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Experimental investigation on gas-liquid mass transfer with fast chemical reaction in microchannel

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ABSTRACT

The gas-liquid two-phase flow and mass transfer with fast chemical reaction in a microchannel were investigated experimentally. Result showed that the bubble volume decreased exponentially with its movement along the channel in a quickly reactable absorbent. The volumetric mass transfer coefficient k_La increased with gas flow rate, while it was insensitive to liquid flow rate under experimental conditions, although the mass transfer coefficient k_L increased with liquid flow rate. In slug-bubbly flow regime, the k_L decreased when increasing gas flow rate, but inverse tendency was found for slug flow pattern. Both k_L and k_La increased with the increase of absorbent concentration. A new correlation for predicting the volumetric mass transfer coefficients k_La was proposed by taking the enhancement factor E of chemical reaction into account.

1. Introduction

In recent years, microfluidic technology has been widely applied in many industrial fields such as chemical, material, biolchemical, pharmaceutical and food engineering, etc., due to its excellent mass and heat transfer performances. The systematical investigation on the mass transfer mechanism inside microchannel is of critical importance for the design and optimization of the microreactor.

Bercic et al. [1] investigated the absorption of methane into water in capillaries under Taylar flow regime. They found that the volumetric mass transfer coefficient relies mainly on the length and velocity of liquid slug, and the contribution of mass transfer in the liquid film is inapparent.

van Baten et al. [2] suggested that the mass transfer coefficient was a sum of two contributions: bubble caps and liquid film surrounding the bubble. A criterion of *Fo* number ($Fo = D/(t_{film}\delta_{film}^2)$, D is the diffusivity of liquid phase, t_{film} the contact time of liquid film with Taylor gas bubble, and δ_{film} the thickness of film surrounding buble.) was proposed to judge the contribution of the mass transfer. If $Fo < 0.1$, both the bubble caps and the film region were important; if $Fo > 1$, the contribution of the bubble caps was dominant due to the saturation of liquid film region.

Yue et al. [3] investigated the absorption of oxygen in the deionized water, but the experimental volumetric mass transfer coefficients were less evidently than the prediction values of Bercic and Pintar [1] and van Baten et al. [2]. Yue et al. [3] argued that the mass transfer between the liquid film and liquid slug was mostly stemmed from the diffusion without the convective mixing, thereby the mixture between the liquid film and liquid slug was incomplete due to the short diffusive time for the short liquid slug. Moreover, the dissolution rate of CO_2 bubble into water also was experimentally investigated [4,5].

The analysis on the mechanism of mass transfer between gas and liquid phases under the Taylor flow has been implemented through a bubble and following liquid slug [6,7]. Kreutzer et al. [6] thought that the mass transfer could be divided into three different steps: (1) From the bubble directly to the liquid film, (2) from the bubble to the vortex region in the liquid slug, (3) from the vortex region to the liquid film.

Sobieszuk et al. [8] investigated the mass transfer of CO_2/N_2 absorption into $KHCO_3/K_2CO_3$ solution and determined the mass transfer coefficients for bubble caps ($k_{L,cap}$) and liquid film ($k_{L,film}$), respectively. They found that the values of $k_{L,cap}$ and $k_{L,film}$ were mutually approximate. Yao et al. [9] investigated the flow and mass transfer of CO_2-water system under elevated pressures in a microchannel. On the basis of the dissolution rate of gas bubbles, the mass transfer coefficients were calculated using a unit cell model. The difference of dissolution rates in the main channel at

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different flow rates was very little for a short contact distance. In addition, many reseachers [10–12] have found that the mass transfer during the formation stage of bubbles is relatively large in the total mass transfer process.

Mikaelian et al. [13,14] studied the gas-liquid mass transfer of spherical bubbles in square and circular microchannels through CFD analysis, and found the recirculation between two successive bubbles. Furthermore, they proposed a model for describing the dissolution of a chain of spherical pure gas bubbles into a non-volatile liquid. Jia and Zhang [15] numeically investigated the mass transfer of Taylor flow in a microfluidic T-junction using three-dimensional Volume of Fluid, and found the vortices in both the concentration and velocity fields in the liquid slug. In addition, the simulation results showed that the mass transfer through the thin liquid film around the Taylor bubble was dominant during the dissolution, and the dominance gradually disappeared with the increase of the liquid film thickness, the maximum of mass transfer rate appeared at the top end of the caps in the cap region.

Up to now, many efforts have been devoted to the research on gas-liquid mass transfer in the microchannel [16–18]. However, the understanding on the mechanism of mass transfer in microfluidic device remains still far from sufficient, especially in the system involving chemical reaction. In this paper, the mass transfer between gas-liquid two phases with fast chemical reaction was mainly concerned. Experiments of CO_2-NaOH aqueous solution two-phase flow were conducted in a square microchannel. The evolution of bubble volume along the microchannel was recorded. Furthermore, the overall liquid mass transfer coefficients and the overall liquid volumetric mass transfer coefficients were determined to investigate the mass transfer performance of the whole microchannel, and a correlation, considering the enhancement factor E of chemical reaction on mass transfer, was proposed to predict the volumetric mass transfer coefficients.

