Viscous Finger Widening with Surfactants and Polymers

D. Bonn,[1] H. Kellay,[2] M. Ben Amar,[1] and J. Meunier[1]

[1]*Ecole Normale Supérieure, Laboratoire de Physique Statistique, 24 Rue Lhomond, 75231 Paris Cedex 05, France*
[2]*Centre de Physique Moleculaire Optique et Hertzienne, Université Bordeaux 1 351, Cours de la Libération, 33405 Talence, France*

(Received 1 March 1995)

We study the viscous fingering instability which results from a competition between capillary and viscous forces. We show that for two different systems the instability is modified drastically by changing the surface tension or viscosity by means of surfactants or polymers. For both systems the width of the finger can increase with increasing velocity before settling at a plateau value larger than half the channel width. A numerical study shows that the large deviations from the classical result can be attributed to a velocity dependence of the dynamic interfacial tension and viscosity.

PACS numbers: 47.20.Hw, 61.25.Hq, 68.10.–m,

The Saffman-Taylor instability [1] arises when a less viscous fluid pushes a more viscous fluid in a thin linear channel or Hele-Shaw cell. The interface between the fluids develops an instability leading to the formation of fingerlike patterns, called viscous fingers. It has received much attention as an archetype of pattern-forming systems [2], a model system for flow through porous media [1], and as a limiting factor in the recovery of crude oil [3]. The problem originates from the oil industry: Petroleum engineers reported "tongues of water in oil" in secondary oil recovery.

The width of the viscous fingers is determined by the capillary number $\mathrm{Ca} = \Delta\mu U/\gamma$, which represents the ratio of viscous forces to capillary forces: $\Delta\mu$ is the viscosity difference, U the finger velocity, and γ the surface tension. The viscous forces tend to narrow the finger, whereas the capillary forces tend to widen the finger. As a result, the width of the finger w relative to the channel width W, $\lambda = w/W$, decreases with increasing finger velocity. For large values of the capillary parameter, λ reaches a limiting value of about $\frac{1}{2}$ [1,2]. The dimensionless control parameter is $1/B = 12\,\mathrm{Ca}(W/b)^2$, where b ($= 0.25$ mm, $W = 4$ cm) is the distance between the plates of the Hele-Shaw cell. When scaled on $1/B$, measurements of λ for different systems all fall on the same universal curve. Changing the viscosity or surface tension thus only leads to a displacement on the universal λ vs $1/B$ curve. We show here that for two different complex fluids, by acting on the surface tension and viscosity, results are obtained that differ greatly from the classical result.

In the first experiment, a surfactant is present at the interface between water and an oil. The anionic surfactant AOT is dissolved in pure water at concentrations above the critical micellar concentration (CMC ≈ 2.5 mM) so that the interface is covered with a saturated surfactant monolayer. The less viscous, driving fluid is heptane. Figure 1(a) depicts the results for two AOT concentrations. The surprise is that the finger width settles at a plateau value significantly above the classical $\lambda = \frac{1}{2}$ value. For the 3 mM AOT solution, it can be observed that λ goes through a minimum and then increases before reaching a high-velocity plateau value.

