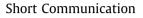
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## **Chemical Engineering Science**

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



# Dynamics of bubble formation in highly viscous liquids in a flow-focusing device



CHEMICAL

ENGINEERING SCIENCE

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#### HIGHLIGHTS

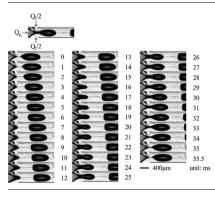
- Bubble formation in highly viscous liquids is studied.
- Liquid viscosity and gas-liquid flow rate ratio affect the bubble formation dynamics.
- The linear expansion stage and breakup stage for bubble formation are highlighted.
- A scaling law is proposed to predict the size of bubbles.

#### ARTICLE INFO

Article history: Received 23 November 2016 Received in revised form 14 June 2017 Accepted 16 June 2017 Available online 24 June 2017

Keywords: Microchannels Viscous liquids Gas-liquid two-phase flow Bubble Dynamics

#### G R A P H I C A L A B S T R A C T



#### ABSTRACT

This article reports the dynamics and mechanism for N<sub>2</sub> bubble formation in highly viscous glycerolwater mixtures in a flow-focusing device by using a high-speed digital camera. The evolution of the volume for the gaseous thread during bubble formation is highlighted. A square microchannel with 400  $\mu$ m × 400  $\mu$ m is used. The bubble formation process can be divided into an expansion stage and a breakup stage. The volume of the gaseous thread increases linearly with time in the two different stages, however, the growth rate in the breakup stage is always greater than that in the expansion stage. The growth rate for the evolution of the gaseous thread is controlled by the capillary number and the gasliquid flow rate ratio, with varied exponents for the two stages. The turning point of the two stages occurs at the moment when the width of gaseous neck reaches its maximum, and the dimensionless time at the turning point is related to the gas-liquid flow rate ratio. Finally, the volume of generated bubbles in highly viscous liquids in the flow-focusing device is scaled with the capillary number and the gasliquid flow rate ratio.

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#### 1. Introduction

Microchemical engineering and technology has attracted increasing attention because of its high rates for the mass and heat

\* Corresponding authors. E-mail addresses: ttfu@tju.edu.cn (T. Fu), ygma@tju.edu.cn (Y. Ma). transfer, the numbering-up facility, safety and flexible controllability (Elvira et al., 2013). The dynamics of multiphase flows in confined spaces is one of the key problems in the microchemical technology, such as the formation of bubbles and droplets in microchannels. Microbubbles have numerous applications in medicine (Lindner, 2004; Rodríguez-Rodríguez et al., 2015), food engineering (Zuniga and Aguilera, 2008), and materials synthesis



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$d_b$	diameter of bubble ( $\mu$ m)	V	volume of gaseous thread during bubble formation ( $\mu m^3$ )
$k_1$	growth rate in the expansion stage	$V_{f}$	final volume of the generated bubble ( $\mu m^3$ )
$k_2$	growth rate in the breakup stage	Ŵc	width of the channel (µm)
L	length of bubble (µm)	$W_{max}$	maximum width of the gaseous neck ( $\mu$ m)
l	length of gaseous tip (µm)	w(l)	width of gaseous tip (µm)
$P_g$	pressure of gas (Pa)	α	prefactor of gas-liquid flow rate ratio
$P_1$	pressure of liquid (Pa)	$\sigma$	surface tension (mN·m <sup>-1</sup> )
$Q_g$	volumetric flow rate of gas phase $(mL \cdot h^{-1})$	$\mu_{g}$	viscosity of gas (mPa·s)
$Q_1$	volumetric flow rate of liquid phase $(mL \cdot h^{-1})$	$\mu_l$	viscosity of liquid (mPa·s)
r	radius of bubble or droplet (µm)	$\varphi$	gas-liquid flow rate ratio ( $\varphi = Q_g/Q_l$ )
t	time (ms)	$ ho_g$	density of gas (kg·m <sup>-3</sup> )
Т	formation period of bubbles (ms)	$\rho_l$	density of liquid (kg·m <sup>-3</sup> )
$T_c$	dimensionless time at the turning point, $T_c = t/T$	Са	capillary number ( <i>Ca</i> = $\mu_l u / \sigma$ )
и	superficial velocity of liquid (m·s <sup>-1</sup> )	Re	Reynolds number ( $Re = w_c \rho_l u / \mu_l$ )

(Suslick and Price, 1999). Therefore, many contributions have been devoted to the formation and manipulation of bubbles in microchannels (Cao et al., 2006; Castro-Hernández et al., 2011; Hoeve et al., 2011; Marín et al., 2009; Xu et al., 2006; Chen et al., 2013).

