Measurements of interfacial film thickness for immiscible liquid–liquid slug/droplet flows

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Abstract

A novel method for measuring the interfacial liquid film thickness between immiscible liquids of a slug/droplet in a micropipe is proposed. This method is capable of measuring an oil slug/droplet in water with the relative refractive index (m) between the immiscible liquids very close to one in a capillary tube. Unlike the measurement configuration for an air slug in water, an optical orifice for optimizing the refracted fringes pattern by the liquid–liquid interface is introduced. Measurements of film thicknesses of a kerosene oil droplet/slug in a water wetted capillary tube ($m > 1$) and a water droplet/slug in a kerosene oil wetted capillary tube ($m < 1$) were demonstrated. This diagnostic method is easy to implement and it will have great potential for immiscible liquid flow research in microfluidic channels.

Keywords: optic diagnostics, film thickness, immiscible liquids, microfluidics

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Research on immiscible liquid–liquid slug/droplet dynamics and mass or heat transfer in micropipes is an important area in microreactors, microfluidics and lab-on-a-chip devices, such as the formation of patterns in flows far from equilibrium [1] and the effects of wettability on the patterns generated in two-phase flows [2]. Droplets have been used in microchannels to perform and enhance two-phase chemical reactions [3]. In this paper, we refer to droplets that contact the wall of the capillary tube but do not wet the walls as slugs. This is because even though slugs press against the wall of the capillary tube, they may be prevented from intimate contact with the walls by a thin film of carrier fluid [4, 5]. For numerical analysis and modelling, it is crucial to understand whether a thin liquid exists between a liquid droplet/slug and the wall. It is also very important to know how thick the liquid film is if it does exist. In the past, however, due to lack of a measurement technique, experimental studies have mainly focused on visualization of

flow pattern and measurement of temperature and effective thermal conductivity. The measurement technique for liquid film thickness between the liquid droplets/slugs and the capillary tube has not been well established.

Several optical techniques have been used to measure the flat film thickness. For example, Wu *et al* [6] developed an infrared (IR) technique that relies on light transmission to measure the soap film thickness in a 60 mm $\times$ 1200 mm channel. Hidrovo and Hart [7] presented an emission re-absorption laser induced fluorescence (ERLIF) method for measurement of fluid film thickness from 5 to 400 μm with 1% accuracy. Browne [8] developed an optical technique to determine the fluid film thickness distribution in conjunction with the moiré fringes. However, all these techniques are applicable to a plate film surface and are difficult to use for the immiscible liquid–liquid film formed inside a micro capillary tube.

The recently developed optical diagnostic method by Wang and Qiu [9] demonstrated a promising fringe probing

Figure 1. Schematic diagram of experimental apparatus.

technique of liquid film thickness of an air plug bubble in a micropipe. This method, with the extension of the previous optical techniques [10, 11], utilized the total reflected interference fringe pattern to estimate the liquid film thickness between the gas–liquid–glass interfaces. A well matched liquid surrounding the micro glass pipe was used to reduce the complication of the calculations. Unfortunately, this method may not be directly applied to immiscible liquid–liquid droplet/slug condition due to some constraints. The first constraint is that unlike the air-plug/liquid interface, the refractive index of the surrounding liquid (the film) may be smaller than the slug's which causes a problem in finding the total reflection angle from the incident surface. For example, a kerosene droplet/slug surrounded by a water film will have no total reflection angle when the ray is incident from water to kerosene. Furthermore, because the relative refractive index of the droplet/slug and that of the glass wall may happen to be very close to 1, it may cause no fringe scattering from the interface or too many different refraction/reflection patterns mixed at certain receiving angles. Therefore, the scattering angles and the optimized receiving angle need to be addressed.

In this paper, the effect of optical orientation angles was discussed in the measurements of liquid film thickness of immiscible liquids. Both relative refractive indices greater (kerosene slug/droplet surrounded by water film) and less than 1 (water slug/droplet surrounded by kerosene film) were studied. This technique can provide measurement results that should also be important for studies that utilize slug-based microfluidic platforms to manipulate proteins, biological fluids and organic reagents [12, 13].

2. Experimental set-up

As shown in figure 1, a uniphase mode 1135P, 15 mW He–Ne laser ($\lambda = 632.8$ nm), a split lens and a lens (L1) with 200 mm focus are utilized for producing interference fringes. The distance between the two parallel split beams is 50 mm. The fine parallel fringes formed are projected onto the glass–film–slug multi layers, and the multi refracted

fringe pattern is recorded at an optimized orientation angle by an image system. The scattered fringes at the liquid/liquid (droplet/slug) interface are received into a JVC TK-C1381 CCD camera, which is coupled with a Nikon L37c 52 mm optical lens. According to the geometrical optics on light refraction, the viewing angle of the image system can be optimized.