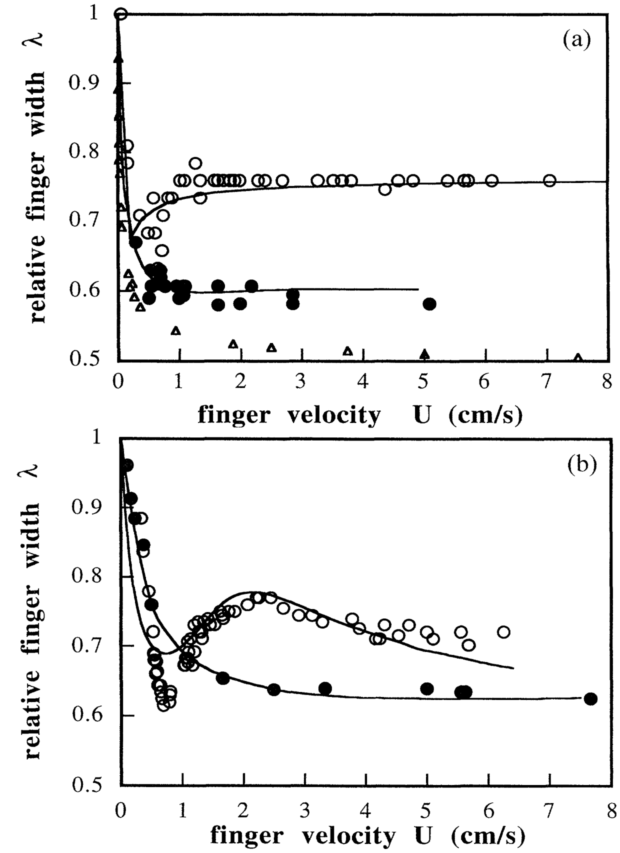

FIG. 1. The relative width of the finger as a function of the finger velocity. The symbols are the experimental results and the drawn lines are the result of the numerical calculations. (a) For the oil/water/surfactant system for 3 mM AOT (open circles) and for 15 mM (filled circles). Also shown is the classical result for a water/glycerol mixture with air as the driving fluid (triangles). (b) For the polymer solution, 20 wppm (closed circles) and 500 wppm (open circles).

In a second experiment, we use a polymer solution of polyethyleneoxide (Polyox WSR 301, $M \approx 4 \times 10^6$ a.m.u.) in water as the fiscous fluid and compressed air as the less viscous, driving fluid. In Fig. 1(b), we show two measurements; we again find plateau values with $\lambda > \frac{1}{2}$ and also, for the more concentrated polymer solution, a finger width that increases with increasing velocity.

Figure 2 shows the shape of three observed fingers compared to the classical result for the form of the finger. For the surfactant system, the form of the observed fingers agrees well. However, for the polymer solution at low velocities narrow, pointed fingers are found. For somewhat higher velocities we observe blunt and wider fingers.

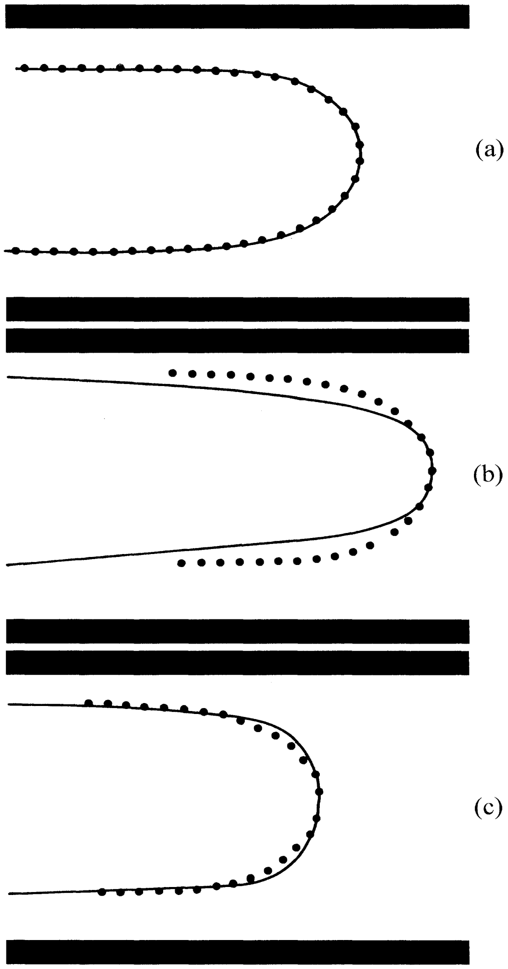

FIG. 2. The shape for three of the observed fingers with approximately the same limiting width compared to the classical result for the form of the finger with inclusion of surface tension effects, shown here as the dots. (a) For the surfactant system, the form of the fingers is in agreement with the classical theory. (b), (c) Two fingers for the 500 wppm polymer solution [corresponding to the measurement in Fig. 1(b)] with velocities 0.7 and 1.3 cm/s, respectively.

The experimental observation then is that for the two completely different systems the results are very similar and deviate strongly from the classical result: Stable, very large fingers can be obtained. This deserves some attention, since in previous experiments either narrow [4,5] or unstable fingers [6] were found by disturbing the finger. Also, the form of the fingers for the polymer solution is aberrant. How can these results be understood?

