Chemical Engineering Science 109 (2014) 306–314

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Mass transfer characteristics of bubbly flow in microchannels

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HIGHLIGHTS

- The mass transfer characteristics of bubbly flow were first studied.
- The mass transfer coefficients were determined by an online measurement method.
- The mass transfer coefficients of bubble forming and flowing stages were studied.
- The effect factors on mass transfer coefficient were discussed.
- The empirical correlations were established to predict mass transfer coefficients.

ARTICLE INFO

Article history:
Received 27 October 2013
Received in revised form
2 February 2014
Accepted 4 February 2014
Available online 11 February 2014

Keywords:
Microfluidic device
Bubbly flow
Absorption
Mass transfer
Microstructure
Transport process

ABSTRACT

The mass transfer characteristics of bubbly flows in co-flowing and T-junction microchannels are described in this study. A CO_2–N_2 gas mixture and a monoethanolamine–ethylene glycol solution were used as the gas and liquid phases, respectively, to generate gas/liquid bubbly microflows. The bubble dimensions changed obviously during the bubble-forming and bubble-flowing stages. The overall mass transfer coefficients (K_L) at these stages were determined by developing an online measurement method through which time-dependent changes in bubble volume were analyzed. Investigation of the effects of phase flow rate and concentration of the two phases on K_L showed that K_L was in the range of 1×10^{-4}–4.9×10^{-3} m/s. In addition, empirical correlations were established to predict K_L for different mass transfer stages.

1. Introduction

Microfluidic devices, which have undergone rapid development over the last two decades, exhibit many attractive advantages as compared with conventional large-scale devices (Gunther and Jensen, 2006; Kobayashi et al., 2006; Mae, 2007). Microfluidic devices have been widely applied in various research areas, including chemical synthesis, separation, material preparation, and biological engineering (Huebner et al., 2008; Kobayashi et al., 2004; Seemann et al., 2012; Wang et al., 2011). Gas/liquid systems play important roles in scientific and industrial fields, and a number of gas/liquid reaction processes and separation processes have been successfully realized in microfluidic devices (Leclerc et al., 2008; Tan et al., 2011, 2012). In general, adequate reduction of the mass transfer limitation to greatly enhance the process is desired. Therefore, investigating the mass transfer characteristics in various flow regimes and microfluidic devices is very essential.

In recent years, several experimental investigations, in which the mass transfer characteristics of gas/liquid systems were examined in microfluidic devices, have been published. Yue et al. (2009) studied the mass transfer performance of air/water Taylor flow in Y-shaped microchannels. H.J. Su et al., 2010 investigated the mass transfer process of H_2S absorption by methyldiethanolamine (MDEA) solution in a cross-flow T-junction microchannel. Aoki et al. (2011) studied the mass transfer coefficient of slug flow in miniaturized channels and established a correlation between the Sherwood number and the Peclet number. Roudet et al. (2011) examined the process of oxygen mass transfer from gas phase to water phase in straight and meandering millimetric channels for both Taylor flow and slug-annular flow regimes. In general, the mass transfer coefficients in the previous studies are usually determined by analyzing the solute concentration of samples collected at the outlet of microfluidic devices. However, the sample collection time and phase separation time are often longer than the fluid residence time in microfluidic devices, which might result in inaccurate characterization of mass transfer (Kashid et al., 2011a). A simple approach to overcome this problem in gas/liquid systems is to establish an online measurement method that determines the quantity of mass transfer and the solute

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http://dx.doi.org/10.1016/j.ces.2014.02.004
0009-2509

concentration by analyzing the volume changes in the gas slugs. By using this approach, Abolhasani et al. (2012) obtained the volumetric mass transfer coefficient and Henry's constant for CO_2 dissolving in physical solvents. Li et al. (2012) investigated the process of CO_2 chemical absorption and determined the reaction rate constant. In our group, Tan et al. (2012a) also used this approach and obtained the overall mass transfer coefficient of segmented flow with CO_2 chemical absorption.

The aforementioned studies focused mainly on the mass transfer performance of gas slug flow, but it is a challenge to determine whether mass transfer occurs in the liquid film between the gas slug and the channel wall (Roudet et al., 2011; Sobieszuk et al., 2011). However, few studies have reported the mass transfer performance of bubbly flow in microfluidic devices. Considering the advantages of bubbly flow, such as its well-defined specific interfacial area and greater stability, here we developed an online measurement approach and studied the mass transfer characteristics of bubbly flow with CO_2 chemical absorption for the first time. Moreover, owing to the great impact of mass transfer stages on process control and equipment design, distinguishing different stages of mass transfer is considered to be important (Aoki et al., 2011; Shao et al., 2012a; Wang et al., 2009; Xu et al., 2012). In the case of liquid/liquid systems, Xu et al. (2008) investigated the mass transfer rules in the droplet-forming and droplet-flowing stages. For gas/liquid systems, Tan et al. (2012b) studied the mass transfer performance of segmented flow during the forming stage in T-junction microchannels with different contact angles. In this study, we calculated the overall mass transfer coefficient (K_L) at the bubble-forming and bubble-flowing stages. In addition, the effects of phase flow rate and concentration of the two phases on the mass transfer coefficients were also clarified, and correlation equations were developed to predict the mass transfer coefficients in different stages.

