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Pressure drop of gas-liquid Taylor flow in square microchannels

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Abstract

The pressure difference in gas-liquid Taylor flow in square microchannels with three different hydraulic diameters (190, 298, and 505 μm) were measured. Three liquids (Water and two types of glycerol-water solutions) were used as the liquid phase, and N_2 gas was used as the gas phase. The experiments were carried out for the ranges of $1.98 \times 10^{-3} < Ca_T < 0.132$ and $2.61 < Re_T < 661$, where Ca_T and Re_T are the capillary and Reynolds numbers based on the total volumetric flux, respectively. The measured pressure drop of Taylor bubbles based on the unit cell model was compared with the existing models, and large discrepancies could be recognized. Dimensionless pressure drop of a Taylor bubble increased with the capillary number, and depends on the liquid viscosity and the hydraulic diameter at the same capillary number in the middle and high capillary number region, i.e., the inertia effect. Hence, a new pressure drop model including these effects as dimensionless numbers was developed based on the relationship between the pressure drop and liquid film thickness of a Taylor bubble, and it could show good agreement with the measured data. The pressure drop of Taylor flow could also be evaluated using the unit cell model with the developed model. In addition, a pressure drop model for Taylor flow based on only the total volumetric flux was developed in terms of convenience of prediction.

Keywords: Taylor flow, Taylor bubble, square microchannel, pressure drop, unit cell model

1. Introduction

Taylor flow also called as slug flow or elongated bubble flow is one of the typical flow patterns in gas-liquid two-phase flow in microchannels (Triplett et al., 1999; Kawahara et al., 2002; Serizawa et al., 2002; Chung and Kawaji, 2004; Yue et al., 2008; Choi et al., 2011a). Knowledge of Taylor flow in microchannels is essential for the design and development of microdevices such as microreactors, heat exchangers and fuel cells. In particular, accurate prediction of the pressure drop of Taylor flow in microchannels is of great interest since the frictional pressure drop remarkably increases with decreasing channel hydraulic diameter.

Various cross sectional shapes of microchannels such as circular, rectangular, triangle, and so on, are adopted in microdevices, in particular, circular and rectangular shapes are major. Many studies on the pressure drop of gas-liquid Taylor flow in circular microchannels, hence, have been carried out, and several pressure drop models have been developed (Kreutzer et al., 2005; Kawahara et al., 2009; Warnier et al., 2010; Minagawa et al., 2013; Langewisch and Buongiorno, 2015; Kurimoto et al. 2017). With regard to rectangular microchannels, ease of channel fabrication by molding and lamination of channels are advantages in practical applications. It is also necessary to obtain knowledge and develop an accurate model for the pressure drop of Taylor flow in rectangular microchannels.

The unit cell model is of great use for the prediction of the pressure drop of Taylor flow (Kreutzer et al., 2005; Warnier et al., 2010). The unit cell model is composed of a single Taylor bubble and a single liquid slug (Fig. 1). Since characteristics of uniform Taylor flow are governed by dynamics of the single unit, it can be assumed that the whole pressure drop of Taylor flow in a microchannel is equal to the sum of the pressure drop of single unit cells. The unit cell model is basically represented as follows:

$$\Delta P_{UC} = \Delta P_L + \Delta P_G \quad (1)$$

where ΔP_{UC} is the pressure drop in a unit cell, ΔP_L the frictional pressure drop in a liquid slug and ΔP_G the pressure drop due to the presence of a Taylor bubble, respectively. The pressure gradient dp/dz is represented by

$$\frac{dp}{dz} = \frac{\Delta P_{UC}}{L} \quad (2)$$

where L is the length of the unit cell (Fig. 1). ΔP_L can be represented by

$$\Delta P_L = \lambda_S \frac{L_L}{D_h} \frac{\rho_L j_T^2}{2} = \frac{c_\lambda}{Re_T} \frac{L_L}{D_h} \frac{\rho_L j_T^2}{2} \quad (3)$$

where λ_S is the friction factor for single phase flow, L_L the liquid slug length in a unit cell (Fig. 1), D_h the hydraulic diameter of a channel, ρ_L the liquid density, j_T the total volumetric flux, c_λ the friction coefficient, Re_T the Reynolds number based on j_T , respectively. It is well-known that the values of c_λ for laminar flows is 64 in a circular channel and 56.9 in a square channel (Shah and London, 1978). The total volumetric flux j_T is the sum of gas

volumetric flux j_G and liquid volumetric flux j_L ($j_T = j_G + j_L$). Re_T is defined by

$$Re_T = \frac{\rho_L j_T D_h}{\mu_L} \quad (4)$$

where μ_L is the liquid viscosity. By substituting Eqs. (1) and (3) into Eq. (2) and rearranging the equation, the following equation can be obtained:

$$\frac{dp}{dz} = \lambda_S \frac{L_L}{LD_h} \frac{\rho_L j_T^2}{2} + \frac{\Delta P_G}{L} = \lambda_{TP} \frac{L_L}{LD_h} \frac{\rho_L j_T^2}{2} \quad (5)$$

where λ_{TP} is the friction factor for two-phase flow. A model of ΔP_G or λ_{TP} , therefore, is required to predict the pressure drop of Taylor flow.

Bretherton (1961) analytically derived the following pressure drop model for a Taylor bubble in a circular capillary:

$$\Delta P_G = 9.04 \frac{\sigma}{D} (3Ca)^{2/3} \quad (6)$$

where σ is the surface tension, D the diameter of a circular channel, and Ca the capillary number based on the velocity of a Taylor bubble u_G , respectively. Ca is defined by

$$Ca = \frac{\mu_L u_G}{\sigma} \quad (7)$$

Equation (6) can be applied in the limit of $Ca \rightarrow 0$. Kreutzer et al. (2005) proposed the following friction factor model for Taylor flow in circular microchannels:

$$\lambda_{TP} = \frac{64}{Re_T} \left[1 + c_K \left(\frac{L_L}{D} \right)^{-1} \left(\frac{Re_T}{Ca_T} \right)^{1/3} \right] = \frac{64}{Re_T} \left[1 + c_K \left(\frac{L_L}{D} \right)^{-1} \left(\frac{\rho_L D \sigma}{\mu_L^2} \right)^{1/3} \right] \quad (8)$$

where c_K is the constant and Ca_T the capillary number based on j_T , respectively. Ca_T is defined by

$$Ca_T = \frac{\mu_L j_T}{\sigma} \quad (9)$$

They obtained c_K values of 0.17 for their experimental results and 0.07 for their numerical results, and concluded that c_K value difference was caused by the Marangoni effect due to the presence of impurities in the liquid phase. Warnier et al. (2010) extended the Bretherton's pressure drop model by replacing $Ca^{2/3}$ with $Ca^{2/3}/(1+3.34 Ca^{2/3})$ to predict the pressure drop for a wide range of the capillary number:

$$\Delta P_G = 7.16 \frac{\sigma}{D} \frac{(3Ca)^{2/3}}{(1+3.34Ca^{2/3})} \quad (10)$$

The constant 7.16 is different from the constant 9.04 in Eq. (6). Langewisch and Buongiorno (2015) presumed that the difference was caused by confusion of the abstract in Bretherton (1961), i.e., Bretherton showed the pressure drop of the front shape of a bubble as the total pressure drop of a bubble in the abstract. The pressure drop due to the presence of a Taylor bubble is caused by bubble front and rear shapes, especially the front shape. Since the bubble front shape is correlated with liquid film thickness, the contribution of the bubble shape to the pressure drop can be expressed using the same functional form as the liquid film thickness (Bretherton, 1961; Kurimoto et al., 2018). Warnier et al., therefore, introduced $Ca^{2/3}/(1+3.34 Ca^{2/3})$ based on the liquid film thickness model proposed by Aussillous and Quéré (2000).

Wong et al. (1995) analytically derived the pressure drop of a long gas bubble in a square capillary in the limit of $Ca \rightarrow 0$ under the condition of neglecting the inertia force, which is represented by

$$\Delta P_G = c_W \frac{\sigma}{D_h} Ca^{2/3} \quad (11)$$

where c_W is the constant. c_W depends on $2L_G/D_h$ which is the ratio of bubble length L_G to D_h . Hazel and Heil (2002) carried out the finite element simulation of an air finger in a square channel under the condition of neglecting the inertia force. Their numerical results showed that the pressure drop at a bubble tip increased with Ca and its tendency was similar to Wong et al.'s analytical solution. Good agreement between them, however, was shown only in the range of $Ca \leq 10^{-3}$. Fuerstman et al. (2007) proposed the following pressure drop model for Taylor bubbles in rectangular microchannels:

$$\Delta P_G = \frac{c_{F1}\mu_L j_L L_G}{H^2} + \frac{c_{F2}\sigma Ca_L^{2/3}}{H} \quad (12)$$

where c_{Fi} ($i = 1$ and 2) are the constants, H the channel height, and Ca_L the capillary number based on j_L , respectively. Ca_L is defined by

$$Ca_L = \frac{\mu_L j_L}{\sigma} \quad (13)$$

In their experiment, they used four types of liquid (Water and Glycerol water solutions) and ten microchannels with various aspect ratios of the height to the width, and the order of the capillary number was less than 10^{-3} . The first and second terms in Eq. (12) represent the pressure drop in the liquid phase between a Taylor bubble and channel corners, i.e. liquid film at channel corners, and the pressure drop due to the front and rear shapes of a Taylor bubble, respectively. The contribution of the first term to the total pressure drop is very small in a clean system, i.e. without surface active agents in the liquid phase. c_{F1} and c_{F2} values were determined as 1.3 and 12 based on their experimental data, respectively. Yue et al. (2009) measured pressure difference in air-water Taylor flow in square microchannels with the hydraulic diameters of 200 and 400 μm . They proposed the following friction factor model by extending the friction factor model for Taylor flow in circular microchannels proposed by Kreutzer et al. (2005):

$$\lambda_{TP} = \frac{56.9}{Re_T} \left[1 + c_K \left(\frac{L_L}{D_h} \right)^{-1} \left(\frac{\rho_L D_h \sigma}{\mu_L^2} \right)^{1/3} + 5.5 \times 10^{-5} \left(\frac{L_L}{D_h Re_T} \right)^{-1.6} \right] \quad (14)$$

