Lattice Boltzmann study of mass transfer for Bretherton/Taylor bubble train flow

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Abstract

This work presents a thorough procedure of how to determine the volumetric mass transfer coefficient in the context of lattice Boltzmann simulations for the Bretherton/Taylor bubble train flow. In this work we address the situation when the hydrodynamic pattern changes from having a vortex in slug ($Ca < 0.7$) to not having it ($Ca > 0.7$) [1]. For the latter case the bubble shape is not symmetric and cannot be approximated through flat surfaces and circumferences as it is done in literature [3, 4]. For $Ca < 0.7$ where there is a vortex in the slug the tracer is well mixed. Thus, it is common to use periodic boundary conditions and the inlet/outlet averaged concentration as the characteristic concentration, Eq. 4. The later is not valid for flows where tracer is not well mixed, i.e. $Ca > 0.7$. This work through examination of different boundary conditions and different characteristic concentration definitions establishes the procedure to determine the volumetric mass transfer coefficient for flows with the capillary number $0.1 < Ca < 1.0$. With simulations of a few unit cells the connection between time (used in simulations) and space (used in experiments) domains is established. The following boundary conditions were examined: periodic boundary conditions, open boundaries, and a few unit cells open boundary simulations. It was shown that the time dependent average concentration to be a characteristic concentration produce robust results, though a bit overpredicted. The inlet/outlet flux averaged concentration [3] to be a characteristic concentration produce unreliable results in the range of capillary numbers specified. In comparison with works [3, 4] where only one unit cell was simulated, a few unit cells open boundaries conditions resemble physical situation most closely. However, they are harder to be conducted but provide the volumetric mass transfer coefficient based only on the spatial information for the capillary number $Ca > 0.7$. We show that all presented in literature strategies are extreme limits of one equation, Eq. 17. Finally, simulation results for different Peclet numbers are compared with analytical predictions presented by van Baten and Krishna [3] and shows a good agreement. The work is of use to people performing mass transfer numerical simulations for bubble train flow in microchannels and/or within the lattice Boltzmann model context.

1 Introduction

The monolith reactors during last years are getting more attention as a promising alternative to slurry reactors and trickle bed reactors [4, 5]. Reactors usually operate in the Bretherton-Taylor regime [6, 7] which describes a flow through a liquid medium of equally sized, long air bubbles, see Fig. 1. This flow regime is characterized by surface tension dominance over inertia and viscous effects, and by comparatively small gas flow velocities [8]. Due to surface tension dominance bubble train flow exhibit advantageous properties which can not be achieved in macroscopic counterparts: liquid thin films [6] between bubbles and walls remarkably enhance mass transfer from gas and walls to liquid; the plug flow regime occurring in monolith reactors allows to perform chemical reactions in slugs only [4]. Moreover, the low slip velocity between gas and liquid is utilized in the experiments to measure liquid velocity [7]: bubbles travelling with approximately the same velocity as liquid can be captured with a camera. Overall, nowadays one can find a large number of applications of the Bretherton-Taylor bubble train flow: continuous flow analyzers to measure liquid velocity, lung openings, chemical reactions as hydrogenation of nitroaromatics, 2-ethylhexenal, Fischer-Tropsch synthesis, etc. Extensive reviews of Kreutzer et al. [4], Yue et al. [8], Gupta et al. [9] cover a significant number of applications.

This work is focused on the gas to liquid mass transfer study for the Bretherton/Taylor flow. The good understanding of mass transfer and how it depends on the parameters as the capillary number, the Reynolds number, slug and bubble lengths allows to properly manufacture a microchannel with necessary properties to ensure that chemical reactions are performed in the best manner. The mass transfer coefficient characterizing mass transfer is defined as the flux from the surface divided on the concentration difference between the concentration imposed on the body and the characteristic concentration in the domain. The characteristic concentration in the domain significantly depends on underlying hydrodynamics fields. For example, experimental studies [5, 8] show a complex dependency of the mass transfer coefficient on flow parameters: bubble and slug lengths, bubble velocity, which are in turn depend on the capillary number Ca and the Reynolds number Re. Yue et al. [8] established an experimental correlation for the volumetric mass transfer coefficient for a bubble train as a function of the diffusion coefficient, slug and bubble lengths, and bubble velocity:

$$k_L\, a = \frac{2}{d_h}\Big(\frac{D U_{\text{bubble}}}{L_{\text{bubble}} + L_{\text{slug}}}\Big)^{0.5}\Big(\frac{L_{\text{bubble}}}{L_{\text{bubble}} + L_{\text{slug}}}\Big)^{0.3}, \qquad (1)$$

where $k_L\, a$ is the volumetric mass transfer coefficient, d_h is the hydrodynamic capillary diameter, L_{bubble} is the bubble length, L_{slug} is the slug distance (between bubbles), U_{bubble} is the bubble velocity, D is the diffusion coefficient.

