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On Diffuse Interface Modeling and Simulation of Surfactants in Two-Phase Fluid Flow

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Abstract. An existing phase-field model of two immiscible fluids with a single soluble surfactant present is discussed in detail. We analyze the well-posedness of the model and provide strong evidence that it is mathematically ill-posed for a large set of physically relevant parameters. As a consequence, critical modifications to the model are suggested that substantially increase the domain of validity. Carefully designed numerical simulations offer informative demonstrations as to the sharpness of our theoretical results and the qualities of the physical model. A fully coupled hydrodynamic test-case demonstrates the potential to capture also non-trivial effects on the overall flow.

AMS subject classifications: 76T30, 65M60, 65Z05

Key words: Phase-field model, Cahn-Hilliard equation, surface active agent, Ginzburg-Landau free energy, well-posedness.

1 Introduction

The presence of surface active substances may greatly affect the physical properties of fluid mixtures. Indeed, these effects are used critically in many important applications in everyday life; detergents and oil-water emulsions in food are two immediate examples.

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The fact that surfactants *lower* the surface tension is exploited in both of these cases: detergents make the water more ″wet″ and an emulsifying agent stabilizes an emulsion by preventing small droplets to coalesce.

For humans, probably the most critical everyday usage of surfactants is made in the *alveoli* in the lung, where *pulmonary surfactant*, amongst other things, prevents lung collapse at the end of expiration [22,26].

An interesting and very striking realization *in vivo* was reported recently in [32] where *chemotaxis* was implemented for small droplets of fluid in a bulk solution, physically contained in a maze. The net transport at the millimeter-scale and the subsequent solution to the maze, was achieved by a clever usage of surfactant and a pre-existing pH-gradient. Quite likely, such a constructive set-up could find applications in lab-on-a-chip manufacturing.

Consisting of hydrophobic ″heads″ and hydrophilic ″tails″, surfactant molecules have a strong preference to occupy sites at the water-fluid or water-gas interfaces. Below the *critical micelle concentration* (CMC), surfactants therefore adsorb efficiently to the interfaces where their physical effects become prominent. Above the CMC, additionally, spontaneous formation of stable groups of surfactants — *micelles* — occurs in the bulk solution [38].

Given the ″thermodynamical″ signature of surfactants; that is, the diffusion-limited flow into the interfaces and the spontaneous creation of highly regular micelles from an unordered state in the bulk, modeling through some kind of system's energy assumption is a tempting approach.

Ariel, Diamant and Andelman [1,9] have successfully postulated free-energy terms with theoretically convincing properties. Their approach is inherently *sharp* in that they set up equations for the *interface*, the *sub-surface region* and the *bulk*. A later work [38] shows that this methodology can be extended to include also the region above the CMC. A great feature with this type of modeling is the fact that all properties of the model result from a single postulated entity; the system's free energy.

Given the multitude of scales present and the complex coupling to hydrodynamics, numerical experiments become important as tools to gain a better understanding and a fuller physical insight.

In a so-called sharp interface method, the interface is considered to be infinitesimally thin and its exact location is represented either explicitly (front-tracking with e.g., Lagrangian markers), or implicitly (e.g., level-set [55] and volume of fluid (VOF) methods [28]). The evolution of the surfactant concentration on each interface can be described by a partial differential equation on this time-dependent manifold [47]. Techniques for solving this PDE and include insoluble surfactants in multiphase flow simulations have been developed based on several different interface representation techniques [3,28,31,33,42,55].

When the surfactants are soluble also in the bulk, source terms due to adsorption and desorption terms enter this PDE and this must be coupled to a PDE for the bulk concentration of surfactants with appropriate boundary conditions for the surfactant flux. See