ΔP_G of Yue et al.'s model can be represented using Eqs. (1-3), (5) and (14):

$$\Delta P_G = \Delta P_L \left[c_K \left(\frac{L_L}{D_h} \right)^{-1} \left(\frac{\rho_L D_h \sigma}{\mu_L^2} \right)^{1/3} + 5.5 \times 10^{-5} \left(\frac{L_L}{D_h Re_T} \right)^{-1.6} \right] \quad (15)$$

For c_K , they adopted a value of 0.07 based on the study by Kreutzer et al. The second term in the square bracket of Eq. (14) is the same expression as that in Eq. (8), and the third term in the square bracket of Eq. (14) represents the influence of recirculation in very short liquid slugs. Yue et al.'s pressure drop model could evaluate their experimental data within $\pm 20\%$ errors. Choi et al. (2010) measured the pressure difference in N_2 -water Taylor flow in four rectangular microchannels with three hydraulic diameters of 143, 322 and 490 μm and four

different aspect ratios. They proposed two types of correlations for the pressure drop of Taylor bubbles, one of which is for the experimental result in each rectangular microchannel with the different hydraulic diameter and the aspect ratio, and the other is for all experimental results. The proposed correlations are the following, respectively:

$$\Delta P_G = c_{C1} Ca^{c_{C2}} \quad (16)$$

$$\Delta P_G = \rho_L j_T^2 \left(0.07 + \frac{106.8}{Re_T} \right) \quad (17)$$

where c_{Ci} ($i = 1$ and 2) are the constants, which depend on the hydraulic diameter and the aspect ratio. The values for a square microchannel are $c_{C1} = 3.17 \times 10^4$ and $c_{C2} = 1.15$. Note that Choi et al. adopted effective bubble length which is represented by $L_G - D_h$ instead of L_G . Choi et al. (2011b) measured pressure difference in N_2 -water Taylor flow in a rectangular microchannel with hydraulic diameter of $490 \mu m$ and proposed the following model for the pressure drop of Taylor bubbles:

$$\Delta P_G = 6.845 \times 10^4 Ca^{1.36} \quad (18)$$

As summarized above, the pressure drop data of Taylor flow in rectangular microchannels have been obtained and some prediction models for Taylor bubbles have been developed. Only water, however, was used as the liquid phase in the experimental studies except for the study by Fuerstman et al., i.e. effects of liquid property on the pressure drop have not been investigated adequately. In fact, Han & Shikazono (2009) experimentally showed that the liquid film thickness of Taylor bubbles in square microchannels depends on liquid property, and the pressure drop of a Taylor bubble is correlated with the liquid film thickness (Bretherton, 1961; Wong et al. 1995). With regard to the channel aspect ratio of the height to the width, the pressure drop of a Taylor bubble increases with decreasing the channel aspect ratio at the same Ca (Choi et al., 2010), i.e., Taylor flow data in a square microchannel can be regarded as the data in the limiting case of rectangular microchannels. Hence, the knowledge of Taylor flow in a square microchannel is essential for understanding effects of the channel aspect ratio on the pressure drop of Taylor flow. With regard to the existing pressure drop models in rectangular microchannels, most of the models have a simple function form of Ca with an exponent, Eqs. (11), (12), (16) and (18). As Warnier et al., (2009) extended the Bretherton's pressure drop model by adopting a new functional form, discussion about the functional

form of the pressure drop model in square microchannels would be required to deal with the pressure drop in wide ranges of dimensionless numbers.

For obtaining knowledge and discussing pressure drop models, the pressure difference in gas-liquid Taylor flow in square microchannels, hence, were measured using a pressure gauge in this study. Three square microchannels with channel hydraulic diameter $D_h = 190$, 298, and 505 μm and three liquids (water and two glycerol-water solutions) were used to obtain the experimental data for wide ranges of dimensionless numbers. The measured data were compared with the above-mentioned pressure drop models to investigate the applicability of the models. After the comparisons, a new pressure drop model was discussed for more reliable prediction.

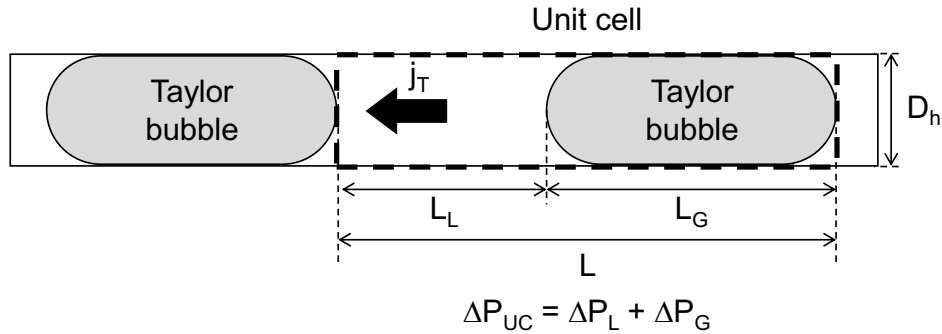


Fig. 1 Taylor flow in a microchannel

2. Experimental method

The experimental setup is shown in Fig. 2. The experimental setup is similar to our previous study (Kurimoto et al., 2017), and the most significant difference is naturally microchannel shape, i.e. square microchannel or circular microchannel. Microchannel plates with the square microchannels of 190, 298 and 505 μm in hydraulic diameter were used. The liquid temperature was kept at 298 ± 2 K. Deionized water (FUJIFILM Wako Pure Chemical Corporation) or a glycerol-water solution (52 or 64 wt%) was used as the liquid phase, and nitrogen gas was used as the gas phase. The kinematic viscosities of the 52 and 64 wt% glycerol-water solutions were about five and ten times that of water, respectively. Deionized water and glycerol (FUJIFILM Wako Pure Chemical Corporation) were used for the glycerol-water solutions. The density and surface tension were measured at least five times using a densimeter and capillary tubes (glass tube, 700 μm i.d.), respectively. Uncertainties estimated at 95% confidence level in the measured density and surface tension were 0.16% and 2.5%, respectively. Since the measured values of density agreed well with literature data (Ishikawa, 1968), the values of viscosity were obtained in literature (Ishikawa, 1968). Fluid systems examined are summarized in Table 1. The

difference in the types of the liquids mainly affects the liquid viscosity.

A syringe pump (YMC CO. LTD., YSP-202) supplied the liquid and controlled its flow rate. A gas cylinder supplied nitrogen gas, and a mass flow controller (FUJIKIN Inc., FCST 1005(M)L) controlled its flow rate. Gas-liquid Taylor flow was generated at a Tee junction in the microchannel plate and flowed in the microchannel. Pressure after the Tee junction was measured using a pressure gauge. Either one of two pressure gauges with different range was used to obtain experimental data for a wide range of the pressure (NAGANO KEIKI CO. LTD., GC50 or GC 55). A high-speed video camera (PHOTRON Ltd., FASTCAM Mini UX50, frame rate = 500-8000 frame/s, spatial resolution $\simeq 0.990\text{-}2.81 \mu\text{m/pixel}$) with back illumination by a LED light source was used to obtain Taylor flow images for calculation of bubble velocity, bubble length and liquid slug length. The images were taken after Taylor bubbles reached a constant length. For the estimation of uncertainties, the gas and liquid flow rates were measured in advance of the experiment. Uncertainties estimated at 95% confidence level in the measured gas and liquid flow rates were 2.4% and 6.7 %, respectively.

A schematic diagram of the microchannel plate is shown in Fig. 3. The microchannel plates were made of transparent acrylic resin and consisted of two flat plates, one of which had microchannels cut by a machining center and endmills. Bolts around the microchannel joined the flat plates together. W_{tap} , H_{tap} , z_m , and z_t are the width and the height of pressure tap, the distance between the mixing section and the outlet, and the distance between the pressure tap and the outlet, respectively. Table 2 gives detailed values of these factors. The visualization section by the high speed camera is basically the same position as the pressure tap.

