Interfacial Dynamics of Immiscible Liquid-Liquid Displacement and Breakup in a Capillary Tube

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Abstract The interfacial liquid film dynamics of two immiscible fluids in a micropipe is of interest in microfluidics and two-phase flows. Liquid film displacement is essential for fluid dynamics, heat and mass transfer. Understanding interfacial dynamics and liquid film thickness in two immiscible fluids is important in two-phase flow research because the interfacial area of fluid film and its thickness have a significant impact on the heat and mass transfer processes. However, the processes of liquid film formation, development and breakup are complex which involve the interfacial tension between fluid-fluid and fluid-solid, the beating process of fluid dynamics, the thermal effect and the capillary induced instability of thin liquid film breakup. As a consequence of the small length scale, the capillary forces play a fundamental role in the physics of the phenomena. For the high surface area and volume ratio will affects the evaporation of the film fluid. In this paper, annual type thin water film displacement, velocity field and breakup in a two immiscible fluid system in a capillary tube are presented. A novel method for measuring the interfacial liquid film thickness between immiscible liquids of annual oil liquid and an oil slug/droplet (oil-water-oil) in a capillary tube is proposed. A water plug/droplet was introduced into kerosene oil and pushed forward under a constant pressure. Water droplet adhesion to the glass capillary wall was observed and the dynamic changes of interfacial liquid film thickness were visualized and measured against different capillary numbers by using light absorption techniques utilizing high speed CCD camera and Beer-Lambert law. The mean film propagation velocity was measured by micro resolution particle image velocimetry (μPIV). The experimental results show the film thickness against the capillary number at certain cross-section obeys a power law. The measured velocity can also be used to demonstrate the liquid film break up processes. Furthermore, a ripple pattern film thickness was presented when a long annual water film was formed.

1. Introduction

Interfacial dynamics and liquid film thickness in two immiscible fluids is important in two-phase flow research (Coney 1973). An important application is in oil recovery from porous media, where the liquid film of the displaced oil that remains attached to the rocks has significance in the efficiency of the recovery during water-flooding operation (Edson et al. 2009). However, the processes of liquid film formation, development and breakup are complex which involve the interfacial tension between fluid-fluid and fluid-solid, the beating process of fluid dynamics, the thermal effect and the capillary induced instability of thin liquid film breakup. As a consequence of the small length scale, the capillary forces play a fundamental role in the physics of the phenomena. The relationship between liquid film thickness and the flow boiling heat transfer in microscales has been studied (Kenning et al. 2006 and Thome et al. 2004). Taylor (1961) first gives the mean liquid film thickness remaining on the wall by measuring the difference of bubble velocity and the mean velocity. About the heat transfer characteristics, Wang and Qiu (2005) and Qiu and Wang (2005) developed novel optical fringe probing techniques for film thickness measurements in capillary tubes (Wang and Qiu 2005, Qiu et al. 2005). Utaka (2006) measured the liquid film thickness by a vapor bubble in a narrow gap mini-channel and the heat transfer phenomena base on it.

For the thin film blocking the object or heat transfer, the typical ones happened in the membraneless microfluidic fuel cells. There will be a depletion fluid film area attached on the cathode and anode. The depletion film thickness dominates the performance of microfuel cell by Chang et al. (2006). Choban et al. (2004) gives the data about the film thickness and the different concentration of fuel in micro fluidic fuel cells. And, Lee et al. (2007) optimize the microfluidic fuel cell parameters to minimize the affection of the
Different methods to measurement the thin film thickness base on different physical phenomena have been constantly proposed. Normally, these methods can divide into 3 groups: acoustic method, electrical methods and optical methods. Lu et al. (1993) measured liquid film thickness of R113 and FC-72 inside a horizontal rectangular channel. By measure the different arriving time of echo ultrasonic waves by different interfacial reflection, the film thickness can be calculated. Wang et al. (2012) present an analytical solution to estimate the film thickness in two-phase annular flow utilizing electrical resistance tomography. The resistances were measurement after they fixed two electrodes on the pipe wall. Both the relationship between resistance and water film thickness for concentric case and eccentric case were considered to determine the film thickness. For optical methods, interference and pixel counting are two common ways. Ji and Qiu (2009) presented an interference fringe film thickness measurement method by 2D spatial fringe scattering technique. They utilize the relationship between film thickness and the spatial frequency of scattered fringes. Steinbrenner et al. (2007) measurements the thin film thickness in air-fluid two phase flow by counting pixel number after seeding fluorescence molecule into the water.

