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Effect of microchannel junction angle on two-phase liquid-gas Taylor flow

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HIGHLIGHTS

- Effects of fluid flow rates and junction angle θ on liquid-gas Taylor flow, hitherto not well studied, were investigated theoretically and experimentally for $20^\circ \leq \theta \leq 160^\circ$.
- Theoretical model to predict gas bubble and liquid slug sizes for different θ and flow rates was established.
- For $\theta = 20^\circ$, the unit cell volumes decreased significantly with decreasing liquid or increasing gas flow rate.
- Although having similar trends, for $\theta = 45^\circ, 90^\circ, 135^\circ$ and 160° , the unit cell volumes were less sensitive to fluid flow rate changes and are all less than that of $\theta = 20^\circ$.

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ABSTRACT

Two-phase liquid-gas Taylor flow triggered by blocking-squeezing mechanism was studied with different junction angle θ microchannels, i.e. $20^\circ, 45^\circ, 90^\circ, 135^\circ$ and 160° , at various liquid (ethanol) and gas (He) flow rates. We experimentally investigated the effects of flow rates and θ on the gas bubble V_B and liquid slug V_S volumes. A theoretical model was formulated for the quantitative predictions of bubble and slug sizes for different θ and flow rates. Good agreements were obtained between theoretical predictions and experimental observations. The unit cell volume V_U ($V_B + V_S$) decreased pronouncedly for the 20° channel with decreasing liquid or increasing gas flow rate, due to the slight increase in V_B and large decrease in V_S . In comparison, for the $45^\circ, 90^\circ, 135^\circ$ and 160° channels with increasing liquid or decreasing gas flow rate, V_U were less sensitive to fluid flow rate changes, due to the approximate cancellation between V_B decrease and V_S increase. For the 20° and 45° channels, it produced larger V_U , due to larger V_B and V_S , when compared to the 90° channel. This is caused by the larger gas bubble throat width D_N at the junction when $\theta < 90^\circ$. As for the 135° and 160° channels ($\theta > 90^\circ$), D_N is approximately equal to the gas channel width, with V_B, V_S and V_U approximately the same as the 90° channel. With $\theta \geq 90^\circ$ (i.e. $90^\circ, 135^\circ$ and 160° channels), as evident from the smaller V_U , higher gas bubble density can be obtained when compared to $\theta < 90^\circ$ (i.e. 20° and 45° channels). Hitherto, this observation has not been realized, and the mechanics is first investigated here with the employment of extreme θ (i.e. 20° and 160°). A thorough understanding of the underlying mechanics affecting Taylor flow can facilitate its exploitation for controlled gas bubble and liquid slug generation. Our theoretical model facilitates the tuning of the channel designs and fluid flow rates to achieve the desired gas bubble and liquid slug sizes for specific applications.

1. Introduction

Microfluidic technology for multiphase flow has received a great deal of attention of many researchers over the past decades. It has been applied in a wide range of fields such as chemistry, material science, optofluidics and biomedical (Theberge et al., 2010; Huebner et al., 2008; Seemann et al., 2011; Teh et al., 2008). The common microfluidic devices are based on cross-flow (Thorsen et al., 2001), co-flow (Cramer et al., 2004) and focused-flow (Anna et al., 2003). In a cross-flow device, two immiscible fluids meet at an angle between the inlets, while in a co-flow device, they meet in parallel streams; for a focused-flow device, the fluids are focused for the generation of droplets or bubbles (Haase, 2017).

Taylor flow is a common two-phase flow configuration that consists of elongated gas bubbles separated by liquid slugs (see Fig. 1). The bubbles almost completely fill the cross-section of the microchannel with a thin liquid film separating them from the channel wall (Abadie et al., 2012). This flow pattern is often achieved with a cross-flow T-junction device, i.e. 90° junction (inlet) angle (Garstecki et al., 2006; van Steijn et al., 2007; Yue et al., 2008), which was first employed to produce segmented liquid-liquid flow (Thorsen et al., 2001), and later to produce segmented liquid-gas flow (Gunther et al., 2004).

Fig 1. Schematic diagrams of (a) blocking process, (b) squeezing process, and (c) gas bubble and liquid slug lengths for two-phase liquid-gas Taylor flow.

Precise distributions of the gas bubbles and liquid slugs are desirable because it ensures predictable outcomes for enhancing the performance of critical processes such as heat and mass transfer in chemical reactors (Ward et al.,

2005; Kiwi-Minsker and Renken, 2005; Dahl et al., 2015), extractors (Kashid et al., 2007) and biocatalytic reactors (Bolivar and Nidetzky, 2013). Hence, the ability to accurately predict and control the gas bubble and liquid slug sizes is the key motivation for this investigation.

Various investigations have been conducted to study the dynamics of gas bubble generation process in Taylor flow (Garstecki et al., 2006; De Menech et al., 2008; Abate et al., 2012; Zhang et al., 2015; Soh et al., 2016; Nekouei and Vanapalli, 2017; Khan et al., 2018). Garstecki et al. (2006) demonstrated that the gas bubble formation for Taylor flow in T-junction microchannel is controlled by the blocking-squeezing mechanism at low capillary number ($Ca < 10^{-2}$). The gas bubble break-up process can be separated into two parts, namely blocking and squeezing. The "blocking" process refers to the blocking of liquid flow path (continuous phase) by the emerging gas bubble (dispersed phase) at the junction (see Fig. 1a). The subsequent "squeezing" process is caused by the increasing pressure drop across the gas bubble, which pinches the bubble neck (see Fig. 1b), eventually breaking it off.

De Menech et al. (2008) had discovered three regimes dependent on the Ca number for two-phase liquid-gas flow, namely the blocking-squeezing, dripping and jetting regimes. The blocking-squeezing mechanism for Taylor flow at low Ca has been affirmed by Abate et al. (2012), who utilized Laplace sensors to perform a detailed analysis of the pressure fluctuations during the bubble break-up in two-phase liquid-gas flow. They also concluded that the squeezing process is still prevalent even at high Ca number ($Ca = 0.21$), where the shear stress is significant.

