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Gas-liquid flow and mass transfer in a microchannel under elevated pressures

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HIGHLIGHTS

- The sizes of CO₂ bubbles and liquid slugs were determined under different pressures.
- An online method was used to study the bubble dissolution and mass transfer rate.
- $k_{\rm L}a$ and $k_{\rm L}$ increase with the increase in system pressure.
- The maximum dissolution of CO₂ bubbles is linear to the liquid slug size.
- Gas absorption during bubble formation was measured.

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Flow and mass transfer of gas-liquid slug flow under elevated pressures up to 3.0 MPa in a microchannel are investigated with CO_2 -water system. The results show that the ratio of the initial bubble length to the unit cell length is linear with the injection gas volume fraction under each pressure condition, but the slope decreases with an increase in the system pressure. The mass transfer coefficients are calculated with a unit cell model based on the dissolution rate of gas bubbles. Increasing pressure leads to larger mass transfer coefficients, as well as higher amount of gas absorption during the bubble formation. But the fraction of gas absorption during the bubble formation stage is only about $1.5 \sim 4.0\%$ of feeding gas. For the bubble dissolution in the main channel, the dissolution rates at different flow rates differ very little for short contact distances from the T-junction, whereas the balance limitation of dissolution at large contact distances only depends on the amount of liquid in a unit cell.

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1. Introduction

Microreaction technology holds great promises for process intensification in diverse chemical engineering applications due to the capabilities of providing high mass and heat transfer rate, improved process control and overall equipment size reduction (Chen et al., 2008; Jensen, 2001). These merits make the implementation of this technology much safer in practice because of not only its excellent thermal management (Chao et al., 2009, 2010), but also low material hold-up. Moreover, the numbering-up methodology for its production throughput increase largely reduces the time from lab research to industrial application.

With controllable and very high specific surface area, microreactors are especially suitable for multiphase processes. For gas-liquid two-phase systems, such as absorption (Ye et al., 2013) and reaction (Keybl and Jensen, 2011; Trachsel et al., 2008; Zhao et al., 2013b), many researches have shown that microreactors have great advantages (Hessel et al., 2005). Generally, gas-liquid two-phase flow patterns in microchannels include bubbly, slug, unstable slug, slugannular, annular and churn flows (Shao et al., 2009; Triplett et al., 1999). Among these flow patterns, slug flow is the most studied one due to its remarkable features. It is dominant under wide operating conditions with regular dispersion of gas bubbles and liquid slugs. The gas bubble and liquid slug move alternatively in the channel, rendering low back mixing and narrow residence time distribution. A large surface area, along with the inner recirculation in the liquid slug, makes it possibly to substantially improve heat and mass transfer processes. (Zaloha et al., 2012). Therefore, slug flow appears to be a very promising operational mode for the intensification of gas-liquid reactions.

Gas-liquid mass transfer within a slug flow mainly contains two zones: (1) between bubble caps and the neighboring liquid slugs and (2) between the bubble body and the adjacent liquid film around the channel wall (Sobieszuk et al., 2012; van Baten

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and Krishna, 2004), as shown in Fig. 1. The first mass transfer zone is closely related to the mixing in the liquid slug due to recirculation (Zaloha et al., 2012), whereas the latter one is affected largely by the effectiveness of the liquid film (Pohorecki, 2007). For example, when very long bubbles are generated, the liquid film can be easily saturated and becomes ineffective due to long gasliquid contact time. Mass transfer in the above two zones will increase with increasing flow rates because of higher surface renewal rates. As a result, the overall mass transfer performance depends on many factors, such as bubble length, slug length and bubble velocity, etcetera. The overall mass transfer performance is also affected by the mixing in the liquid phase, which generally occurs between the liquid slug and the liquid film. It favors high bubble velocity, and can also be enhanced by channel bends and special inner channel structures (Muradoglu, 2010; Su et al., 2009; Zaloha et al., 2012). Up to now, many researches have been devoted to the mass transfer characteristics of slug flow (Berčič and Pintar, 1997; Ganapathy et al., 2013; Sobieszuk et al., 2011; Vandu et al., 2005; Yue et al., 2009). However, the existing studies are all under atmospheric conditions, which do not represent the real conditions in the majority of gas-liquid reactions in the chemical industry.