First, consider the surfactant system. The presence of a surfactant film at the interface between the oil and water introduces supplementary restoring forces, caused by the advection of surfactant molecules from the tip to the sides of the finger by the hydrodynamic flow [7]. This results in a dynamic surface tension at the tip which is higher than the equilibrium tension and increases with velocity. The finger reaches a steady state as neither its form nor its width changes during the experiment. The nonequilibrium adsorption Γ can then be calculated from the equation for mass conservation for the interface [7]: $\mathrm{div}(\Gamma U_t) = -D\partial c/\partial z$; here the first term accounts for the convection of the surfactant molecules by the (tangential component of) hydrodynamic flow U_t, and the second term accounts for the diffusion from the bulk to the interface. D is the bulk diffusion coefficient, and the concentration gradient is in the direction perpendicular to the interface [7]. We have neglected the surface diffusion of the surfactant molecules, which is usually very small. Assuming not too large departures from equilibrium, the local surfactant concentration can be written $\Gamma = \Gamma_e[1 - (1/\alpha)dU_t/dS]$, with S the dimensionless arclength and α a characteristic velocity for the diffusion of surfactant molecules from the bulk to the surface. Expanding the surface tension around its equilibrium value, we obtain a dynamic tension $\gamma = \gamma_e + d\gamma/d\Gamma(\Gamma - \Gamma_e)$; the subscript e denotes an equilibrium value [8], and $\gamma_e \approx 2$ mN m^{-1} for this system.

For the polymer solutions, we measured the surface tension using the Wilhelmy ring method. We find $\gamma = 63 \pm 0.5$ mN m^{-1} virtually independent of the polymer concentration over three decades in concentration: $1 < c < 1000$ wppm. Thus there is an effect of polymer adsorption (a 14% change with respect to the surface tension of water), but it is small and concentration independent. The observed effects, on the other hand, are rather large and concentration dependent. We believe the more important effect to be that the polymer solution is a shear thinning, i.e., a non-Newtonian liquid (its viscosity depends on the local shear rate ω). The main shear in the Hele-Shaw cell occurs in the direction perpendicular to the plates; the shear rate can be estimated as the fluid velocity V divided by the plate spacing, $\omega \approx V/b$. One may attempt to model μ in terms of a single relaxation time $\mu(\omega) = \mu_\infty + (\mu_0 - \mu_\infty)/(1 + \omega^2\tau^2)$, where τ represents the largest relaxation time in the system, i.e., the Zimm relaxation time of the polymer [9]; μ_0 and μ_∞ are the zero- and infinite-shear viscosities.

With these simple models, we proceed by a direct numerical integration of the two-dimensional hydrodynamic equations, with the local tension (surfactants) or viscosity (polymers) as given above. We adapt the McLean-Saffman method [10]. For the surfactant system the calculations are straightforward, since only the local surface tension has to be modified compared to the standard case [11]. For the polymer, however, the local viscosity now depends on the local hydrodynamic velocity. As a consequence, the velocity field is no longer Laplacian. It can, however, be shown that the pressure field remains Laplacian so that the conformal mapping techniques [10] can be applied to this field, which entails a modification of the continuity equation [12]. Only the one adjustable parameter is added to the standard problem in this way: the amplitude of the effect determined by $d\gamma/d\Gamma$ for the surfactants and by the ratio μ_0/μ_∞ for the polymer. The results of the calculation are shown as the drawn curves in Fig. 1, from which it may be concluded that these simple models reproduce the essential physics of the problem.

The pertinent question is then whether the parameters [13] that were varied to fit the data make any physical sense. The results are shown in Fig. 3. For the surfactant system we obtain roughly $\alpha \approx 0.1$ cm/s and $\Gamma_e/\gamma_e \, d\gamma/d\Gamma \approx 10^{-3}$. The parameter α gives the flux of surfactant molecules from the bulk to the surface. Knowing the bulk diffusion coefficient [7], and writing the concentration gradient as a typical concentration difference (of the order of the bulk concentration) over a characteristic diffusion length δ, one obtains $\delta \approx 10$ μm, which seems to be of the right order of magnitude [7]. The relation between the surface tension and the adsorption $\Gamma_e/\gamma_e \, d\gamma/d\Gamma \approx 10^{-2}$, in principle, provides us with an equation of state for the surfactant system. However, comparing with, for instance, the two-dimensional gas equation of state, one finds that a typical value would be on the order of unity. There can be a number of reasons for this large discrepancy. First, in making this comparison the (perhaps too strong) assumption is that locally the equilibrium relation between the surface tension and adsorption still applies. Second, in the hydrodynamic calculation three-dimensional effects have been neglected. Although this is usually a fairly good approximation [14], it may be that the presence of the surfactant alters this. A third possibility is, of course, that our model for the surfactant advection may be oversimplified; the fairly good agreement with experiment, however, does not appear to be not entirely fortuitous.

