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Simulation and experiments of droplet deformation and orientation in simple shear flow with surfactants

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Abstract

The deformation and orientation behavior of three-dimensional (3D) viscous droplets with and without surfactants is studied in simple shear flow using simulations and experiments. Two added amounts of surfactants are considered, along with a range of viscosity ratios and capillary numbers. The numerical method couples the boundary integral method for interfacial velocity, a second-order Runge–Kutta method for interface evolution, and a finite element method for surfactant concentration. The algorithm assumes a bulk-insoluble, nonionic surfactant, and uses a linear equation of state to model the relationship between the interfacial tension and the surfactant concentration on the drop surface. The algorithm was validated by comparison with other numerical results and good agreement was found. The experiments are performed in a parallel-band apparatus with full optical analysis of the droplet. The simulated and measured 3D steady-state shape of the ellipsoidal drops and their orientation are in reasonably good agreement. It was found that the surfactants have a greater effect on drop geometry for smaller viscosity ratios and that the deformation increases as the transport of surfactant becomes more convection dominated. It was also found that surfactants cause the drops to align more in the flow direction and that, for both clean and surfactant-covered drops, this alignment increases with viscosity ratio. Finally, simulations showed a wider distribution of surfactant on the interface for smaller viscosity ratios.

Keywords: Surfactants; Drop deformation; Emulsions; Simulations; Shear flow

1. Introduction

Immiscible multiphase fluid systems, such as emulsions, foams and polymer blends, are encountered in engineering applications in many industries such as foods, pharmaceuticals and plastics. During the processing of these systems, the droplets of the disperse phase are deformed, broken up, and oriented in the continuous phase, producing an evolving morphology or microstructure defined in part by the number density, size distribution, shape and orientation of the disperse phase droplets. The final morphology of the system helps to determine the material, mechanical, chemical, thermal and sensory properties of the finished product. An important step in improving and controlling manufacturing processes involving multiphase fluid systems is the investigation of single droplets under various conditions. The literature contains a wealth of theoretical, experimental and computational studies which establish the dependence of single drop behavior on a variety of flow, fluid and interfacial properties, such as the type of flow, the capillary number (i.e., the ratio of viscous forces to interfacial tension), and the ratio of disperse to continuous phase viscosity.

Studies in steady homogeneous flow fields include those of Taylor (1934), Rallison and Acrivos (1978), Rallison (1981), Grace (1982), Bentley and Leal (1986), Stone and Leal (1989a), Pozrikidis (1994), Cristini et al. (1998), Guido and Villone (1998), Guido et al. (1999), Li et al. (2000), Guido and Greco (2001), Cristini et al. (2003), Kaufmann et al. (2004), Megias-Alguacil et al. (2005), and Cristini and Renardy (2006). Assuming constant interfacial tension, Newtonian fluid phases with equal densities, and Stokes flow conditions, these studies (and many others) show that there is a critical capillary

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number above which a drop no longer reaches a steady shape, but continues to deform until it breaks up. Critical capillary numbers have been determined as a function of viscosity ratio in both shear and extensional flow fields. Not only are critical capillary numbers higher in shear flow than in extensional flow, but there is a critical viscosity ratio in shear flow, above which it is very difficult or impossible to break up a droplet in shear flow under the above assumptions. Moreover, as shown by Cristini et al. (2003), the critical capillary number in shear flow also becomes infinite as the viscosity ratio tends to zero.

When the homogeneous flow field changes in time, the drop behavior changes as well, and is generally more complicated. For example, sudden changes in strain rates can cause droplets to break up at lower strain rates, or capillary numbers, than is expected in steady flow (e.g., Rallison, 1980). Other studies of the effects of transient flow or complex flow on drop behavior include those of Stone et al. (1986), Stone and Leal (1989b), Tjahjadi and Ottino (1991), Bigio et al. (1998), Godbille and Picot (2000), Guido et al. (2000), Feigl et al. (2003), Windhab et al. (2005), Zheng et al. (2005), and Sibillo et al. (2006). The effects of inertia on a drop behavior have also been studied (e.g., Renardy and Cristini, 2001a,b; Renardy et al., 2002b) as have the effects of non-Newtonian fluid phases (e.g., Greco, 2002; Guido et al., 2003a,b; Dressler and Edwards, 2004a,b; Maffetone and Greco, 2004). Review articles include those of Cristini and Tan (2004) and Guido and Greco (2004).