To demonstrate the capability of this newly developed technique, a validating experiment was performed with a kerosene oil slug while it was immersed in water and arranged to transport vertically along the micro tube (see figure 1). Two capillary tubes with 750 μm and 500 μm inner diameters (OD = 1500 μm and 1000 μm , respectively) were employed in the experiments. Distilled water and kerosene oil were used as the wetting and droplet/slug liquids, respectively. The capillary tube was first filled with water, and then the kerosene oil was injected into the tube to form the test slugs. A DV camera was used to capture the fringe pattern, and the data were transferred to image frames with a SONY Video Walkman (GV-D900E PAL). Finally, a highly accurate signal processing technique [14] and an improved fast Fourier transform algorithm were used to determine the spatial frequency of the fringe pattern. By assuming that the droplet/slug is rotationally symmetric about its centreline, the film thickness of the liquid slug can be obtained.

3. Theoretical approach

Because total reflection is not possible when the ray is incident from water to kerosene, other optical orientations must be considered. Considering the refracted ray shown in figure 2, the angle β of the scattering ray refracted by a liquid/liquid slug inside a capillary glass tube can be expressed as

$$\beta = 2\theta + 2(\varphi - \theta_r) + 2(\alpha - \varphi_r) - 2\alpha_r. \quad (1)$$

Figure 2. Schematics of geometrical optics of a scattered ray by a liquid-liquid slug system.

According to the geometrical optics of light refraction and the geometrical relationships as shown in figure 2, we can have

$$\begin{aligned}
 n_{l_3} \sin \theta &= n_g \sin \theta_r \\
 \frac{R}{\sin \theta_r} &= \frac{R + h_1}{\sin \varphi} \\
 n_g \sin \varphi &= n_{l_2} \sin \varphi_r \\
 \frac{R - h}{\sin \varphi_r} &= \frac{R}{\sin \alpha} \\
 n_{l_2} \sin \alpha &= n_{l_1} \sin \alpha_r
 \end{aligned} \quad (2)$$

where n_{l_1} , n_{l_2} , n_{l_3} and n_g are the refractive indices of the slug liquid, wetting liquid, tube glass and matching liquid, respectively. R is the inner radius of the capillary, h is the liquid film thickness and h_1 is the glass wall thickness. Equation (2) also can be rewritten as

$$\begin{aligned}
 \theta_r &= \arcsin \left(\frac{n_{l_3}}{n_g} \sin \theta \right) \\
 \varphi &= \arcsin \left(\frac{R + h_1}{R} \frac{n_{l_3}}{n_g} \sin \theta \right) \\
 \varphi_r &= \arcsin \left(\frac{R + h_1}{R} \frac{n_{l_3}}{n_{l_2}} \sin \theta \right) \\
 \alpha &= \arcsin \left(\frac{R + h_1}{R - h} \frac{n_{l_3}}{n_{l_2}} \sin \theta \right) \\
 \alpha_r &= \arcsin \left(\frac{R + h_1}{R - h} \frac{n_{l_3}}{n_{l_1}} \sin \theta \right)
 \end{aligned} \quad (3)$$

while

$$d\beta = 2 d\theta + 2(d\varphi - d\theta_r) + 2(d\alpha - d\varphi_r) - 2 d\alpha_r. \quad (4)$$

Let $c_1 = n_{l_3}/n_g$, $c_2 = n_{l_3}/n_{l_2}$, $c_3 = n_{l_3}/n_{l_1}$, the differential form of equation (3) will be

Figure 3. Relation between imaged fringe space and viewing angle.

$$\begin{aligned}
 d\theta_r &= \frac{c_1 \cos \theta}{\sqrt{1 - c_1^2 \sin^2 \theta}} d\theta \\
 d\varphi &= \frac{c_1 \cos \theta}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_1^2 \sin^2 \theta}} d\theta \\
 d\varphi_r &= \frac{c_2 \cos \theta}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_2^2 \sin^2 \theta}} d\theta \\
 d\alpha &= \frac{c_2 \cos \theta}{\sqrt{\frac{(R-h)^2}{(R+h_1)^2} - c_2^2 \sin^2 \theta}} d\theta \\
 d\alpha_r &= \frac{c_3 \cos \theta}{\sqrt{\frac{(R-h)^2}{(R+h_1)^2} - c_3^2 \sin^2 \theta}} d\theta \\
 d\theta &= \frac{\delta}{(R + h_1) \cos \theta}
 \end{aligned} \quad (5)$$

By considering that the incident fringe rays are scattered and projected onto a far field screen as shown in figure 3, the following relation can be derived:

$$\tan(\Delta\beta) \approx \frac{\Delta x + \delta}{L} \quad (6)$$

Figure 4. Schematics of geometrical optics of different scattered rays.