Figure 4 shows the cross sections of the microchannels at the outlet. The microchannels have nearly square shapes, although some burrs are visible at the edge of the outlet. Table 3 shows the hydraulic diameter, width and height of the microchannels. The widths of the microchannels were measured from images with stainless steel spheres whose diameters were 0.15 and 0.3 mm. These images were perpendicular to the horizontal microchannels as well as the Taylor flow images. The heights of the microchannels were calculated from the cross sectional images. The measurement errors of them were estimated less than $\pm 3 \%$. The ratios of the width to the height of the microchannels were at most 1.09. The microchannels, hence, could be regarded as being square.

The gas volumetric flux at the pressure measurement point j_G was calculated using the Boyle's law. The Boyle's law between the pressure measurement point and the outlet of the microchannels could be represented as $p_a j_{Ga} = p j_G$, where p_a is the atmospheric pressure, j_{Ga} the gas volumetric flux at the atmosphere, and p the measured pressure using the

pressure gauge, respectively. Since j_{Ga} could be calculated from the gas flow rate controlled by the mass flow controller, j_G was obtained.

Taylor bubble velocity u_G was calculated from the front position of a Taylor bubble and time interval in images taken by the high speed video camera. The Taylor bubble length L_G and the liquid slug length L_L were calculated from the front and rear positions of Taylor bubbles displayed in the images. The front and rear positions were identified by image processing. The variability of the measured u_G , L_G and L_L were at most 2.1, 5.9 and 12 %, respectively.

Taylor flow implies that the velocity of a Taylor bubble does not depend on the sphere-volume equivalent diameter of a bubble, i.e. the characteristic length is the channel hydraulic diameter and Taylor bubble velocity does not depend on bubble length (Hayashi et al., 2011). When the sphere-volume equivalent diameter is smaller than the critical diameter, the bubble velocity depends on the sphere-volume equivalent diameter (Nakahara and Tomiyama, 2003; Kurimoto et al., 2013; Khodaparast et al., 2015). However, it is difficult to identify the critical diameter since it is affected by fluid properties and the hydraulic diameter. Bubbles whose length were 1.2 times longer than the channel hydraulic diameter, hence, were regarded as Taylor bubbles in the present study.

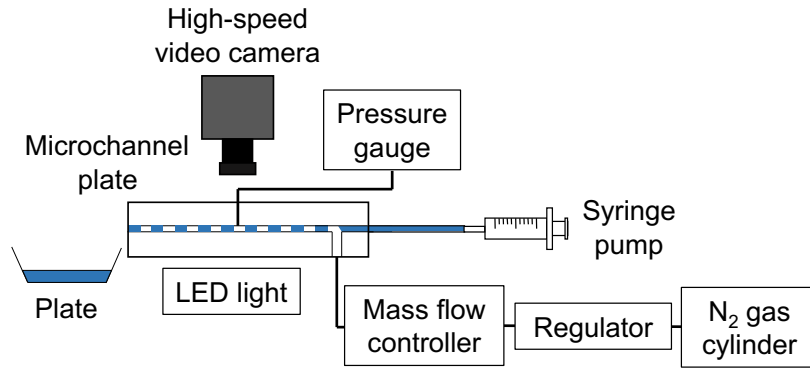


Fig. 2 Experimental setup

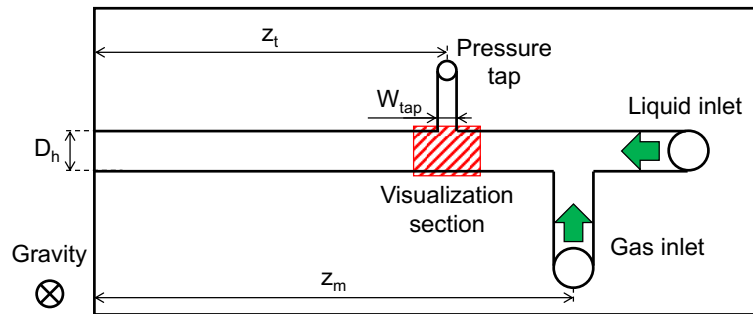
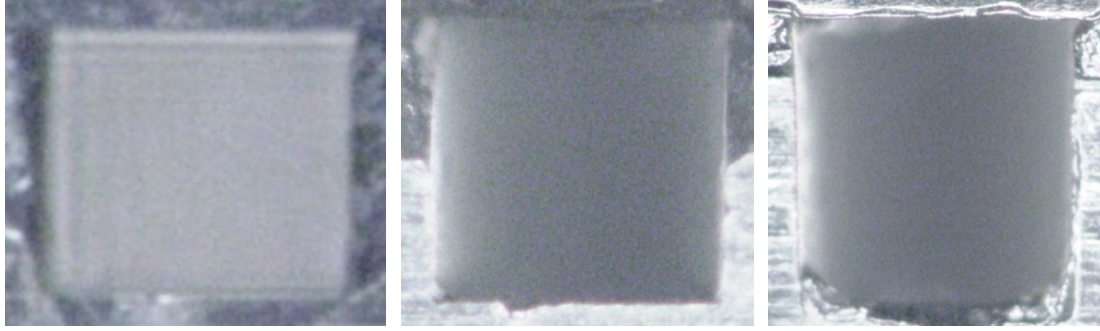


Fig. 3 Schematic diagram of microchannel plate



(a) $D_h = 190 \mu\text{m}$

(b) $D_h = 298 \mu\text{m}$

(c) $D_h = 505 \mu\text{m}$

Fig. 4 Cross section of microchannels at outlet

Table 1 Experimental conditions

Liquid	Deionized water	Glycerol-water solution (52 wt%)	Glycerol-water solution (64 wt%)
Gas	N ₂	N ₂	N ₂
ρ_L [kg/m ³]	997	1129	1164
ρ_G [kg/m ³]	1.19	1.19	1.19
μ_L [mPa·s]	0.890	5.62	11.6
μ_G [mPa·s]	0.018	0.018	0.018
ν [mm ² /s]	0.893	4.98	9.97
σ [mN/m]	71.9	68.7	67.3
D_h [μm]	190, 298, 505	190, 298, 505	190, 298
J_L [m/s]	8.78×10^{-2} -0.924	6.49×10^{-2} -0.828	1.69×10^{-2} -0.268
J_G [m/s]	6.98×10^{-2} -1.06	5.70×10^{-2} -0.889	3.33×10^{-2} -0.418
Ca_T	1.98×10^{-3} - 2.45×10^{-2}	1.02×10^{-2} -0.132	8.84×10^{-3} -0.109
Re_T	72.9-661	7.45-97.1	2.61-32.3

Table 2 Size of microchannel plate

D_h [μm]	W_{tap} [μm]	H_{tap} [μm]	z_m [mm]	z_t [mm]
190	100	200	50	35
298	100	300	50	35
505	100	500	155	135

Table 3 Hydraulic diameter, width and height of microchannels

D_h [μm]	Width [μm]	Height [mm]
190	199	182
298	301	294
505	508	503

3. Results and discussions

3.1 Pressure difference in liquid single phase flow

Pressure difference in liquid single phase flow was measured before the experiment of gas-liquid Taylor flow to validate our experimental setup and method. Figure 5 shows a comparison of the production of the Reynolds number Re_S and the friction factor λ_S between the measured data and the theoretical value ($\lambda_S Re_S = 56.9$) (Shah and London, 1978) in single phase flow. Re_S is defined by

$$Re_S = \frac{\rho_L u_S D_h}{\mu_L} \quad (19)$$

where u_S is the mean velocity in liquid single phase flow. The measured data do not depend on Re_S , and are equally scattered both above and below the theoretical value regardless of the channel diameter and the liquid properties. The experimental setup and method in the present study, therefore, can measure the pressure difference in liquid single phase flow within $\pm 10\%$ errors for the theoretical value.

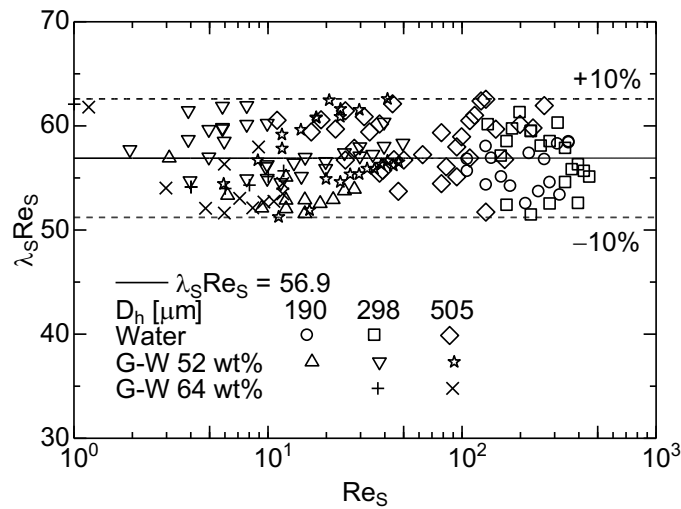


Fig. 5 Comparison of $\lambda_S Re_S$ between measured data and theoretical value in liquid single phase flow

3.2 Pressure difference in gas-liquid Taylor flow

For recognizing necessity of the pressure drop model for Taylor flow, the measured dp/dz of gas-liquid Taylor flow are compared with the following theoretical pressure drop equation for single phase flow based on j_T , i.e. Darcy-Weisbach equation:

$$\left. \frac{dp}{dz} \right|_{S,j_T} = \frac{56.9}{Re_T D_h} \frac{\rho_L j_T^2}{2} \quad (20)$$

Figure 6 shows the comparison between the measured dp/dz of Taylor flow and Eq. (20). Equation (20) naturally cannot evaluate dp/dz of Taylor flow and several measured data are out of the range of $\pm 50\%$ errors. The pressure drop model for Taylor flow in square microchannels, hence, is required for accurate prediction.

