Influence of Wettability on Slug Flow in Micro Channels

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Summary

Two phase flow in micro channel has been of research interest recently due to its high heat and mass transfer rate, and in this study, experimental investigation is performed on the influence of wettability of the tube in slug flow. The liquid film thickness and flow behavior of water-air slug flow are investigated by using laser focus displacement meter (LFDM) and high-speed camera, respectively. Circular glass tubes with inner diameters of 0.76mm and 1mm are prepared to be hydrophilic or hydrophobic by applying surface chemical treatment (using chemical surfactant: piranha solution and toluene). The experimental results are compared and discussed to obtain the influence of wettability, and as a result, the liquid film thickness is found to be similar in hydrophobic and hydrophilic micro channels, which is compatible with theoretical analysis. However, the flow characteristics is influenced by wettability at low capillary number region, at which no liquid film exists. The critical capillary number for film deposition on the wall is influenced by the hydrophobicity of the tube, consequently, the critical capillary number is higher in a hydrophobic tube than that in hydrophilic one.

Keywords: Heat transfer, Liquid film thickness, Wettability, Slug flow, Critical capillary number, Contact angle

1. Introduction

With the advancement in microfabrication techniques, micro-structured devices such as micro channel exchangers and microfluidics have gained importance in a range of industrial applications. To design these micro devices better, it’s significant to understand two phase flow characteristics in micro channels. Slug flow is a prevailing flow pattern in micro channels due to the dominance of surface tension. For slug flow in micro/mini channel, there is usually liquid film around the bubble, and the liquid film thickness is an important parameter in predicting heat transfer performance in micro channel heat exchangers and micro-reactors. In this paper, the liquid film thickness in hydrophilic and hydrophobic micro tubes (inner diameter are 0.76mm and 1mm) is investigated experimentally.

Moreover, it is observed that the liquid film drains away when the capillary number is low, and there exists a critical capillary number, above which, the bubble is lubricated by liquid film. And the study on influence of hydrophobicity on critical capillary number is performed to better understand slug flow behavior in micro channels.

2. Experimental Investigation

2.1 Experimental instruments

The schematic diagram of the experimental apparatus is shown as Fig. 1. Water and air are injected by syringe pumps (Harvard Apparatus, accuracy within 0.35%, reproducibility within 0.05%), then they are mixed in the T-junction and introduced into the test section (circular Pyrex tube with diameters of 0.76mm and 1mm). The flow pattern is recorded by high speed camera (Keyence, VW-600C), and the liquid film thickness is measured by laser focus displacement meter (LFDM) (Keyence, LT9010). The resolution of LFDM is 0.01µm, the response time is 640µs, the diameter of the laser spot is 2µm, and the focal length of the lens in the LFDM is about 5mm. Measured liquid film thickness is transformed to DC voltage signal in the range of ±10 V, and the data is transferred to a PC through the data logger.

<table>
<thead>
<tr>
<th>Fluid</th>
<th>ρ (kg/m³)</th>
<th>µ (µPa·s)</th>
<th>σ (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>997</td>
<td>894</td>
<td>71.97</td>
</tr>
<tr>
<td>Air</td>
<td>1.18</td>
<td>18.4</td>
<td></td>
</tr>
</tbody>
</table>

Table. 1 Fluid properties at 25°C

2.2 Surface treatment method and experimental condition

The fluid property of the working fluid is shown in Table 1. The wettability of the tubes can be evaluated

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by contact angle, which is described by Young’s equation:

$$\cos \theta = \frac{\sigma_{SL} - \sigma_{SG}}{\sigma_{LG}}$$

(1)

For the surface treatment in this experiment, the surface tension of solid-liquid interface is modified to change the contact angle. The inner surface of the tube is flushed with piranha solution (the mixture of 3 parts of concentrated sulfuric acid and 1 part of 30% hydrogen peroxide solution) for 5 minutes, consequently, the organic matter adhering to the surface is removed, and the OH group is exposed, thus glass surface becomes hydrophilic. While for hydrophobic tube, flushing the tube with toluene (C₆H₅CH₃) repels the hydrophilic OH group on the glass surface, then the hydrophobic tube is obtained. As a result, the contact angle of the hydrophilic and hydrophobic after surface treatment is 25° and 109°, respectively. (As shown in Fig. 2) All the experiments were done at the room temperature of 25°C.

The above measurement is under the condition of relative large capillary numbers, however, as the capillary number decreases, the level of difficulty of keeping the bubble moving at a constant velocity increases, thus it’s nearly impossible to obtain the liquid film thickness data at very low capillary number region. Therefore, the investigation is implemented by visual observation for low capillary number region.

