Multiscale computations of mass transfer from buoyant bubbles

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ABSTRACT

Mass transfer in the liquid phase of gas–liquid multiphase flows usually takes place at a considerably slower rate than the transfer of momentum, so mass flux boundary layers are much thinner than momentum boundary layers. In Direct Numerical Simulations (DNS) the resolution requirement for flows with mass transfer are therefore significantly higher than for flow without mass transfer. Here we develop a multiscale approach to compute the mass transfer from buoyant bubbles, using a boundary-layer approximation next to the bubble and a relatively coarse grid for the rest of the flow. This approach greatly reduces the overall grid resolution required.

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1. Introduction

Bubble columns are used in the chemical industry for a large number of processes that involve a gaseous and a liquid phase. Those include, for example, hydrogenation and oxygenation in many processes, as well as the synthesis of methanol (Deckwer, 1992; Furusaki et al., 2001). In bubble columns a gas is generally injected at the bottom of the column into a liquid or slurry (liquid and suspended particles) and as the bubbles rise, the gas diffuses into the liquid and reacts with dissolved reactants. The yields of these processes generally depend sensitively on the flow characteristics, including the behavior of the gas bubbles, and bubble column design, particularly the scale-up from laboratory results to production plants, remains difficult. Large amount of chemicals are processed in bubble columns and even a modest increase in yields can result in significant savings. Numerical simulations, where all continuum length and time scales are fully resolved, are starting to have a profound impact on our understanding of bubbly flows and the chemical reactions taking place in bubble columns. Raffensberger et al. (2003) examined a stationary bubble and a two-step reaction where a gas diffusing into the liquid reacts with dissolved species, followed by a reaction between the product of the first reaction and the dissolved

species to form an undesirable product, and showed that the yield and selectivity are strongly dependent on the details of the flow. For large deformable bubbles with a stationary wake, for example, products from the first (desirable) process are trapped in the wake and therefore have a strong tendency to react with gas from the bubble (forming the undesirable by-product). Including mass transfer and chemical reactions in numerical simulation is challenging due to the large disparity between the time-scales for diffusion of mass and diffusion of momentum in liquids. Liquids generally have a high Schmidt number so mass diffusion is much slower than momentum diffusion. Thus, mass boundary layers are typically much thinner than the flow scales and the resolution requirement for full simulations is generally directed by the mass transfer, rather than the fluid mechanics. Nevertheless, simulations using simplified two-dimensional flows have shown that the sensitivity seen for stationary bubbles carries over to flows with freely moving bubbles, although the exact dependency on the governing parameters is even more complex (Koynov et al., 2005).

A number of authors have recently computed the mass transfer from moving bubbles and drops. Those include Wang et al. (2008), Yang and Mao (2005), Ganguli and Kenig (2011a,b), and Hayashi and Tomiyama (2011), who used a level set methods to compute the bubble motion, and Davidson and Rudman (2002), Bothe et al. (2003), Onea et al. (2009), Francois and Carlson (2010), and Gupta et al. (2010), who used a volume of fluid (VOF) method. Other authors, such as Mao et al. (2001) and Figueroa and Legendre (2010) have used body fitted grids or

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unstructured meshes (Jung and Sato, 2005) to allow them to concentrate grid points near the bubble. In most of these papers the volume of the bubbles is assumed to remain constant, but Hayashi and Tomiyama followed the shrinkage of the bubbles as gas diffused to the liquid. Computations of mass transfer with more complex physics, such as soluble surfactant (Zhang et al., 2006; Muradoglu and Tryggvason, 2008; Booty and Siegel, 2010) and reactions are also starting to appear. In addition to the papers by Khinast and collaborators referenced above, simulations with reactions have been described by Bauer and Eigenberger (2001), Haroun et al. (2010), Wylock et al. (2011), and Dani et al. (2007). In spite of the challenges, Radl et al. (2008) have conducted simulations of the catalytic hydrogenation of nitroarenes under reasonably realistic conditions. While the study included a number of freely moving bubbles, the resolution requirements limited the simulations to two-dimensional systems. To accurately capture the mass transfer, a finer grid was used for the mass transfer and the reactions than for the fluid flow. Darmana et al. (2006) implemented the same approach in three-dimensional calculations of mass transfer from bubbles with relatively modest Schmidt number, $Sc = 1$.

