

Don't know what it is

Final Report for Grant DE-FG02-84ER45131

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In this Final Report we will survey the results of the entire project and add more detail on the completion of work since the last progress report. Similarly, we append to this report only the publications that have appeared since the last progress report.

The work performed on this grant has contributed to many aspects of our rapidly evolving understanding of the properties and behavior of driven interfaces. To present it in summary form, it seems best to organize it into two broad categories: 1. Interfaces driven in Hele-Shaw cells and variants thereof, and 2. Gels, colloids, and polymer solutions as complex media for interface growth and motion.

I. Interfaces driven in pure and perturbed Hele-Shaw cells.

In our earliest work, we and others showed that various regularities could be observed as an initially flat interface developed a Saffman-Taylor fingering pattern in a Hele-Shaw cell and that those regularities could in many features be collapsed onto more general pattern evolution by using suitable dimensionless numbers. Thus these flows could be understood through variations in driving force, density contrast, and wetting properties even though no analytic solutions existed for the flows as they developed from the initially flat interface to a steady state (rectangular cell) or toward the very late time continuous tip-splitting (radial cell). Numerical calculations of these flows done by computational groups around the world were able to reproduce many of the observed features of our flows, although again it was not possible to develop analytic solutions to the important transient behavior we were observing.

Once the properties of the pure Hele-Shaw flows were carefully studied, and the limits of our knowledge of their universal features established, we proceeded to introduce perturbations to these flows to measure the changes induced in the pattern development with as much control as we could establish. Our "race track", which formed a very regular array of steady state channel fingers and then introduced them to an open rectangular channel, allowed us to view finger competition under much better control than had been achievable in other systems and led to observation of a number of interesting regularities as the patterns developed in the race down the length of the cell. When we introduced a gradient in the size of the cell gap, we found non-universal features that could be attributed to the wetting properties of the cell plates and that greatly altered the development of patterns, with a strong dependence on whether the gap was decreasing or increasing. The theorists working with us were able to show significant restoration of understandable behavior by addressing the wetting issues. These studies generally showed remarkable regularity under a wide variety of perturbations, good news

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for the hope of ultimately understanding liquid-phase processing where interfaces are present and both liquids are simple liquids.

We then went on to investigate pattern formation in flows involving non-Newtonian fluids and flows over "microscopic" etched patterns (the latter mimicking what one might expect from some features present in crystal growth from a melt). The viscoelastic fluids chosen for non-Newtonian flows were all polymer solutions and all of them showed much richer patterns than are exhibited by simple liquids. Aqueous solutions of linear homopolymer driven into water in a radial Hele-Shaw cell showed a wide variety of patterns which could be organized according to polymer density, phase viscosity, cell gap and driving force to show significant regularity despite the complexity of the patterns. In all cases, these were clearly viscous fingering patterns. When polymer architecture was changed, however, it became possible to observe distinctly different behavior. Patterns that showed many of the features of brittle fracture could be induced above some threshold of driving force, and in fact the threshold for transition from viscous fingering to fracture could be understood over a wide variety of fluid viscosity, cell gap and polymer molecular weight as long as the polymer contained associating groups. Much higher dimensionless driving forces failed to show fracture in solutions of homopolymer. In some of our most recent work (D.H. Vlad, Jordi Ignés-Mullol and J.V. Maher, Phys. Rev. E60, 4423(1999)), simultaneous measurement of finger tip velocity and pressure at the injection point allowed us to relate the tip velocity to the pressure gradient through a "finger mobility" and to model the flow discontinuities (jumps in finger tip velocity characteristic of fracture propagation) by using a nonmonotonic dependence between a characteristic shear stress and the shear rate at the finger tip. The simple model captured most features of the data with surprising accuracy, again boding well for eventually using this level of understanding to improve liquid phase processing of interesting materials. In another recent experiment, we have separated yield stress features from viscoelasticity (both of which might have been present in all our associating polymer solutions) by using a Boger fluid. No fracturelike instabilities were observed, suggesting that the fracture-like features observed in associating polymer solutions were produced by extreme shear thinning rather than from viscoelasticity. This recent work was reported in D.H. Vlad and J.V. Maher, Phys. Rev. E61, 5439(2000).

