Chemical Engineering Science 100 (2013) 486–495

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Science

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

Generating microbubbles in a co-flowing microfluidic device

Kai Wang, Lisi Xie, Yangcheng Lu *, Guangsheng Luo **

The State Key Lab of Chemical Engineering, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China

HIGHLIGHTS

- Microbubble generation mechanism is explained.
- Two bubble generation phenomena are discovered and discussed.
- Gas pressure fluctuation and interface movement are analyzed.
- Both organic and inorganic gases are tested as the dispersed phase.
- Correlation equation for the average bubble diameter is established.

ARTICLE INFO

Article history:
Received 21 August 2012
Received in revised form
5 February 2013
Accepted 7 February 2013
Available online 26 February 2013

Keywords:
Co-flowing microchannel
Microbubble
Organic and inorganic gases
Generation mechanism
Gas-liquid interface
Pressure fluctuation

ABSTRACT

This paper introduces an experimental study on the microbubble generation mechanism in a co-flowing microfluidic device. Using propane, butane and air as the dispersed phases and different SDS–PEG solutions as the continuous phases, two kinds of bubble generating phenomena, named "single-bubble generation" flow and "dual-bubble generation" flow, were observed in experiment and uniform bubbles with average diameters ranging from 391 μm to 713 μm and polydispersity less than 2.9% were successfully prepared. The pressure fluctuation of gas phase is carefully analyzed with the Young–Laplace equation and the bubble generation mechanism is summarized from the dynamic movement of gas–liquid interface. The main factors impacting the bubble size variation are illustrated for both the organic and inorganic gases. A universal correlation is established to predict the average bubble diameters.

1. Introduction

Gas–liquid two-phase systems are very important items in the scientific research and industrial application. In recent years, the growing trend in the miniaturization of chemical engineering process and the development of high efficiency micromixing and microreaction devices have facilitated the application of gas–liquid systems into sub-millimeter scale or micrometer scale (Tan et al., 2011a, 2012a). Microscaled bubbles in immiscible liquids have several unique properties, including larger specific surface area, smaller transport distance and higher surface energy, comparing with the millimeter or larger scaled bubbles (Shui et al., 2007; Zhao and Middelberg, 2011). These scaling effects have attracted the researchers in some other areas except of chemical engineering, such as applied physics (Garstecki et al., 2005), bio-engineering (Chen et al., 2007), material science (Abate and Weitz, 2011) etc.

In the last decade, microfluidic technology has been demonstrated as one of the most effective methods for the generation of microbubbles. Monodispersed bubbles with average diameters ranging from several micrometers to hundreds of micrometers have been successfully prepared in microchannels (Xu et al., 2006; Marmottant and Raven, 2009). These uniform microbubbles are good items for the basic research of mass-transfer and chemical reaction processes at micrometer scale (Onal et al., 2005; Tan et al., 2012b, 2012c). For the controllable application of microbubbles, their generation mechanisms in different microfluidic devices are very important. Many relevant studies have been reported in literatures involving the T-junction (Tan et al., 2009), Y-junction (Pohorecki and Kula, 2008), cross-junction (Fu et al., 2011), flow-focusing (Hashimoto et al., 2008) and some other kinds of microfluidic devices (Hashimoto and Whitesides, 2010; Castro-Hernandez et al., 2011). For example, Garstecki et al. reported a classic experimental work on the generation of slug shaped microbubbles in the T-junction microchannels (Garstecki et al., 2006). The squeezing effect from continuous phase was

* Corresponding author. Tel./fax: +86 10 62773017.
** Corresponding author. Tel./fax: +86 10 62783870.
E-mail addresses: luyc@tsinghua.edu.cn (Y. Lu), gsluo@tsinghua.edu.cn (G. Luo).

0009-2509/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ces.2013.02.021

demonstrated affecting the movement of gas–liquid interface and the bubble length was determined by dynamic interface break-up, which is a function the volume ratio of two-phase fluids. Xu et al. (2012), working in our research group, successfully prepared round shaped microbubbles using a capillary embedded T-junction microchannel (Xu et al., 2006). In his study, the average bubble diameters were found varied with the shearing velocity (u_C) and the viscosity of continuous phase (μ_C) at $\mu_C u_C > 10^{-2}$, demonstrating the viscous shearing force of continuous phase is also important to control microbubble generation process. According to this shearing ruptured principle, the capillary embedded microchannel was improved in the subsequent studies, using a protruded capillary tip to intensify the shearing force from continuous phase (Wang et al., 2010, 2011). As a result, smaller bubbles were successfully prepared with less consumption of liquid phase. Pohorecki et al. introduced a switching mechanism for the bubble generation in different kinds of Y-shaped microchannels (Sobieszuk et al., 2010). This mechanism was similar with the shearing ruptured mechanism in T-junction microfluidic device. The capillary number (Ca) presenting the ratio of viscous force to interfacial tension was introduced in Pohorecki's correlation equations as a key factor to characterize the bubble length and the distance between neighboring bubbles. The same as T-junction and Y-junction microchannels, the microbubble generations in the flow-focusing and cross-junction microfluidic devices are also driven by the dynamic interface break-up and the shearing effect from continuous phase (Marmottant and Raven, 2009; Castro-Hernandez et al., 2011), thus the generation mechanisms of microbubbles in these microfluidic devices have similarities. However, since the detailed characters of gas–liquid interface movement and rupturing processes as well as the strength of shearing effect are different in different microfluidic devices, individual investigations are still indispensable.