Figure 5. Analysis of receiving angles with different scattering rays.

where δ and Δx are the fringe spaces of the incident and the received light, respectively. L is the distance between the capillary tube and the image screen. Since $L \gg \Delta x$ and $\Delta x \gg \delta$, equation (6) can be rewritten as

$$\Delta\beta \approx \frac{\Delta x}{L}. \quad (7)$$

Let $f = \frac{1}{\Delta x}$; equation (4) can be rewritten as

$$\frac{1}{Lf} \approx d\beta = \frac{2\delta}{R+h_1} \left[\frac{1}{\cos\theta} + \frac{c_1}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_1^2 \sin^2\theta}} - \frac{c_1}{\sqrt{1 - c_1^2 \sin^2\theta}} + \frac{c_2}{\sqrt{\frac{(R-h)^2}{(R+h_1)^2} - c_2^2 \sin^2\theta}} - \frac{c_2}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_2^2 \sin^2\theta}} - \frac{c_3}{\sqrt{\frac{(R-h)^2}{(R+h_1)^2} - c_3^2 \sin^2\theta}} \right]. \quad (8)$$

There are two ways to determine the film thickness, one is to directly measure the value of L according to the experiment set-up, and the other is to regard the slug full filled case (no

Figure 6. The relationship between film thickness h and the relative frequency f/f_0 .

wetting liquid film) as the reference one. In the latter way, i.e. $h = 0$, the following equation can be derived:

$$\frac{1}{Lf_0} \approx d\beta_0 = \frac{2\delta}{R+h_1} \left[\frac{1}{\cos\theta} + \frac{c_1}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_1^2 \sin^2\theta}} - \frac{c_1}{\sqrt{1 - c_1^2 \sin^2\theta}} - \frac{c_3}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_3^2 \sin^2\theta}} \right] \quad (9)$$

where f_0 is the spatial frequency of the received fringes related to the slug full filled case. Dividing equation (8) by equation (9) on both sides, we can obtain

$$h = R - c_2(R+h_1) \sqrt{\sin^2\theta + \frac{1}{[A \frac{f_0}{f} - B]^2}} \quad (10)$$

where

$$A = \frac{1}{\cos\theta} + \frac{c_1}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_1^2 \sin^2\theta}} - \frac{c_1}{\sqrt{1 - c_1^2 \sin^2\theta}} - \frac{c_3}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_3^2 \sin^2\theta}}$$

Figure 7. Raw data and typical FFT results related to kerosene oil full filled case.

Figure 8. Raw data and typical FFT results related to kerosene oil slug immersed in water.

$$B = \frac{1}{\cos \theta} + \frac{c_1}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_1^2 \sin^2 \theta}} - \frac{c_1}{\sqrt{1 - c_1^2 \sin^2 \theta}} - \frac{c_2}{\sqrt{\frac{R^2}{(R+h_1)^2} - c_2^2 \sin^2 \theta}} - \frac{c_3}{\sqrt{\frac{(R-h)^2}{(R+h_1)^2} - c_3^2 \sin^2 \theta}}.$$

The relationship between h and the relative frequency, $\frac{f_0}{f}$, can be numerically solved if the incident angle θ can be eliminated from equation (10). This can be done by substituting equations (2) and (3) into equation (1) if the receiving angle β can be pre-determined.

There are several considerations in selecting the receiving angle β . Because unlike the previous method [9], there will be no total reflection angle available under the condition for relative refractive index greater than 1 ($m > 1$, in this paper, the relative refractive index is defined as the ratio of refractive indices between the slug/droplet and the film), e.g., for kerosene surrounded by water. Furthermore, the previously used matching liquid method for air-liquid plug ($m < 1$) [9] is also not applicable because the scattering angle will be too close to the diffraction location which causes poor signal-to-noise ratio. Therefore, no matching liquid has been proposed in the experiments and all the refractive/reflective rays must be considered in the determination of β . Figure 4 shows the possible scattering ray paths of measurement schematics. To have high signal-to-noise ratio in the measurements, it is anticipated that only ray 1 should be detected in the direction of the correct receiving angle. To do that, the scattering angles for

all the rays have been calculated on the basis of the geometrical optics approach and are shown in figure 5. From figure 5, we can see that there is a region of β between 20.4° and 34.9° where only ray 1 exists. Furthermore, for the $h = 0$ case, i.e. for film thickness zero, the largest scattering angle β is about 25° . To be able to measure very thin interfacial film thickness, very close to zero such as $0.1 \mu\text{m}$ or thinner, the selected β should be less than 25° . Therefore, 24° receiving angle was selected according to the above analysis. The relationship between h and the relative frequency, $\frac{f_0}{f}$, can then be numerically determined from equations (1)–(3) and (10) as shown in figure 6. Thus, with the experimental data of $\frac{f_0}{f}$, the practical thickness of a liquid film can be easily obtained.