Thus, the understanding of mass transfer for the bubble train flow is not possible without deep understanding of hydrodynamic patterns. There are a number of studies available for the hydrodynamic study of the bubble train flow: experimental [10–12] and numerical [1, 13–15]. For the flow of long bubbles between parallel plates chosen here as the study case it is indicated that there

exists a vortex in the liquid slug for $Ca < 0.7$, see Fig. 8. As well, the bubble shape for low capillary numbers ($Ca < 0.1$ [12]) is symmetric. Thus, in the case of flow between plates for $Ca < 0.1 \div 0.7$ the bubble shape can be represented as two hemicircles and two planar interfaces with the vortex existing in the liquid slug. This is heavily utilized for mass transfer analytical estimations.

As mentioned, the mass transfer coefficient is defined as the mass flux from certain area, Eq. 4. Thus, analytical estimations [4, 16] are based on decomposition of the bubble shape. The mass transfer coefficient is calculated through two separate contributions from two films and two hemicircles. For both contributions the Higbie penetration theory [17] is utilized, which states that the mass transfer coefficient for simple geometry flow depends on the average time of liquid packet interaction with a geometrical feature. It can be calculated as $\sqrt{\frac{\pi D}{t_{\text{char}}}}$, where t_{char} is the interaction characteristic time, ~~D is the diffusion coefficient~~. As the example for the Higbie penetration theory, the mass transfer coefficient for the flow of bubbles between parallel plates is calculated as (similarly to work of van Baten and Krishna [3]):

$$k_L = 2\sqrt{\frac{\pi D}{t_{\text{film}}}} + 2\sqrt{\frac{\pi D}{t_{\text{circle}}}}, \tag{2}$$

where $t_{\text{film}} = \frac{L_{\text{film}}}{U_{\text{bubble}}}$ states for the interaction time of liquid traveling through planar part of the bubble, $t_{\text{circle}} = \frac{\pi R_{\text{circle}}}{U_{\text{bubble}}}$ is the time which liquid in the slug travels the distance of half bubble circumference.

Despite the simplicity, analytical expressions work well for flows with low capillary numbers $Ca < 0.1$ [5] where the bubble shape is symmetrical and can be nicely approximated. Moreover, because of the hydrodynamic pattern in the slug, i.e. there is a vortex in slug, one can estimate the time for fluid batch to travel whole circumference. However, with the change of the capillary number the situation drastically changes. The symmetrical bubble shape changes and resembles a bullet [18]. With the change of the shape and for flows with $Ca > 0.7$ there is no vortex in the liquid slug. In this case the Higbie theory fails to estimate contribution from bubble caps. Thus, the need of numerical simulations where all hydrodynamics fields and complicated bubble shapes are taken in the account is obvious.

However, available numerical studies of mass transfer [3, 4] are lacking the simulation of bubble shapes. The usual simulation of the mass transfer is performed as follows:

I The bubble shape is calculated either through analytical correlations [6] or experimental correlations [12] without directly resolving bubbles shapes through multiphase simulations. The expressions for bubble shapes are available only for flows with the capillary number $Ca < 0.1$.

II Hydrodynamics fields are then obtained by performing simulations of one-component flow around the droplet by imposing the bubble velocity on the walls. Thus, the simulations are performed in the reference frame moving with the bubble. The stress free condition is imposed at the bubble surface.

III The mass transfer simulations are performed in the reference frame moving with the bubble. The saturation concentration is imposed at the bubble surface. Only one unit cell with one bubble in it is used for simulations. Periodic boundary conditions are utilized to determine the volumetric mass transfer coefficient, which is calculated through the following equation [3]:

$$k_L\, a = \frac{\overline{\text{Flux}}}{C_{\text{bubble}} - \langle C_{\text{outlet}} \rangle} \frac{\text{bubble surface area}}{\text{unit cell volume}}, \tag{3}$$

where $\langle C_{\text{outlet}}(t) \rangle = \int C U_{\text{outlet}} \mathrm{d}A / \int U_{\text{outlet}} \mathrm{d}A$ is the averaged in space outlet concentration as the function of time, used as the characteristic concentration in the definition, Eq. 4. The averaged in time concentration flux ($\overline{\text{Flux}}$) is calculated as the difference between the overall average concentration in the whole domain ($\langle C_{\text{overall}} \rangle = \int_V C \mathrm{d}V / V$) at time t_1 and at time t_2 divided on the time difference $t_2 - t_1$. The agreement between numerical simulations [3] and correlations of Bercic and Pintar [5] was good.

