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## Effect of bubble interactions on mass transfer in bubbly flow

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### ABSTRACT

The effect of bubble interactions on mass transfer in a multi-bubble system is examined by numerical simulations. Since mass transfer in the liquid phase of gas-liquid multiphase flows usually takes place at a considerably slower rate than the transfer of momentum, the mass flux boundary layers are much thinner than the momentum boundary layers. In direct numerical simulations the resolution requirements for flows with mass transfer are therefore considerably higher than for flows without mass transfer. Here, we use a multiscale approach for the computations of the mass transfer near the bubble surface, in order to reduce the cost, and examine the effect of void fraction and bubble Reynolds number on the mass transfer from bubbles in periodic domains. Specifically, we compare results for a single bubble in a periodic domain with results for several bubbles in a larger domain with the same void fraction. It is shown that even though the average Reynolds number of freely moving bubbles drops after a while, in most cases the mass transfer from the bubbles increases slightly. When the bubbles start to wobble, in most cases the increase in bubble–bubble interactions compensate for the reduction in Reynolds number. © 2014 Elsevier Ltd. All rights reserved.

#### 1. Introduction

Bubble columns are one of the most important chemical processing units in the petro-chemical industry and are used to process millions of tons of material every year [10,14]. Bubble columns generally are simple vertical cylinders containing liquids with bubbles injected at the bottom. The bubbles rise due to buoyancy, interact and mix the liquid and the dissolved gases transferred from bubbles into the surrounding liquid, increasing the reaction rate. In spite of their simple design, the physics in bubble column reactors is complex. Today, we rely mostly on experimental data and semi-analytical correlations for the design of bubble columns, but with increased computational power and the ability to use direct numerical simulation for understanding of multiphase flow, computational studies should be able to help improve the design and optimization of bubble columns.

Considerable research has already been done on the mass transfer from a single bubble, see [9,11,13,15–17,19,20,23,29,30]. Most of those studies focused on two-dimensional or axisymmetric flow. Because of the large disparity between the length and time scale for the mass transfer compared to the momentum transfer, fully resolved three-dimensional calculations of mass transfer are challenging undertakings and those available in the literature are done for very low Schmidt number, Sc = O(1), [8,22]. In order to resolve the mass transfer at the interface of moving bubbles, and make the computational requirement of simulation of high Schmidt number bubbly flows manageable, several authors have presented different approaches; [3] 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 for Stokes flow, [4] 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 [2,1] solved a mass boundary layer equation at the interface for the evolution of the mass concentration next to the bubble and transferred the mass to the surrounding fluid when the boundary layer grew over a certain limit. This approach will be discussed further in the rest of paper.

While much has been done to understand mass transfer from a single bubble, both analytically and numerically, only a limited number of studies have looked at the interaction of bubbles and the effect of void fraction on the mass transfer. [21] studied mass transfer and reactions in a multi-bubble system and concluded that for mixing-sensitive reaction networks the interaction of the bubbles impacts the reaction selectivity significantly, and [24] performed simulations of single and multiple bubbles in order to investigate the effect of different Hatta and Schmidt numbers on the catalytic hydrogenation of nitroarenes. Both of these studies examined only two-dimensional systems.

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#### Nomenclature

$\delta_0$	boundary layer limit	$f_0$	mass fraction on the bubble surface	
0	boundary layer thickness	$J_{\infty}$	magnitude of f outside boundary layer, $f_{\infty} = 0$	
γ Εο	Fötvös number $Fo = \Lambda \log d^2 / \omega$	$\int \delta_0$	$a_0$	
LU	Nexton number $M_{a} = \Delta \rho g u_{b}^{2} / \gamma$	g V	mass transfer coefficient	
IVIO	Morton number, $Mo = g\mu_1 \Delta \rho / \rho_1^2 \gamma^3$	к т	ratios of the viscosity m ///	
Sc	Schmidt number, $Sc = \mu_l / \rho_l D$	III	Tatios of the viscosity, $m = \mu_g / \mu_l$	
Sh	Sherwood number, $Sh = kd_b/D$	$M_0$	total amount of f in the boundary layer	
$\mu$	viscosity	п	coordinate assigned normal to bubble surface	
$\rho$	density	$n_b$	number of bubbles	
$\sigma$	strain rate	r	ratios of the densities, $r= ho_g/ ho_l$	
Α	bubble surface area			
D	mass diffusivity	Subscrip	Subscripts	
$d_{b}$	diameter of the initially spherical bubble	g	gas	
f	mass fraction	ī	liquid	

Here, we use the multi-scale method presented in [2] and further validated with experimental data in [1] to study the mass transfer in bubble clusters, including the effect of different void fractions and Reynolds numbers.