The studies cited above have been conducted for droplets with a clean interface. If surfactants are present, then drop behavior also depends on the evolving distribution of surfactant on the interface, which produces temporal and spatial gradients in interfacial tension. While there have been many studies of drop behavior without surfactants, the behavior of droplets with surfactants has been much less studied. Experimental studies include those of Phillips et al. (1980), Janssen et al. (1997), and Megias-Alguacil et al. (2006), and theoretical and numerical studies include those of Stone and Leal (1990), Milliken et al. (1993), Milliken and Leal (1994), Pawar and Stebe (1996), Li and Pozrikidis (1997), Eggleton and Stebe (1998), Eggleton et al. (1999), Eggleton et al. (2001), Renardy et al. (2002a), Drumright-Clark and Renardy (2004), Kruijt-Stegeman et al. (2004),and Vlahovska et al. (2005).

Many of the numerical studies use the boundary integral method to compute interfacial velocity, coupled with a finite difference method for computing surfactant concentration. Other methods that are used include those of Kruijt-Stegeman et al. (2004), who use the finite element method for both quantities, and Li et al. (2000) and Renardy et al. (2002a), who use a volume-of-fluid method to track the interface, with a continuous surface stress formulation to describe the interfacial tension, and a projection method to solve the Navier–Stokes equations. Moreover, most numerical studies have been performed for insoluble surfactants (e.g., Stone and Leal, 1990; Pawar and Stebe, 1996; Li and Pozrikidis, 1997; Eggleton et al., 1999; Drumright-Clark and Renardy, 2004; Kruijt-Stegeman et al., 2004; Vlahovska et al., 2005) where there is no net movement of surfactant molecules to or from the interface, while the effect of surfactant solubility has been considered in Milliken and Leal (1994) and Eggleton and Stebe (1998). For computational efficiency and to reduce the parameter space, most of the numerical studies above have taken a viscosity ratio of one. The exceptions are Milliken et al. (1993) and Milliken and Leal (1994), who considered multiple viscosity ratios, and Renardy et al. (2002a), who considered a viscosity ratio of 0.05.

Finally, the majority of studies have considered drops in axisymmetric extensional flow. There have been far fewer studies of three-dimensional (3D) surfactant-covered drops in simple shear flow. One of the earlier studies was that of Li and Pozrikidis (1997), who simulated drop deformation in shear flow using the boundary integral method and a linear equation of state for an insoluble surfactant. The viscosity ratio was restricted to one and inertia was neglected. More recently, Renardy et al. (2002a), Drumright-Clark and Renardy (2004) and Vlahovska et al. (2005) also simulated 3D surfactant-covered drops in simple shear flow using a linear equation of state for an insoluble surfactant. Each of these studies also looked at a single viscosity ratio. Vlahovska et al. (2005) considered a viscosity ratio of one, and compared the results of boundary integral simulations with the results of a perturbation solution which they derived for the deformation of a surfactant-covered drop in any linear flow field. The effect of inertia on surfactant-covered drops in simple shear flow was considered by Renardy et al. (2002a) and Drumright-Clark and Renardy (2004) for a viscosity ratio of 0.05 and 1, respectively, using a volume-of-fluid approach.

The above studies have yielded valuable information about the effect of surfactants on droplet deformation and breakup. For example, surfactants have the greatest effect on droplets with a lower viscosity ratio, since the Marangoni stresses hinder the interfacial velocity, causing the system to act essentially as if the drop had a higher viscosity (e.g., Milliken et al., 1993; Milliken and Leal, 1994). On the other hand, surfactants have little effect on droplets with higher viscosity ratios (e.g., Milliken et al., 1993; Milliken and Leal, 1994; Janssen et al., 1997). In general, it has been found that, once surfactant is present on a drop, there are two competing processes which govern its effect on drop deformation: surfactant dilution, which works to hinder drop deformation, and surfactant convection, which works to enhance drop deformation. The former is due to the increase of interfacial area as the drop deforms, which tends to decrease surfactant concentration, resulting in increased interfacial tension and hence smaller drop deformation. Surfactant convection is due to the transport of surfactant along the interface by flow. This tends to increase surfactant concentration toward the tips of the drop, which decreases interfacial tension there, resulting in increased drop deformation. Whether the surfactant effect is dilution-dominated or convection-dominated depends on several factors, such as magnitude of Marangoni stresses, initial interfacial tension, surface saturation, surfactant interactions, viscosity ratio, and capillary number (e.g., Stone and Leal, 1990; Pawar and Stebe, 1996; Li and Pozrikidis, 1997; Kruijt-Stegeman et al., 2004).

The goal of the present study is to investigate, through simulations and experiments, the effect of surfactants on the