2. Experiment

As shown in Fig. 1, a square cross-section microchannel with depth 400 μm, width 400 μm and length 40 mm was adopted in the experiment. The channel was fabricated in a polymethyl methacrylate (PMMA) plate, and then sealed with another PMMA plate by screws. The gas flow rate Q_G and liquid flow rate Q_L were controlled respectively by pumps (PHD 2000, Harvard Apparatus, America. The accuracy is ±0.35%.).

The CO_2 (the mass fraction purity ≥99%, Tianjin Liufang Gas Station, China) and NaOH (the mass fraction purity is 99.4%, Aladdin, China) solution were feeded into the gas and liquid inlets of the microchannel, respectively. After the flow stabilized about 5 min for a new experimental condition, the formation and movement of the bubble in microchannel were recorded by a high-speed digital camera (MotionPro Y5, IDT, USA) at 2000 frames per second. The pressure of gas phase at inlet was measured by a piezometer (ST3000, Honeywell, USA. The precision is 0.02%), and the pressure at outlet is atmospheric pressure. CO_2 was used as the dispersed phase and a series of NaOH aqueous solutions with 0.3wt% sodium dodecyl sulfate were used as the continuous phase. The density ρ, viscosity μ_L and surface tension σ of solution were measured respectively using densimeter (DMA4500, Anton Paar, Austria), automatic ubbelohde viscometer (iVisc, LAUDA, Germany) and tensiometer (OCA15ECm Data Physics Instruments GmbH, Germany) at 293.15 K, and the data were given in Table 1. The experiments were carried out at 293.15 ± 1 K and atmosphere pressure, the ranges of Q_G and Q_L are separately (10–320) mL·h^{-1} and (20–80) mL·h^{-1} for mass transfer, and Q_L with (10–80) mL·h^{-1} for flow pattern.

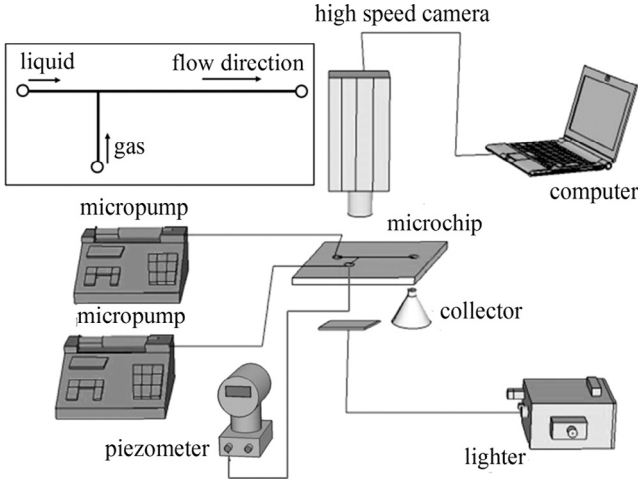

Fig. 1. Schematic diagram of the experiment setup.

3. Results and discussion

3.1. Evolution of the bubble volume

Three main flow regimes were observed as shown in Fig. 2: bubbly regime, slug regime and slug-annular regime, analogous to those reported by Cubaud [4,20]. However, the slug flow could evolve into bubbly flow along the microchannel due to the fast absorption as shown in Fig. 2b. Cubaud [4] also observed the evolution of dissolving CO_2 bubble in water using long serpentine microchannels. This indicated the absorption played a key role on the gas-liquid two-phase flow.

When the bubbles in the channel are less than the channel width, the regime is named as bubbly flow, and the bubble could be regarded as a sphere. When the bubbles in the channel are larger than the channel width, the regime is slug flow, and the slug bubble includes two semispheroid ends and a body. In our experimental, the capillary number $Ca = \mu_L u/\sigma$, (u is superficial velocity of gas-liquid flow, $u = (Q_G + Q_L)/w^2$, w is the width of channel) is less than 0.003, thus we assume that the cross section of slug bubble body is approximately flat on the side regions and constant curve with radius r in the corners in square microchannel [21], and its area was 90% of cross section of microchannel [22]. The volume V_B and surface area A_B of a slug bubble could be separately calculated by:

$$V_B = \pi w^3/6 + 0.9w^2(l_B - w) \quad (1)$$

$$A_B = \pi w^2 + [2\pi r + 4(w - 2r)](l_B - w) \quad (2)$$

$$r = w\sqrt{1/(40 - 10\pi)} \quad (3)$$

where l_B is the length of bubble, which is obtained from the image recorded by the high-speed digital camera using the software MATLAB, and the average bubble length of at least three images was used to calculate the area and volume of bubble. The relative error of l_B is 2%.

Experiment in this work showed that the bubble volume decreases exponentially and finally approaches to zero except Q_G = 100 mL h^{-1} with the movement of bubble in microchannel as shown in Fig. 3. With the increase of gas flow rate Q_G, the volume of bubbles formed at inlet of microchannel (t = 0) increases. The pressure drops for experiments in Fig. 3 are (0.46, 0.73, 1.06, 1.43 and 1.81) kPa, respectively. The effect of pressure on volume was negligible.