Zhang et al. (2015) had observed identical two-phase flow regimes in cylindrical T-junction microchannels as in planar geometries (e.g. rectangular cross-sections) with the same stability. Soh et al. (2016), with a refined volume-of-fluid (VOF) model, had numerically investigated the velocity fields during the gas bubble formation for the squeezing and dripping regimes, and found that large recirculating flows ensued following the bubble break-off.

Nekouei and Vanapalli (2017) had also employed simulations based on VOF method for the investigation of the effect of viscosity ratio (dispersed phase to continuous phase) on the dispersed drop size, and reach the conclusion that there is only strong dependency when the ratio is more than 1. Similarly with VOF method, Khan et al. (2018) discovered that the pressure drop across the gas bubble during the squeezing process is affected by the flow velocity, wall contact angle and surface tension, which in turn affect the generated bubble size.

Although there are abundant investigations (Garstecki et al., 2006; De Menech et al., 2008; Abate et al., 2012; Zhang et al., 2015; Soh et al., 2016; Nekouei and Vanapalli, 2017; Khan et al., 2018) on the dynamics of gas bubble generation for two-phase liquid-gas Taylor flow, limited investigations have been performed to study the effect of microchannel junction angle (Dietrich et al., 2008; Fries and Rudolf von Rohr, 2009; Tan et al., 2009; Ngo et al., 2016). Dietrich et al. (2008) investigated the gas bubble

formation in focused-flow micro-devices (with one main channel and two side channels) for various junction angles, i.e. 90° (cross shape), 45° and 30° (convergence shape). It was revealed that the bubble formation depends on the geometry of the focusing section, with the bubble size increases with decreasing junction angle.

Based on VOF method, Ngo et al. (2016) numerically investigated the effect of junction angle (ranging from 30° to 90°) on droplet generation under alternating digitized pattern formation (ADPF) in focused-flow configuration. They discovered that the size of the alternating droplets decreases with increasing junction angle, and the droplet size decreases more significantly when the junction angle is less than 60°.

With polydimethylsiloxane (PDMS) channels, Fries and Rudolf von Rohr (2009), using a similar cross-flow configuration as shown in Fig. 1a, demonstrated that short unit cell length (defined as the combined length of gas bubble and liquid slug), can be achieved when the gas phase is flowing in the direction of the liquid phase, i.e. at a channel junction angle less than 90° as defined in our investigation.

In contrast, using polymethyl methacrylate (PMMA) channels, Tan et al. (2009) demonstrated that the gas bubble length decreases exponentially with increasing channel junction angle (> 90°). However, it has to be highlighted that different inlet conditions were employed; inlets of gas and liquid were interchanged as compared to that shown in Fig. 1. This configuration causes the gas (continuous phase) flow to be blocked by the emerging liquid (dispersed phase) slug instead.

Both investigations (Fries and Rudolf von Rohr, 2009; Tan et al., 2009) proposed correlation equations to describe the change of gas bubble length with respect to junction angle. However, their equations contain fitting parameters which can only be obtained from experiments. There are no clear physical interpretations of these parameters. Furthermore, hitherto, the effect of junction angle with fluid flow rate interaction is not well-investigated, and an in-depth understanding of the underlying mechanics will be beneficial.

Our investigation attempts to unravel the fundamental mechanics of gas bubble formation at different microchannel junction angles. The effects of fluid flow rates and junction angle on the gas bubble and liquid slug sizes were experimentally observed over a wide range of junction angles. In addition, a theoretical model on the gas bubble formation process was formulated to predict the gas bubble and liquid slug sizes at different junction angles, gas and liquid flow rates.

2. Materials and methods

2.1 Design and fabrication of microchannels

Fig. 2 shows the three-dimensional (3D) schematic microfluidic device for liquid-gas Taylor flow, with 90° junction angle channel used as an illustration. An example of a microfluidic junction angle chip is shown in Fig. S1 of the Supplementary Material. Due to its promising physical and

chemical properties (Nunes et al., 2010), such as optical transparency, low gas permeability, temperature tolerance and chemical resistance, cyclic olefin polymer (COP) has recently emerged as a potential alternative to the commonly used substrate materials, e.g. PDMS and PMMA. Thus, COP (Zeonor 1420R, 2mm thickness) was employed as the substrate material in our investigation, which is well-suited for chemical and biological applications.

The fabrication of the microfluidic chip was performed with computer numerical control (CNC) micromilling machine Mini Flat P40 from isel Germany AG. Rectangular microchannels of different junction angles, i.e. 20°, 45°, 90°, 135° and 160°, were milled on the bottom COP slab as shown in Fig. 2. The widths of the gas and liquid channels were $185 \pm 5 \mu\text{m}$ and $286 \pm 6 \mu\text{m}$ respectively, and the heights of the microchannels were $152 \pm 10 \mu\text{m}$ as measured by Nanovea CHR 150 profilometer (see Fig. 1a). The roughness arithmetic mean R_a of the milled microchannels were $= 2.0 \pm 0.6 \mu\text{m}$.

Fig 2. Schematic diagram of experimental setup with 3D microfluidic device for liquid-gas Taylor flow, with 90° junction angle channel used as illustration.

The channel size difference between the gas and liquid inlets has been shown to affect the bubble generation process in Taylor flow by others (Garstecki et al., 2006; Fries and Rudolf von Rohr, 2009; van Steijn et al. 2010). The blocking-squeezing mechanism can be enhanced when the liquid channel width is larger than the channel height, and the gas channel is at least equal to half of the liquid channel (Garstecki et al., 2006; Fries and Rudolf von Rohr, 2009). This will result in less variation of the bubble and liquid slug sizes in the experiments, which will produce significantly smaller standard deviations (Fries and Rudolf von Rohr, 2009). Thus, we have chosen our channel dimensions in a similar fashion to minimize the experimental variation of the bubble and liquid slug sizes.