2.3 Experimental results
2.3.1 Liquid film thickness measurement at high capillary number region

The liquid film thickness is measured by LFDM, and the results for tubes of different wettability and diameters are shown in Figs. 3 and 4, respectively. The capillary number is defined as Eq. 2:

$$Ca = \frac{\mu U_b}{\sigma}$$

(2)

Unlike the results in Yoshinaga et al (2015) ³), the liquid film thickness in hydrophilic tube is found to be slightly thicker than that in hydrophobic tube (Fig.3, inner D=0.76mm). However, in the tube diameter of 1mm (Fig.4), the liquid film thickness is undistinguishable in hydrophobic and hydrophilic tubes. And the small difference of results probably comes from the measurement uncertainty of bubble velocity, because it is very difficult to keep the bubble travelling at a constant velocity. Compared to liquid film thickness measurement (accuracy is 0.01µm), the uncertainty in bubble velocity measurement accounts for the majority of the measuring uncertainty.

Visual observation is employed to investigate the influence of wettability at low capillary region (as circled in Fig. 3 & Fig. 4). As a result, an interesting phenomenon is observed, when the capillary number is low, there is no liquid film around the bubble, and the bubble is directly in contact with the inner wall of the tube. This conclusion is based on the following observation: 1) There is a small droplet on inner wall (Fig. 5a & Fig. 5b), which indicates the liquid film does not exist. Because a droplet cannot lie on liquid film, otherwise they will merge together. 2) Contact lines are observed at the bubble head and tail (Fig. 5a & Fig. 5b). And the contact line is the intersection of three phases (solid-liquid-gas), indicating there is no liquid film around the bubble. Because if liquid film
exists, it will separate solid phase (inner surface of the tube) and vapor phase (air in the bubble), and in this case, only two-phase intersection exists (vapor-liquid interface and liquid-solid interface), which is contradictory with the observation. 3) Due to the existence of contact line, a dry bubble has sharp transition part in bubble head and tail (Fig.5 a), but the bubble head and bubble tail becomes smooth and nearly spherical (Fig. 5 d) when it's lubricated by liquid film.

Fig.5 shows the liquid film formation process in tube of moderate wettability (static contact angle=60°). By the way, the hydrophilic tube is not used to illustrate the liquid film formation process, because liquid film can easily form on a hydrophobic tube, therefore, this process is not so apparent. In addition, the distinction between a dry bubble and a wetted bubble in a hydrophilic tube is so slight, and that makes visual observation more challenging.

The bubble is stationary initially (Fig. 5 a), and the bubble is dry without liquid film around it. As the pressure difference at the two ends of the bubble increases, in the first stage, the bubble is still stationary. However, the contact angle at bubble head (receding contact angle) decreases (Fig. 5b) and the contact angle at bubble tail (advancing contact angle) increases, which is called contact angle hysteresis. With further increase in the pressure difference between two ends of the bubble, the bubble starts to move, and the contact angles dynamically changes with respect to the contact line velocity, which is called dynamical contact angle4). The receding contact angle decreases with increasing contact line velocity while the advancing contact angle increases with increasing contact line velocity. And there exists a critical capillary number, above which, liquid film will be left behind as the bubble head travels downstream, whereas, there will be no liquid film if the capillary number is lower than the critical value.

As shown in Fig.5 c, at bubble velocity of 25mm/s (Ca=3.13×10⁻⁴), the front part of the bubble is wetted, bubble head becomes smooth and spherical, and the contact line lies at the center of the bubble, which means half of the bubble is wetted while the latter half of the bubble is still dry. With further increase in capillary number, the entire bubble is wetted by liquid film (Fig.5 d), and both the bubble head and tail is smooth and spherical.

The above mentioned critical capillary number is determined by visual observation, the inlet mass flux pumped by the syringe is gradually increased as well as the bubble velocity. When the capillary number is increased to a point that the front part of the bubble is wetted by liquid film while the rear part of the bubble remains dry (Fig.5 c), it's considered that the bubble
is at the critical state, and the corresponding capillary number is regarded as the critical capillary number. The critical bubble velocity can be calculated from recorded video, thus the critical capillary number is obtained as well. For example, the bubble velocity calculated from Fig. 5 c is 25 mm/s, therefore, the critical capillary number of this tube is $3.13 \times 10^{-4}$.