Processes that take place on a scale much smaller than the dominant scale (defined below) are common in multiphase flows. For multiphase flows the "natural" or dominant small-scale is usually set by the balance of surface tension, viscosity and inertia. For bubble columns, for example, this scale determines the average bubble size. In most cases, the dominant scale corresponds to roughly where the appropriately chosen nondimensional numbers, such as Weber, Capillary, Ohnsorge, and Reynolds, are O(1) (and the key word here is obviously "appropriately"). Bubble collisions, coalescence and breakup can, however, lead to thin films, threads, and tiny drops that are much smaller than the average bubble size. Additional physical processes, such as mass transfer and reactions can also result in processes that take place on much smaller scales than the dominant ones. For small-scale processes, surface tension and viscosity generally dominate the dynamics, resulting in a relatively simple geometry and flow. These are exactly the situations where analytical or semi-analytical descriptions work well and the obvious strategy is to use such descriptions for these processes and couple them with the numerical description of the rest of the flow. This is, of course, an old idea. Boundary layer theory allows us to compute viscous drag for panel methods; the Hadamarad–Rybczynski solution for small drops in Stokes flows allows us to treat drops as point particles; and thin film models have been used to account for small-scale motion before (see Bossis and Brady, 1984; Davis et al., 1989; Ge and Fan, 2006, for example). It seems likely that coupling analytical description for small-scale process with numerical solutions may have a much broader applicability, but before a general procedure can be established, it is likely that additional examples are needed. In Thomas et al. (2010) we developed such an approach for the thin film between a drop sliding down an inclined wall.

Here we describe the development of a multiscale approach intended to allow us to incorporate mass diffusion into a direct numerical simulation (DNS) of bubbly flows. The approach is based on the observation that the mass concentration changes very rapidly near the gas–liquid interface, forming a thin mass-boundary layer that controls the diffusion of chemical species from the bubble to the liquid. Since the structure of this boundary layer is relatively simple, its shape and thickness can be predicted accurately with a boundary layer description. Thus, we use the model to predict how much mass diffuses from the bubble into the liquid and then solve an advection–diffusion equation for most of the domain, to determine where the mass goes. For mass transfer, Alke et al. (2010) and Bothe et al. (2011) have used the

exact solution of a one-dimensional diffusion problem, fitted to the computational results close to the bubble, to find the mass flux at the surface and Booty and Siegel (2010) have computed the evolution of a soluble surfactant by incorporating a singular perturbation analysis of the fluid next to the interface into a numerical solution of the interface motion. The approach of Booty and Siegel, although limited to Stokes flow, appears to be more closely related to the method presented here than that of Alke et al. In this paper we focus on the mass transfer only, leaving the reactions for later. Although the approach developed here is intended for use in DNS of many bubbles in turbulent flows, such as those presented by Lu and Tryggvason (2006, 2007, 2008), here we present results for two-dimensional flows only, focusing mostly on the initial rise of a single bubble.

2. Numerical method and problem setup

For buoyant bubbles the governing nondimensional numbers are the Eötvös number, the Archimedes number, and the ratios of the densities and the viscosities. These numbers are defined in the Nomenclature. For mass transfer we also need to include the Schmidt number, which measures how fast mass diffusion takes place compared to viscous diffusion of momentum. For liquids the Schmidt number is generally very large so mass boundary layers are thin compared to viscous boundary layers.

The simulations discussed in this paper are done using a front-tracking/finite-volume method where the governing equations are solved on a fixed, regular, mesh, covering both the ambient liquid and the bubbles (Unverdi and Tryggvason, 1992). The interface is marked by connected marker points that are advected with the fluid velocity and a marker function, constructed from the location of the interface, is used to set the density and viscosity of the different fluids. The marker points are also used to compute the surface tension. The method has been used earlier for a large number of simulations of multiphase flows, and both the method and various validation tests have been described in detail in several publications. For applications to bubbly flows, see Tryggvason et al. (2001), Bunner and Tryggvason (2002a,b, 2003), and Esmaeeli and Tryggvason (2005), for example. For other implementation of this method, see van Sint Annaland et al. (2005) and Hua et al. (2008), for example. The computational domain is fully periodic and we add a force equal to the weight of the fluid to prevent uniform acceleration in the direction of gravity. Here we present computations only for two-dimensional domains. We believe that extending the approach to three-dimensional flows is relatively straightforward.

The mass transfer is governed by an advection–diffusion equation for the mass fraction f

$$\frac{\partial f}{\partial t} + u \cdot \nabla f = D\nabla^2 f, \qquad (1)$$

and we take the value of f on the bubble boundary, f_0, to be given, without loss of generality. We assume that the fluid inside the bubble is well mixed and retains a uniform concentration throughout the simulation. For our case, where the mass diffusion in air is much higher than in water, this should be a reasonable assumption. Thus, it is only the solution outside the bubble that is of interest. As discussed by numerous authors (see Yang and Mao, 2005; Onea et al., 2009, for example) the solution to the original equations for the mass concentration in the gas and the liquid is discontinuous at the bubble surface, with the discontinuity given by Henry's law. However, by rescaling the concentration and the diffusion coefficient in the liquid, the solution can be made continuous. We also assume that the transfer of mass out of the bubble is so small that any change in the bubble volume can be