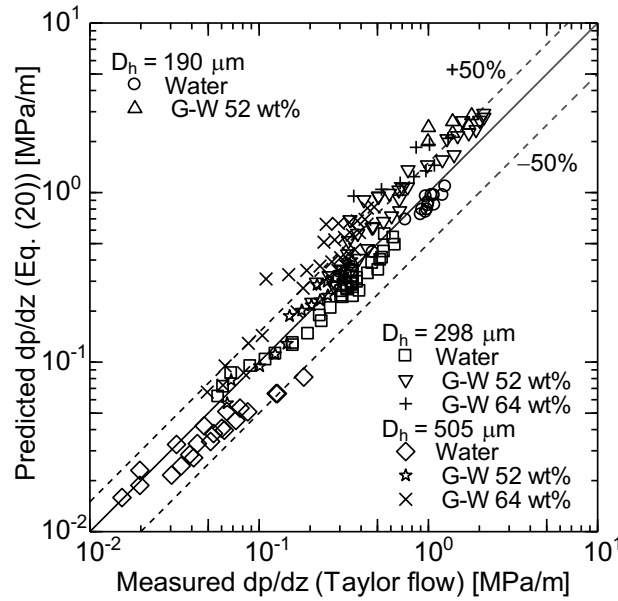


Fig. 6 Comparison between measured dp/dz of Taylor flow and predicted dp/dz based on single phase flow (Eq. (20))

For discussion of ΔP_G from our experimental data, ΔP_G was calculated using the following equation based on the unit cell model:

$$\Delta P_G = \frac{dp}{dz} L - \frac{56.9}{Re_T} \frac{L}{D_h} \frac{\rho_L j_T^2}{2} \quad (21)$$

Figure 7 shows comparisons of ΔP_G between the measured data and the prediction models in rectangular microchannels. The predicted ΔP_G in Fig. 7(a)-(f) are calculated using Eq. (11) (Wong et al., 1995), Eq. (12) (Fuerstman et al., 2007), Eq. (15) (Yue et al., 2009), Eqs. (16-17) (Choi et al., 2010), and Eq. (18) (Choi et al., 2011b), respectively. The value of c_W in Eq. (11) is regarded as 7.106 based on $2L_G/D_h$ of all experimental data in the present study. Equation (11) underestimates ΔP_G for all experimental data (Fig. 7(a)). Equation (12) can evaluate some ΔP_G (Fig. 7(b)). This is because the second term of Eq. (12) is similar functional form to Eq. (11) and c_{F2} is larger than c_W . Equations (15-18) can evaluate some ΔP_G in N₂-water cases, while large differences between the measured data and the models can be observed in N₂-Glycerol-water solution cases. This is because their models are based on their experimental data in N₂-water cases, i.e. their models do not include effects of the liquid viscosity on ΔP_G appropriately. Taylor bubble shapes are deformed by the difference in the liquid viscosity as shown in Fig. A1, which causes the difference in ΔP_G . It can be concluded that the existing ΔP_G models are not appropriate for the estimation of ΔP_G in the present experimental range.

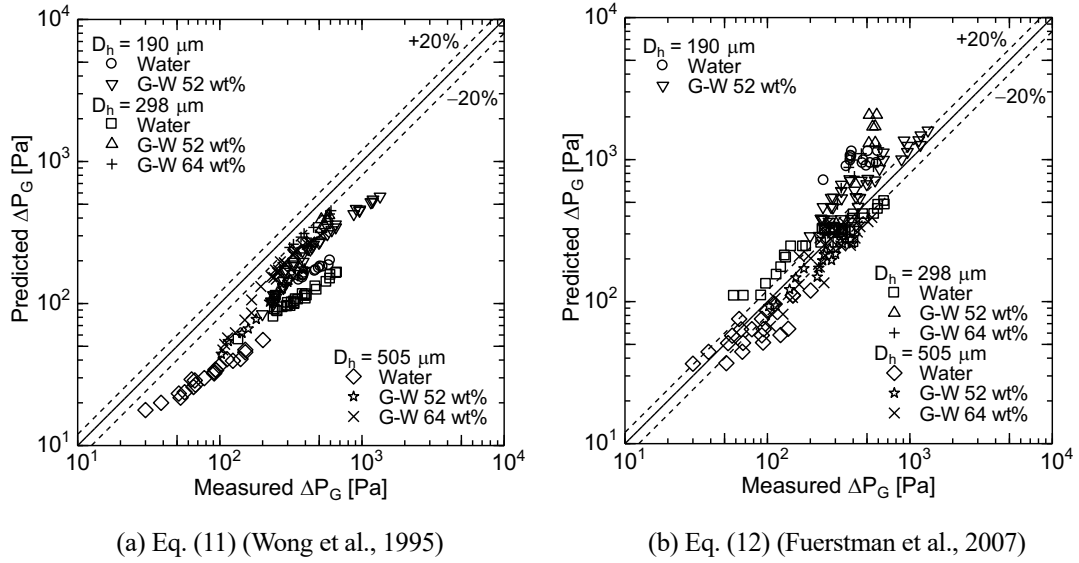
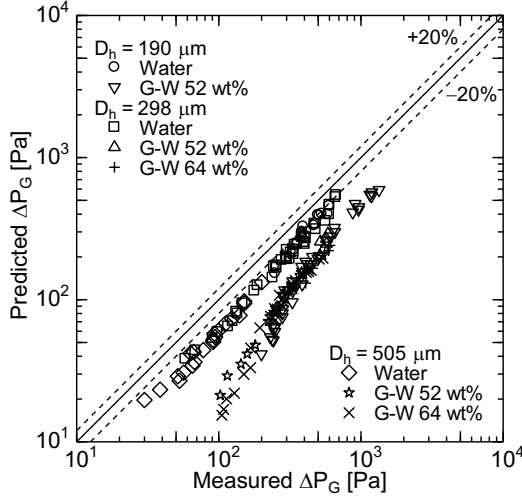
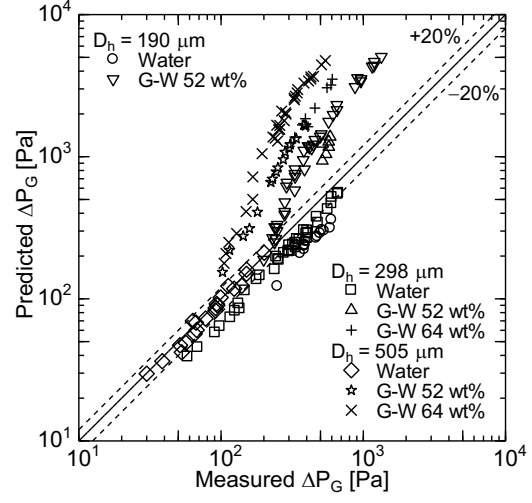


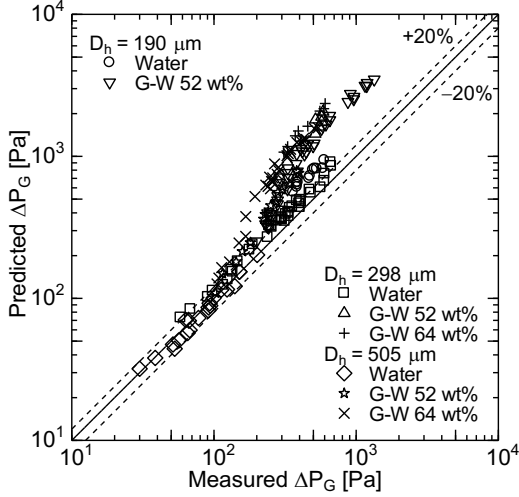
Fig. 7 Comparisons of ΔP_G between models and measured data



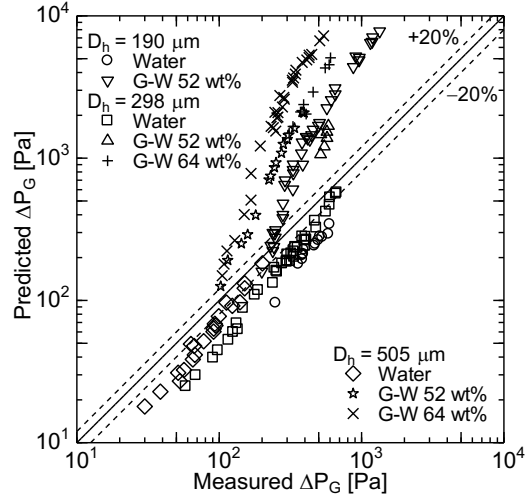
(c) Eq. (15) (Yue et al., 2009)



(d) Eq. (16) (Choi et al., 2010)



(e) Eq. (17) (Choi et al., 2010)



(f) Eq. (18) (Choi et al., 2011b)