It is well known that high pressure is beneficial for gas-liquid reactions due to the increase of gas solubility in the liquid phase and feed throughput for the same reactor size. Many studies have already been focused on developing applications of high-pressure microfluidics (Keybl and Jensen, 2011; Marre et al., 2010; Trachsel et al., 2008; Verboom, 2009). But the design is usually arbitrary. Therefore, there is a growing need to study the transport phenomena involved in gas-liquid processes in microchannels. Zhao et al. (2013a) studied the gas-liquid flow patterns under elevated pressures up to 5.0 MPa. They found a shift of transition line between bubbly and slug flows to higher gas-phase Weber number and low liquid-phase Weber number when system pressure is elevated. Yao et al. (2014a) investigated the effect of system pressure on gas-liquid slug flow. A strong leakage flow was found to increase with the increase in system pressure, leading to a bubble formation shift from the transition regime to the squeezing regime. Apart from these studies, there is still a lack of detailed knowledge about the transport phenomena under different pressures in microchannels, especially concerning the gas-liquid mass transfer characteristics.

The present work aims at improving the fundamental understanding into gas-liquid flow and mass transfer under elevated pressures. Taylor flow of CO_2 -H₂O mixture was investigated, where flow characteristics such as bubble and slug lengths were measured by imaging method. And mass transfer information was extracted from the dissolution of CO_2 bubbles. Finally, mass transfer coefficients and the amount of absorption during bubble formation were calculated and compared under different system pressures up to 3.0 MPa.

2. Experimental section

2.1. Microchannel devices

Fig. 2 shows the schematic diagram of the gas-liquid microchannel contactor used in this study. Gas-liquid slug flow was generated at the T-junction and moved downstream the channel.

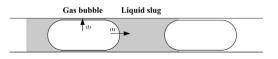


Fig. 1. Mass transfer in the gas-liquid slug flow.

The contactor module was composed of transparent polyaryl sulfone (PASF), polycarbonate (PC) and stainless steel materials. The channel with a cross section of 600 μ m × 300 μ m (width × depth) was fabricated on a PASF substrate by using milling technology. The machined PASF plate was covered by another smooth PASF plate. The two plates were sandwiched by two PC plates, then by two stainless steel plates. With steel screws, the microchannel contactor was tightly sealed so that no leakage occurred at 5 MPa gas-tightness experiments. The contactor houses an observation window that enabled complete record of flow patterns in the meandering microchannel.

2.2. Experimental setup

Pure CO₂ and deionized water were used as test fluids to study the flow and mass transfer under elevated pressures. The experimental setup was shown in Fig. 3. CO₂ was provided from a cylinder and controlled by a series of mass flow controllers with different flow ranges (D07-7B, Beijing Seven Star Electronics, China, accuracy of 0.5% full scale). Deionized water was pumped by a high-precision digital piston pump (Beijing Satellite Manufacturing Factory, measurement range: $0 \sim 5$ mL/min, precision: 0.3%). The system pressure was

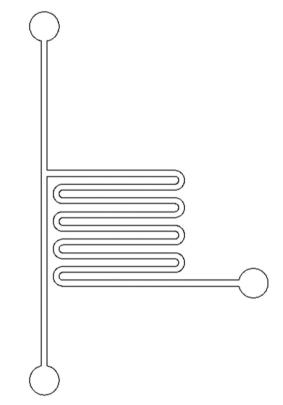


Fig. 2. Schematic diagram of the gas-liquid microchannel contactor.

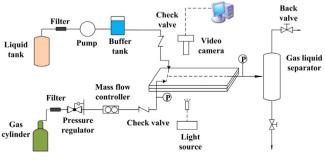


Fig. 3. Schematic of the experimental setup.

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