Recently, some researchers have used tracer particles seeded into liquid phase to enhance the detection of two phase flow interface (Oriol 2007). At the same time, PIV or Micro-PIV can be applied to examine the velocity distribution in the liquid phase. The velocity field in the liquid phase has strong relationship with the film thickness by Fouilland (2009). Tian and Qiu (2002) developed a method for eliminating background noise in micro resolution PIV measurements.

In this paper, we present a new thin film generation method happened in fluid-fluid two phase fluid in a capillary. And we give another optical method to measurement the thin film thickness. At the same time, by seeding particles in one liquid phase, the velocity field inside the film calculated by PIV. Because the PIV method is not a high precision method for thickness measurement, we combine these two to a new measurement method to give the thin film thickness and inter velocity filed at same time.

2. Experimental Setup

Working Principle

The phenomenon happened when Kerosene (continue phase) and DI water (dispersed phase) two phase flow in the capillary. The typical figure is shown in Figure 1.

![Figure 1 Liquid film formation (a) Initial condition, (b) Film formation, (c) Film breakup](image)

The dark rectangle is the injected DI water droplet dyed with blue ink. The droplet will attach completely on the capillary wall in several seconds. Once the compressed air pushes kerosene into the capillary, the droplet will move forward. At the same time, the attached droplet part is still binding on the wall. Then a thin film will be generated to connect the droplet body and the attached part. After several milliseconds, the thin film
will be break up at the end. In the drawing process, the initial attached part maybe removes with the kerosene flow, as shown in Figure 1. But this short distance remove won’t affect the thickness distribution in the droplet’s tail.

The droplet with the most volume of initial one is defined by ‘Droplet Body’; the thin film behind the droplet body is defined by ‘Droplet Tail’ in this thesis; the initial attached position with some water residual is defined by ‘Residual’. The film thickness has relationship with several factors, such as the press in the capillary, the initial volume of droplet, the viscosity of droplet. As shown in Figure 2, the water droplet is extended like a spring in the film generation (several miliseconds). The cross-section at A is a 3 layer flow with one layer water and 2 layers kerosene. The kerosene layer (attached on the wall) is similar with the films in slug flow. The reason for the water film generation is because the surface intension and the press given by the kerosene.

![Diagram of droplet and film](image)

**Figure 2** The schematic cross sections of liquid film in a capillary tube

The interfacial area of water and kerosene is enlarged by the thin film. If we replace water and kerosene into two immiscible reactants, the reaction of these two fluids will be accelerated when the thin film generated. The film thickness will affect one type reactant’s mass. And, the water thin film will change the equivalent heat coefficient of the two phase flow.

To figure out the thin film thickness, we designed an experiment system. As the inter velocity field in the film will affect the thickness, velocity filed inside the film is also measurement at the same time.

### Experimental Method

The experiment contains several parts: driver module, droplet delivery module, control module, capillary, light sources, high speed cameras and disposal bottle. The schematic diagram is shown in Figure 3. The driver module has a compressed air source and a fluid bottle. The 1L fluid bottle could keep the pressure, from the compressed air source, to a steady one to drive fluid into the capillary. A normally closed solenoid valve (SMC, VX2130A-02-5G1) is utilized to connect the driver module and the delivery module. The delivery module is consists by 2 syringes (HAMILTON, 87919#). Syringe 1 is in charge of supporting marked ink for the experiment. And, by syringe 2, the droplet could be delivered to an accurate position. The tank (50*40*200mm, PMMA) is full of glycerol with concentration 80% to match the refractive index of capillary glass (m=1.47). The capillary (Inter Diameter: 0.86mm, Length: 150mm) is injected into the tank. At the end of capillary, a proposal bottle is set to collect all the proposal fluid. The system employs two image capturing high speed cameras. The No.1 high speed camera (REDLAKE, MOTIONXTRA HG-100K) will record the flowing video by 3000ps in 800*1128 pixels with a 650W lamp (ARRILITE 600). The lamp is set at the same side with the camera facing the capillary from above. To enhance the contrast, a white diffuse reflector is set at the bottom of tank. The angle between lamp and the vertical line is about 30°. To simple the high speed camera tripod, a front-slivered mirror is set on the top of tank to reflect the light to
No.1 high speed camera, which is set horizontally. The capillary, the high speed camera and the lamp is kept on the same vertical plane. The No.2 high speed camera (Phantom v7.3) will record the flowing video by 24000ps in 1200*900 pixels with a 300W*8 quartz halogen light (PALLITE VIII). The quartz halogen light is set at the different side with No.2 high speed camera facing the capillary. The angle between the camera and the light is about 30°. And, another white diffuse reflector is set at the light side, too.