Two vertical through holes (diameter of 1.58mm) on the top COP slab were milled to enable fluid access of the microfluidic system (see Fig. 2). Alignment studs and slots were also milled to facilitate the alignment of the top and bottom COP slabs during bonding. With appropriate milling, upon bonding of the top and bottom COP slabs, a horizontal square slot (height and width of 1.58mm, and length

of 5mm) was formed, to facilitate the insertion of the outlet port.

The top and bottom COP slabs (see Fig. 2) were bonded via force-assisted thermal bonding process (Lim et al., 2017; Lim et al., 2018) with a Specac Atlas manual hydraulic press with heated platens. Two thin PDMS layers (~3mm thick each) were employed to sandwich both the top and bottom pieces between the hot press to compensate for the non-uniformities in the platen flatness. This facilitated the application of uniform pressure across the chip surface to ensure even bonding. The temperature was set at 136°C with a piston force of 0.5kN for 5min.

Inlet/outlet ports (N-333 NanoPort Assembly, IDEX Health & Science) were integrated to the microfluidic chip for tubing connections in the experiments (see Fig. 2) (as discussed in Sec. 2.2). The nanoports were glued to the chip via cyanoacrylate adhesive (Contact VA 2500 HT, Weicon) with a set time of 12h. Then, they were sealed with Teflon tape and encapsulated by epoxy steel adhesive (Pro-Spec) at 1:1 mixing ratio. Lastly, the device was left in room conditions for 24h for the adhesives to reach its full strength.

2.2 Experiment setup and procedure

Schematic diagram of the experimental setup is shown in Fig. 2. The image of the experimental setup is shown in Fig. S2 of the Supplementary Material. The experiments were observed at 10x magnification (spatial resolution of 1.95µm per pixel) with an inverted microscope (Nikon Eclipse Ti-U) connected to a high speed camera (Fastcam SA5, Photron) for capturing the images of the gas bubbles and liquid slugs during the experiment. A high intensity light source was required to capture the images at high frame rate and short exposure time. An ultra-high pressure mercury lamp (Nikon Intensilight 130W) with an ultraviolet (UV) cut-off filter was employed to supply the white light illumination.

The liquid phase (Ethanol 96%, GCE laboratory chemicals) contained in a 5ml glass syringe (Hamilton Gastight 1005 TLL glass syringe) was delivered via a syringe pump (KDS Legato 200 series), and the delivery of the gas phase (helium (He)) from a gas tank was controlled by a mass flow controller (Bronkhorst) (see Fig. 2).

Table 1. Density, dynamic viscosity and surface tension of experimental fluids at room conditions.

Fluids	Density (kg/m ³)	Dynamic viscosity (Pa.s)	Surface Tension (mN/m)
Helium gas	0.179	1.96×10^{-5}	-
Ethanol 96%	809	1.2×10^{-3}	25.87 ± 0.06 (measured)

The physical properties of the experimental fluids (i.e. He gas and Ethanol 96%), namely density, dynamic viscosity and surface tension (measured via pendant drop method), at room conditions are summarized in Table 1. The static contact angle of the ethanol solution on the surface of

COP substrate (Zeonor 1420R) was measured to be $12.3 \pm 0.4^\circ$ (high wettability). Experimental measurements for the surface tension and contact angle (measured by DataPhysics OCA 35 optical contact angle system) can be found in Section S2 of the Supplementary Material.

Fluorinated ethylene propylene (FEP) tubing (internal diameter (ID) = 0.25mm and outer diameter (OD) = 1.58mm, VICI-Jour) was used to connect the liquid and gas supplies to the inlet channels, and the outlet channel to a sponge for the gradual absorption of the exiting liquid.

The experiments were performed with three different liquid (ethanol) flow rates, namely 100µl/min, 180µl/min and 260µl/min, at a constant gas (He) flow rate of 100µl/min. In addition, experimental runs with another two different gas flow rates, namely 180µl/min and 260µl/min, at a constant liquid flow rate of 260µl/min were also conducted. All the experiments were carried out at room temperature of ~20°C and atmospheric pressure of 1atm.

Before commencement of an experiment, the FEP tubing and microchannel were flushed with ethanol. The syringe pump and mass flow controller were then turned on to deliver the liquid phase and gas phase into the channel. Images were only captured after the bubble generation reached a steady-state condition (after ~2min). The microchannel junction and the downstream region of the junction (see Fig. 1) were the areas of interest for the current investigation. The high speed camera was set at a frame rate between 4000fps and 7000fps depending on the liquid and gas flow rates to acquire images (1024 x 512 pixels) for analysis. The experiment was performed three times (triplicate) for a given set of liquid and gas flow rates and a particular junction angle to ensure repeatability.

2.3 Measurements of gas bubble and liquid slug lengths

The images obtained from the liquid-gas Taylor flow experiments as discussed in Sec. 2.2 were utilized for the measurements of the gas bubble and liquid slug lengths. A MATLAB program was written to process and analyze the images. The experimental gas bubble length $L_{B(Exp)}$ and liquid slug length $L_{S(Exp)}$ were determined from the grayscale images based on the extremes of the gas-liquid menisci/interfaces (see Fig. 1c).

Ten gas bubble and ten liquid slug lengths were measured from different images for each experimental run. Since the experiments were conducted in triplicate, a total of 30 gas bubble and 30 liquid slug lengths were measured for a given liquid and gas flow rates, and junction angle.

The experimental gas bubble volume $V_{B(Exp)}$ and liquid slug volume $V_{S(Exp)}$ were calculated by substituting their experimentally measured lengths into Eqn. 1 and 2 respectively, which will be discussed in Sec. 2.4.