2. Experiments and methodology

2.1. Microfluidic devices

Fig. 1 shows schematic diagrams of the microfluidic chips used in this study. Two different microfluidic devices – a co-flowing microchannel and a T-junction microchannel with embedded capillaries – were designed to generate monodispersed microbubbles. These devices were fabricated on polymethyl methacrylate (PMMA) plates by precision milling and sealed by a high-pressure thermal sealing machine (A274, Techson). The microchannels have a square cross section with an area of $A=0.36\ mm^2$. Glass capillaries with a tapered tip were used as the dispersed phase inlet channel to prevent large-area contact of gas and liquid phases before bubble formation. The capillaries were pulled by a flaming micropipette puller (Sutter Instrument Company) to form cuspate tips with a diameter of 45 μm. The meandering channels located downstream provided sufficient time to accomplish the mass transfer process.

2.2. Operation and observation

A high-speed CMOS camera (DK-2740, Dantec Dynamics) and microscope (BXFM, Olympus) were used to record the bubble-forming and bubble-flowing stages with frame frequencies of 2000 and 1000 fps, respectively. Both gas and liquid phases were delivered and controlled by syringe pumps (LSP01-1A, Longer). The gas flow rate (Q_G) was controlled at 75 μL/min or 100 μL/min, and the liquid flow rates (Q_L) varied from 225 μL/min to 700 μL/min. The corresponding u_G (Q_G/A) was in the range of 3.5–4.6 mm/s, while u_L (Q_L/A) ranged from 10 mm/s to 32 mm/s. As a result, the average residence time of two-phase fluids varied from 2.4 s to 6.5 s.

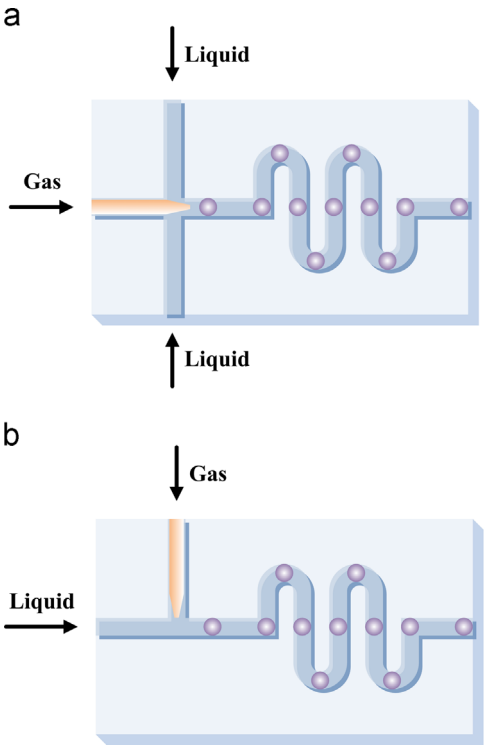

Fig. 1. Schematic diagrams of microfluidic devices: (a) co-flowing microchannel and (b) T-junction microchannel.

The Reynolds number Re_G was in the range of 0.2–0.3, and Re_L ranged from 0.4 to 1.4. In our experiments, the polydispersity index for microbubbles was less than 3%, indicating that the microbubbles were highly uniform (Xu et al., 2006).

2.3. Experimental system

In this study, the experimental system comprised a CO_2–N_2 gas mixture and a MEA–ethylene glycol solution (Sada et al., 1985). The gas (purity 99.99%) was purchased from Beijing Huayuan Company, and the analytically pure MEA and ethylene glycol (≥ 99.0%) were provided by Beijing Chemical Plant. Different CO_2 volume fractions of CO_2–N_2 gas mixture (17 vol%, 37 vol%, and 56 vol%) and different MEA concentrations' solutions (5 wt% and 15 wt%) were prepared before the experiments. The viscosity was measured with an Ubbelohde viscometer for all liquid phases, and the surface tension of each system was measured with a pendant drop interfacial tension meter (OCAH200, DataPhysics Instruments GmbH). The viscosity of each gas mixture and the diffusion coefficient of CO_2 in each solution were calculated by the mixing method (Dubois and Thomas, 2009; Yue et al., 2004). The physical properties of the different experimental systems are listed in Table 1.

2.4. Methodology

In this study, an online measurement method through which time-dependent changes in bubble volume were analyzed was developed to determine the mass transfer coefficients at the bubble-forming and bubble-flowing stages. This method was based on the following premises:

(1) The CO_2–N_2 gas mixture can be taken as ideal gas.
(2) N_2 is considered as an inert component, and its solubility in liquid solutions is negligible. The evaporation of liquid solutions can be ignored as well.