The two image capturing systems locate on different two planes with angle 90° to minimize the interaction. The video taken by No.1 system is utilized to measure the film thickness, and the video by No.2 system is used to calculate the velocity field during the film.

![Experimental setup of measurement facility](image)

Figure 3 Experimental setup of measurement facility

Two kinds of fluid are utilized in the capillary: Kerosene and water marked by blue ink (Parker, QUINK) with concentration 10% and particles (DANTEC, 9080R6011) 0.012g/mL. The blue water with particle is the dispersed phase, and the kerosene is the continue phase in the experiment when flowing. To measure the film thickness, Beer–Lambert law is taken. The normal equation is like this:

\[ I = I_0e^{-\alpha l c} \quad (1) \]

where, \( \alpha \) is the attenuation coefficient; \( l \) is the adsorption length; \( c \) is the concentration of adsorption material.

The most common utilization Beer-Lambert law is to measurement the concentration of liquid solutions (Liu et al. 2005). Researchers fill a transparent tank with known length with that kind of solution, and calculate the concentration. In this experiment, the concentration of blue ink is fixed, we measurement the light intensities to calculate the adsorption length, which is the film thickness. If all the parameters can be measurement in the Beer-Lambert Law, however, in this experiment, we did not measure the exactly value of each parameter. The concentration and the absorption coefficient of blue ink are not important. The
calculate method is to take the initial darkest point (which adsorption length is the capillary diameter, 0.86mm) as reference substance, and try to find the ratio of interesting point thickness and capillary diameter. The final equation is like this:

\[
\frac{I_1}{I_2} = \ln\left(\frac{I_1}{I_0}\right)/\ln\left(\frac{I_2}{I_0}\right)
\]  

(2)

In which \(I_1\) is the interesting point thickness, \(I_2\) is the capillary diameter, \(I_0\) is the light intensity before inject into the interesting point, \(I_1\) is the light intensity after inject into the interesting point, \(I_2\) is the light intensity before inject into the darkest point, \(I_2\) is the light intensity after inject into the darkest point.

Actually, the light intensity before inject into the capillary is difficult to measure, so we take the average light intensity outside the capillary as the intensity before inject into the capillary in the video frame. At the same time, we can minimum the other parameters’ effect for the thickness measurement. After reading the four light intensities from the video and calculating, the thickness will be got. Because the droplet tail is a long ring which has the same axis with the capillary, the film thickness is not the values calculated directly by the pixels of the video frame.

As shown in Figure 4, after projecting to the CMOS of the high speed camera, the detected film thickness has some relationship with the real one. The shadow part is the cross section of droplet tail (film); \(R\) is the out-diameter; \(r\) is the inter-diameter; \(\alpha\) and \(\beta\) are the angle which original side is the horizontal line. From this figure, the real thickness of the film is the one located at the 0 degree. To deduce the error, we try to utilize more date to calculate the real film thickness. So, the function of detected thickness and real film thickness is given:

\[
I_d = \begin{cases} 
R \cos(\theta) & |\theta| \geq \arcsin\left(\frac{R-l}{R}\right) \\
R \cos(\theta) - \sqrt{(R-l)^2 - (R \sin(\theta))^2} & |\theta| < \arcsin\left(\frac{R-l}{R}\right) 
\end{cases}
\]

(3)

In which, \(I_d\) is the detected thickness; \(l_r\) is the real thickness; \(R\) is the out-diameter; \(\theta\) is the angle of original side and detected point. By this equation, we could take all the detected data to calculate. In this method, the refractive occurring on the interface of different fluid is neglected. Although the different between refractive of kerosene (\(m=1.44\)) and water (\(m=1.33\)) is small (0.11), the error of sampled method will be there. The real experiment data and the calculation base on this method can fit each other very well after comparison. This error can be neglected.

