# Interfacial Dynamics in Complex Fluids

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## 1 Overview of the Field

Complex fluids refer to those with internal microstructures whose evolution affects the macroscopic dynamics of the material, especially the rheology [7]. Examples include polymer solutions and melts, liquid crystals, gels and micellar solutions. Such materials often have great practical utilities since the microstructure can be manipulated via processing flow to produce outstanding mechanical, optical or thermal properties. A good example is main-chain liquid-crystalline polymers (LCPs). Their molecular backbone is rodlike, with a degree of rigidity, such that the polymer assumes an anisotropic orientational order due to spontaneous alignment of the molecules. This order, further enhanced by extensional flows, leads to exceedingly high strength and modulus in the Kevlar fiber, a commercially successful product of du Pont.

An important way of utilizing complex fluids is through composites. By blending two immiscible components together, one may derive novel or enhanced properties from the composite, and this is often a more economical route to new materials than synthesis. Moreover, the properties of composites may be tuned to suit a particular application by varying the composition, concentration and, most importantly, the interfacial morphology. Take polymer blends for example [11]. Under optimal processing conditions, the dispersed phase is stretched from drops into a fibrillar morphology. Upon solidification, the long fibers act as in situ reinforcement and impart great strength to the composite. The effect is particularly strong if the fibrillar phase is liquid crystalline [2]. The dispersed phase may also be solid as in colloidal dispersions, or gas as in thermoplastic foams. From a scientific viewpoint, the essential physics in all such composites is the coupling between interfacial dynamics and complex rheology of the components.

Despite their practical importance, our current knowledge of two-phase complex fluids is very limited. The main difficulty is that these materials have a myriad of internal boundaries, which move, deform, break up and reconnect during processing. This leads to a seemingly intractable mathematical problem, and also hampers experimental observation and measurement. A secondary difficulty is that the rheology of each component alone is highly complex, with the internal microstructure coupled with the flow field. Thus, these materials feature dynamic coupling of three disparate length scales: molecular conformation inside each component, mesoscopic interfacial morphology and macroscopic hydrodynamics. An understanding of the interfacial dynamics in complex fluids is a major fundamental challenge as well as a significant practical need. The problem involves several traditional disciplines: mathematical modeling, numerical computation, soft-matter physics, fluid mechanics, material science and engineering. An objective of the workshop is to explore new research directions in the context of multi-disciplinary interactions.

# 2 Recent Developments and Open Problems

To date, mathematical modeling and numerical simulations of two-phase flows have followed two distinct approaches, each with its own advantages and limitations. In the first, and conceptually straightforward, approach, an interface is treated as a sharp boundary of zero thickness on which boundary conditions are matched to couple the two phases. The interfaces are handled by employing a moving mesh that has grid points on the interfaces, and deforms according to the flow on both sides of the boundary [4]. A practical difficulty of this approach is to reconcile the Eulerian framework naturally suitable for the bulk flow and the Lagrangian description of the moving boundaries. Therefore, keeping track of the moving mesh entails a computational overhead, and large displacement of internal domains causes mesh entanglement as happens when one drop overtakes another. Besides, topological changes during interfacial rupture and reconnection cannot be handled in a rational way.

The second approach seeks to regularize the interface so the problem can be solved in a purely Eulerian framework on fixed grids. These include the volume-of-fluid method, the front-tracking method, the level-set method and the phase-field method [6, 8, 10, 14]. Instead of formulating the flow of two domains separated by an interface, these methods represent the interfacial tension as a body-force or bulk-stress spread over a narrow region covering the interface. Then a single set of governing equations can be written over the entire domain, and solved on a fixed grid in a Eulerian framework. The computational bottleneck is usually resolution of the interfacial structure. So far, the application of these methods has been mostly limited to Newtonian fluids.

Therefore, although a general mathematical formalism for treating two-phase complex fluids is not yet available, the basic elements for constructing such a methodology have emerged in recent years. The open problems are to integrate such elements into versatile and efficient computational algorithms, and to apply these to interfacial flows of complex fluids that are of interest to physicists and engineers. The latter may include, for instance, impact of a liquid on an interface, coalescence and rupturing of interfaces, the effects of surfactants and macroscopic manifestations of mesoscopic morphology in emulsion rheology.