2.4 Evaluation of gas bubble and liquid slug volumes

Liquid-gas flow system has been extensively exploited in various microscale reaction and separation processes, such as synthesis of advanced materials (Sebastian Cabeza et al., 2012; Rahman et al., 2013), enhancement of mass transfer performance (TeGrotenhuis et al., 2000; Jähnisch et al.,

2004; Sauzade and Cubaud 2013), fast measurement of gas solubility (Tan et al., 2010), liquid-gas heterogeneous reactions (Jähnisch et al., 2000; de Mas et al., 2001; Inoue et al., 2007) and deoxyribonucleic acid (DNA) sequencing (Abate et al., 2013).

Quantitative analysis for the aforementioned applications requires the volumes of the gas and liquid segments to be known. However, only two-dimensional (2D) images of the gas bubbles and liquid slugs can be observed with an optical microscope. Knowing the geometry of the channel, mathematical models can be utilized to calculate their volumes based on the measured bubble and slug lengths.

Most researchers (Garstecki et al., 2006; Cubaud et al., 2012) conveniently describe the shape of the gas bubble/liquid slug as a block with length $L_{B/S}$, for which the volume can be obtained from the simple estimate $V_{B/S} = HWL_{B/S}$, where H and W are the height and width of the rectangular microchannel respectively. However, a more accurate estimation can be arrived at by taking into consideration that the gas bubble/liquid slug has rounded caps at the front and rear of the gas bubble/liquid slug and does not conform to the four corners of the microchannel.

Although there are several models (Leclerc et al., 2010; Li et al., 2012; Sauzade and Cubaud 2013) that account for the abovementioned concerns, Musterd et al. (2015) provides an universal model which enables the calculation of the gas bubble and liquid slug volumes from the 2D microscope images for a wide variety of microchannel cross-sectional geometries. As such, the model developed by Musterd et al. (2015) was adopted in this investigation.

Since the microchannel cross section employed in our investigation was rectangular, the gas bubble volume can be calculated as follows (Musterd et al., 2015):

$$V_B = \left[HW_L - (4 - \pi) \left(\frac{2}{H} - \frac{2}{W_L} \right)^{-2} \right] \left(L_B - \frac{W_L}{3} \right), \quad (1)$$

where H and W_L are the height and width of the liquid channel respectively, and L_B is the gas bubble length.

Fig 3. Schematic diagram showing subtraction of two small bubbles from one big bubble (where liquid slug is sandwiched in between two small bubbles) to obtain liquid slug.

In the investigation by Musterd et al. (2015), the expression to calculate the liquid slug volume is not derived. By considering the liquid slug and two gas bubbles (where the

liquid slug is sandwiched in between) (see Fig. 3) as a big gas bubble and subtracting the two small gas bubbles from it, the liquid slug volume can be obtained:

$$V_S = V_{S+2B} - 2V_B = \left[HW_L - (4 - \pi) \left(\frac{2}{H} - \frac{2}{W_L} \right)^{-2} \right] \left(L_S + \frac{W_L}{3} \right), \quad (2)$$

where V_{S+2B} is the combined volume of a liquid slug and two gas bubbles (treated as a big gas bubble), and L_S is the liquid slug length.

The liquid film surrounding the gas bubble is assumed to be negligible as the film is infinitely thin for capillary number $Ca < 10^{-2}$ (Garstecki et al., 2006; Yue et al., 2008; Guo and Chen, 2009). The capillary number $Ca = \mu_L U_{TP} / \sigma_L = \mu_L (u_L + u_G) / \sigma_L = \mu_L (Q_L + Q_G) / A_L \sigma_L$, where A_L is the cross sectional area of liquid channel, U_{TP} is the superficial two-phase velocity, σ_L is the surface tension and μ_L is the dynamic viscosity of liquid phase, and u_L , u_G , Q_L and Q_G are the average speeds and flow rates of the liquid and gas phases respectively. For our experimental parameters, the most extreme case, namely He and ethanol flow rates = 260 $\mu\text{l}/\text{min}$ and liquid microchannel cross sectional area = 43500 μm^2 , gave a capillary number $Ca = 9.3 \times 10^{-3}$ ($< 10^{-2}$).

3. Results and discussion

3.1 Theoretical model for prediction of gas bubble and liquid slug sizes

Fig. 4 shows the image sequence for the gas bubble generation in microchannels with different junction angles. With that, a theoretical model to study the effect of channel junction angle on liquid-gas Taylor flow was formulated by extending the scaling relation developed by Garstecki et al. (2006) for the quantitative prediction of the gas bubble/liquid droplet size for a T-junction (i.e. 90° junction angle) channel for the current configurations.

The gas bubble volumes for the different junction angle microchannels versus capillary number Ca and Weber number We are shown in Fig. 5a and 5b respectively. The Weber number $We = Ca \times Re$, where $Re = \rho_L U_{TP} D_H / \mu_L$ is the Reynolds number; $D_H = 2HW_L / (H + W_L)$ is hydraulic diameter of the liquid channel and ρ_L is the density of the liquid phase. Fig. 5a shows that the bubble volumes decreased exponentially with increasing Ca , whereby the 20° channel had the largest volume followed by the 45° channel, and then the 90°, 135° and 160° channels. Indeed, the 45°, 90°, 135° and 160° channels had approximately similar bubble volumes. Similar observations are shown in Fig. 5b for increasing We .

As the highest Ca number in our experiments was 9.3×10^{-3} ($< 10^{-2}$) (see Section 2.4), the blocking-squeezing regime applied (Garstecki et al., 2006). As such, the break-up of the gas bubble is caused by the pressure drop across the emerging bubble. Our theoretical model does not consider the effect of fluid properties such as the surface tension; as within this blocking-squeezing regime, the fluid properties can be omitted as earlier demonstrated and discussed by Garstecki et al. (2006) and van Steijn et al. (2007).

