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# An online method to measure mass transfer of slug flow in a microchannel



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## HIGHLIGHTS

- Bubble length increase is compared with the two-phase pressure drop.
- A unit cell model is proposed to study the absorption of CO<sub>2</sub> bubbles.
- Bubble size reduction is used to determine the mass transfer coefficient.
- Mass transfer during bubble formation process is measured.
- The effect of fluid properties on the mass transfer is studied.

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## ABSTRACT

Physical absorption of CO<sub>2</sub> bubbles under slug flow has been studied in a T-junction microchannel. Based on the absorption rate of gas bubbles, an online photographic method is proposed to determine the mass transfer coefficients. Validity of this method is verified by evaluating bubble expansion due to pressure drop and by comparing the results with literature. The effect of fluid properties on the mass transfer has also been studied with ethanol solutions, which shows that mass transfer coefficient increases with the increase of ethanol concentration. The amount of gas absorbed during the bubble formation process has been measured to be about 2–10% of the inlet gas phase, and is found to linearly scale with the maximum mass transfer rate. For each fluid system, the initial dissolution rates of bubbles differ very little for short contact distance, whereas, the final amount of dissolution only depends on the residence time.

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## 1. Introduction

Microreaction technology has gained increasing attention on the improvement of multiphase processes over the recent years. High mass and heat transfer rate are obtained in microreactors due to large and controllable surface area to volume ratio (Yue et al., 2007; Cao et al., 2010), which is pretty important for multiphase processes. Key features of these novel reactors include miniaturization and numbering up mode, leading to a great reduction in investment of both expenses and time from lab research to industrial application. Additional benefits include better process safety via excellent thermal management (Cao et al., 2010), low material hold-up, sharp residence time distribution (Günther et al., 2004) and high volumetric productivity.

When gas and liquid flow through microchannels, a stable slug flow or Taylor flow (Triplett et al., 1999; Zhao et al., 2013) is obtained within a large range of operating conditions. The slug flow is characterized by sequences of an elongated gas bubble and a liquid slug. If the channel wall is wetted by the liquid phase, the gas bubbles are separated from the wall by a thin liquid layer (Fries et al., 2008b; Thulasidas et al., 1995). Slug flow is considered as a promising flow pattern to improve reaction performance for many reasons: uniformly dispersed gas bubbles, fixed gas–liquid interface, narrow residence time distribution, enhanced mass or heat transfer due to inner recirculation of liquid slugs and flexible operating conditions. Up to now, wide attention has been paid to slug flow on various aspects such as bubble formation process and bubble length (Dang et al., 2013; Garstecki et al., 2006; van Steijn et al., 2007), bubble shape and liquid film distribution in the cross section (Fries et al., 2008b; Han and Shikazono, 2009; Kreutzer et al., 2005a; Thulasidas et al., 1995), gas hold-up or void fraction (Kawahara et al., 2005; Xiong and Chung, 2007), pressure drop (Kreutzer et al., 2005a,b; Yue et al., 2009) and so on. Even for some major drawbacks that hinder the

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large scale application of microreactors such as distribution of multi-channels (Al-Rawashdeh et al., 2012, 2013) and short residence time/reaction time (Hessel et al., 2013), significant progress has been made.

Mass transfer characteristics of gas–liquid slug flow in microchannels have also been a hot subject in literature (Berčić and Pintar, 1997; Ganapathy et al., 2013; van Baten and Krishna, 2004; Vandu et al., 2005; Yue et al., 2007). Earlier work was carried out experimentally in capillaries with different diameters by Berčić and Pintar (1997), who found that the overall liquid side volumetric mass transfer coefficient is related to flow details such as bubble velocity and bubble length. Sobieszuk et al. (2011) used the Danckwerts Plot to simultaneously measure the interfacial area and liquid side mass transfer coefficient, which showed dependency on contact time. As recently reviewed by Sobieszuk et al. (2012), most of the works deal with quantification of mass transfer and building empirical correlations for predicting them while less attention has been paid on the effect of fluid properties on mass transfer characteristics. Fluid properties such as viscosity and surface tension have a large effect on the hydrodynamics, they are due to the effect of mass transfer, too. Therefore, research on influences of fluid properties on mass transfer characteristics is of great importance. The practical meaning is also obvious as fluid properties vary largely in practical gas–liquid processes. However, classic methods, which usually consist of measuring the gas and/or liquid composition, are rather time-consuming since flow rates in single microchannel usually range from a few  $\mu\text{L}/\text{min}$  to several  $\text{mL}/\text{min}$ . Also, these conventional methods still face other problems including saturation problems (Pohorecki, 2007) and serious end effects (Sobieszuk et al., 2011; Yue et al., 2007), which originate from large contribution of the inlet, outlet and gas–liquid separation sections. Online methods serve as good solutions (Dietrich et al., 2013; Tan et al., 2012b) to avoid these problems. Tan et al. (2012b) developed an online method to determine the mass transfer by measuring the change of  $\text{CO}_2$  bubble volume under absorption of  $\text{NaOH}$  solutions. Recently, Dietrich et al. (2013) developed a new colorimetric technique to measure the concentration distribution in the liquid slugs. Through this way, not only overall mass transfer is calculated, but also spatial information is obtained to visualize the locations where the mass transfer from gas bubbles happens. However, the above methods are only suitable for specific fluid systems with chemical reaction and the chemical enhancement factor is not separated. Therefore, developing novel online method that can directly measure the mass transfer coefficients is of great importance.