![Figure 4 Relation of projected thickness and angle at the capillary cross-section](image-url)
As shown in Figure 5(a), the real film thickness value is given. Figure 5(b) is the same time video frame. Because this frame size of video is 1504*100, the total axial pixel number is 1504. The typical thickness of thin film is about 20 µm. The peak value is the droplet body. As shown in Figure 5(b), the size of droplet is shrinking in radial direction, so the value is not exactly the capillary radius, about 0.43mm in Figure 5(a).

![Figure 5](image)

Figure 5 Measurement results (a) visualization of liquid film formation, (b) measured film thickness

**Velocity Field Measurements**

To measure the velocity field of the film, LaVision PIV is used. Normally, particles are needed for the PIV calculation. In this thesis, hollow glass sphere particle with diameter 10µm is selected. The particle density is about 1.1g/cm³, which means the particle could be steady disperse in water. The mass concentration of particle is about 0.012g/ml; the volume concentration of particle is about 1%. According to the thickness measurement data, the typical thickness of film is during 10-50µm. The minimum surface density of particle is 1/10000 µm², which means that there is at least one particle in a 0.1mm length square. As the inter-diameter of capillary is 0.86mm, about 8-9 particles will be located at the vertical cross-section line. Considering the film layer is 2 on the video frame, about 17 particles can be detected during that line. Even though there will be some particles loss in the solution preparation and the particles will deposit as the density 1.1g/cm³, the particle number in the vertical cross-section line will not less than 10. The lens' magnification for the velocity field measurement high speed camera is 1:1. At the best situation, an object
with length 0.02mm will form on the camera CCD as one pixel. That means the whole capillary inter-diameter is consists with 34 pixels. There is one particle bright spot in each 3 pixels. This is enough for the PIV calculation.

To predict the light intensity refracted by particles, Mie theory is taken as the calculating algorithm. The particle diameter is 10µm. Other parameters needed in the calculating are shown in Table 1:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intensity Incident Light</td>
<td>2400W/m²²</td>
</tr>
<tr>
<td>Wavelength</td>
<td>550 nm</td>
</tr>
<tr>
<td>Particle Diameter</td>
<td>10 µm</td>
</tr>
<tr>
<td>Refractive Index: Particle</td>
<td>1.52</td>
</tr>
<tr>
<td>Refractive Index: medium</td>
<td>1.33</td>
</tr>
<tr>
<td>Absorption Coefficient</td>
<td>0</td>
</tr>
<tr>
<td>Elevation angle</td>
<td>0°</td>
</tr>
<tr>
<td>Off-axis angle</td>
<td>10°-180°</td>
</tr>
<tr>
<td>Receiving cone half angle</td>
<td>1°</td>
</tr>
</tbody>
</table>

The calculated results of off-axis angle and refractive light intensity are shown in Figure 6. The forward scattering is much larger than the back scattering. The refractive light intensity is decreasing with the increasing off-axis angle. The calculation indicates that the high speed camera should put on the different side of capillary with the quartz halogen light, and the angle with light transfer should be as small as possible unless the background light affects the refractive light. The final angle of high speed camera and the quartz halogen light is set as 30°.

The typical film formation and breakup processes are shown in Figure 7. The CCD size of No.2 high speed camera is smaller than No.1 high speed camera. So the video frame length is smaller. The whole video frame is 22.76*30.35mm. When the droplet tail break, the whole droplet figure, droplet body and droplet tail, can not be shown at the same time. Because the break is more important, the view field is set on the initial droplet stay position. By changing the magnification of lens to 1:0.85, the resolutions of two high speed cameras are the same that each pixel of video frame is 0.02mm. The darkest area in the frame is the droplet. The bright spots located at the left side are the particles which deposit during the droplet delivery. The particles in the droplet tail could be seen clearly, which enables the PIV measurements.
The interrogation window is 16*16 and 12*12 in PIV calculation. The coordinate system takes the top of the inner tube of the capillary as y zero point, the leftmost pixel in the frame as x zero point. Each velocity value belongs to the original position point in the frame. The velocity field is 0 where is no droplet body and droplet tail. At the head of the droplet body, the speed is quite high. At the droplet body center, the speed is almost 0. This is because the particle is bright spot, the same with the droplet body head. However, in the droplet body center, the fluid thickness is about 0.8mm, which is much thicker than the one of droplet tail, 0.01mm. The first row and the last velocity speed is less than the ones in the middle. It proves that the viscosity is constant in Newton’s fluid. The rightmost velocities are not correct because the particles will move out of the view field. So we just take the middle ones.