For the polymer solution [Fig. 3(b)], the values for the viscosity are of the correct order of magnitude; typically, we find $\mu_0 \approx 5\mu_{\text{water}}$ and $\mu_\infty \approx 0.5\mu_{\text{water}}$. The rather large differences with the viscosity of water (at the low concentrations of polymer) are most probably related to the drag-reducing properties of the polymer. It is well known that solutions of these polymers can have, for instance, an elongational viscosity that is a hundred times that of the solvent, and they can exhibit drag reduction at the same time [9]. The relaxation time of the polymer is also of the correct order of magnitude. Calculating the Zimm relaxation time of the polymer, $\tau_Z \approx 3R^3\mu/k_BT$, with k_B the Boltzmann constant and T the temperature, with a (hydrodynamic) radius $R = 0.2$ μm determined from light scattering, we find $\tau_Z \approx 6 \times 10^{-3}$ s. It is surprising that τ appears to be concentration dependent. For the highest concentrations, it may be that there are some entanglement effects [9]. However, it may also be that, again, the model we have used is somewhat too simple. In the similar experiment of Smith *et al.* [5], narrow (rather than wide) fingers were obtained. These can also be found in our calculation; compared to the present experiment, they occur for a smaller ratio μ_0/μ_∞.

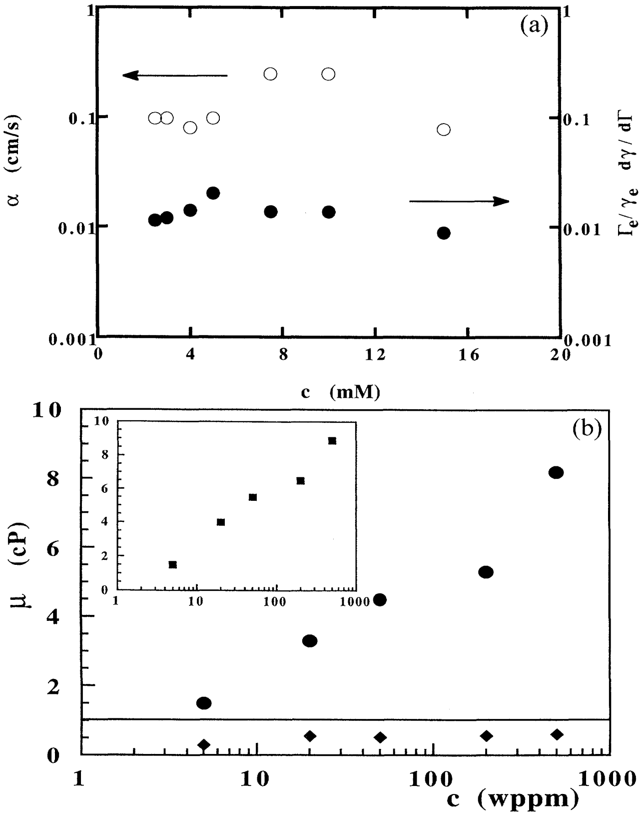

FIG. 3. The results of the fitting procedure. (a) For the surfactant system; the open circles denote the characteristic velocity for the arrival of surfactant molecules α and the filled circles give the derivative of the tension with respect to the adsorption. (b) For the polymer system; filled circles are for the zero-shear viscosity μ_0 and the diamonds are for the infinite shear viscosity μ_∞. The drawn line is the viscosity of water. The inset shows the relaxation time τ (units 10^{-3} s) as a function of the polymer concentration (units wppm).

These results suggest a simple physical interpretation of our observations. The first-order effect of the introduction of a dynamic viscosity or tension is qualitatively the same for both systems: The capillary number $\mathrm{Ca} = \mu U/\gamma$

will no longer be a simple linear function of the finger velocity U, since γ or μ depend on U; this will weaken the dependence of Ca on U. When the velocity dependence of γ or μ annihilates the dependence of Ca on velocity, it does not change anymore by changing the velocity. As there is a unique value of λ assigned to each value of Ca through the selection mechanism [13], this gives rise to the plateau values observed in the experiment. Moreover, this also explains the striking similarity of the findings for the two different systems. Of course, anisotropy effects [4] should also be taken into account. The experimental observations then point to the fact that the velocity dependence of the capillary number has been annihilated effectively by dynamic tension or viscosity and anisotropy effects.