When one plate of a radial Hele-Shaw cell is etched microscopically to use the effect of the etching on the wetting behavior of the interface as a pattern is forced through the cell, it has been shown that patterns very like the dendrites seen under some circumstances in directional solidification can be formed. We have shown that the dendritic tips formed over such lattices can be scaled simply to relate the tip development to a dimensionless flow time. In our most recent work, we introduced random imperfections in the microscopic cells over which the patterns were forced to flow. The more the imperfections the more disorderly and ramified the patterns produced, but the tip scaling persisted with little or no alteration (E.L. Decker, Jordi Ignés-Mullol, A. Baratt and J.V. Maher, Phys. Rev. E60, 1767(1999)).

II. Gels, Colloids and Polymer Solutions as Complex Media for Interface Growth and Motion.

Motivated by a desire to observe interfaces encountering complex barriers as they attempt to travel, grow and evolve, we used a number of test systems. First, we imprisoned a critical binary liquid mixture with a consolute point near room temperature in a gel, attempting to use the polymer network of the gel as a prison to trap the growing regions of separated phases as we cycled the temperature and forced the originally one-phase solvent to phase separate. We found that many of the system's observed features could be understood by assuming, as DeGennes suggested, that the polymer network imposed a random field on the separating liquid mixture. Other features of the behavior of these gels, however, were more complex and reminiscent of wetting systems, leading us to move to other kinds of microscopic wetting—namely, the behavior of colloidal particles in liquid mixtures undergoing phase separation or near critical points. These colloidal systems showed fascinating reversible aggregation near a critical point, holding out the hope of useful insights into aggregation and purification of particles by partitioning, but leaving us with important questions as to the relative importance of polarization of the liquid mixture near the walls of a colloidal particle and reaction of the surface of the particle to the local composition of the mixture. We have not as yet been able to get past this ambiguity. Moving on to attack the issue of how a dissolved polymer can affect the solvent's behavior at an interface, we imprisoned diblock copolymer at fluid-fluid interfaces, in some cases where one phase of fluid was a good solvent for one block and the other for the other block, in other cases where one fluid was a good solvent for one block and the other was better but not a good solvent for the other block. We then observed the behavior of capillary waves at these interfaces as a function of the polymer molecular weight, polymer number density at the interface and capillary wave frequency. We then observed the behavior of similar interfaces when decorated with triblock and pentablock copolymers. In all cases our capillary waves were driven mechanically and it was possible to extract appropriate longitudinal and transverse elasticities and viscosities using the known dispersion relations for capillary waves. These data are intriguing, showing saturation at rather low molecular number densities and thus suggesting that extra polymer rather quickly goes into micelle-like structures. The behaviors of the various interfaces were quite regular and give promise of good predictability for processing issues once more information is gathered. Unfortunately, at present it is not easy to relate the macroscopic interfacial properties to the molecular architecture and conformation of the polymer.

III. Conclusion.

The work done under this grant has contributed very significantly to the international effort to learn about nonlinear and pattern forming systems. Our data have been very influential as theoretical and computational groups have struggled to understand the dynamics of nonlinear processing steps and the structure-property relations of complex materials. The Hele-Shaw cell has been especially productive during this period of intense interest in "simple" nonlinear pattern formation, providing the simplest and best understood pattern forming system which could then be complicated with changes of

boundary condition or changes of fluid property to test in a controlled way the effect on pattern formation of added physical/mathematical complexity.

IV. Papers Published Since the Last Progress Report

1. Effect of Lattice Defects on Hele-Shaw Flow over an Etched Lattice, E. L. Decker, Jordi Iñes Mullol, A. Baratt and J. V. Maher, Phys. Rev. E60, 1767 (1999).
2. Velocity Jump Instabilities in Hele-Shaw Flow of Associating Polymer Solutions, D. H. Vlad, Jordi Iñes Mullol and J. V. Maher, Phys. Rev. E60, 4423 (1999).
3. Tip-splitting instabilities in the channel Saffman-Taylor flow of constant viscosity elastic fluids, D.H. Vlad and J.V. Maher, Phys. Rev. E61, 5439 (1999)