditions. These works treat only the nonlinearities that occur when time progresses but cannot improve on the discrepancies that occur at early times, near the onset of instability.

To improve the comparison with theory, we take three-dimensional effects into account. The agreement is somewhat improved, but there still remains an important discrepancy for small B, that we do not fully understand for the moment. Unfortunately, the full three-dimensional Stokes flow problem is extremely difficult if not impossible to solve, so it will be very hard to do better theoretically. The comparison of the experimental and the theoretical results nevertheless teach the important lesson that the three-dimensional effects are not a small correction in this problem, and therefore need to be taken into account. Another observation that points in this direction is that in the two-dimensional case, all horizontal dimensions should be much larger than the plate spacing. Although this is the case for the initial diameter of the fluid, the wavelength of the fingers is in fact very similar to the gap, and here three-dimensional effects may become very important. These effects are unfortunately very difficult to incorporate in our theoretical treatment of the problem.

For the evolution of the number of fingers with time, the comparison with the linear theory is not very favourable, even if three-dimensional effects are taken into account. Therefore, the mechanism for this exponential behaviour is probably a non-linear one. If we assume that the coalescence time between pairs of fingers is constant, we get a coalescence cascade that explains the exponential behaviour.

In order to go further, probably, not only the curvature so the Laplace relation Eq. (11) has to be modified but also the continuity Eq. (10). The latter is probably the more efficient effect although this is hard to quantify exactly for the reasons mentioned above. As an example, we can assume that there a film of oil is left behind on the plates once the meniscus has passed, as suggested by Fig. 1. For simplicity, we assume a film of constant thickness in the vicinity of the edge, although we know that its thickness may be a function of the 2D curvature. We know that this film exists in classical viscous fingering experiments and that its value is on the order of $C^2$. In this case the continuity equation Eq. (10) is modified, and we obtain $Vn(1-m) = \Omega$. The basic state has a different dynamics $R_0(t) = A_0(b_0/\eta)^{(3-m)/2}$ and finally the number of fingers in the cylindrical model becomes:

$$n^2 = \frac{1}{6\eta} \frac{B(t)}{B(0)} \left( \frac{B(0)(2-\mu/2)}{B(t)(2-\mu/2)} \right) + \frac{1}{3} \left( \frac{A_0^2}{B(t)} \right)^2 (53)$$

so that a constant film thickness does not modify the discrepancy at initial times, but it does modify the curvature-related effects considerably. All these effects remain to be considered if one wants a complete understanding of the fingering instabilities occurring in the debonding of soft adhesives.

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