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Mass transfer characteristics of gas–liquid absorption during Taylor flow in mini/microchannel reactors

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HIGHLIGHTS

- Numerical simulation of mass transfer in Taylor flow microchannel reactors.
- Purely theoretical approach that models channel length and inlet mixing region.
- Reaction system of absorption of carbon dioxide in aqueous sodium hydroxide.
- Effect of length, wettability, inlet mixing, phase concentrations, temperature.
- Predicted results compare favorably with experimental data in the literature.

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ABSTRACT

This paper reports a numerical study of the mass transfer characteristics during Taylor flow in mini/microchannel reactors. A finite-element implementation of the phase field method was used to predict the hydrodynamics of the two-phase flow. The phase distribution thus obtained was used to define the computational domain to model the reactive mass transfer. The reaction system of the absorption of CO_2 into aqueous NaOH solution was considered. Channels with characteristic dimensions ranging from 100 μm to 750 μm were modeled with cross-flow and flow-focusing inlet configurations. The effect of channel length was studied by varying the residence time in the transient simulation. The results indicated that channels having a small characteristic dimension could yield reductions in the residence time, and therefore the reactor size, by as much as 85%. This reduction was further enhanced by higher concentration levels of the liquid reactant and increased temperatures. The inlet mixing region was found to have a significant influence on the total mass transfer. The channel wall wettability was found to affect the mass transfer characteristics negligibly. The predictions from the currently proposed model were compared with available experimental data, as well as with predictions of an earlier unit cell-based model, and a good agreement was obtained.

1. Introduction

Microscale technologies are quickly penetrating new application areas in diverse engineering applications due to their proven potential for process enhancement, improved process control, increased safety and overall size reduction (Ehrfeld et al., 2000; Guangwen et al., 2008). Specifically, multiphase microreactors are of application to various engineering processes and when optimally designed, the inherent higher surface area to volume ratio of these systems provides substantial enhancements in the heat and mass transfer performance, while keeping the pressure drops at moderate levels. A widely accepted classification system for microscale systems was proposed by Kandlikar and Grande (2003) which is based on the hydraulic diameter, D. It was proposed that conventional channels have $D > 3$ mm, minichannels have 200 μm $< D <$ 3 mm, and microchannels have 200 μm $< D <$ 10 μm.

Taylor flow (Davies and Taylor, 1950) is a two-phase regime that is dominated by surface tension forces and is characterized by an alternating liquid slug and gas bubble formation with a thin film of liquid separating the gas phase from the channel wall. Further, convective mixing is enhanced by the recirculating velocity components and hence this flow regime has been of significant interest for heat and mass transport processes and is the focus of the present work.

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Berčič and Pintar (1997) studied the absorption of methane in water during Taylor flow in minichannels and subsequently proposed an empirical correlation for the mass transfer coefficient. However, as has been reviewed and remarked upon in many subsequent works, the notable absence of any functional dependence on channel diameter in their correlation has led to a relatively poor predictive accuracy of the expression for alternative systems.

Fundamental approaches to predict the mass transfer coefficient were developed by Irandoust and Andersson (1989) by separately considering the various contributions to the overall mass transfer from the gas bubble through the liquid film to the channel wall, from the gas bubble to the liquid slugs through the bubble caps, and from the liquid slugs to the channel wall. Likewise, van Baten and Krishna (2004) modeled a single unit cell of Taylor flow in minichannels of varying diameters and accounted for the mass transfer contributions from the two bubble caps and the film region. Modifications to the same in order to consider long contact times (or large unit cell lengths) were suggested by Vandu et al. (2005) in order to match their predictions with experimental data for the absorption of oxygen into water, wherein the film contribution to mass transfer was shown to be the dominant factor.

Yue et al. (2007) reported reactive mass transfer experiments in a rectangular minichannel and proposed an empirical correlation for the mass transfer coefficient. Subsequently, Yue et al. (2009) reported another empirical correlation for non-reactive mass transfer in square minichannels to improve the inaccurate predictions obtained by using certain previous mass transfer correlations, such as that of van Baten and Krishna (2004), for their square minichannel application. Likewise, Su et al. (2010) and Tan et al. (2012a) have also proposed empirical expressions for Sherwood number and mass transfer coefficient, respectively.

While most of the works reviewed above have been based on empirical approaches, these methods are typically limited to specific systems only and are not universal in nature. For this reason, certain studies, including the present one, have focused on the development of numerical approaches.