Fig. 7 Continued

For understanding the discrepancies between the measured data and the existing models in more detail, the relationship between $\Delta P_G D_h / \sigma$ and Ca is shown in Fig. 8. As summarized in Section 1, most of the pressure drop models for a Taylor bubble is represented by the following form regardless of the cross sectional shape of channels, since the order of the pressure jump at the interface is estimated by the surface tension and the characteristic length (Bretherton, 1961; Wong et al., 1995):

$$\Delta P_G = \frac{\sigma}{D_h} f \quad (22)$$

where f is the function consisting of dimensionless numbers. $\Delta P_G D_h / \sigma$ represents dimensionless pressure drop of a Taylor bubble. The solid and the broken lines in the figure mean Eq. (11) (Wong et al., 1995) and Eq. (16) (Choi et al., 2010), respectively. In addition to our experimental data, experimental data of N₂-water Taylor flow in a square microchannel with the hydraulic diameter of 490 μm measured by Choi et al. (2010) are also plotted in the figure. $\Delta P_G D_h / \sigma$ of the measured data mainly increases with Ca . When the liquid phase is water, $\Delta P_G D_h / \sigma$ gently increases with Ca in the range of $Ca < 0.01$ and steeply increases with Ca in the range of $Ca > 0.01$. This tendency agrees well with the experimental data and the correlation by Choi et al. $\Delta P_G D_h / \sigma$ of N₂-water cases slightly decreases with decreasing D_h at the same Ca . When the liquid phase is glycerol water solutions of 52 and 64 wt%, the experimental data exist in high Ca region, i.e. $\Delta P_G D_h / \sigma$ decreases with increasing the liquid viscosity at the same Ca , and the tendencies of $\Delta P_G D_h / \sigma$ on Ca are similar to the data in water cases. Fuerstman et al. (2007) reported that c_{F2} in Eq. (12) was a constant in low capillary number region, i.e. the liquid viscosity and D_h do not affect $\Delta P_G D_h / \sigma$ and f can be represented by the function of Ca with an exponent. This previous experimental fact agrees well with our experimental data for $Ca < 5 \times 10^{-3}$. However, f cannot be represented by the function of Ca with an exponent for $Ca \geq 5 \times 10^{-3}$, since the $\Delta P_G D_h / \sigma$ depends on the liquid viscosity and D_h at the same Ca . This difference in $\Delta P_G D_h / \sigma$ between the liquids in the range of $Ca \geq 5 \times 10^{-3}$, hence, is the main cause of the discrepancies between the measured data and the models. The dependences of $\Delta P_G D_h / \sigma$ on the liquid viscosity and D_h at the same Ca represents the inertia effect. $\Delta P_G D_h / \sigma$ basically decreases with decreasing the Weber number We at $Ca \simeq 2.00 \times 10^{-2}$ as shown in Table 4. The definition of We is

$$We = \frac{\rho_L u_G^2 D_h}{\sigma} \quad (23)$$

With regard to Eq. (11), all measured $\Delta P_G D_h / \sigma$ are higher than Eq. (11). Two causes of this underestimation of Eq. (11) are supposed as follows. One is the inertia effect. Although Eq. (11) was derived under the condition of neglecting the inertia force, the minimum Re in all measured data was 3.26. The other is Ca range. Numerical simulation of an air finger in a square channel under the condition of neglecting the inertia force shows agreement with the Wong et al.'s analytical solution only in the range of $Ca \leq 10^{-3}$ (Hazel & Heil, 2002). It can be concluded that the functional form of f cannot be represented by the function of Ca with an exponent such as Eqs. (11-12), (16) and (18) for dealing with all $\Delta P_G D_h / \sigma$ in the present experimental range, and a new f model is required for accurate prediction.

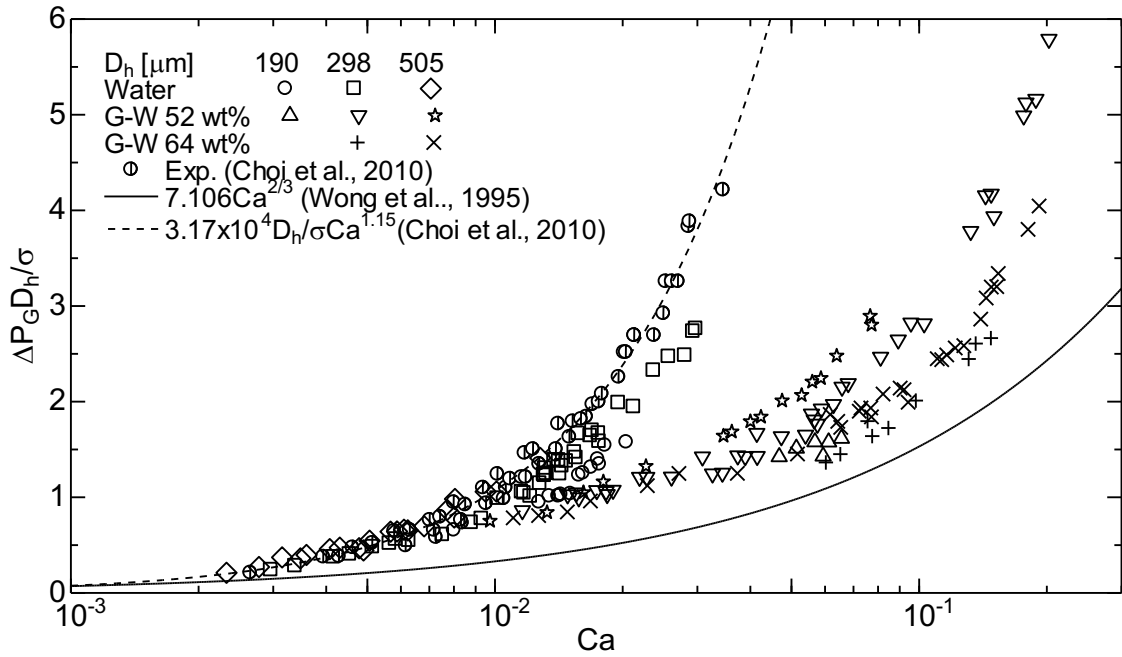


Fig. 8 Relationship between $\Delta P_G D_h / \sigma$ and Ca

Table 4 Relationship between $\Delta P_G D_h / \sigma$ and We at $Ca \sim 2.00 \times 10^{-2}$

D_h [μm]	Liquid	$\Delta P_G D_h / \sigma$	We	$Ca \times 10^{-2}$
490 (Choi et al., 2010)	Water	2.52	18.0	2.01
298	Water	1.99	10.3	1.95
190	Water	1.58	7.17	2.04
505	G-W 52 wt%	1.32	0.641	2.27
298	G-W 52 wt%	1.21	0.381	2.28
505	G-W 64 wt%	1.12	0.155	2.29

Bretherton (1961) theoretically demonstrated that the pressure drop of a Taylor bubble in a circular capillary depended on the front and rear shapes of a Taylor bubble, especially the front shape. Since the front shape is correlated with the liquid film thickness between the Taylor bubble and the channel wall (Bretherton, 1961; Kurimoto et al., 2018), Warnier et al. (2010) extended the Bretherton's pressure drop model using the liquid film thickness model proposed by Aussillous and Qu  r   (2000) to apply for a wide range of the capillary number as mentioned section 1. This relationship between the pressure drop and the liquid film thickness is natural since the magnitude of drag force acting on a Taylor bubble is governed by the cross sectional area of the Taylor bubble. Similarly, Wong et al. (1995)

showed that the liquid film thickness of a Taylor bubble in a square capillary could be represented by the same functional form as ΔP_G in the limit of $Ca \rightarrow 0$ under the condition of neglecting the inertia force, i.e. $Ca^{2/3}$. They showed that the bubble front shape mainly contributed to the pressure drop of a Taylor bubble in a square channel as well as that in a circular channel. Instead of $Ca^{2/3}$, $aCa^{2/3}/(1 + bCa^{2/3})$ where a and b are constants is one of options for the functional form as Warnier et al. adopted it for the pressure drop model in circular microchannels. However, it is not appropriate as the functional form for the evaluation of the present data since the inertia effect is required in the functional form as shown in Table 4. Han and Shikazono (2009) proposed the following liquid film thickness model of Taylor bubbles at channel center and channel corner in square channels for $Ca < 0.4$. The functional form of the model is based on the scaling analysis:

$$\frac{\delta_k}{D_h} = c_{H1} + \frac{c_{H2}Ca^{2/3}}{1 + c_{H3}Ca^{2/3} - c_{H4}We^{c_{H5}}} \quad (24)$$

where δ is the liquid film thickness, c_{Hi} ($i = 1-5$) the constants, and the subscript k denotes center or corner of a channel, respectively. Note that the characteristic length of the Weber number derived by their scaling analysis was originally $D_h/\sqrt{2} - \delta_{corner}$, and they replaced it for D_h for simplicity, i.e., Eq. (23). C_{H1} means a limiting liquid film thickness at a channel corner in a square microchannel in the limit of $Ca \rightarrow 0$. In the point of view of the pressure drop, it can be regarded as the contribution of the curvature of a static bubble and the contribution can be canceled out between the front and rear shapes. Based on the relationship between the pressure drop and the liquid film thickness, the second term of RHS in Eq. (24) is adopted as the functional form of f . It is assumed that the pressure drop of a Taylor bubble agrees with Eq. (11) in the limiting case of $Ca \rightarrow 0$ under the condition of neglecting the inertia force, and we adopt 7.106 as the numerator coefficient. The following ΔP_G model, therefore, is developed using the least squares method for values of C_{Hj} ($j = 3-5$) based on our and Choi et al.'s experimental data:

$$\Delta P_G = \frac{\sigma}{D_h} f = \frac{\sigma}{D_h} \frac{7.106Ca^{2/3}}{1 + 1.25Ca^{2/3} - 0.587We^{0.131}} \quad (25)$$

Figure 9 shows a comparison between the measured $\Delta P_G D_h / \sigma$ and Eq. (25). The developed model can give good agreement with most of the experimental data within $\pm 20\%$ errors. The developed model, therefore, is a more reliable model for the pressure drop of Taylor bubbles than the existing models in the present experimental range.