4. Experimental results

In order to demonstrate the capability of this newly developed method for measurement of immiscible liquid-liquid interfacial film thickness, a validating experiment was performed with a kerosene oil slug immersed in water and arranged to transport vertically along a micro tube (see figure 1). Two sizes of borosilicate glass tubes were considered in the experiments, one with inner diameter $750 \mu\text{m}$ and outer diameter $1500 \mu\text{m}$, respectively. The other smaller tube used had inner diameter $500 \mu\text{m}$ and outer diameter $1000 \mu\text{m}$ to compare the diameter effect. Table 1 gives the constants and parameters employed in this work.

Figure 9. Liquid film thickness measured with moving velocity at $V = 1.169 \text{ m s}^{-1}$.

Figure 10. Liquid film thickness measured with moving velocity at $V = 0.709 \text{ m s}^{-1}$.

Table 1. Constants and parameters.

$\delta \text{ (}\mu\text{m)}$	$R \text{ (}\mu\text{m)}$	n_{l1}	n_{l2}	n_{l3}	n_g	m	$\beta \text{ (deg)}$
2.531	375, 250	1.448	1.333	1.0	1.473	1.086	24

By utilizing the FFT and five point fitting method [14], the spatial frequency, f , can be accurately determined with the captured fringe patterns. In order to reduce the noise of background, the image was pre-processed by averaging the grey level along each column. The individual values were then subtracted by their mean values to eliminate the bright background. After that, this data array was processed by 1D-FFT and the related spatial frequency was determined (see figures 7 and 8). Figure 7 shows the raw data and FFT results obtained with the fringe pattern when the kerosene oil was fully filled with no water film surrounding. Therefore, this case thus was used to determine the reference value of f_0 . Figure 8 shows similar data and results related to a kerosene oil slug surrounded with a water film.

Figures 9 and 10 give the results of film thickness measured at different slug/droplet velocities along the capillary tube with $750 \mu\text{m}$ inner diameter. The slug velocity V was measured using the correlation between two consecutive images recorded by CCD camera. The measured average interfacial film thickness for $V = 1.169 \text{ m s}^{-1}$ and $V = 0.709 \text{ m s}^{-1}$ are $1.5 \mu\text{m}$ and $2.2 \mu\text{m}$, respectively. It is interesting to know that, from the measurement results of

Figure 11. Liquid film thickness measured with slug velocity $V = 4.29 \text{ m s}^{-1}$.

Figure 12. Liquid film thickness measured with slug velocity $V = 2.62 \text{ m s}^{-1}$.

Figure 13. Liquid film thickness measured with water slug surrounded by kerosene oil.

figures 9 and 10, the higher the slug velocity, the thinner the surrounding liquid film. Figures 11 and 12 show similar results for a smaller capillary tube with inner diameter $500 \mu\text{m}$. It seems that the smaller the diameter of the tube, the thicker the interfacial film thickness of the surrounding water. For both cases, the standard deviation of the measured interfacial film thickness is about $0.15 \mu\text{m}$.

Finally, to demonstrate the measurement capability of the method for water immersed in kerosene ($m < 1$), a measurement was also presented as shown in figure 13. In this measurement, total reflective light was used to determine the film thickness as described in [9]. The water slug was in a steady state and the probe was scanning along the slug surface. The measurement results show a much thicker interfacial film thickness when water is surrounded by kerosene in a glass capillary tube. This may be explained by the glass surface energy, the surface tensions of liquids and the interfacial force between the liquids and glass interface.

5. Conclusions

A novel optical method has been further extended to measure the interfacial film thickness of immiscible liquids of a slug/droplet formed inside a capillary tube. The selection of optimized receiving angle has also been proposed. Both the principles and validation experiments have been described in detail in this paper. The validation experiments were conducted with kerosene oil slugs immersed in water and water slugs immersed in kerosene oil. The experimental results show that this newly developed method can obtain the liquid film thickness with high resolution. This diagnostic method is easy to implement and it will have great potential for immiscible liquid flow research in microfluidic channels.

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