There is a certain critics towards the presented numerical approaches [3, 4]. They mainly originate from the bubble shape approximation. It is taken symmetrical, i.e. consisting from hemispheres and cylinder film for the case of flow in circular capillaries. This is valid for small capillary numbers ($Ca < 0.1$.) As it was discussed, for such capillary numbers in the reference frame moving with the bubble there is a vortex in slug. Thus, the tracer is well mixed in slug.The choice of the characteristic concentration needed for the mass transfer coefficient, Eq. 4, in this case is obvious. With small differences in results it can be either averaged in the liquid slug concentration or the outlet space averaged concentration, as used in formulation of van Baten and Krishna [3]. Another critics is towards periodic boundary conditions to calculate the mass transfer coefficient. While it is clear to use periodic boundary conditions for the calculation of hydrodynamics fields for long bubbles motion in the microchannel, it is not the case for mass transfer simulations. Experimental correlations [5] show that the concentration along the streamwise direction changes as the exponential function. However, mass transfer simulations are made only for one unit cell using the periodic boundary conditions with the same concentration at the inlet and at the outlet. The question how one unit cell simulation corresponds to experimental measurements arises where concentration difference is measured at the distances of at least of a few unit cells difference [5]. In other words one needs to understand how the discrete one unit cell simulation corresponds to the continuous picture in experiments where one does not distinguish bubbles but takes measurements of concentration at different locations.

Addressing situations for a rich number of hydrodynamic patterns, shapes, effects for a bubble train we feel that there is a need to examine more carefully the strategies and assumptions which stay behind the mass transfer coefficient numerical simulations. We aim at establishing numerical simulations procedures as to how properly obtain the mass transfer coefficient via a study of different

boundary conditions, different characteristic concentration definitions. The case we want to examine is the two-dimensional case of the bubble train flow between parallel plates. The following issues will be addressed:

I Applicability of periodic boundary conditions to determine the mass transfer coefficient when there is a change of pattern from having vortex in the slug to not having it.

II Validity to use the inlet/outlet averaged or domain averaged concentrations as characteristic concentrations in the definition of the mass transfer coefficient.

III Transition of the continuous experimental picture to numerical simulations of a few unit cells mass transfer. This is the question about connection between time and space domains. In experiments the characteristic concentration is taken as an average in time, but in numerical simulations [3] the characteristic concentration is taken as an average in space.

IV How the change of the hydrodynamic pattern ($Ca > 0.7$) influences mass transfer.

Note that the goal of this paper is to establish procedures and boundary conditions used for the mass transfer coefficient determination. Though at the end of the manuscript we present a comparison of the volumetric mass transfer coefficient on the Peclet number with analytical [16] and experimental correlations [8], the thorough determination of the Sherwood number as a function of other non-dimensional parameters as gas holdup, bubble/slug lenghts, the capillary number is the goal of the future work.

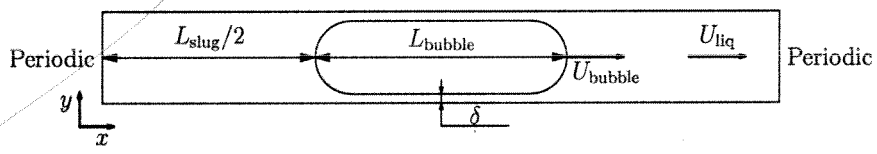

Figure 1: Simplified sketch of the bubble train motion. Using periodic conditions for the velocity field is natural, but needs validation for the mass transfer.

To establish numerical procedures we performed multiphase simulations [14, 18] for the range of capillary numbers $Ca = 0.1 \div 1.0$ to extract bubble shapes. For this range of capillary numbers we are able to capture the bubble shape change and the change of hydrodynamic patterns. The mass transfer simulations were performed with different boundary conditions (open, periodic) and with a few unit cells (1 to 10 unit cells). The numerical approach we take is the lattice Boltzmann method, a relatively new CFD competitor emerged during last 20 years [19–22]. During years the method was applied to simulate not only hydrodynamic problems [23], but as well multiphase flows [24–26], heat transfer [27, 28], ferrofluids [29, 30].