Fig 4. Image sequence on the gas bubble generation for microchannel junction angles of (a) 20°, (b) 45°, (c) 90°, (d) 135° and (e) 160°. Experimental images at liquid (ethanol) flow rate $Q_L = 180\mu\text{l}/\text{min}$ and gas (He) flow rate $Q_G = 100\mu\text{l}/\text{min}$ were shown for illustration.

Fig 5. Gas bubble volumes for different junction angle micro-channels versus (a) capillary number Ca and (b) Weber number We .

The basic principle for deriving the gas bubble length is to determine the rate of growth of the bubble during the blocking and squeezing processes. The total length of the gas bubble L_B can be divided into two parts, namely the part of bubble formed during the blocking process $L_{B(Block)}$,

and the remaining part which is formed during the squeezing process $L_{B(Squeeze)}$ (Garstecki et al., 2006):

$$L_B = L_{B(Block)} + L_{B(Squeeze)}. \quad (3)$$

Garstecki et al. (2006) assumed $L_{B(Block)}$ to be approximately equal to the liquid channel width W_L , implying that the gas phase fully blocks the outlet channel completely. However, our experimental results show that the gas phase only blocks the outlet channel partially during the blocking process (see Fig. 4). The bubble will only completely blocks the liquid channel during the subsequent squeezing process. For a more accurate prediction, this partial blockage effect will be taken into account in our analysis.

The junction width between the gas and liquid channels for any junction angle θ is given by:

$$W_J = \frac{W_G}{\sin\theta}, \quad (4)$$

where W_G is the gas channel width.

To account for the maximum partial blocking of liquid channel, a static parameter will be sufficient to describe the phenomenon, namely the coverage ratio γ :

$$\gamma = \frac{D_C}{W_L}, \quad (5)$$

where D_C is the experimentally measured diameter of bubble circular front at the end of the blocking phase, which is defined as the instant before the gas bubble throat width D_N starts to decrease (see Fig. 6a).

The diameter of the circular front D_c is γW_L and the mean length along the center of the “gas neck” D_L can be obtained via Pythagoras’ theorem as $(D_N^2/4 + \gamma^2 W_L^2/4)^{1/2}$ (see Fig. 6a), where D_N is the gas bubble throat width across the channel junction. Thus, $L_{B(Block)}$ can be estimated as the sum of D_c and D_L , which can be written as:

$$L_{B(Block)} = \gamma W_L + \frac{1}{2} \sqrt{D_N^2 + \gamma^2 W_L^2}. \quad (6)$$

Fig 6. Geometrical analysis to estimate gas bubble length during blocking process for (a) 20° and (b) 160° microchannels, where γ is coverage ratio, θ is junction angle, D_c is bubble front diameter, D_L is mean gas neck length, D_N is gas bubble throat width, W_G is gas channel width, W_j is junction width and W_L is liquid channel width.

For $\theta \leq 90^\circ$, D_N is equal to W_j as the gas bubble spans across the entire junction width. After substituting Eqn. 4 into Eqn. 6, $L_{B(Block)}$ can be expressed as:

$$L_{B(Block)} = \gamma W_L + \frac{1}{2} \sqrt{\frac{W_G^2}{\sin^2 \theta} + \gamma^2 W_L^2}; \text{ for } \theta \leq 90^\circ. \quad (7)$$

For $\theta > 90^\circ$, the gas phase which is flowing against the liquid phase is forced to change flow direction (as seen in 135° and 160° junction angle microchannels in Fig. 4d and 4e) and the bubble does not span across the entire junction width. This causes D_N to be approximately equal to the gas channel width W_G instead of W_j (see Fig. 6b); substituting this into Eqn. 6 gives:

$$L_{B(Block)} = \gamma W_L + \frac{1}{2} \sqrt{W_G^2 + \gamma^2 W_L^2}; \text{ for } \theta > 90^\circ. \quad (8)$$

It should be highlighted here that Eqn. 8 is identical to Eqn. 7 with $\theta = 90^\circ$. This shows that increasing θ beyond 90° does not change the size of gas bubbles, i.e. instead of $\sin\theta$, $\sin\theta$ is replaced by 1 for all θ larger than 90°. The constant bubble size with increasing θ beyond 90° has been verified in our experiments (see Sec. 3.2).

Based on the experimental images for the five junction angles investigated, the coverage ratio γ was measured to be 0.8 ± 0.1 . The measurement of γ was performed at the end of the blocking phase, which is defined as the instant

before the gas bubble throat width D_N starts to decrease. Thus, γ was estimated as 0.8 (mean value) for the theoretical analysis. γ is expected to vary with the widths of the gas and liquid channels. However, as the channel widths were held constant in this investigation, a constant value of γ was employed.

The pressure increase due to the constricted liquid flow causes the “squeezing” of the gas bubble throat (see Fig. 4), which D_N shrinks (see Fig. 1b) at an estimate rate equals to the average speed of the liquid phase u_L (Garstecki et al., 2006):

$$v_{Squeeze} = u_L = \frac{Q_L}{A_L} = \frac{Q_L}{HW_L}, \quad (9)$$

where Q_L is the liquid flow rate, A_L is the cross sectional area of the liquid microchannel and H is the channel height.

During the squeezing process, the gas bubble will completely block the liquid channel. As such, the bubble is assumed to elongate at an estimate rate (Garstecki et al., 2006):

$$v_{Growth} = \frac{Q_G}{A_L} = \frac{Q_G}{HW_L}, \quad (10)$$

where Q_G is the gas flow rate.

Therefore, $L_{B(Squeeze)}$ can be obtained by (Garstecki et al., 2006):

$$L_{B(Squeeze)} = t_{Squeeze} \cdot v_{Growth}, \quad (11)$$

where $t_{Squeeze}$ is the squeezing time.