Similarly, when the bubble decelerates to a velocity below the critical bubble velocity, the liquid film cannot be maintained by the pressure difference between the two ends of the bubble (see detailed analysis in section 3.2.3), and the liquid film around the bubble starts to drain away. From Fig. 6 b to Fig. 6 e, it's observed that the contact line moves back with the dewetting of liquid film. And it's calculated that the dewetting velocity of liquid film is about 22 mm/s, which is close to the critical bubble velocity. Moreover, as the hydrophobicity of tube increases, the dewetting velocity increases as well as the critical capillary number. For a hydrophobic tube with static contact angle of $109^\circ$, the dewetting velocity increases to 95 mm/s, and the critical bubble becomes 100 mm/s. The dewetting velocity and critical bubble velocity of three different wettability is summarized as Table 2.

### Table 2 Dewetting velocity, critical bubble velocity and critical capillary number in different tubes (inner D=1 mm)

<table>
<thead>
<tr>
<th>Wettability</th>
<th>$U_d$ (mm/s)</th>
<th>$U_{cr}$ (mm/s)</th>
<th>$C_{acr}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrophilic</td>
<td>8</td>
<td>7</td>
<td>$8.68 \times 10^{-5}$</td>
</tr>
<tr>
<td>Non-treatment</td>
<td>22</td>
<td>25</td>
<td>$3.10 \times 10^{-4}$</td>
</tr>
<tr>
<td>Hydrophobic</td>
<td>104</td>
<td>100</td>
<td>$1.24 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

3. Discussion

3.1 Liquid film thickness

The slug flow in the micro channel is governed by Navier-Stokes equation, and if the boundary condition is assumed to be no-slip on the inner wall of the tube, the liquid film thickness should be the same for hydrophobic and hydrophilic tubes. In addition, the liquid film thickness is larger than 10 µm, which is far beyond the effective range of van der waals forces (within 100 nm). Therefore, the only possible explanation for the thinner liquid film is the slippage on the wall, which is described by Navier slip boundary condition, Eq. (3).

$$U_s = \bar{U} + \frac{\partial U}{\partial z}$$ (3)
Because the liquid film thickness is determined by capillary number, which is the relative value of surface tension and viscous force. The surface tension of water-air interface remains unchanged against the hydrophobicity of tube, and viscous force becomes smaller due to the smaller velocity gradient caused by the slippage on hydrophobic wall, consequently, the equivalent capillary number becomes Eq. (4). Therefore, liquid film thickness becomes smaller for the same bubble velocity.

\[
Ca = \frac{\mu(U_1 - U_2)}{\sigma} \quad (4)
\]

However, the slip length \( l_s \) is of the magnitude of 1 nanometer, thus the slip velocity is negligible compared to bubble velocity. Therefore, it’s concluded that liquid film thickness is not influenced by the wettability.

3.2 Critical capillary number

As analyzed in section 3.1, the liquid film thickness is not influenced by wettability. However, the wettability of the tube influences critical capillary number. Critical capillary number is the value at which the bubble is only partially wetted, while above which the bubble is totally wetted by liquid film. Moreover, the influence of wettability on critical bubble velocity will be analyzed from three perspectives: dynamic contact angle, dewetting velocity and pressure distribution around the bubble.

3.2.1 Dynamic contact angle

There have been a lot of research about the dynamic contact angle. For liquid moving on a given surface, the contact angle is a function of the contact line velocity or capillary number, \( \theta = f(Ca) \). The receding contact angle (Fig. 5) at bubble head decreases with increasing capillary number, while the advancing contact angle increases with increasing capillary number. The implication of dynamic contact angle is the existence of intersection of three phases (water-air-wall) at this capillary number, in another word, there is no liquid film when bubble moves at this capillary number.

As shown in section 2, the critical capillary number for liquid film deposition is \( 1.24 \times 10^{-3} \) for hydrophobic tube, which is higher than that in hydrophilic tube (\( Ca_c=8.68 \times 10^{-5} \)). To explain it, firstly it’s noticed that the receding contact angle should decrease to a certain level to allow the liquid to intrude in and form liquid film. As for a hydrophobic tube, the initial contact angle is larger than that in hydrophilic tube, which indicates higher capillary number is required to decrease the receding contact angle to a certain contact angle, at which liquid film forms. In another word, the critical capillary number for liquid film formation is higher in hydrophobic tubes.

3.2.2 Dewetting velocity

On the other hand, dewetting velocity is closely related to the critical capillary number. The formation of liquid film can be regard as the result of relative movement of bubble and the contact line (moves at dewetting velocity). As demonstrated in Fig. 7, when the bubble velocity exceeds the liquid film dewetting velocity, the liquid film is left behind, the length of liquid film left behind is roughly the relative displacement between bubble head and contact line.

