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Direct Numerical Simulation of droplet formation processes under the influence of soluble surfactant mixtures

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ABSTRACT

The present study is concerned with Direct Numerical Simulation (DNS) of the formation of droplets under the influence of a multicomponent surfactant system, where coupling effects become important. The presented numerical method is based on an Arbitary Lagrangian Eulerian (ALE) interface-tracking method. Collocated second-order Finite Volume/Finite Area methods are employed to discretise the governing equations. Since strong coupling effects among the surfactant species are present, a block-coupled solution procedure is adopted for interfacial surfactant transport. Various sorption models are included. The method is verified by a set of analytical test cases and validated against experimental results regarding droplet formation processes as they are studied in interfacial science in the context of Profile Analysis Tensiometry (PAT) for determination of surface tension.

1. Introduction

Surfactants are amphiphilic molecules, the structure of which exhibits a soluble head and an insoluble tail. Surfactants are subject to ad- and desorption processes and accumulate at fluid interfaces. Once adsorbed to the interface, they influence the interfacial properties, in particular the surface tension. The distribution of surfactants is generally nonuniform as a consequence of advective and diffusive transport. Due to the changed surface tension, surfactants effectively cause significant changes in the deformation, coalescence and break-up behavior of bubbles and droplets. Moreover, often not a single surfactant but surfactant mixtures are present. The concentrations at the interface are non-dilute, giving rise to strongly coupled multicomponent transport processes.

Since the interfacial transport and the local ad- and desorption processes are not easily assessed by experimental investigations, Direct Numerical Simulation (DNS) can give valuable insights into the local distribution of the adsorbed surfactants and the corresponding local surface tension values.

Interface tracking methods resolve the fluid interface by sets of marker particles or surface meshes. Boundary Integral methods [1,2] have been used for simulation of insoluble [3–10] and soluble [11–15] surfactants on droplet or bubble deformation. Simulations considering insoluble and soluble surfactants on basis of a second-order isoparametric surface Finite Element method on moving meshes have been published by Ganesan and Tobiska [16,17]. Based on the Front Tracking method [18–21], Zhang et al. [22] introduced a 2d-axisymmetric method to account for soluble surfactants. While the surface transport equation for the adsorbed surfactants is solved on a surface grid, the surface source terms due to adsorption/desorption are evaluated on the Eulerian background mesh, introducing a surface indicator function to evaluate density and viscosity fields. Muradoglu and Tryggvason [23] applied a Finite Difference method on top of a surface mesh defined by a set of connected Lagrangian marker particles, which are also advected by a Front Tracking method. The method is also 2d-axisymmetric and has been applied to rising bubble scenarios by Tasoglu et al. [24]. Lai et al. [25] propose the application of an Immersed Boundary method [26–28] for the simulation of insoluble surfactants. A combination of Eulerian flow and Lagrangian interfacial variables is applied, linking the variable corresponding to the reference frames by a Dirac delta function. Arbitrary Lagrangian Eulerian (ALE) methods have been described in [29–31] and applied to interface tracking methods in [32–36]. An extension to handle insoluble surfactants has been published by Li [37] who applies a fractional step Finite Volume method on a surface, which is represented by a set of marker points. Hameed et al. [38] applied this method to investigate the influence of insoluble surfactants on bubble break-up processes. Tuković and Jasak [39] developed a Finite Area method (FAM) on arbitrary unstructured, i.e. polygonal, moving and deforming surface meshes to handle soluble surfactants. The method is second order accurate in space and time and which forms the basis of the present study.

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Tornberg and Engquist [40] showed a Segment Projection method which is capable to handle insoluble surfactants. Khatri and Tornberg [41] extended this in a recent publication to handle soluble surfactants. The method is applicable for two dimensional simulations.