The minimum thread of gas bubble throat width $D_{N(Min)}$ (just before squeeze-off instance) for the different experimental junction angle microchannels were close to zero, in comparison to the initial gas bubble throat width D_N across the channel junctions (see Fig. 7). As such, the squeezing time can be expressed as:

$$t_{Squeeze} = \frac{D_N}{v_{Squeeze}} = \frac{W_j}{v_{Squeeze}}. \quad (12)$$

Knowing that D_N is approximately equal to W_G for $\theta > 90^\circ$, and substituting Eqn. 4, 9, 10 and 12 into Eqn. 11:

$$L_{B(Squeeze)} = \left(\frac{W_G}{\sin \theta} \right) \left(\frac{Q_G}{Q_L} \right); \text{ if } \theta > 90^\circ, \sin \theta \text{ replaces by } 1. \quad (13)$$

Fig 7. Minimum thread of gas bubble throat width $D_{N(Min)}$ (just before squeeze-off instance) for different experimental junction angle microchannels, in comparison to initial gas bubble throat width D_N across channel junctions.

Fig 8. Comparisons between experimental and theoretical (a) gas bubble volumes, (b) liquid slug volumes, (c) unit cell volumes and (d) gas bubble volume to unit cell volume ratios for microfluidic devices with channel junction angles of 20°, 45°, 90°, 135° and 160°, at various liquid (ethanol) flow rates with a constant gas (He) flow rate of 100µl/min.

It is noted from Eqn. 13 that $L_{B(Squeeze)}$ is reduced to $W_G Q_G / Q_L$ when $\theta = 90^\circ$, which is derived by Garstecki et al. (2006). Differing from their investigation, Eqn.13 accounts for the effect of junction angle on the gas bubble development during the squeezing process.

Substituting Eqn. 7, 8 and 13 into Eqn. 3 yields the final expression for the length of the gas bubble L_B :

$$L_B = \gamma W_L + \frac{1}{2} \sqrt{\frac{W_G^2}{\sin^2 \theta} + \gamma^2 W_L^2} + \left(\frac{W_G}{\sin \theta} \right) \left(\frac{Q_G}{Q_L} \right); \quad (14)$$

if $\theta > 90^\circ$, $\sin \theta$ replaces by 1

The theoretical gas bubble volumes $V_{B(Theo)}$ for the experimental conditions as discussed in Sec. 2.2 were calculated by substituting their theoretically predicted lengths $L_{B(Theo)}$ given by Eqn. 14 into Eqn. 1.

To satisfy the law of mass conservation, the theoretical liquid slug volumes $V_{S(Theo)}$ can be obtained via multiplication of $V_{B(Theo)}$ by the gas to liquid flow rate ratios Q_L/Q_G :

$$V_{S(Theo)} = \frac{V_{B(Theo)} Q_L}{Q_G}. \quad (15)$$

Comparisons between the experimental and theoretical results are presented in Sec. 3.2.

3.2 Comparison between experimental and theoretical results

Fig. 8 and 9 show the comparisons between experimental and theoretical results for gas bubble volume (see Fig. 8a and 9b), liquid slug volume (Fig. 8b and 9b), unit cell volume (Fig. 8c and 9c) and gas bubble volume to unit cell ratio (Fig. 8d and 9d), at various liquid (ethanol) and gas (He) flow rates. Good agreements were achieved between theoretical results and experimental results.

Through comparing the produced gas bubble volume with the volume of gas phase injected from a gas tank via a mass flow controller, the variations of the gas bubble volume are less than 12%. Within an experimental run, the monodisperse gas bubbles were measured to be highly uniform with polydispersity index of $2 \pm 1\%$, where $\sigma_i = \text{mean bubble volume} / \text{standard deviation} \times 100\%$ as defined in (Xu et al. 2006).

For our theoretical model (refer to Sec. 3.1), the gas bubble length formed during the blocking process $L_{B(Block)}$ is approximated as the sum of the circular front diameter D_C and the gas neck mean length D_L , with the introduction of coverage ratio γ for the partial blocking of the liquid channel by the gas phase (see Eqn. 6 and Fig. 6a). Based on the experimental images, γ was estimated as 0.8 (mean value).

Fig 9. Comparisons between experimental and theoretical (a) gas bubble volumes, (b) liquid slug volumes, (c) unit cell volumes and (d) gas bubble volume to unit cell volume ratios for microfluidic devices with channel junction angles of 20°, 45°, 90°, 135° and 160°, at various gas (He) flow rates with a constant liquid (ethanol) flow rate of 260 µl/min.

As for the theoretical model by Garstecki et al. (2006), which is applicable to the 90° junction angle microchannel only, full coverage of the liquid channel is assumed, i.e. $L_{B(Block)}$ is approximately equal to the liquid channel width W_L with $\gamma = 1$, without taking into account the presence of gas neck.

Fig. 8a and 9a show that the predictions by the current model have better agreements with the experimental results as compared to the theoretical model of Garstecki et al. (2006). The better agreement is achieved through the inclusion of the coverage ratio and the presence of gas neck in our theoretical model.

We have further compared our theoretical model with the experimental results of Abadie et al. (2012) for liquid-gas (i.e. ethanol-air) Taylor flow in microfluidic T-junction (i.e. 90° junction angle microchannel, $W_G = 525 \mu m$ and $W_L = 1000 \mu m$). This comparison with the experimental results of Abadie et al. (2012) is also conducted against the theoretical model of Garstecki et al. (2006) and the empirical correlation equation (curve fitting) of Fries and Rudolf von Rohr (2009). For the correlation equation by Fries and Rudolf von Rohr (2009), the bubble length is: $L_B/W_L = 1 + (aY^b\beta^c)(Q_G/Q_L)$, where Y is the W_L to W_G ratio, $\beta = 180^\circ - \theta$,

and the fitting parameters $a = 17.2131$, $b = -1$ and $c = -0.5$. The comparison shows that our theoretical model agrees better than the theoretical model of Garstecki et al. (2006) (see Fig. 10). Fig. 10 shows that our theoretical model has better agreement than the correlation equation of Fries and Rudolf von Rohr (2009) only when the gas to liquid flow rate ratio is low, and at higher ratio the correlation equation has better agreement.