The droplet tail thickness and its velocity field could be measure respectively by the two videos. To unit they together, time synchronization and space synchronization are essential problem. The resolutions of video are modified the same as mentioned above. After set the original position as the same, the space synchronization will be done. We set it by comparing the dark points of the initial droplet. As the time synchronization, the two video shooting speeds are different, 3000fps (thickness) and 24000fps (velocity field). Theoretically, every one frame taken for the thickness will be corresponds to 3 frames taken for the velocity field.

The united thickness and velocity field examples are below as shown in Figure 8. The time zero frame is defined when the droplet is start to move. The time of the 3 figures are: 6ms, 13.29ms and 30.24ms.

3. Results and Discussions

To find the relationship between the film thickness and capillary number, we do the experiment with compressed air pressure: 20Kpa and 27.5Kpa; the volumes of initial droplets: 1.0μl and 1.5μl. In the data analysis, we take several cross sections on the film tail. We define the original cross section is the section which one’s mean calculated thickness is the first 5% of the initial droplet thickness from the droplet body to the droplet tail. The cross section interval unit is the capillary diameter. We named the original cross section as “0” and the others as natural number from 1 to n (as shown in Figure 2). In this way, the whole interesting area in the capillary could be divided into 3 parts: the droplet body, the droplet tail, the residual. The droplet tail with capillary diameter axis is the research object.

The measured film thickness at n=1 is shown in Figure 9. The measured film thickness shows a polynomial tendency against the capillary number. The polynomial fitting method is of nonlinear least square method with equation $\text{Thickness (Ca)} = a \cdot C_a^b + c$, which is support by Matlab Curve Fitting Toolbox.

In Figure 9, the equation $\text{Thickness (Ca)} = a \cdot C_a^b + c$ can describe the relationship between speed and thickness perfectly. The initial point of schematic is taken from the film is generated; the final point of schematic is taken just before the film in this cross section is break up.
Figure 8: Instant water film thickness and mean velocity field inside of liquid film.
Figure 9 Measured water film thickness vs. capillary number (20Kpa, 1.5ul, at n=1 cross section)

The film thickness vs. capillary number at different locations is shown in Figure 10.

Figure 10 Measured water film thicknesses vs. capillary number at different locations

The initial thicknesses of different cross section are different: nearer the droplet body, thicker the film. The final thicknesses of different cross section are different, too: nearer the droplet body, thinner the film. It is found that there is a capillary number where the thicknesses of different cross sections are same.

**Capillary wave induced ripple form liquid film**
When the length of droplet tail is longer than 30 mm under the pressure 20kPa to 33kPa, there will be some varicose-like instability generated at the middle of the deposited film as shown in Figure 11. The thickness range of the varicose-like film thickness is from several micrometers to forty micrometers. The reason for the varicose-like film deposition is caused by Rayleigh instability (Rayleigh 1902). The wavelength $\lambda$ in Figure 11 is found to be about $\lambda/R_i=6$, where $R_i$ is the radius of the liquid film.

![Diagram of capillary wave induced ripple form liquid film](image)

**Figure 11** The diagram of capillary wave induced ripple form liquid film

### 4. Conclusions

In the present work, a new phenomenon of droplet film generation is described. An experiment system is established to measurement the film thickness and the inter-liquid velocity field. By Beer-Lambert law and PIV, each point thickness and inter velocity in the film can be calculated. Analysis is given to describe the relationship of film thickness in fixed cross-section and the velocity. The velocity field before and after film...
break up is discussed, too. The break up can be predict according to the unusual change happened in velocity field. The phenomenon of varicose-like film caused by Rayleigh instability is visualized.

Acknowledgements

The work described in this paper was supported by a grant from the Research Grants Council (RGC) of the Hong Kong Special Administration Region, China (Project Nos. 618210 and 617812).

References
