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Diffuse-interface simulations of drop coalescence and retraction in viscoelastic fluids

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Abstract

Drop dynamics plays a central role in defining the interfacial morphology in two-phase complex fluids such as emulsions and polymer blends. In such materials, the components are often microstructured complex fluids themselves. To model and simulate drop behavior in such systems, one has to deal with the dual complexity of non-Newtonian rheology and evolving interfaces. Recently, we developed a diffuse-interface formulation which incorporates complex rheology and interfacial dynamics in a unified framework. This paper uses a two-dimensional implementation of the method to simulate drop coalescence after head-on collision and drop retraction from an elongated initial shape in a quiescent matrix. One of the two phases is a viscoelastic fluid modeled by an Oldroyd-B equation and the other is Newtonian. For the parameter values examined here, numerical results show that after drop collision, film drainage is enhanced when either phase is viscoelastic and drop coalescence happens more readily than in a comparable Newtonian system. The last stage of coalescence is dominated by a short-range molecular force in the model that is comparable to van der Waals force. The retraction of drops from an initial state of zero-velocity and zero-stress is hastened at first, but later resisted by viscoelasticity in either component. When retracting from an initial state with pre-existing stress, produced by cessation of steady shearing, viscoelasticity in the matrix hinders retraction from the beginning while that in the drop initially enhances retraction but later resists it. These results and the physical mechanisms that they reveal are consistent with prior experimental observations.

Keywords: Phase-field method; Interfacial dynamics; Film drainage; van der Waals force; Disjoining pressure; Dynamic interfacial tension

1. Introduction

Drop dynamics is the key to understanding interfacial morphology in two-phase materials[1]. In nature and in industrial processes, many such materials have components that are complex fluids themselves, with internal microstructures whose evolution affects the macroscopic dynamics of the material, especially the rheology. Examples include polymer blends [2], polymer-dispersed liquid crystals [3] and various biological fluids [4].

Theoretical and numerical analysis of drop dynamics in complex fluids has to struggle with the dual difficulties of moving interfaces and microstructure-dependent rheology. Specifically, there is the interplay among microscopic, mesoscopic and macroscopic scales: (a) the internal microstructure, e.g., molecular conformation, inside each component; (b) the interfaces and (c) the flow field. The coupling between (b) and (c) alone is well studied for Newtonian drops [5]. Similarly, the (a)–(c) coupling is the subject of molecular constitutive theories in rheology (e.g., [6]). Having both (a) and (b) present in a flow problem is the novelty of this work.

Methods for solving moving-interface problems fall into two broad categories: interface tracking and interface capturing [7,8]. The former uses a moving mesh with grid points residing on the interface. The latter determines the position of the interface by using a scalar function, whose evolution is typically represented by an advection equation on a fixed

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grid. Conceptually, both treat the interface as a zero-thickness surface, though an essential ingredient in the fixed-grid methods is a numerical regularization that spreads the interfacial force over a volume.

Recently, Yue et al. [9] proposed a fixed-grid *diffuse-interface model* for two-phase flows of complex fluids. This model differs from other fixed-grid methods in that the interface is treated as *physically* diffuse (e.g., [10,11]). The interfacial position and thickness are determined by a phase-field variable whose evolution is governed by a mixing energy. This way, the structure of the interface is rooted in molecular forces; the tendencies for mixing and demixing are balanced through the nonlocal mixing energy. This contrasts the level set and volume-of-fluid methods, which replace the surface tension by a body force or stress as a *numerical* device to regularize the singularity. The significance of this physical root will become apparent when we discuss the rupture of the thin film separating two coalescing drops. When the interfacial width approaches zero, the diffuse-interface model becomes identical to a sharp-interface level-set formulation. It also reduces properly to the classical sharp-interface model.