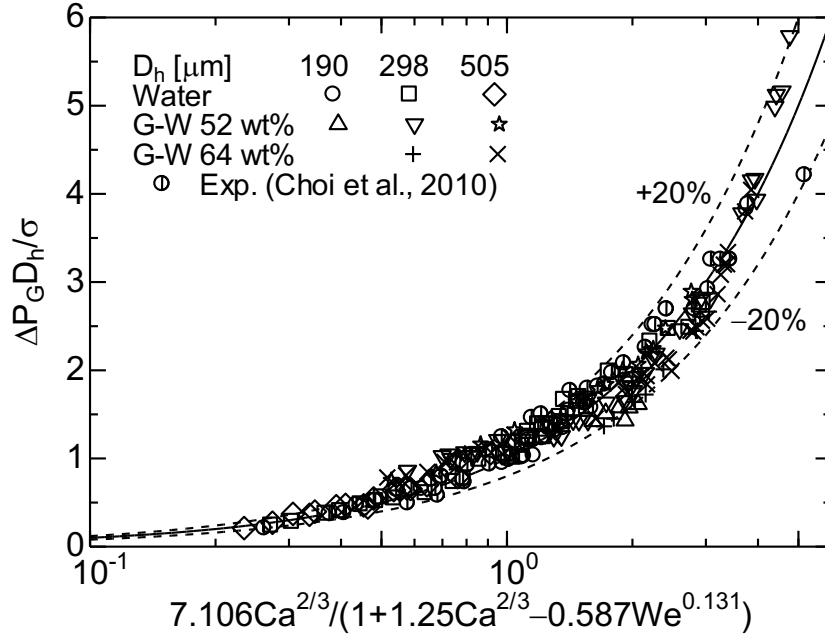


Fig. 9 Comparison between measured $\Delta P_G D_h / \sigma$ and f (Eq. (25))

By combining Eqs. (1-3) and (25), the following dp/dz model for Taylor flow can be obtained:

$$\frac{dp}{dz} = \lambda_{TP} \frac{L_L}{D_h L} \frac{\rho_L j_T^2}{2} = \frac{L_L}{D_h L} \frac{\rho_L j_T^2}{2} \left\{ \frac{56.9}{Re_T} \left[1 + \frac{D_h}{L_L} \frac{0.250 Ca^{2/3}}{Ca_T (1 + 1.25 Ca^{2/3} - 0.587 We^{0.131})} \right] \right\} \quad (26)$$

A comparison of dp/dz of Taylor flow between the measured data and Eq. (26) is shown in Fig. 10. Equation (26) can evaluate most of the measured dp/dz within $\pm 15\%$ errors.

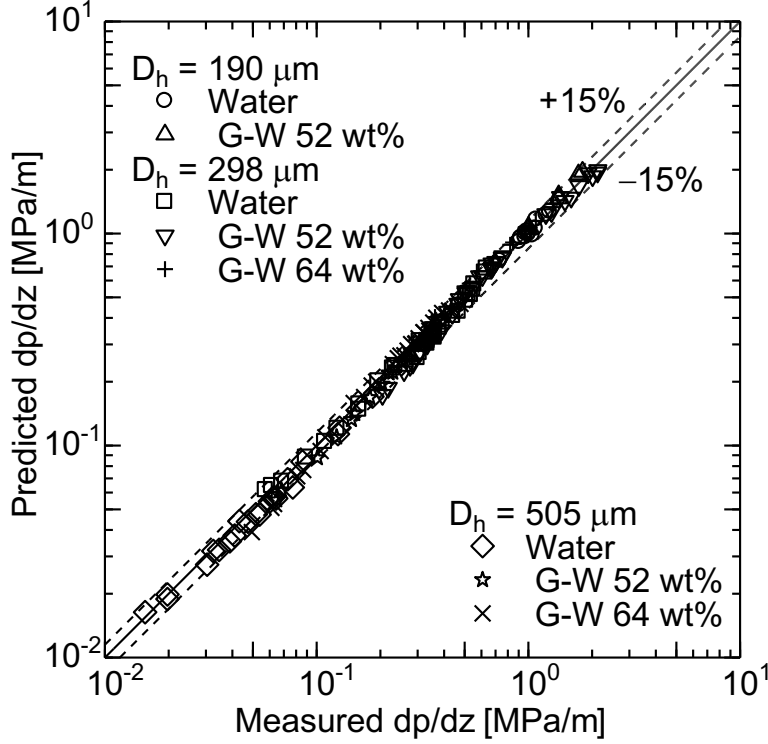


Fig. 10 Comparison of dp/dz of Taylor flow between measured data and Eq. (26)

Although Eqs. (25) and (26) can accurately predict the pressure drop of a Taylor bubble and Taylor flow in square microchannels respectively, Eq. (26) requires both dimensionless numbers based on u_G and j_T , i.e., Ca , We , Re_T and Ca_T . A pressure drop model using only dimensionless numbers based on j_T is more convenient, since a reliable model for predicting u_G is not required when predicting dp/dz (Langewisch and Buongiorno, 2015). The following pressure drop model for a Taylor bubble based on Ca_T and the Weber number $We_T (= Re_T Ca_T)$, hence, is developed using the same functional form as Eq. (25):

$$\Delta P_G = \frac{\sigma}{D_h} f = \frac{\sigma}{D_h} \frac{7.106 Ca_T^{2/3}}{1 + 0.744 Ca_T^{2/3} - 0.623 We_T^{0.120}} \quad (27)$$

By combining Eqs. (1-3) and (27), dp/dz model based on Re_T , Ca_T and We_T can be obtained:

$$\frac{dp}{dz} = \lambda_{TP} \frac{L_L}{D_h L} \frac{\rho_L j_T^2}{2} = \frac{L_L}{D_h L} \frac{\rho_L j_T^2}{2} \left\{ \frac{56.9}{Re_T} \left[1 + \frac{D_h}{L_L} \frac{0.250 Ca_T^{-1/3}}{1 + 0.744 Ca_T^{2/3} - 0.623 We_T^{0.120}} \right] \right\} \quad (28)$$

A comparison between the measured $\Delta P_G D_h / \sigma$ and Eq. (27) and a comparison of dp/dz between the measured data and Eq. (28) are shown in Figs. 11 and 12, respectively.

Equations (27) and (28) can evaluate most of the measured data as well as Eqs. (25) and (26), respectively. The difference in the predicted dp/dz by the models are evaluated by the root mean square error (RMSE) calculated using $\sqrt{\sum^{N_d} (x_p - x_e)^2 / N_d}$, where N_d is the number of data, x_p the each predicted data, x_e the each measured data, respectively. Since the RMSE of Eqs. (26) and (28) is 4.0×10^{-2} and 4.8×10^{-2} respectively, Eqs. (26) and (28) can predict dp/dz with the same level of accuracy.

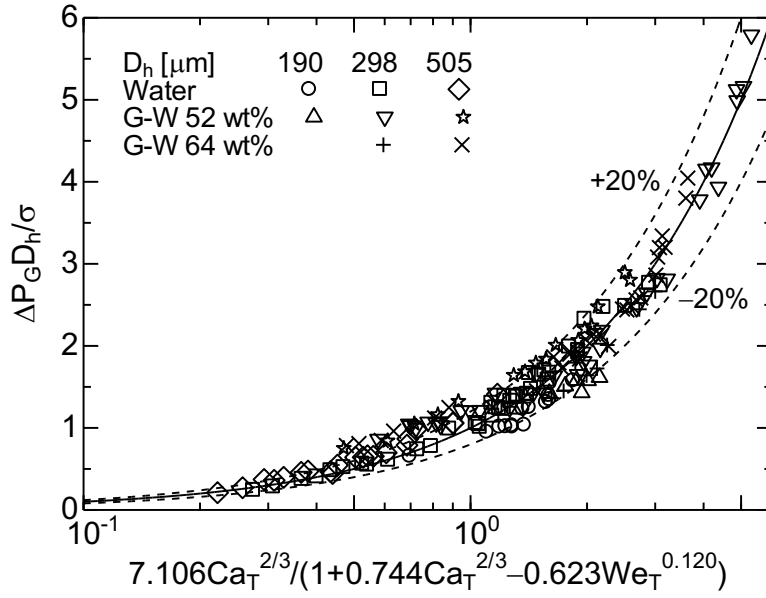


Fig. 11 Comparison between measured $\Delta P_G D_h / \sigma$ and f (Eq. (27))