Salman et al. (2004) modeled a unit cell of the Taylor flow regime using a finite volume method-based code. An initial tracer concentration was applied to the cell, and changes to its concentration were tracked. The model was reported to be suitable for low Bodenstein numbers only, and a simplified analytical solution was proposed for higher Bodenstein numbers, typically greater than 10. A similar approach was presented by van Baten and Krishna (2004) considering only the liquid slug and treating the Taylor bubble as a void with a constant initial tracer concentration at the interface.

Raimondi et al. (2008) performed direct numerical simulation (DNS) of liquid–liquid mass transfer during Taylor flow in square microchannels. The hydrodynamics were obtained by the volume of fluid (VOF) method. The concentration field was governed by using transformational parameters to modify the conventional convention–diffusion equations in order to derive equations that were compatible with the one-fluid approach used for predicting the hydrodynamics of the flow. It is worth noting that Raimondi et al. (2008) decoupled the hydrodynamics from the mass transfer and therefore their method is accurate only when the transport of species negligibly affects the hydrodynamics of the flow. In other words, interfacial deformations due to mass transfer are assumed to be negligible here. These issues were outlined by Onea et al. (2009), who reported a qualitative study of the mass transfer across deforming interfaces. The model was based on the VOF approach and considered a continuous numerical concentration across the interface by implementing Henry's law on one side and a reference concentration on the other. The discontinuities in mass flux across the interface were overcome by effecting appropriate modifications to the cell face diffusivities.

Shao et al. (2010) noted that some previous modeling approaches are not well suited for applications involving chemical reactions for which a highly accurate prediction of the interfacial concentration is required. These include approaches that consider only the bubble boundary and apply a tracer concentration at the interface. They proposed a numerical model which considered both phases and simultaneously solved for the concentration change in each phase. The gas concentration at the interface was continuously updated in a method very similar to that of Onea et al. (2009), by using Henry's law to obtain a continuous concentration profile. As in most previous studies, the Taylor flow regime was modeled using the unit cell approach, with mass transfer occurring across the interface to both the liquid slugs and the thin liquid film region between the gas bubble and channel wall. They favorably compared their predictions against experimental data while the disagreement with existing empirical correlations was noted.

From the brief review above, which was largely limited to mass transfer modeling approaches in mini/microchannel reactors, the following points can be inferred. First, in spite of the common interest in the development of empirical correlations, these correlations are generally specific to certain conditions only and their universal applicability as yet remains to be established. Second, relatively few studies have focused on microscale Taylor flows involving reactive mass transfer as compared to non-reacting flows. In this regard, the numerical model of Shao et al. (2010) can be considered a definite advancement, although it is clearly limited by its need for experimental data for defining the unit cell domain. Unfortunately, the use of empirical correlations for obtaining this data limits the universal applicability of the model. In other words, the computational domain for mass transfer would vary depending on the correlation being used, as was demonstrated by Shao et al. (2008), while comparing experimentally measured Taylor bubble lengths against multiple empirical correlations in literature. While this variation can certainly be avoided by developing purely numerical approaches, one must take into consideration the associated increase in the computational requirements. Third, the unit-cell based approaches have been quite popular on account of the periodic nature of the Taylor flow regime. While these approaches do reduce the computational time, they are grossly limited by their inability to model and thus account for the mass transfer that occurs in the inlet mixing region present in most microchannel reactors. The significance of the same limitation for reacting flows requires investigation, especially considering that enhancement factors as high as 1.6 were reported by Fries and von Rohr (2009) depending on the inlet mixing region. Likewise, Tan et al. (2012b) demonstrated that up to 30% of the total mass transfer could occur during the formation stage of Taylor flow in the inlet region. Lastly, it is worth noting that one of the primary goals of an accurate mass transfer model is its potential use in the design of scaled-up microchannel reactors having complex channel geometries and optimized channel dimensions. In view of these limitations, the following points are addressed in the present paper.

A numerical approach to simulate gas–liquid mass transfer with chemical reactions during Taylor flow in a mini/microchannel reactor is proposed in this paper. The modeling methodology that governs the hydrodynamics of two-phase flow and the mass transfer phenomena is based on purely theoretical (non-empirical) approaches. The hydrodynamics of the flow are decoupled from the mass transport phenomena. The computational domain considers an entire microreactor that includes the channel length as well as the inlet mixing region. The numerical predictions are validated with experimental and theoretical data reported in the