In our context of two-phase complex fluids, another attraction of the diffuse-interface method is its capability of easily incorporating the rheology of microstructured fluids. This is by virtue of its energy-based variational formalism. As long as the conformation of the microstructure is describable by a free energy, this energy can be added to the mixing energy to form the total free energy of the multi-phase system. Then a formal variational procedure applied to the total free energy will give rise to the proper constitutive equation for the microstructured fluids in addition to the evolution equation of the phase field variable. Using the Frank distortion energy for a liquid crystal, Yue et al. [9] illustrated how interfacial dynamics and complex rheology can be included in a *unified* theoretical framework. Dissipative effects such as viscous stresses, of course, have to be accounted for separately, e.g., via the standard irreversible thermodynamic procedure [12] or by including Brownian motion in Hamilton's principle [13,14].

Yue et al. [9] have implemented the diffuse-interface method using a spectral representation, and presented preliminary numerical results to validate the theoretical model and the numerical method. The goal of this paper is to apply the method to physically interesting problems where it generates new insights into the physics. We will investigate two problems: drop coalescence after head-on collision and drop retraction from an elongated initial shape. The far-field matrix fluid remains quiescent in both problems.

2. Theory and numerical method

Yue et al. [9] have given a detailed derivation of the theoretical model, discussed its strengths and weaknesses and its relationship with other fixed-grid methods, and described the numerical scheme using spectral discretization. In this section, we will specialize the formalism for a mixture of a Newtonian and an Oldroyd-B fluid, and summarize the main features of the numerical procedure.

The Newtonian and Oldroyd-B components are immiscible except in a very thin interfacial region. This diffuse interface has a small but non-zero thickness, inside which the two components are mixed and store a mixing energy. An Oldroyd-B fluid consists in a dilute suspension of linear Hookean dumbbells in a Newtonian solvent [15]. The total free energy of the mixture thus comprises two parts: mixing energy of the interface and elastic energy for the dumbbells.

We introduce a phase-field variable ϕ such that the concentrations of the Oldroyd-B and Newtonian components are $(1+\phi)/2$ and $(1-\phi)/2$, respectively. For the mixing energy, we adopt the familiar Ginzburg-Landau form:

$$f_{\text{mix}}(\phi, \nabla\phi) = \frac{1}{2}\lambda|\nabla\phi|^2 + \frac{\lambda}{4\epsilon^2}(\phi^2-1)^2, \tag{1}$$

where λ is the mixing energy density with the dimension of force, and ϵ is a capillary width that scales with the thickness of the diffuse interface. As $\epsilon \to 0$, the ratio λ/ϵ produces the interfacial tension in the classical sense [16,9]. The evolution of ϕ is governed by the Cahn-Hilliard equation:

$$\frac{\partial\phi}{\partial t} + \boldsymbol{v}\cdot\nabla\phi = \gamma\lambda\nabla^2\left[-\nabla^2\phi + \frac{\phi(\phi^2-1)}{\epsilon^2}\right], \tag{2}$$

where $\gamma\lambda$ determines the relaxation time of the interfacial profile [9].

For a single dumbbell with a connector $\boldsymbol{Q}$, its elastic energy is $\frac{1}{2}H\boldsymbol{Q}\cdot\boldsymbol{Q}$, where H is the elastic constant. For an ensemble of dumbbells with configuration distribution $\Psi(\boldsymbol{Q})$, the average energy can be written as

$$f_{\text{d}} = \int_{R^3}\left(kT\ln\Psi + \frac{1}{2}H\boldsymbol{Q}\cdot\boldsymbol{Q}\right)\Psi\,\text{d}Q, \tag{3}$$

where k is the Boltzmann constant and T is the temperature, and the integration is over all possible configurations of $\boldsymbol{Q}$. Now the total free energy density of the two-phase system is:

$$f = f_{\text{mix}} + \frac{1+\phi}{2}nf_{\text{d}}, \tag{4}$$

where n is the number density of the dumbbells.

A variational procedure applied to the total free energy will yield the elastic stress tensor for the system. The stress tensor due to f_{mix} has been derived by Yue et al. [9], and here we will only consider the elastic stress due to the dumbbell energy f_{d}. We impose a virtual displacement $\delta\boldsymbol{x}$ on the material, which takes place instantaneously so that the dumbbells deform affinely with no slip between the bead and the surrounding fluid:

$$\delta\boldsymbol{Q} = \boldsymbol{Q}\cdot\nabla\delta\boldsymbol{x}. \tag{5}$$