Moreover, the dewetting velocity is found to be close to the critical bubble velocity. And the dewetting velocity is higher in hydrophobic surface than that in hydrophilic surface. Therefore, the critical bubble velocity (or critical capillary number) is higher in hydrophobic tubes than that in hydrophilic tubes.

3.2.3 Pressure distribution

At the capillary number slightly larger than critical capillary number, the bubble (Fig. 5d) head and tail are nearly spherical (at low capillary number), and the bubble is lubricated by liquid film. According to Laplace pressure, the pressure in the liquid film is higher than the liquid pressure at bubble head and tail, because it has only one principal radii of curvature in the film region, and two principal radii of curvature at bubble head and tail, the pressure relations are described by Eqs. (5)-(7):

\[
P_{\text{film}} + \sigma / R = P_{\text{head}} \quad (5)
\]

\[
P_{\text{head}} + 2\sigma / R = P_{\text{bubble}} \quad (6)
\]

\[
P_{\text{tail}} + 2\sigma / R = P_{\text{bubble}} \quad (7)
\]
Therefore, the liquid in the film tend to drain out when the bubble is stationary or moves at a low velocity. Because the pressure difference (right hand side of Eq. (8)) at the two ends of the bubble is too small to maintain the high pressure in film region, thus no liquid film exists. And in this case, the pressure difference is balanced by the capillary pressure (left hand side of Eq. (8)).

\[
2\pi R \sigma \cos \theta_a - \cos \theta_{adv} = \int_{S_1} P_z \cos \vec{n}_z \cdot \vec{a} \, dS_z - \int_{S_2} P_z \cos \vec{n}_z \cdot \vec{a} \, dS_2 \tag{8}
\]

(\(R\) is the radius of the tube, \(\sigma\) is the surface tension of water-air interface, \(\vec{n}_1\) and \(\vec{n}_2\) are the local surface normal vectors of bubble head and bubble tail, and \(\vec{a}\) is a vector along the axis of the tube)

However, \((\cos \theta_a - \cos \theta_{adv})\) is upper bounded, and as the pressure difference increases, eventually, the capillary pressure induced by contact angle hysteresis (left hand side of Eq. (8)) can no longer balance the pressure difference between the two ends of the bubble, then the liquid intrudes in and forms liquid film. Following simplified formula is used to obtain the pressure difference caused by capillary force:

\[
\Delta P = \frac{2\pi R \sigma (\cos \theta_a - \cos \theta_{adv})}{\pi R^2} \tag{9}
\]

Referring to the experimental data (Fig. 5), when bubble is at critical velocity, the receding contact angle \(\theta_a\) is 30\(^\circ\) and the advancing contact angle \(\theta_{adv}\) is 90\(^\circ\). And referring to Eq. (9), the resulting pressure drop between two ends of the bubble is 249.4 Pa. It is the maximum pressure difference that can be balanced by capillary force, thus if the pressure difference between the two ends of the bubble exceeds the above value, there forms liquid film around the bubble.

In addition, it’s observed that the difference of receding contact angle and advancing contact angle is larger in hydrophobic tubes, therefore, higher pressure drop is required to surpass the capillary force and form liquid film.

4. Conclusions

The liquid film thickness of air-water slug flow is measured by LFDM in hydrophobic and hydrophilic tubes. And wettability is found to have no influence on liquid film thickness. However, when the capillary number is lower than a critical value, liquid film does not exist. And the critical capillary number is influenced by the wettability of tubes. As a result, the critical capillary number for liquid film formation is about 1.24×10\(^{-3}\) and 8.68×10\(^{-5}\) for hydrophobic (static contact angle=109\(^\circ\)) and hydrophilic tubes (static contact angle=25\(^\circ\)) respectively. In another word, the bubble is easier to be lubricated by liquid film in hydrophilic tubes.

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Nomenclature

\[\begin{align*}
Ca & : \text{capillary number} (= \mu U_b / \sigma) \\
Ca_{cr} & : \text{critical capillary number} (= \mu U_{cr} / \sigma) \\
D & : \text{inner diameter, m} \\
p & : \text{pressure, Pa} \\
\sigma & : \text{surface tension of water-air interface, N/m} \\
U_{cr} & : \text{critical bubble velocity, m/s} \\
U_b & : \text{bubble velocity, m/s} \\
U_d & : \text{dewetting velocity} \\
\delta & : \text{liquid film thickness, m} \\
\rho & : \text{density, kg/m}^3 \\
\mu & : \text{viscosity, Pa} \cdot \text{s} \\
\theta & : \text{contact angle, degree (°)}
\end{align*}\]

References