Commun. Comput. Phys.
doi: 10.4208/cicp.090310.020610a

Vol. **10**, No. 2, pp. 453-473
August 2011

Numerical Study of Surfactant-Laden Drop-Drop Interactions

Jian-Jun Xu[1,*], Zhilin Li[2], John Lowengrub[3] and Hongkai Zhao[3]

[1] *School of Mathematical and Computational Sciences, Xiangtan University, Xiangtan, Hunan 411105, China.*
[2] *Department of Mathematics, North Carolina State University, Raleigh, NC, 27695, USA.*
[3] *Department of Mathematics, University of California Irvine, Irvine, CA, 92697, USA.*

Received 9 March 2010; Accepted (in revised version) 2 June 2010

Communicated by Pingwen Zhang

Available online 28 April 2011

Abstract. In this paper, we numerically investigate the effects of surfactant on drop-drop interactions in a 2D shear flow using a coupled level-set and immersed interface approach proposed in (Xu et al., J. Comput. Phys., 212 (2006), 590–616). We find that surfactant plays a critical and nontrivial role in drop-drop interactions. In particular, we find that the minimum distance between the drops is a non-monotone function of the surfactant coverage and Capillary number. This non-monotonic behavior, which does not occur for clean drops, is found to be due to the presence of Marangoni forces along the drop interfaces. This suggests that there are non-monotonic conditions for coalescence of surfactant-laden drops, as observed in recent experiments of Leal and co-workers. Although our study is two-dimensional, we believe that drop-drop interactions in three-dimensional flows should be qualitatively similar as the Maragoni forces in the near contact region in 3D should have a similar effect.

PACS: 47.55.D-, 47.55.dk, 47.55.N-

Key words: Surfactants, drops, coalescence, surface tension, Marangoni effect, level-set method, immersed interface method, Stokes flow.

1 Introduction

The microstructure of emulsions and polymer blends depends strongly on the coalescence events that occur during processing. Surfactants and compatibilizers (block co-polymers) are often used to prevent coalescence events and stabilize drop size distributions.

*Corresponding author. *Email addresses:* jjxu21@xtu.edu.cn (J. Xu), zhilin@math.ncsu.edu (Z. Li), lowengrb@math.uci.edu (J. Lowengrub), zhao@math.uci.edu (H. Zhao)

Flow induced coalescence of drops in a viscous fluid has historically been modeled and described as taking place in three consecutive stages (see e.g., [7,12,15,19]). The first stage is the approach and collision of drops, starting from an initially large separation. The second stage is film drainage, which commences when the separation distance is asymptotically small compared to the drop radii. During this stage, the relative translational motion of the drops is largely arrested, and the motion consists of rotation of the two-drop pair in the flow, with simultaneous thinning of the thin film that separates the two drops. As the film thins, the distance between the two drops may become small enough for non-hydrodynamic attractive forces such as van der Waals attractive force to trigger an instability leading to film rupture. When this occurs, this third stage leads rapidly to coalescence. The theory of film drainage for clean drops was reviewed by Chesters [3]. In the absence of van der Waals forces, the interacting drops reach a minimum distance as a consequence of a balance of the compressional force imposed by the flow and the repulsive lubrication force in the thin film separating the drops. The drops then separate without coalescing. Coalescence therefore requires non-hydrodynamic forces such as the van der Waals force to rupture the film when the film is sufficiently thin.

The presence of surfactants introduces three main mechanisms during the drop interactions: the average reduction in surface tension associated to the presence of surfactants; nonuniform capillary pressure (normal stresses) on the drop surface due to non-uniform distribution of surfactants; Marangoni forces associated variation of surface tension along the interface.

The effect of surfactants on drop-drop collisions and coalescence has been investigated theoretically (e.g., [4, 5, 29]) and experimentally (e.g., [7, 12, 15, 19, 26, 47]). In a series of experiments, Leal and co-workers [12,15,19,47] found that surfactants can have surprising and non-monotonic effects on the coalescence of two drops for a wide range of viscosity ratios. In particular, for a small amount of surfactants, the critical Capillary number below which coalescence occurs decreases significantly compared to that for clean drops. When additional surfactant is added, the critical Capillary number increases towards that for clean drops. Further, a range of Capillary numbers, with a nonzero lower bound, is found for which coalescence occurs. When the initial offset of the drops is above a critical threshold, no coalescence is found below or above this range.

There has been much work on simulating the motion of a single surfactant-laden drop (or bubble) using various numerical methods including boundary integral (e.g., [2,9,23, 28,33,38]), volume of fluid (e.g., [8,13,20]), arbitrary Lagrangian-Eulerian (e.g., [10,11, 46]), front tracking (e.g., [16,17,30,39,48]), level-set [25,43,44], and diffuse interface [1,40] methods. However there are few numerical studies on surfactant-laden drop-drop interactions. Recently, Dai and Leal [7] used a boundary integral method to study head-on collisions of surfactant-laden drops in a biaxial extensional flow. In addition to finding that the interface is immobilized in the near contact region, they also found that the hydrodynamic force pushing the drops together is increased by the Marangoni immobilization of the interface outside of the thin film of fluid separating the drops. This causes