In contrast to interface tracking methods, Interface capturing methods use implicit interface representations by introducing so-called marker or color functions. Level-Set methods [42–45] have been enhanced to handle both soluble and insoluble surfactants. Adalsteinson and Sethian [46] and Xu and Zhao [47] independently describe a method for solving partial differential equations on moving interfaces, based on the idea of extending the surface quantity within a narrow band around the interface. In a subsequent study [48], the interfacial transport method was coupled to a flow solver for incompressible Stokes flow. This method has been applied to simulate the influence of a insoluble surfactant on droplet–droplet interactions [49] and has been extended for three-dimensional problems in [50]. While the aforementioned Level-Set methods utilized an Immersed Interface method to directly enforce the jump condition at the interface, Xu et al. [51] propose a Level-Set Continuum Surface Force (CSF) approach to handle insoluble surfactants. Solving the surface partial differential equations on discrete levels representing the interface has been accomplished by Olshanskii and Reusken [52]. Volume-Of-Fluid methods [53–55] involving piecewise linear interface re-construction [56] have been extended by Renardy et al. [57] for an insoluble surfactant, which is defined in a narrow band around the interface. The method is applied to three-dimensional droplet deformation processes. Drumright-Clarke and Renardy [58] applied the same method to investigate droplet break-up processes. In both publications a linearized surface tension equation of state has been applied. James and Lowengrub [59] show an axisymmetric Volume-Of-Fluid method, incorporating a Langmuir surface tension equation of state. Instead of solving a transport equation for the surfactant concentration, the authors track the surfactant mass to enforce mass conservation. Alke and Bothe [60] construct an iso-surface based on the volume fraction field. Soluble surfactants are considered for which the sorption process is diffusion controlled, utilizing a Langmuir sorption isotherm. The method is applied to a rising bubble. All Volume-Of-luid methods mentioned so far use the so-called Continuum Surface Force model [61] to evaluate the surface tension force. Davidson and Harvie [62] combine the method of James and Lowengrub [59] with the axisymmetric Volume-Of-Fluid method of Rudman [63]. They also separate surfactant mass and the surface area but do not solve an evolution equation for the surfactant area; instead they utilize the surface area gained by the reconstruction algorithm. Recently, Fleckenstein and Bothe [64] proposed a simplified model, deriving a Navier-type jump condition including the effects of the Marangoni stresses and incorporating this into the one-field formulation of the momentum balance.

Hybrid methods combine either methods within each group or between groups, in an attempt to make use of the advantages of both. Regarding surfactants, Yang and James [65] use a Coupled Level-Set Volume-Of-Fluid (CLSVOF) method, initially introduced by Sussman and Puckett [66]. The evolution of the surfactant mass is tracked by applying an Arbitrary Lagrangian–Eulerian method. As in [59], surfactant mass instead of the surfactant concentration is tracked. Cernicos [67] proposes a hybrid Level-Set/Front-Tracking methods, which is capable to handle insoluble surfactants incorporating a linear surface tension equation of state.

An extension to handle soluble surfactants based on Smoothed Particle methods [68] has been proposed by [69]. Diffuse Interface methods [70] have also be enhanced to handle surfactants by [71–73] just to mention a few.

All above described methods focus on the influence of single surfactants, while the technical relevant aspect of surfactant mixtures has not been addressed. In the present study a Finite Volume based Arbitrary Lagrangian Eulerian (ALE) interface tracking framework [36] is utilized in combination with a Finite Area method [39] for the simulation of interfacial transport processes, focusing on the proper accounting of sorption processes. The study is based on the open source Computational Fluid Dynamics library OpenFOAM. We distinguish between slow (kinetic controlled) and fast (diffusion controlled) sorption processes as different implementation strategies for fast and slow sorption processes are required (and explained in detail below). Both strategies are validated by means of comparisons to analytical test cases. The method is applied to a droplet formation process under the influence of different surfactants. Droplet formation process describe a realistic experimental scenario from surface science [74] to determine the integral surface tension value for a surfactant contaminated droplet. The applicability of the method to describe droplet formation processes has been shown in [75]. Additional physical effects to be regarded for contaminated droplets are effects due to diffusion and convection as well as dilution effect due to the change in surface area.

2. Governing equations

The mathematical modeling for two-phase systems under the influence of soluble surfactants is based on a sharp interface representation. The interface is described by a surface of zero thickness, holding surfactant molar mass. The interface Σ divides the domain Ω into two sub-domains Ω_1 and Ω_2 (cf. Fig. 1). We assume incompressible fluids, isothermal conditions, the absence of phase change and chemical reactions as well as local thermodynamic equilibrium at the interface. The surfactant concentrations are assumed to be dilute in the bulk, while at the interface non-dilute concentrations need to be considered. In Section 2.1 we address the continuum mechanical balances, while in Sections 2.2 and 2.3 the model is closed by introducing the constitutive equations modeling the sorption processes.

2.1. Continuum mechanical modeling

The flow in the bulk phases is described by the incompressible Navier–Stokes equations [76]

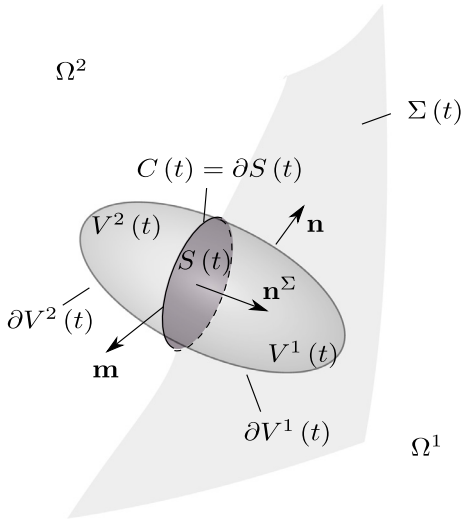

Fig. 1. Control volume for a two-phase system.