A Volume-of-Fluid-based method for mass transfer processes at fluid particles

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HIGHLIGHTS

- Rigorous derivation of the interfacial jump condition for chemical potentials.
- Conservative numerical method for DNS of conjugate mass transfer at fluid interfaces.
- 3D unsplit subgrid-scale model for mass transfer simulation at high Schmidt numbers.
- New validation case for mass transfer coupled to convection and diffusion.
- Computation of scaling law for local concentration boundary layer thickness.

ARTICLE INFO

ABSTRACT

This paper presents a VOF-based numerical method for simulating mass transfer across deformable fluid interfaces. The underlying mathematical model is based on continuum thermo-mechanics and allows for different solubilities of the species in the respective fluid phases, while volume changes due to mass transfer are neglected. The discontinuous changes in species concentrations at the interface are modeled by means of Henry's law which is shown to be a good approximation to the jump conditions at the interface in many cases. Within the numerical approach the concentration of the transfer component is represented by two separate variables, one for each phase. This is crucial in order to avoid artificial mass transfer. The physical mass transfer is modeled by an interfacial exchange term accounting for Henry's law, where two variants are compared. In addition, a subgrid-scale model is introduced for capturing steep concentration gradients appearing at the interface in convection dominated cases. The approach is validated by comparison of 3D numerical results for a spherical bubble rising at small Reynolds number in a glycerol–water mixture with an accurate solution based on the Hadamard–Rybczynski velocity field. Results obtained from 3D simulations for oxygen transfer from bubbles at moderate Reynolds numbers are compared with known Sherwood number correlations, showing good agreement. Finally, the dependence of the local concentration boundary layer thickness on Schmidt number is investigated.

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1. Introduction

Many industrial applications based on two-fluid systems involve mass transfer operations such as extraction, gas scrubbing and waste water treatment in bioreactors. For the design of efficient two-phase mass transfer reactors detailed knowledge of, say, bubble sizes and shapes, rise velocities, internal circulation, swarm behavior, bubble induced turbulence, bubble size distributions (including coalescence and break up) as well as the influence of contamination by surface active species are of fundamental importance. Optimization of mass transfer units additionally

requires profound understanding of the underlying mass transport and mass transfer processes.

Any detailed mathematical model for mass transport and mass transfer fails to admit exact analytical solutions since these processes depend on the complex flow patterns. Existing analytical studies rely on approximate solutions of the two-phase Navier–Stokes equations in axisymmetric cases, like axisymmetric Stokes flow around spherical bubbles; see Brauer, 1971, Sadhal et al., 1997, Clift et al., 1978 and the references given there. Even for such known velocity fields, exact solutions of the species equation are not available. Hence rigorous investigation of detailed mass transport and mass transfer requires numerical simulations. In fact, there are several numerical results concerning simulations of mass transfer in such simplified situations, like spherical bubbles/drops or axisymmetric domains, that lead to

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two-dimensional problems (Mao et al., 2001; Schulze and Kraume, 2002; Henschke and Pfennig, 1999; Piarah et al., 2001). While these studies are not sufficient to treat the general case, they are nevertheless useful for basic understanding and also for validation purposes. Experiments by Schlüter (2002) and Bork et al. (2003) show asymmetric distributions of the dissolved component in the wake of single bubbles rising in stagnant liquids in the case of sufficiently large Reynolds numbers. Consequently, a deeper understanding of mass transfer phenomena in general situations requires three-dimensional numerical simulations of the two-phase balance equations for mass, momentum and additional species equations without simplifying assumptions concerning, e.g., fluid flow or bubble shape. Due to the increase in computational power so-called Direct Numerical Simulations (DNS) become more and more feasible in which all relevant time and length scales of the solutions of the detailed continuum mechanical model are resolved.

The focus of the present work is on single bubbles rising in a Newtonian fluid with mass transfer from the bubble to the surrounding liquid. However, the numerical method presented here is – with the exception of the subgrid-scale technique – also applicable to mass transfer in liquid–liquid systems and to systems with multiple fluid particles. The concentrations of dissolved chemical components are not continuous at the interface. To handle this jump discontinuity numerically is a challenging task, since any relative movement between the interface and this discontinuity surface will result in artificial mass transfer.