In the investigation by Fries and Rudolf von Rohr (2009), they examined experimentally liquid-gas (i.e. ethanol-nitrogen) Taylor flow in microchannels of 45°, 90° and 135° junction angles and other geometrical parameters. For the purpose of investigating the effect of junction angle, we compared predictions of our theoretical model and the empirical correlation equation (curve fitting) by Fries and Rudolf von Rohr (2009), and their experimental results using their mid-range channel geometry reported, i.e. inlet ratio $Y = 2$ and $W_L = 200 \mu m$. These comparisons are representative for the whole set of results of Fries and Rudolf von Rohr (2009) as similar trend was observed for other geometrical configurations investigated by them.

Indeed, as the correlation equation of Fries and Rudolf von Rohr (2009) is obtained based on curve fitting on their

own results, it is reasonable to expect that their equation will be an excellent fit to their data. However, our theoretical model also predicts reasonably well their experimental data even though the trend is opposite from their correlation equation and data (see Fig. 11a). A major reason for this is that over this narrow range, there were relatively small variations versus junction angles.

Fig 10. Comparisons with experimental results of Abadie et al. (2012) for liquid-gas (i.e. ethanol-air) Taylor flow in T-junction (90°) microchannel against our theoretical model, theoretical model of Garstecki et al. (2006) and correlation equation of Fries and Rudolf von Rohr (2009).

Comparisons of predictions of the correlation equation by Fries and Rudolf von Rohr (2009) and our theoretical model, with our experimental results are also presented. As observed from Fig. 11b, the correlation equation of Fries and Rudolf von Rohr (2009) does not agree well with our experimental results. With the experimental trend clearly demonstrated by the inclusion of the 20° results, the trend they predicted is clearly opposite of our experimental trend. In contrast, our theoretical model can accurately predict our experiment results and trend (see Fig. 8, 9 and 11b), but also predict reasonably well the experimental data of Fries and Rudolf von Rohr (2009) (see Fig. 11a).

One of the main applications of microfluidics is for enhancing liquid-gas reactions, e.g. palladium (Pd)-mediated carbon-11 (^{11}C)-labelled reactions (Miller et al., 2011; Dahl et al., 2015). The high surface-to-volume ratio is the major advantage of employing microfluidic platform for flow chemistry. In these reactions, the liquid and gas flow rates are adjusted to tune the gas bubble and liquid slug sizes for achieving suitable surface-to-volume ratio to enhance the reactions.

The gas bubble and liquid slug sizes can be altered either by increasing the liquid flow rate at a constant gas flow rate or decreasing the gas flow rate at a constant liquid flow rate. Increasing the liquid flow rate produced smaller V_B but larger V_S (see Fig. 8a and 8b). Similarly, decreasing the gas flow rate led to smaller V_B at the expense of larger V_S (see Fig. 9a and 9b).

Fig 11. Comparisons of predictions of our theoretical model and correlation equation of Fries and Rudolf von Rohr (2009) with (a) their experimental results, and (b) our experimental results.

The gas bubble to unit cell volume ratio is defined as $\gamma_{B/U} = V_B/V_U$, with the unit cell volume $V_U = V_B + V_S$. Through manipulating the liquid flow rate at a constant gas flow rate or changing the gas flow rate at a constant liquid flow rate, $\gamma_{B/U}$ can be tuned precisely allowing the reagents to be controlled accurately for the generation of the chemical products.

It can be observed from Fig. 8d and 9d that $\gamma_{B/U}$ increased with decreasing liquid flow rate or increasing gas flow rate. For a given liquid and gas flow rates, as expected, to satisfy the law of mass conservation, the experimental $\gamma_{B/U(Exp)}$ for the different junction angle microchannels were almost constant, i.e. independent of the junction angle (see Fig. 8d and 9d).

A small V_U means that the gas bubbles and liquid slugs are densely packed (high bubble density), indicating a high surface-to-volume ratio that will aid in the enhancement of chemical reactions. The variation of V_U in response to the liquid or gas flow rate change was more pronounced for the 20° junction angle channel, with V_U decreased with decreasing liquid flow rate at a constant gas flow rate or increasing gas flow rate at a constant liquid flow rate (see Fig. 8c and 9c). This is because decreasing the liquid flow rate or increasing gas flow rate results in only slight increase in V_B (see Fig. 8a and 9a), with large decrease in V_S (see Fig. 8b and 9b).

In comparison, for the 45°, 90°, 135° and 160° channels with increasing liquid flow rate at a constant gas flow rate or decreasing gas flow rate at a constant liquid flow rate, V_U were less sensitive to fluid flow rate changes (see Fig. 8c and 9c) due to the approximate cancellation between V_B decrease (see Fig. 8a and 9a) and V_S increase (see Fig. 8b and 9b).

The microchannel junction angle θ provides another dimension for the manipulation of the gas bubble and liquid slug sizes in addition to the liquid and gas flow rates. For the 20° and 45° junction angle channels, it produced larger V_B and V_S when compared to the 90° channel (see Fig. 8a, 8b, 9a and 9b). This is caused by the larger junction width W_J when $\theta < 90^\circ$, leading to a larger gas bubble throat width D_N at the junction for the 20° and 45° junction angle channels (see Eqn. 4 and Fig. 6a).

Even though the 160° junction angle channel has the same W_J as the 20° channel (see Eqn. 4), D_N does not span across the entire junction width. The change of flow direction for $\theta > 90^\circ$ produces D_N similar to the gas channel width W_G , which is similar to that of the 90° channel (see Fig. 6b). Therefore, V_B and V_S for the 135° and 160° junction angle channels were similar to the 90° channel (see Fig. 8a, 8b, 9a and 9b). This has never been observed previously until the employment of extreme junction angle channels (i.e. 20° and 160° channels) in this investigation. The mechanics is first investigated here with the associated theoretical model developed in Sec. 3.1 to describe this observation.