# 3 Highlights of the Scientific Program

The workshop attracted the mathematical authorities on each class of methods as well as leading physicists and engineers known for their expertise in complex fluids and interfacial dynamics. In the meantime, we have also included a significant number of younger researchers, with the aim of providing them with essential tools and techniques that they would not normally encounter within their own institutions and research groups. Aside from the formal presentations and discussions, attendants took advantage of the many opportunities for informal interactions, which many considered a major benefit of the workshop over the usual scientific conferences. The presentations fall into six topical areas, within each we give a brief description of one especially interesting talk, with references where possible.

## 3.1 Drop dynamics in complex fluids

The major numerical methods for computing drop deformation were represented at the workshop: volume-of-fluid, level-set, phase-field and the sharp-interface boundary-integral method. The presenters discussed the start-of-the-art of these methods and simulations in drop deformation, coalescence and breakup in viscoelastic, liquid crystalline and surfactant-laden fluids. In addition, experimental observations were reported on coalescence between drops and interfaces, and comparisons with numerical simulations were made for the partial coalescence cascade.

As a highlight of this group of talks, Jianjun Xu discussed his work on *A level-set method for interfacial flows with surfactant* [13]. Using a level-set representation for fluid interfaces with surfactants, the method is based on an Eulerian formulation and couples a semi-implicit discretization of the surfactant equation with the immersed interface method for the flow-solver. 2D simulations reveal the effects of surfactants on single drops, drop-drop interactions and interactions among multiple drops in Stokes flow under an applied shear.

## 3.2 Microfluidics in complex fluids

Microfluidic devices manipulate small amounts of liquids through micron-sized channels, and have attracted a great deal of interest for their applications in analytical chemistry and biological analysis [1]. The talks in this area examined the interfacial instabilities that cause jet breakup and formation of uniform-sized micro-droplets in various flow geometries, as well as control strategies for rheologically complex fluids. For example, Boris Stoeber described *Visco-thermal flow instabilities of thermally responsive fluids* in microfluidic channels [9]. Using thermally responsive polymers that undergo reversible gelation as a result of viscous heating, the author observed unusual flow instabilities in pressure-driven flows through a microchannel. The reversible phase change of these thermally responsive fluids and their particular rheological behavior promise novel microflow control applications including active valving and passive microflow control based on viscous heating within the flow.

## 3.3 Mathematics and physics of the moving contact line

The moving contact line may be one of the most challenging problems in two-phase flows. In the conventional Navier-Stokes sharp-interface formulation, the contact line constitutes a non-integral singularity. In physical reality, the flow in the vicinity depends on the interplay between microscopic molecular forces and continuum-level hydrodynamics. The contributions on this topic ranged from experimental and theoretical studies of short-range interfacial forces to large-scale numerical simulations of forced wetting and two-fluid displacement in channel flows.

David Jacqmin discussed *Phase-field calculations of wetting failure and instabilities*, which employ a phase-field model to coarse-grain the microscopic physics and integrate it into a continuum simulation [5]. The model eliminates the apparent singularity at the wetting line and also captures the gross energetics of wetting. Two- and three-dimensional phase-field-Navier-Stokes calculations of liquid-liquid systems show wetting failure through tipstreaming and splitting instabilities. It is hypothesized that tipstreaming can be understood in part as a quasi-two-dimensional phenomenon.

## 3.4 Structural evolution in polymers and micelles

This topic attracted some ten contributions that dealt with liquid crystalline polymers, wormlike micellar solutions, block copolymers, surfactant solutions and monolayers. The methods of investigation included constitutive modeling, numerical simulation of flow-structural coupling, and experimental measurements. A particularly interesting talk came from Pam Cook entitled *Wormlike micellar solutions: A model and its predictions* [12]. Those so-called "living polymers" continuously break and reform their structure in solutions and exhibit a characteristic breakage time in addition to the network relaxation time. As a result, their rheology features a plateau in the steady state flow curve associated with the development of spatial inhomogeneities in the flow. The talk outlined a network-based constitutive model that incorporates the breaking and reforming of the micelles. Each micellar species is modeled as a nonlinear bead-spring dumbbell. Predictions of the model in transient and steady state shear and in extension are shown to agree with experiments.

