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Viscous fingering with imposed uniaxial anisotropy

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We study viscous fingering in the radial Hele-Shaw cell with a set of parallel grooves on one of the plates in order to obtain the morphological phase diagram of a system with imposed uniaxial anisotropy. Varying the pressure of air injected into glycerin and the distance between the plates, a rich variety of phases can be obtained corresponding to all of the possible combinations of stable or splitting tips in directions parallel and perpendicular to the grooves. We demonstrate that the effective anisotropy governing the stability of the tips is a result of a complex interplay between the local anisotropy and the driving force.

The role of anisotropy in the formation of diffusionlimited interfacial patterns has recently become one of the central questions in the studies of growing structures. Both the theoretical models and the related experiments indicate that the rich variety of morphologies observed in such phenomena can be interpreted in terms of the interplay of the anisotropy, fluctuations, and the driving force controlling the velocity of the moving phase boundary.

The study of viscous fingering in the radial Hele-Shaw cell^{1,2} has been shown to be a very useful tool for the experimental investigation of the formation of patterns under various conditions, including anisotropy.^{3,4} Recent experiments demonstrated that qualitatively different patterns can be obtained if the parameters of the system are changed. The splitting of the fingers' tips can lead to both fractal⁵ and homogeneous⁶ structures, while the imposed³ or inherent⁴ anisotropy results in dendritic shapes with stable tips. Similar results have been found in recent experiments on electrodeposition^{7,8} and two-dimensional solidification.⁹ The theoretical interpretation of this complex behavior is far from being complete, and new experimental findings are expected to be helpful in the construction of a unified picture of diffusion-limited pattern formation.

Recent results¹⁰⁻¹² in the theory of dendritic growth—a related problem¹³—show that a sufficient amount of anisotropy in the surface tension can stabilize the tips of the growing patterns. Since the above-mentioned experiments^{3,4} and computer simulations^{14,15} have demonstrated that diverse sources of anisotropy may lead to similar results, the concept of effective anisotropy has been introduced,^{4,14} which treats anisotropy irrespective of its origin. The effective anisotropy A_{eff} is defined as follows:

$$A_{\text{eff}} = 1 - v_n' / v_n \quad (1)$$

where v_n and v'_n are the normal interfacial velocities at the tip and at a distance r from it, respectively, with r being the radius of curvature at the tip. If A_{eff} is large enough, the tip is stabilized by the effective anisotropy: It advances faster, without letting the finger become flat enough for splitting. A_{eff} is a complicated function of the local anisotropy (which is present in the surface tension, in the local environment, or in the liquid itself) and the driving force. It is also influenced by the amount of fluctuations.^{9,14} In the viscous fingering experiments with isotropic liquids the surface of the plates of the Hele-Shaw cell has been found to play a particularly important role. Ben-Jacob *et al.*³ introduced engraving a grid on one of the plates of the Hele-Shaw cell and found that as a function of the pressure (the driving force) with which the air was injected into the viscous liquid a sequence of morphological phase transitions took place. In particular, when they increased the pressure, the disordered patterns associated with the geometry of diffusion-limited aggregates¹⁶ (DLA's) were observed to cross over into a snowflakelike regular structure with stable tips, suggesting that a larger driving force has a stabilizing effect.

In this paper, we study viscous fingering in the radial Hele-Shaw cell with uniaxial anisotropy in order to provide additional information about the role of anisotropy versus the driving force in the growth of diffusion-limited patterns. One of the questions we address experimentally is concerned with the stabilizing effect of the driving force: Does increasing the pressure generally result in the enhancement of anisotropy? According to our experiments, uniaxially engraved plates give rise to a variety of unexpected patterns and transitions, including a crossover from stable tips to tip splitting as the driving force is increased. We found a new kind of behavior with stable growth in a direction which is perpendicular to the direction of the easy growth. The morphological phase diagram obtained for our model system demonstrates the complexity of the problem of viscous fingering between two close engraved plates.

The motion of the interface between air and a viscous fluid placed between two close parallel plates is determined by the pressure distribution in the liquid p, satisfying the Laplace equation

$$\nabla^2 p = 0 \tag{2}$$

(if the fluid is incompressible) and by the following boundary conditions. At the interface the normal velocity v_n is governed by the pressure gradient

$$v_n = -k\nabla p \quad , \tag{3}$$

where k is a constant depending on the distance between the plates and the viscosities of the fluids. In addition, the pressure drop at the interface Δp depends on the local cur-

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$$\Delta p = -\alpha \kappa \quad , \tag{4}$$

where α is a combination of material constants, including the surface tension. The anisotropy usually enters the problem through the angular dependence of α .

The experimental setup consisted of a horizontal radial Hele-Shaw cell made of 320×320×5-mm glass plates and a pressure controller. The lower plate contained parallel grooves having a depth h = 0.4 mm and a width of 0.8 mm. The grooves were separated by 1.2 mm. The distance between the glass plates b was controlled by metallic holders within the accuracy of 0.01 mm. A heavy iron frame was used to prevent the lifting of the upper plate. The pressure controller contained a buffer that was large compared to the volume inside the cell, and the pressure could be varied in the region 0.5-200.0 mm Hg with an accuracy of 0.1-0.5 mm Hg. We used glycerin with 13 vol% water as the viscous fluid. Since its viscosity is sensitive to the thermal changes, the temperature was kept at 25.0°C $(\pm 0.5 \,^{\circ}\text{C})$ at which temperature the viscosity was ~150 cP and the air-liquid surface tension ~ 65 dyn/cm. Records of the developed patterns were taken by using photographics and video methods.