Similarly, V_U for the 20° and 45° junction angle channels ($\theta < 90^\circ$) was larger than the 90° channel (see Fig. 8c and 9c), which is caused by the increase in both V_B and V_S (see Fig. 8a, 8b, 9a and 9b). As a result of the approximately same V_B and V_S (see Fig. 8a, 8b, 9a and 9b), V_U for the 135° 160° junction angle channels ($\theta > 90^\circ$) was approximately the same as the 90° channel (see Fig. 8c and 9c).

For the enhancement of chemical reaction, a high surface-to-volume ratio is desirable; this means that a high density of gas bubble and liquid slug train is required (i.e. small unit cell volume). To realise this, the microchannel junction angle should be larger or equal to 90°. Although the gas bubble to unit cell volume ratios are almost constant for various channel junction angles, junction angle

larger or equal to 90° is a better choice due to the smaller unit cell volume.

4. Conclusions

In this investigation, two-phase liquid-gas Taylor flow was experimentally examined with different junction angle microchannels, i.e. 20°, 45°, 90°, 135° and 160°, at various liquid (ethanol) and gas (He) flow rates. The effects of flow rates and junction angle on the gas bubble and liquid slug volumes were investigated. Since the experimental conditions were in the blocking-squeezing regime ($Ca < 10^{-2}$), we formulated a theoretical model by extending the scaling relation developed by Garstecki et al. (2006), for the quantitative predictions of gas bubble and liquid slug sizes for channels with different junction angles, gas and liquid flow rates.

The theoretical predictions agree well with the experimental results. As expected, for a given ratio of gas and liquid flow rates, to satisfy the law of mass conservation, the gas bubble to unit cell volume ratios were almost constant regardless of the junction angle. The unit cell volume decreased pronouncedly for the 20° channel with decreasing liquid or increasing gas flow rate, due to the slight increase in gas bubble volume and large decrease in liquid slug volume. In comparison, for the 45°, 90°, 135° and 160° channels with increasing liquid or decreasing gas flow rate, the unit cell volumes were less sensitive to fluid flow rate changes, due to the approximate cancellation between the decrease in gas bubble volume and increase in liquid slug volume.

For the 20° and 45° junction angle channels, it produced larger unit cell volume, due to larger gas bubble and liquid slug volumes, when compared to the 90° channel. This is caused by the large junction width when the junction angle is less than 90°, leading to a larger gas bubble throat width at the junction. Even though the 160° junction angle channel has the same junction width as the 20° channel, the gas bubble throat width does not span across the entire junction width. The change of flow direction for junction angle more than 90° produces gas bubble throat width similar to the gas channel width, which is similar to that of the 90° channel. Therefore, for the 135° and 160° channels, the gas bubble volume, liquid slug volume and unit cell volume were similar to that of the 90° channel.

As evident from the smaller unit cell volumes, with larger junction angle microchannels (i.e. 90°, 135° and 160° channels), higher gas bubble density can be attained when compared to the smaller junction angle channel (i.e. 20° and 45° channels). The high surface-to-volume ratio will aid in the enhancement of chemical reactions. Future investigations will be conducted by applying the conceptual framework developed from this investigation for flow chemistry reactions.

Nonmenclature

A_L	Cross sectional area of liquid channel (m ²)
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Ca	Capillary number ($Ca = \mu_L U_{TP} / \sigma_L$), dimensionless
D_C	Experimental diameter of bubble circular front at end of blocking phase
D_H	Hydraulic diameter of liquid channel ($D_H = 2HW_L / (H + W_L)$) (m)
D_L	Mean gas neck length (m)
D_N	Gas bubble throat width (m)
$D_{N(Min)}$	Minimum thread of bubble throat width (m)
H	Height of channel (m)
$L_{B(Exp)}$	Experimental gas bubble length (m)
$L_{B(Theo)}$	Theoretical gas bubble length (m)
$L_{B(Block)}$	Gas bubble length for blocking process (m)
$L_{B(Squeeze)}$	Gas bubble length for squeezing process (m)
$L_{S(Exp)}$	Experimental liquid slug length (m)
Q_G	Gas flow rate ($\mu\text{L}/\text{min}$)
Q_L	Liquid flow rate ($\mu\text{L}/\text{min}$)
Ra	Roughness arithmetic mean (m)
Re	Reynolds number ($Re = \rho_L U_{TP} D_H / \mu_L$), dimensionless
u_L	Average speed of the liquid phase (m/s)
u_G	Average speed of the gas phase (m/s)
U_{TP}	Superficial two-phase velocity ($u_L + u_G$) (m/s)
v_{Growth}	Bubble growth velocity during squeezing process (m/s)
$v_{Squeeze}$	Squeezing velocity (m/s)
$V_{B(Exp)}$	Experimental gas bubble volume (m^3)
$V_{B(Theo)}$	Theoretical gas bubble volume (m^3)
$V_{S(Exp)}$	Experimental liquid slug volume (m^3)
$V_{S(Theo)}$	Theoretical liquid slug volume (m^3)
V_U	Unit cell volume (m^3)
W_G	Gas channel width (m)
W_J	Junction angle width (m)
W_L	Liquid channel width (m)
We	Weber number ($We = Ca \times We$), dimensionless

Greek letter

γ	Coverage ratio at end of blocking phase/instant before D_N decreases, dimensionless
$\gamma_{B/U}$	Gas bubble to unit cell volume ratio, dimensionless
Υ	Liquid channel to gas channel width ratio (W_L/W_G), dimensionless
θ	Microchannel junction angle ($^\circ$)
μ_L	Dynamic viscosity of liquid (Pa.s)
ρ_L	Density of liquid phase (kg/m^3)
σ_i	Polydispersity index (%)
σ_L	Surface tension of the liquid (N/m)

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