The mass transfer problems in the lattice Boltzmann framework were mainly addressed in series of works of Ginzburg and co-authors [31–33]. However, all these works are of general nature to simulate the advection-diffusion equation via the lattice Boltzmann framework. In comparison, this work focuses on the application side as to establish the procedure of how to obtain the volumetric mass transfer coefficients for bubble train flow. One should mention the work of Yoshino and Inamuro [34] about heat and mass transfers in porous media and the work of Derksen [2] simulates lateral mixing in cross-channel flow. While two last works are focused at particular mass transfer problems, both problems are of homogeneous nature and do not guide as how to obtain the mass transfer coefficient for non-homogenous problems.

The paper is organized as follows. We start with definitions of the volumetric mass transfer coefficient and apply them for the bubble train flow to derive expressions to connect space and time domains. Then the lattice Boltzmann model used to simulate mass transfer is presented followed by benchmarks. Finally, numerical simulations of different boundary conditions and a few unit cells simulations for different hydrodynamic patterns are presented to establish the thorough procedure to determine the volumetric mass transfer coefficient. The comparison with analytical correlations is also presented.

2 Mass transfer definitions

By the definition the mass transfer coefficient from the surface with the imposed constant concentration C_{bubble} is the following:

$$k_L = \frac{\dot{m}}{P\Delta C}, \tag{4}$$

where $\dot{m}$ is the mass rate $\left[\frac{kg}{s}\right]$, P is the area of the surface $\left[m^2\right]$, ΔC is the concentration difference between the surface and the surrounding medium $\left[\frac{kg}{m^3}\right]$. Therefore, k_L has a dimension of velocity $\left[\frac{m}{s}\right]$. Usually, the surrounding medium concentration is taken at the infinite distance from the bubble. However, in the case of complicated geometries and non-homogeneous concentrations, the medium concentration can be the average concentration in the domain or the flux averaged concentration at the inlet or outlet, etc. Thus, one needs to establish a thorough definition of the volumetric mass transfer coefficient in the case of complex geometries and non-trivial hydrodynamic velocity patterns.

There are different methods to estimate the mass transfer coefficient k_L. We first examine the theoretical definitions of the mass transfer in case of point mass sources.

2.1 Point mass sources

In what follows we will present three approaches to calculate point mass transfer coefficients (by point source we assume the source with the infinitesimally small

surface area P):

1. Let us look at the infinitesimal small domain of the volume $A\Delta x$ not moving and with the point mass source. Then the concentration difference can be found as $\Delta C = C^* - C(t)$, where C^* is the imposed point source concentration, $C(t)$ is the time dependent concentration, which do not depend on the location due to homogeneity. Therefore, one can write the time dependent ODE for the concentration in the domain:

$$\dot{m} = A\Delta x \frac{\mathrm{d}C}{\mathrm{d}t} = k_L P(C^* - C(t)), \tag{5}$$

with the initial condition $C(0) = 0$ The solution can be found by solving ODE:

$$C(t) = C^*(1 - \exp(-k_L\, at)), \tag{6}$$

where $k_L\, a$ is the volumetric mass transfer coefficient defined as:

$$k_L\, a = k_L \frac{P}{A\Delta x} = k_L \frac{P}{V}, \tag{7}$$

where P is the source surface, V is the unit cell volume.

2. Let us predict mass transfer happening in moving with the velocity U liquid, see Fig. 2.

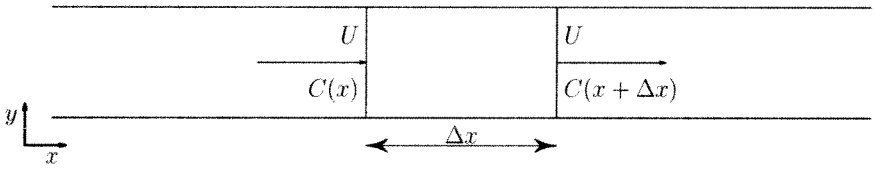

Figure 2: The mass transfer in the moving liquid.

If one can assume that the mass point sources are distributed in the whole medium, then the mass accumulated in the unit volume $V = A\Delta x$ can be calculated as the difference of mass fluxes entering and leaving domain $U\big(C(x + \Delta x) - C(x)\big)$. The accumulated mass should be proportional to the mass transfer coefficient:

$$U\big(C(x + \Delta x) - C(x)\big) = k_L P\big(C^* - C(x)\big), \tag{8}$$

giving the same equation but only in the coordinate domain:

$$C(x) = C^*\Big(1 - \exp\big(-k_L a \frac{x}{U}\big)\Big). \tag{9}$$

Note that the concentration $C(x)$ does not depend on time.

3. If one transfers to the frame moving with the liquid velocity U, then the situation will be the same as in the first case. However, one can connect the time and spatial location with the velocity U ($t = \frac{x}{U}$) to obtain the same equation as in the case 2.