## 3.5 Modeling of membranes and other biological systems

Theoretical models and numerical simulations were presented on the mechanical behavior of electro-elastic and biological membranes, charge-selective vesicles and white blood cells passing through the capillary network. The phase-field model, with a properly designed energy functional, was shown to yield a promising description for elastic membranes. In his talk titled *Diffusive interface modeling and numerical simulation of lipid membranes* [3], Qiang Du presented a series of works on the phase-field modeling and simulations of vesicle membrane deformations under the elastic bending energy. These include studies of full three dimensional energy minimizing configurations, effect of spontaneous curvature, interaction with background fluid flows, and multicomponent and open membranes. In addition, he also introduced an ingenious technique for retrieving topological information within the diffusive interface framework which may have broad applications.

### 3.6 Advances in numerical algorithms

This series of talks covered the latest progress in numerical algorithms for computing interfacial motion as well as the bulk flow of microstructured fluids. On the former, several types of methods have been discussed above in relation to simulating drop dynamics. The use of adaptive meshing to resolve diffuse interfaces has produced impressive results. Besides these, several lecturers expounded on techniques based on a mixed Eulerian and Lagrangian formulation. For the latter, the contributions ranged from a study of the well-posedness of viscoelastic flows as related to the high-Weissenberg number problem to closure approximation for the FENE fluid.

A highlight of this group of talks is perhaps *A compromise between the Eulerian and ALE approach to free boundary problems* by Peter Minev (http://www.math.ualberta.ca/ pminev/). His work attempts to produce a scheme that comprises the advantages of the two main approaches for discretization of free boundary problems in fluid dynamics: the Eulerian and the Arbitrary Lagrangian-Eulerian (ALE) approaches. His scheme resembles an ALE scheme because at each time step the free boundary is aligned with finite element faces. On the other hand, the structure of the grid remains unchanged. The algorithm uses a fixed background grid, but at each time step it finds the closest to the free boundary grid points and projects them onto the boundary without changing the structure of the grid. The time discretization is done by means of a velocity correction scheme which utilizes non-conforming elements for the projection step. The mass conservation is implemented by means of one Lagrange multiplier per each distributed phase. Finally, the scheme is validated on several well-known free boundary flows.

## 4 Outcome of the Meeting

The problem of interfacial dynamics in complex fluids is multi-disciplinary. But the work done by mathematicians, engineers, physicists and material scientists has largely been independent of one another. Researchers across the disciplines have so far seldom had the chance to interact and learn of each other's work. The greatest achievement of this workshop may have been its facilitating the exchange of ideas among researchers across traditional disciplines. More specifically, we can list the outcomes of the meeting as follows.

(1) A survey of the state of the art of "Interfaces in Complex Fluids" that highlights it as an important emerging area in mathematics and physical science. Experimentalists have described recent observations and measurements on processes such as moving contact-lines, interfacial rheology, Marangoni flows and self-assembly on interfaces. Theoreticians and numerical analysts have summarized the predictive capabilities of their models, including fixed- and moving-grid numerical methods, incorporating continuum and molecular rheological theories.

(2) Identification of the most pressing scientific issues. Experimentalists have listed phenomena that cannot be readily rationalized or explained, including interfacial rupture, thermally and chemically modulated tip-streaming and surface-directed self-assembly. Modelers and simulators have highlighted the need for multi-scale modeling of singular events such as moving contact-lines and topological changes.

(3) Facilitating inter-disciplinary exchange and collaborations. Out of the broad and thorough discussions of interfacial dynamics in complex fluids came a more comprehensive context for the individual problems each researcher has been tackling. In particular, younger scientists have benefited from interactions with more established experts and from the opportunity to integrate or distinguish themselves in this exciting multi-disciplinary community. Collaborations will develop, we hope, between researchers with complementary skills and expertise.

## 5 List of Participants

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