Comparing with the microbubble generation studies on the T-junction, Y-junction, cross-junction and flow-focusing microfluidic devices, the two-phase flow and the bubble size laws are still less reported regarding the co-flowing microchannel, another important platform of microfluidic application. Only Marin et al.'s (2009) investigation was referred by us before preceding this work's study. Besides the unclear bubble generation rules in the co-flowing microchannel, so far as we know, all the working systems in the gas–liquid microflow studies are exclusively carried out with air, N_2 or CO_2 as the gas phase, but few talks about the organic gases, which are more important for the energy, environment and material fields. Microbubbles can be used in those processes, for example in the alkane gas cleaning processes to extrude sulfur contained impurities, to increase the apparent reaction rates.

Microfluidic devices are good platforms in the basic researches of microbubble generation, microbubble flow and gas–liquid mass transfer performances at microscale. This paper introduces a preliminary study on the microbubble generation mechanism in a co-flowing microfluidic device with organic and inorganic gases in the gas–liquid working systems. The pressure fluctuation in gas phase and the movement of gas–liquid interface are discussed and the main factors affecting the bubble size variation are introduced. Based on the bubble generation mechanism, a correlated equation is established to predict the average bubble diameters.

2. Experiment

2.1. Setup and method

The experiment setup was assembled by two syringe pumps (LSP02-1B, Longer, China), three gas tight syringes (5 mL, Gaoge, China), three coiled stainless still pipes (inner diameter 600 μm, outer diameter 1 mm), a temperature controlled heating chamber, a microscope (View Solutions, USA), a bottom light source (View Solutions, USA), a high speed COMS camera (PL-A741, PixeLINK, Canada) and a microfluidic chip as shown by Fig. 1a. The syringes and pumps were used to feed the two-phase fluids and control their flow rates. In the experiment, the volume flow rates of gases were varied from 50 μL/min to 200 μL/min and the volume flow rates of water phases were changed from 100 μL/min to 1200 μL/min. The heating chamber had an inner structure of 160 mm × 120 mm × 40 mm and it was placed on the microscope platform. The glass window on the heating chamber made the microchip visible from the outside. Two pieces of ceramic heating plate (220 V, 100 W) were placed in the heating chamber to heat the atmosphere in it. The temperature in the heating chamber was monitored by a temperature sensor and controlled by a commercial PID (proportion–integration–differentiation) controller. In the experiment, the temperature controlling system was turned on first. About 30 min later the temperature became stable and the two-phase fluids were subsequently pumped in. The 1.5 m long coiled pipes provided large heating area, helping fluids increase temperature. Tested by an infrared radiation thermometer (AR-350, Sigma Tech., Hong Kong), the temperature fluctuation in the heating room was less than 1.5 °C around the setting value.

The microfluidic chip was fabricated on a polymethyl methacrylate (PMMA) plate with precise end mills and sealed with another PMMA plate at 75 °C, 0.4 MPa with a high-pressure thermal sealing machine (A274, Techson, China). As shown in Fig. 1, the microchannel was fabricated to a cross junction and a tapered glass capillary was embedded in it to organize a co-flowing structure. The capillary tip was fabricated with a micropipette puller (P-97, Sutter, USA). The inner and outer diameters (d_{in} and d_{out}) of the capillary tip were 80 μm and 120 μm. Following the microchannel, a wide monitor channel was placed, used to observe the generated microbubbles at a relative low velocity. The microchannels in the PMMA chip had square cross-section with width and height equaling to 600 μm ($w=h=600$ μm). The length of the channel in the downstream of co-flowing microstructure was 15 mm. The width and length of the monitor channel was 2.0 mm and 10 mm. It had a same depth as the microchannel. Due to the short length of the microchannel, the pressure drop in the microfluidic chip was omitted, comparing with the atmosphere pressure. This assumption was easily proved in the experiment from the very short waiting time (<10 s) from the pump running to the bubble generating.

The microbubble generation process and the gas–liquid flow in the microchannel and monitor channel were recorded by the microscope and the CMOS camera. The camera worked at an exposure time of 10 μs and recording rate of 40 frames/s. Some pictures of the microbubbles in the co-flowing microchannel, the downstream microchannel and the monitor channel are provided in Fig. 2. According to the experimental result, not all the flowing bubbles in the microchannel had round shapes, bring difficulty for the size measurement. However, the two-phase fluids decelerated in the monitor channel and the bubbles changed to round shape. The bubble diameter (d) in the monitor channel was measured from the pixel number along the diameter direction with a microscope ruler of 158 pixels/mm. A self-made MATLAB code was used to calculate the average bubble diameter (Eq. (1)) and the bubble polydispersity (Eq. (2)) with at least 50 samples ($n>50$). Another important parameter characterizing the movement of gas–liquid interface – the maximum distance of retracting interface (backward movement of gas–liquid interface in the capillary tip, l_{inter}) in a bubble generation period—was also measured from the recorded videos as the shown in Fig. 2a.