VOF-based simulations of purely physical mass transfer across deforming interfaces without chemical reaction have been reported in Davidson and Rudman (2002), where a two-scalar method for mass transfer, including a jump discontinuity in concentration at the interface, has been developed for 2D planar and axisymmetric flow, and in Bothe et al. (2004). In the latter paper, transfer of oxygen from air bubbles rising in water or aqueous solutions is simulated, taking into account the realistic jump discontinuity of the oxygen profiles at the interface using a transformation of the concentration fields and a single transport variable. Darmana et al. (2006) performed 3D simulations of mass transfer at rising fluid particles for using the Front Tracking method. There, the transport resistance inside the fluid particle is neglected, i.e. a constant concentration value inside the bubble is assumed. Radl et al. (2007) performed 2D simulations of deformable bubbles and bubble swarms with mass transfer in non-Newtonian liquids using a semi-Lagrangian advection scheme. To prevent stability problems, a reduced density ratio between gas and liquid was used. 2D simulations of mass transfer using a front tracking approach were presented in Aboulhasanzadeh et al. (2012). In this article a 2D boundary layer approximation of the concentration boundary layer was used to reduce computational effort. Simulations with the approximated concentration profile are compared to fully resolved concentration fields and qualitative results are presented. Rather recently, first papers on numerical simulation of reactive mass transfer appeared. In Khinast (2001) and Khinast et al. (2003), the impact of single bubble wake dynamics on the reaction-enhanced mass transfer and on the yield and selectivity of the cyclohexane oxidation reaction is studied numerically for fixed shapes in 2D. In Khinast et al. (2005), reactive mass transfer at deformable interfaces is examined using a 2D front tracking/ front capturing hybrid method. In Deshpande and Zimmermann (2006a), a level set based method is used to simulate mass transfer across the interface of a moving deformable droplet. This method is extended to reactive mass transfer in Deshpande and Zimmermann (2006b), where an instantaneous chemical reaction occurs inside a moving droplet which leads to a quasi-stationary problem for the mass transfer. In Radl et al. (2008), 2D simulations are performed using a front-tracking method to investigate the effect of different Hatta and Schmidt numbers on the catalytic

hydrogenation of nitroarenes for single bubbles and bubble clusters. Based on the numerical approach in Bothe et al. (2004), reactive mass transfer with parallel consecutive reactions at rising bubbles has been analyzed in Bothe et al. (2009) by using local selectivities. The approach from Bothe et al. (2004) employs a single scalar field for every transfer component representing a normalized concentration for which the jump discontinuity is removed. The same approach has been adopted by Onea et al. (2009) for mass transfer from Taylor bubbles at low Schmidt numbers. While conceptually simple, this method has disadvantages concerning artificial mass transfer. More recently, 2D and 3D simulations of physical mass transfer have been presented in Francois and Carlson (2010). In this article, similar to Bothe et al. (2004), a single auxiliary variable was used holding a scaled concentration field also admitting representation of the species jump at the interface. Alternative to a single-scalar approach with a scaled concentration field, Marschall et al. (2012) derived a single-field model for interfacial species transfer starting from two species transport equations with corresponding jump conditions by using the conditional volume-averaging technique.

In Alke et al. (2009), a new VOF-based approach has been introduced which employs two scalar fields for each transfer species in order to avoid the drawbacks of the single-scalar methods. Fleckenstein and Bothe (2011) combined this method with an unsplit computation of the interfacial mass transfer flux and applied it to mass transfer at high Schmidt numbers. In the present paper, we provide the details required to implement the unsplit two-scalar approach together with an extensive validation. The method allows for different solubility of the dilute dissolved component in the respective fluid phases, given that local chemical equilibrium at the interface is attained and is determined by Henry's law. For the computation of the mass transfer across the interface, two principally different variants are employed. One is based on local equilibration of the chemical potentials in interface-carrying grid cells, while the other one exploits the diffusive flux to or from the interface. For increased accuracy in the case of thin concentration boundary layers at the surface of rising bubbles, a subgrid-scale model is added which performs a nonlinear flux correction in underresolved simulations. The overall method can be used for DNS of mass transport in general two-phase flows with deformable interfaces, including droplets, bubbles, falling films, etc. Here, the method is performed with an extended version of the highly parallelized code free surface 3D (FS3D, Rieber, 2004), which employs an advanced Volume of Fluid method.

The present paper is confined to physical mass transfer, while the method applies to reactive mass transfer as well; cf. Bothe et al. (2011a).

2. Sharp interface continuum mechanical model

We employ a continuum thermo-mechanical sharp interface model in which we consider a fluid particle which is immersed in a liquid. The deformable interface between the two phases is presented as a mathematical surface of zero thickness.

2.1. Two-phase Navier–Stokes system

We assume incompressible isothermal flow of two Newtonian fluids of constant density which are immiscible on the molecular scale. Inside the phases, balance of mass and momentum then leads to the Navier–Stokes equations, i.e.

$$\nabla \cdot \mathbf{u} = 0 \quad \text{in } \Omega \backslash \Sigma, \tag{1}$$

$$\partial_t(\rho \mathbf{u}) + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) + \nabla p = \nabla \cdot \mathbf{S} + \rho \mathbf{g} \quad \text{in } \Omega \backslash \Sigma \tag{2}$$