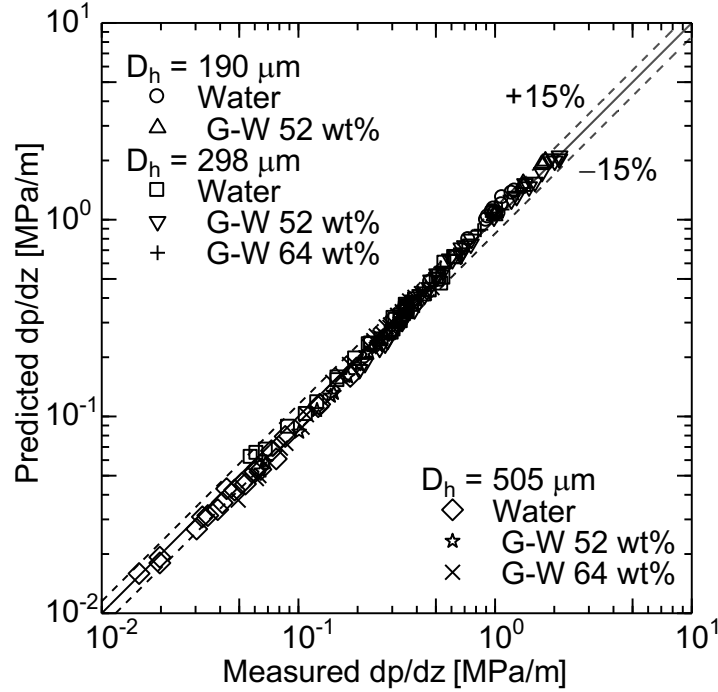


Fig. 12 Comparison of dp/dz between measured data and Eq. (28)

4. Conclusion

The pressure difference in gas-liquid Taylor flow in square microchannels was measured to obtain knowledges and discuss pressure drop models. The experiments were carried out for ranges of $1.98 \times 10^{-3} < Ca_T < 0.132$ and $2.61 < Re_T < 661$, where Ca_T and Re_T are the capillary and Reynolds numbers based on total volumetric flux, respectively. Experimental setup and method were validated with the theoretical model in single phase flow. The existing pressure drop models for a Taylor bubble could not evaluate the measured data. For understanding the discrepancies between the measured data and the existing models, we showed relationship between the measured dimensionless pressure drop of a Taylor bubble $\Delta P_G D_h / \sigma$ and the capillary number. While $\Delta P_G D_h / \sigma$ increases with the capillary number as well as the existing models, dependence of $\Delta P_G D_h / \sigma$ on the liquid viscosity and the hydraulic diameter at the same capillary number in the middle and high capillary number range, i.e., the inertia effect, caused the discrepancies. A new pressure drop model for a Taylor bubble in square microchannels, hence, was developed by adopting the liquid film thickness model proposed by Han and Shikazono (2009) as the functional form. The developed model could accurately evaluate the measured $\Delta P_G D_h / \sigma$. The unit cell model consisting of the developed pressure drop model and the pressure drop equation in single phase flow could also accurately evaluate the measure pressure drop of Taylor flow. In addition, a pressure drop model for a Taylor bubble based on the total volumetric flux was

developed. It could be adopted to the unit cell model for predicting the pressure drop of Taylor flow, which is more convenient in terms of consisting of only the total volumetric flux as the characteristic velocity.

Acknowledgements

We would like to thank Mr. Kota Ozasa for his contribution to the experiments and image processing.

Appendix A

The measured Taylor bubble shapes are shown in Fig. A1. All measured data of u_G , j_G , j_L , dp/dz , L_G and L_L are summarized in Table A1.