In this work, an online method based on the physical dissolution rate of bubbles is proposed to measure the mass transfer of slug flow in a rectangular microchannel. In the first part, the principle of the method is illustrated and verified by comparing the mass transfer coefficients with literature. Then pure  $\text{CO}_2$  is used to analyze absorption process with different liquid absorbents in the microchannel. Information about mass transfer coefficients, bubble and slug length, absorption during bubble formation process, dissolution rate will be presented and discussed.

## 2. Experimental section

Gas–liquid slug flow was generated in a rectangular cross-section microchannel with a crossing T-junction. The main channel has a meandering form with right angled bends as shown in Fig. 1. All the channels have the same width of  $600\ \mu\text{m}$  and depth of  $300\ \mu\text{m}$ . The entire length of the main channel is about  $150\ \text{mm}$ . The channels were fabricated on a polymethyl methacrylate substrate (PMMA, A grade, 92% of light transmittance, ShenZhen HuiLi Acrylic Products Co., Ltd.) using micromachining technology (FANUC KPC-30a) in our CNC Machining Center. The reactor was sealed by screws.

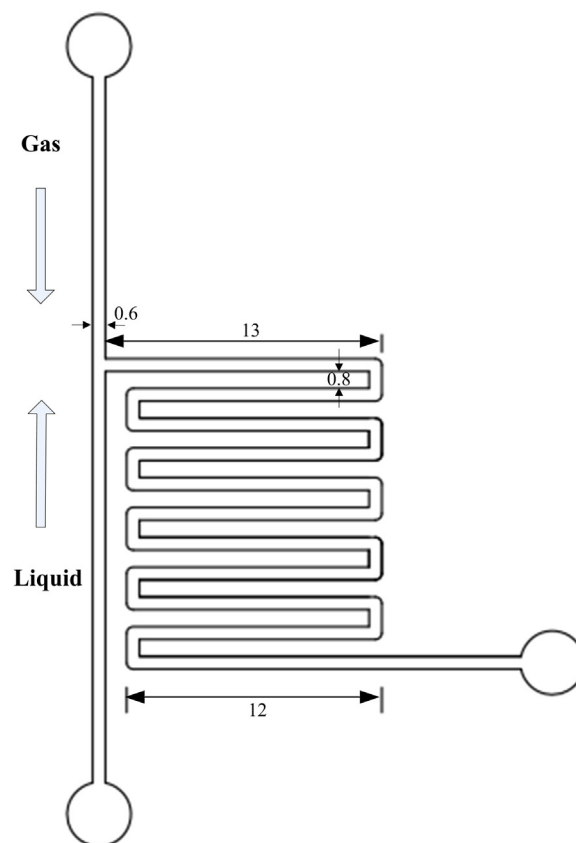


Fig. 1. Schematic diagram of the gas–liquid microchannel contactor.

Gas flow was provided via a pressure regulator and controlled by a mass flow controller (D07-19B, Beijing Sevenstar Electronics Co., Ltd., China) with an accuracy of 0.5% full scale. Liquid flow was pumped by a high precision digital piston pump (Series II, Chrom. Tech. Inc.). The actual flow rate under each run was determined by a weighing method. In order to eliminate the pulsation of the liquid flow rate, a buffer tank was introduced before the micro-reactor inlet. After flowing through the microchannel contactor, gas and liquid were separated in a gas–liquid separator. The pressure drop between the gas inlet and the outlet was directly recorded by a differential pressure transducer. All experiments were conducted under ambient conditions. Detailed information about the experimental setup is available in our previous work (Yao et al., 2013). The liquid phase used here was water–ethanol mixtures with different content. The fluid properties are displayed in Table 1.

To study the flow and absorption characteristics of slug flow, the flow pattern was recorded by a CMOS high-speed camera system (BASLER A504kc) with a macrolens (Nikon AF Micro-Nikkor 2,8/60 mm). The CMOS camera was placed above the visual window and strong light was provided by a cold light source. In all experiments, the CMOS camera was set to work at a recording rate of 640 frames/s and a resolution of  $1280 \times 800\ \text{pixel}^2$ . The shutter time was set as  $80\ \mu\text{s}$ . A moderate amplification of the images was chosen to have one pixel representing about  $15\ \mu\text{m}$ . With a Matlab program (Yao et al., 2013), we can obtain flow information such as bubble lengths, bubble velocities, bubble locations and so on. Under each operating condition, a sequence of at least 200 images was analyzed and the data were averaged to obtain the final value. The relative standard deviation (RSD) of the bubble and slug length did not exceed 5%. In our work, the picturing zone can only cover the first 8 channels with a length of about  $100\ \text{mm}$ . So the flow pattern in the last channel length (4–11 channels,

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