The final question concerns the form of the fingers. For the surfactant system, anisotropy effects [4] are probably not very important. For the polymers, the viscosity depends on the local shear rate, which varies along the finger. For low velocities [Fig. 2(b)], the larger viscosity gradient is near the tip so that the forward direction is a direction of "easy growth," which then leads to smaller and more pointed fingers [4,5]. For higher velocities [Fig. 2(c)], the effect saturates at the tip. The gradient is largest at the sides of the finger, which "grow" more easily; blunt and wider fingers are found. The finger forms from our calculation reproduce this effect qualitatively, but are close to the classical form, which points to the possible importance of three-dimensional effects.

In conclusion, we have shown that the viscous fingering instability is modified drastically when one uses complex fluids to act on the surface tension or viscosity. The effects on the instability can be understood in terms of a velocity dependence of the dynamic tension or viscosity. Moreover, we have shown that the above findings may be applicable to determine the dynamic surface tension or viscosity simply by measuring the width of the finger: The instability could be used to obtain material properties of the system. Finally, the stable, very large fingers that we found were part of the original motivation for studying the instability; larger and more stable fingers improve the efficiency of oil recovery.

LPS de l'ENS is URA 1306 and CPMOH is URA N283 of the CNRS. We thank W. van Saarloos, Y. Couder, P. Tabeling, D. Bensimon, and A. Simon for helpful discussions.

[1] P. G. Saffman and G. I. Taylor, Proc. R. Soc. London A **245**, 312 (1958); D. Bensimon *et al.*, Rev. Mod. Phys. **58**, 977 (1986); G. M. Homsy, Annu. Rev. Fluid Mech. **19**, 271 (1987).

[2] D. Kessler, J. Koplik, and H. Levine, Adv. Phys. **37**, 255 (1988).

[3] Improved Oil Recovery, Norwegian Petroleum Directorate Report, 1993 (unpublished).

[4] E. Ben-Jacob *et al.*, Phys. Rev. Lett. **55**, 1315 (1985).

[5] D. E. Smith *et al.*, Phys. Rev. A **45**, 2165 (1992).

[6] M. Ben Amar, R. Combescot, and Y. Couder, Phys. Rev. Lett. **70**, 3074 (1993).

[7] V. G. Levich, *Physicochemical Hydrodynamics* (Prentice-Hall, Englewood Cliffs, NJ, 1962); D. D. Joseph, Eur. J. Mech. **B9**, 565 (1990). The observation that α does not depend on the bulk concentration is rather surprising. This may be due to the existence of an adsorption barrier for the surfactant molecules. It is not the arrival of molecules from the bulk, but rather the time it takes them to adsorb that is the rate-determining step; this can be due to an electrostatic repulsion of the surfactant molecules that approach the interface [A. Bonfillon-Colin, thesis, Paris, 1994 (unpublished)].

[8] Because of the spatial surface tension gradient, the surface stiffness has also to be taken into account. The pressure drop over the interface is then given by $\Delta p = (\gamma + \gamma'')/R$; the derivative is taken with respect to the angle between the surface normal and the forward direction, and R is the radius of curvature.

[9] R. B. Bird, R. C. Armstrong, and O. Hassager, *Dynamics of Polymeric Liquids* (Wiley, New York, 1987).

[10] J. W. McLean and P. G. Saffman, J. Fluid Mech. **1**, 455 (1981).

[11] A. T. Dorsey and O. Martin, Phys. Rev. A **35**, 3989 (1987).

[12] D. Bonn, H. Kellay, M. Bräunlich, M. Ben Amar, and J. Meunier (to be published).

[13] $d\gamma/d\Gamma$ for the surfactants and μ_0/μ_∞ for the polymer are varied in the fitting procedure. τ for the polymer and α for the surfactants are obtained by imposing that the curves fit the data as a function of both $1/B$ (calculated from the equilibrium values) and the velocity. For the polymers, the absolute value of μ_∞ can be found by noting that this parameter determines the high-velocity plateau value for λ.

[14] P. Tabeling, G. Zocchi, and A. Libchaber, J. Fluid Mech. **177**, 67 (1987); C. W. Park and G. M. Homsy, J. Fluid Mech. **139**, 291 (1984); S. Tanveer, Proc. R. Soc. London A **428**, 511 (1990).