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Bubble formation in microchannel was found to be mainly controlled by two mechanisms (Garstecki et al., 2006; Thorsen et al., 2001): the shearing mechanism in unconfined flow and the squeezing mechanism in completely confined flow. The shearing mechanism indicates that the bubble breaks up in the equilibrium of the surface tension and viscous force, and the liquid viscosity plays an important role in bubble formation. Thorsen et al. (2001) proposed that the radius *r* of the generated droplet in oil in the T-shaped microchannel is inversely proportional to the capillary number *Ca*:  $r \propto Ca^{-1}$ , with capillary number *Ca* representing the ratio of the viscous force over the surface tension force, and  $Ca = \mu_i u / \sigma$ .  $\mu_i$ , u and  $\sigma$  represent the viscosity and velocity of the continuous phase and the surface tension between the two phases, respectively. The squeezing mechanism implies that the bubble formation is controlled by the accumulated pressure in the blocked liquid phase around the growing gaseous thread, and the bubble size is determined by the gas-liquid flow rate ratio and the geometry of the channel. In this case, the bubble size is not dependent of the liquid viscosity. Garstecki et al. (2006) proposed that the bubble size was proportional to the gas-liquid flow rate ratio in a Tshaped microchannel in completely confined flow at low capillary numbers ( $Ca \le 10^{-2}$ ):  $L/w_c = 1 + \alpha \varphi$ , where L,  $w_c$  and  $\varphi$  represent the bubble length, channel width and the gas-liquid flow rate ratio, respectively; and  $\alpha$  is an adjustable parameter related to the geometry of the channel. In addition, some studies showed that the bubble formation was controlled by the joint shearing mechanism and the squeezing mechanism for partly confined breakup of gaseous thread during bubble formation (Christopher et al., 2008; De Menech et al., 2008; Fu et al., 2010; Xu et al., 2008). Castro-Hernández et al. (2011) reported a new regime for bubble formation in low viscous fluids in a flow-focusing microfluidic device. and scaled the bubble size with the flow rate ratio and the viscosity ratio of gas and liquid phases:  $d_b/w_c = 2.75(\mu_g/\mu_l)^{1/12}\varphi^{5/12}$ , in which,  $d_b$  and  $\mu_g$  represent bubble diameter and viscosity of gas phase, respectively.

Although a lot of efforts have been devoted to the bubble formation mechanism and the prediction of bubble size in microfluidic devices, most of these studies have focused on bubble formation in low viscous fluids (Elvira et al., 2013). Highly viscous fluids are frequently encountered in polymer engineering and food engineering (Bolaños-Jiménez et al., 2009; Thoroddsen et al., 2007), but there are few reports for bubble formation in highly viscous fluids in microdevices (Lu et al., 2014). Furthermore, the literature mainly considers the final volume of the generated bubbles, however, the mechanism and detailed information for bubble formation process have not been fully explored yet. This work investigates the dynamics for bubble formation in highly viscous liquids in a flow-focusing device. The evolution of the volume of the gaseous thread during bubble formation is clarified, and the effect of the operating conditions such as the gas-liquid flow rate ratio, and the liquid viscosity on the dynamics of bubble formation is highlighted.

#### 2. Experimental procedures

Experimental facilities include a microfluidic flow-focusing device, a fluid control system and an image acquisition system, as shown in Fig. 1(a). Gas is fed from a  $N_2$  cylinder and the gas flow rate is controlled by a micrometering value (KOFLOC, Japan). The pressure  $P_{\alpha}$  at the exit of the N<sub>2</sub> cylinder is maintained at a constant value of 0.7 MPa. Thus, the actual volumetric gas flow rate is estimated by the ratio of the volume of generated bubbles to the bubble formation period as the actual flow rate varied with the change of the resistance in the downstream channel, according to the Hagen-Poiseuille relationship (Garstecki et al., 2005). Liquid is pumped into the horizontal microchannel by a syringe pump (PHD 2000, Harvard Apparatus, USA) through polyethylene rubber tube. The square microchannel with a cross-section of  $400\,\mu m \times 400\,\mu m$  is fabricated in a polymethyl methacrylate (PMMA) plate (45 mm  $\times$  27.5 mm  $\times$  2 mm) by milling. The dispersed gas phase is introduced from the main channel with a volumetric flow rate of  $Q_g$ , and the continuous phase liquid is fed from the two lateral channels with a volumetric flow rate of  $Q_1/2$ , as shown in Fig. 1(b). The process of the bubble formation is magnified by a microscope (ECLIPSE Ti-U, Nikon, Japan) and recorded by a high-speed digital camera (MotionProY5, IDT, USA). The recording rate of the camera is 4000 fps (frames per second). Halogen lamp is placed at the other side of the microfluidic device to provide sufficient light for the image acquisition. The images for bubble formation are captured when the flow reaches stable after a new flow condition is set. All the experiments are conducted at room temperature and atmospheric pressure.

Highly viscous glycerol and 99%, 93%, 90%, 87%, 80%, 75% glycerol-water solutions are used as the continuous phase. The density of liquids  $\rho_l$  is measured by using a pycnometer. The surface tension of the liquid in air  $\sigma$  is measured by the pendant drop method via a tensiometer (OCAH200, Data Physics instruments GmbH, Germany). The viscosity of the liquid  $\mu_l$  is measured by

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