Fig. A1 Measured Taylor bubble shapes

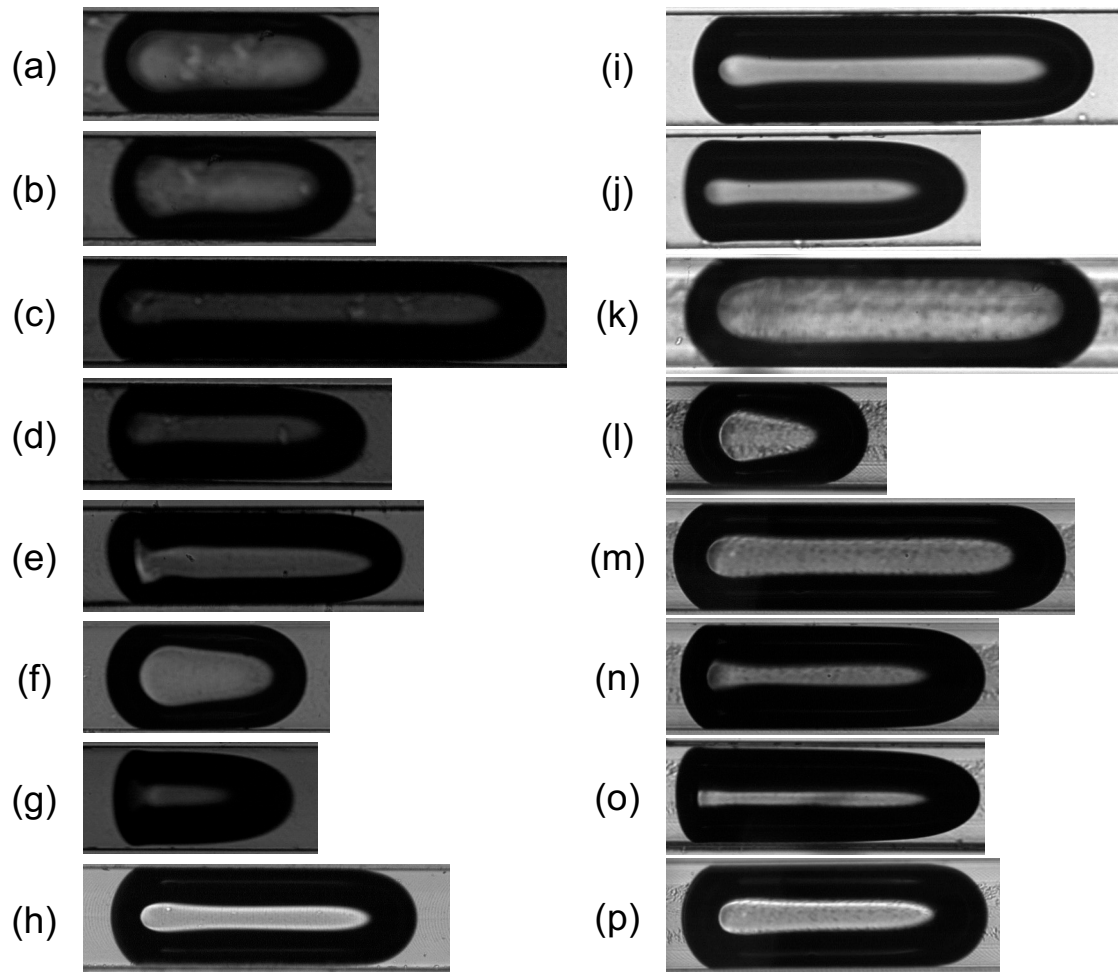


Table A1 Measured data

Fig. A1	u_G [m/s]	j_G [m/s]	j_L [m/s]	dp/dz [MPa/m]	L_G [mm]	L_L [mm]	Fig. A1	u_G [m/s]	j_G [m/s]	j_L [m/s]	dp/dz [MPa/m]	L_G [mm]	L_L [mm]	Fig. A1	u_G [m/s]	j_G [m/s]	j_L [m/s]	dp/dz [MPa/m]	L_G [mm]	L_L [mm]
$D_h = 190 \mu\text{m}$, Water							$D_h = 298 \mu\text{m}$, G-W soln. (52 wt%)							$D_h = 505 \mu\text{m}$, G-W soln. (52 wt%)						
	1.47	0.755	0.626	1.05	0.541	0.242		0.752	0.342	0.316	0.229	0.733	0.486		0.623	0.340	0.176	0.0644	1.80	0.598
	1.36	0.770	0.585	0.967	0.510	0.218		0.421	0.183	0.166	0.472	0.773	0.480		0.409	0.138	0.205	0.0516	1.12	1.13
	1.28	0.573	0.626	0.999	0.428	0.245		0.693	0.362	0.166	0.507	1.15	0.371		0.493	0.205	0.205	0.0601	1.28	0.877
	1.41	0.748	0.626	1.08	0.465	0.213		0.662	0.259	0.248	0.674	0.752	0.518		0.651	0.335	0.205	0.0772	1.62	0.644
	1.09	0.392	0.668	0.903	0.362	0.354		0.770	0.342	0.248	0.702	0.859	0.405		0.769	0.189	0.468	0.126	0.806	0.810
	1.30	0.572	0.668	1.01	0.433	0.285		0.994	0.505	0.248	0.758	1.00	0.313		0.396	0.0698	0.264	0.0431	0.932	2.30
	1.15	0.385	0.710	0.967	0.321	0.375		1.09	0.475	0.331	0.987	0.774	0.325	(l)	0.475	0.136	0.264	0.0623	1.02	1.25
	1.14	0.383	0.752	0.986	0.318	0.366		1.17	0.449	0.414	1.21	0.639	0.345		0.550	0.135	0.322	0.0729	0.872	1.29
	1.22	0.373	0.835	1.09	0.287	0.331		1.62	0.735	0.414	1.29	0.855	0.286		0.648	0.132	0.381	0.0857	0.666	1.59
	1.43	0.542	0.835	1.22	0.360	0.283	(g)	1.26	0.427	0.497	1.42	0.525	0.373		0.815	0.189	0.468	0.128	0.630	1.11
	1.65	0.715	0.835	1.26	0.434	0.282		1.76	0.701	0.497	1.49	0.781	0.300		1.03	0.296	0.527	0.183	0.681	0.748
(a)	0.647	0.221	0.413	0.469	0.495	0.647		2.19	0.889	0.580	1.59	0.999	0.383		$D_h = 505 \mu\text{m}$, G-W soln. (52 wt%)					
(b)	1.03	0.410	0.578	0.733	0.464	0.475		1.81	0.663	0.580	1.73	0.646	0.302		0.119	0.0340	0.0574	0.0647	1.36	1.887
	1.16	0.390	0.743	0.921	0.357	0.475		1.84	0.638	0.663	1.91	0.542	0.290	(m)	0.163	0.0676	0.0574	0.0684	1.87	1.309
	$D_h = 190 \mu\text{m}$, G-W soln. (52 wt%)							2.16	0.813	0.663	2.01	0.663	0.273		0.198	0.0650	0.0861	0.101	1.34	1.281
	0.696	0.386	0.162	1.00	1.04	0.330		2.31	0.794	0.745	2.13	0.661	0.326		0.221	0.0633	0.115	0.124	1.10	1.662
	0.750	0.350	0.243	1.39	0.756	0.391		2.48	0.795	0.828	2.12	0.816	0.484		0.424	0.184	0.115	0.151	1.65	0.728
(c)	0.573	0.290	0.162	0.994	0.860	0.340		0.182	0.0581	0.0994	0.285	0.835	1.02		0.278	0.0617	0.144	0.146	1.02	1.839
	0.629	0.263	0.243	1.39	0.637	0.401		0.225	0.0963	0.0994	0.305	0.891	0.710		0.443	0.179	0.144	0.178	1.39	0.804
	0.728	0.244	0.324	1.72	0.524	0.493		0.224	0.0570	0.133	0.347	0.690	1.15		0.490	0.174	0.172	0.204	1.25	0.857
(d)	0.804	0.319	0.324	1.80	0.595	0.468		0.269	0.0938	0.133	0.389	0.758	0.761	(n)	0.648	0.285	0.172	0.220	1.47	0.630
	$D_h = 298 \mu\text{m}$, Water							0.142	0.0597	0.0649	0.205	1.08	1.02		0.519	0.169	0.201	0.229	1.11	0.902
	1.16	0.563	0.444	0.353	0.829	0.437		0.193	0.0990	0.0649	0.218	1.30	0.680		0.686	0.276	0.201	0.252	1.31	0.675
	1.26	0.651	0.444	0.380	0.830	0.401		0.213	0.0977	0.0812	0.259	1.14	0.721		0.583	0.165	0.230	0.254	1.05	0.910
	1.37	0.749	0.444	0.360	1.12	0.440		0.191	0.0583	0.0974	0.276	0.896	1.03		0.719	0.268	0.230	0.279	1.17	0.699
	0.936	0.380	0.461	0.313	0.689	0.554		0.458	0.183	0.162	0.465	0.898	0.539		0.784	0.260	0.258	0.310	1.12	0.707
	1.07	0.472	0.461	0.333	0.808	0.499		0.721	0.362	0.162	0.507	1.19	0.363		0.940	0.358	0.258	0.328	1.31	0.590
	1.12	0.562	0.461	0.360	0.845	0.470		0.477	0.179	0.195	0.543	0.758	0.624		0.947	0.351	0.287	0.352	1.17	0.629
	1.25	0.657	0.461	0.353	0.967	0.438		0.706	0.353	0.195	0.588	0.981	0.421		$D_h = 505 \mu\text{m}$, G-W soln. (64 wt%)					
	1.42	0.751	0.461	0.353	1.08	0.439		0.509	0.176	0.227	0.606	0.680	0.676		0.158	0.0647	0.0465	0.104	1.99	1.05
	0.981	0.381	0.478	0.306	0.687	0.567		0.807	0.345	0.227	0.669	0.963	0.402		0.217	0.0602	0.0929	0.168	1.26	1.48
	1.05	0.472	0.478	0.333	0.806	0.511		0.580	0.172	0.260	0.676	0.663	0.679		0.301	0.119	0.0929	0.182	1.53	0.915
	1.15	0.563	0.478	0.353	0.782	0.447		0.834	0.337	0.260	0.753	0.860	0.433		0.421	0.176	0.0929	0.191	1.87	0.662
	1.06	0.468	0.495	0.360	0.712	0.485		0.399	0.188	0.130	0.382	1.01	0.507		0.448	0.169	0.116	0.229	1.57	0.722
	0.819	0.386	0.342	0.266	0.804	0.444		0.683	0.372	0.130	0.416	1.44	0.339		0.479	0.163	0.139	0.269	1.38	0.767
	1.27	0.763	0.342	0.299	1.37	0.400	(h)	0.233	0.0962	0.0974	0.308	0.903	0.729		0.526	0.157	0.163	0.303	1.24	0.817
	1.06	0.571	0.342	0.306	0.979	0.353		0.279	0.143	0.0974	0.333	1.02	0.587		0.360	0.193	0.0465	0.110	3.29	0.561
	1.14	0.567	0.410	0.333	0.945	0.400		0.378	0.191	0.0974	0.337	1.22	0.453		0.381	0.184	0.0697	0.150	2.29	0.626
	0.947	0.375	0.479	0.360	0.560	0.441		0.508	0.285	0.0974	0.344	1.48	0.360		0.644	0.279	0.116	0.242	1.96	0.544
	1.19	0.558	0.479	0.380	0.788	0.401		0.320	0.0924	0.166	0.438	0.690	0.766		0.815	0.387	0.116	0.249	2.26	0.449
	1.03	0.371	0.547	0.393	0.554	0.454		$D_h = 298 \mu\text{m}$, G-W soln. (64 wt%)							0.375	0.109	0.139	0.260	1.17	1.03
	1.36	0.548	0.616	0.440	0.772	0.537		0.352	0.189	0.0670	0.362	1.51	0.398		0.659	0.268	0.139	0.279	1.68	0.583
(e)	1.58	0.725	0.616	0.467	0.942	0.483	(i)	0.380	0.180	0.101	0.528	1.09	0.474		0.839	0.370	0.139	0.294	2.00	0.484
	1.42	0.537	0.684	0.508	0.650	0.466		0.441	0.172	0.134	0.681	0.942	0.509		0.425	0.106	0.163	0.292	1.08	1.06
	2.27	0.862	0.855	0.635	0.850	0.421		0.452	0.166	0.168	0.820	0.768	0.613		0.678	0.258	0.163	0.319	1.48	0.616
	2.41	1.04	0.855	0.622	1.02	0.420		0.763	0.329	0.168	0.846	1.11	0.383		0.863	0.354	0.163	0.341	1.76	0.523
	2.37	1.06	0.924	0.555	1.21	0.679		0.494	0.159	0.201	0.966	0.670	0.680		0.450	0.103	0.186	0.321	0.958	1.17
	1.72	0.713	0.684	0.521	0.809	0.405		0.793	0.314	0.201	1.02	0.957	0.419		0.537	0.152	0.186	0.337	1.10	0.885
	2.07	0.796	0.770	0.548	1.01	0.446		0.573	0.154	0.235	1.08	0.649	0.678		0.709	0.250	0.186	0.355	1.35	0.656
	1.91	0.755	0.684	0.535	0.961	0.399	(j)	0.861	0.292	0.268	1.31	0.757	0.492		0.896	0.342	0.186	0.380	1.60	0.544
	0.472	0.200	0.239	0.159	0.734	0.582		$D_h = 505 \mu\text{m}$, Water							0.549	0.148	0.209	0.367	0.979	0.990
	0.711	0.292	0.316	0.235	0.636	0.462		0.255	0.0710	0.146	0.0304	1.18	1.96		0.743	0.242	0.209	0.391	1.22	0.695
(f)	0.608	0.197	0.316	0.195	0.589	0.665	(k)	0.188	0.0724	0.0878	0.0155	1.99	1.84		0.889	0.329	0.209	0.422	1.39	0.577
	0.273	0.155	0.0947	0.0614	1.21	0.571		0.281	0.144	0.0878	0.0197	2.83	1.33		1.05	0.418	0.209	0.436	1.62	0.530
	0.336	0.206	0.0947	0.0686	1.44	0.474		0.225	0.0720	0.117	0.0197	1.84	2.25	(o)	1.12	0.404	0.232	0.478	1.52	0.536
	0.367	0.204	0.126	0.0889	1.14	0.533		0.387	0.212	0.117	0.0325	2.06	0.816		0.0739	0.0341	0.0225	0.0611	1.98</	

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