#### 2. Computational method and multiscale model

#### 2.1. Numerical method

For buoyant bubbles the governing non-dimensional numbers are the Eötvös number,  $Eo = \Delta \rho g d_b^2 / \gamma$ , the Morton number,  $Mo = g\mu_1^4 \Delta \rho / \rho_l^2 \gamma^3$ , and the ratios of the densities,  $r = \rho_g / \rho_l$ , and the viscosities,  $m = \mu_{g}/\mu_{l}$ , where  $\rho$  and  $\mu$  are the density and the viscosity, respectively,  $\gamma$  is the interface surface tension,  $d_b$  is the bubble diameter, and g is the magnitude of gravity. Subscripts g and *l* show the respective properties of gas and liquid. For mass transfer we also need to include the Schmidt number.  $Sc = \mu_1 / \rho_1 D$ , where D is the mass diffusivity. The Schmidt number measures how fast mass diffusion takes place compared to viscous diffusion of momentum. For the liquid side of most gas-liquid multiphase flows, it is generally very large so mass boundary layers are thin compared to the viscous boundary layers. We also use two other non-dimensional parameters for the representation of the results, the Reynolds number,  $Re = \rho_l u_b d_b / \mu_l$ , and the Sherwood number,  $Sh = kd_b/D$ , where k is the mass transfer coefficient,  $k = (dm/dt)/(f_0 - f_\infty)A = (dm/dt)/f_0 n_b \pi d_b^2$ . Here, A is the bubbles surface area,  $f_0$  is the mass fraction at bubble interface,  $f_\infty$  is the mass fraction outside the mass boundary layer, dm/dt is the rate of mass transfer from the bubbles into the domain, and  $n_b$  is the number of bubbles. Time is non-dimensionalized with  $\sqrt{d/g}$ .

The simulations discussed in this paper are done using a threedimensional front-tracking/finite-volume method where the governing equations are solved on a fixed, regular mesh, covering both the liquid and the bubbles [28]. The interface is marked by connected marker points that are advected with the fluid velocity. 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. This method has been validated and used extensively for a large number of simulations of multiphase flows. For applications to bubbly flows, see [27,5–7,12], for examples. For other implementation of this approach, see [26,18]. The computational domain is fully periodic, and we add a force equal to the weight of fluid to prevent uniform acceleration in the direction of gravity.

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

r	ratios of the densities, $r =  ho_g /  ho_l$
Subscrip	ts
g !	gas liquid

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

and we take the value of f inside and 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 most cases, where the mass diffusion in the gas phase is much higher than in the liquid phase, 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, for example, [31,22]) 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. Including the change of volume of bubbles for gases with large solubility can be important and can be easily added to our method. However, here we do not include it in order to be able to simulate a statistically steady state, where the system characteristics does not change, for a long time.

#### 2.2. Subscale model for mass transfer

In order to implement an analytical description that resolves the boundary layer, captures its evolution, and allows us to predict the transfer of mass, we divide the simulated domain into two regions: inside the boundary layer and outside the boundary layer (rest of the domain). We use (1) for both region with the addition of a sink term in the boundary layer equation and a corresponding source term in the equation for the rest of the domain. These terms are zero except when the mass boundary layer thickness  $\delta$ , becomes larger than a boundary layer limit  $\delta_0$ , which is a predefined constant that we take to be equal to 2.5 times the grid spacing, h. When the boundary layer gets thicker than  $\delta_0$ , we transfer mass using the source term into the mass-grid field and follow the mass using the advection diffusion equation on the grid. The solution is essentially independent of  $\delta_0$  and the effect of changing  $\delta_0$  from 1 × *h* to 6 × *h* on the mass transfer is less than 1% [2, see]. Overall our approach is similar in spirit to [3] except that we solve the boundary layer equations using an approximate method resulting in a relatively simple (and fast) method. More details about the implementation of this embedded analytical description can be found in our previous paper, [1].

Here we assume that the mass concentration outside the bubble is sufficiently low so that it can be neglected when we compute the mass diffusion out from the bubble. In general the mass transfer should depend on the concentration in the surrounding liquid

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