The results are summarized in Fig. 1 in the form of a morphological phase diagram. This figure shows a number of unexpected features of pattern formation in our system. Depending on the values of the parameters, i.e., the pressure of the air and the distance b between the plates, the direction of the stable growth changes and virtually all of the possible geometries can be achieved, including tip splitting in the "easy" (parallel) direction with a simultaneous stable growth in the perpendicular direction. As the distance between the plates is changed some of the patterns do not appear, while others become observable. The morphological phase diagram is similar to the phase diagrams of two-component alloys including the presence of a "metastable" phase IA with a phase boundary indicated by a dashed line. In this phase the shape of the observed patterns depends on minor details like the distance of the walls or position of the injecting pipe relative to the grooves. Similarly, the thickness of the phase boundaries is caused by the sensitivity of the system to such details in the transition regions. The typical patterns corresponding to the various morphological phases are displayed in Fig. 2.

As a function of increasing pressure a general tendency from perpendicular (phase I) to parallel stability (phase II) can be observed with sometimes (e.g., for b = 0.7 mm and p = 15 mm Hg) strikingly sharp transitions. For other values of b and p a new phase appears between phases I and II, either with simultaneous stable tips in the parallel and the perpendicular directions (phase III) or with no stable tips (phase IV). During the transitions $I \rightarrow II$, $III \rightarrow II$, or $I \rightarrow IV$ the perpendicular direction becomes unstable as the driving force (the pressure) is increased. This behavior is different from the prediction of the local models, 10,11 where increasing the driving force drives the system toward more regular anisotropic patterns, i.e., tip stabilization. Our experiments suggest that there is no reason to believe that the dependence of the effective an-



FIG. 1. Morphological phase diagram of the interfacial patterns in a uniaxially engraved radial Hele-Shaw cell. The parameters varied were the pressure (p) and the thickness of the liquid layer (b). The phases are denoted by characteristic patterns obtained for parameter values indicated by the filled circles. Phase IA (within the dashed line) is "metastable" in the sense that minor changes in the experimental conditions change the patterns qualitatively. The direction of the grooves is shown by the arrow.

isotropy on the driving force is generally monotonic.

As a possible explanation for our experimental results the following qualitative picture can be put forward. Because of the particular geometry we used there are two different kinds of couplings, parallel and perpendicular, which can produce anisotropy in the system. At low pressure, the bubble advances in the perpendicular direction by jumping from row to row: This mechanism makes the growth in the perpendicular direction easy. If the plate separation is relatively large (b/h > 1), the curvatures at low pressures are small and, consequently, the motion of the interface in the parallel direction is not affected very much by the grooves: The parallel coupling is small in this case. By increasing the pressure the difference between the velocities v and v' of Eq. (1) at a perpendicular tip decreases because the role of the grooves as perpendicular hindrance decreases. On the other hand, the characteristic curvatures in the parallel direction can become larger, comparable with 1/h, and the coupling in this direction increases, resulting in a larger effective anisotropy. If the separation of the plates is small (b/h < 1) the coupling in the parallel direction can be large for small pressures as well, because the fluid is forced to flow in the channels. The different coupling mechanisms in the two directions demonstrate that the effective anisotropy can depend on its

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(a) (b) (c) (d) (e) (f)

FIG. 2. Typical patterns obtained by injecting air into glycerin in a radial Hele-Shaw cell with imposed (left-right) uniaxial anisotropy. (a) phase I, (b) phase II, (c) phase IA (stable), (d) phase IA (metastable), (e) phase III, (f) phase IV. The parameter values corresponding to these patterns are indicated by filled circles in Fig. 1.

variables in a nontrivial way and this is the origin of the complicated morphological phase diagram of Fig. 1.

The concept of effective anisotropy is expected to be useful in the further studies of tip instability. As an example, one should mention the very recent experiments by Couder *et al.*,¹⁷ who obtained stable finger tips by putting a small separate bubble on the tip of a growing finger in a nonprepared Hele-Shaw cell. In this case the small bubble introduces the anisotropy and, again, a new kind of coupling is responsible for the sufficiently high value of A_{eff} . Our results indicate that the role of local anisotropy versus the driving force in realistic systems is perhaps extremely complex and it is the details of the given growth process which determine the shape of the actual patterns. Very recent results on electrodeposition support this conclusion. In the experiments reported by Sawada¹⁸ increasing the undercooling (the driving force) resulted in dendritic growth of the originally DLA-like zinc deposits, while Kaufman, Nazzal, Melroy, and Kapitulnik¹⁹ found a crossover from the stable dendritic to a tip-splitting mode as the driving force was increased in the experiments on polymerization by electrochemistry. These results together with the surprising variety of morphological phases observed in our experiments demonstrate that the explanation of the role of driving force requires further theoretical and experimental efforts. 2356

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