TRANSIENT PHENOMENA OF DYNAMIC CONTACT ANGLE IN MICRO CAPILLARY FLOW

J. SUNG[1,c], Y.B. KIM[2], M.H. LEE[1]

[1]Department of Mechanical & Automotive Engineering, Seoul National University of Science and Technology, Seoul 139-743 Korea
[2]Graduate School of Energy & Environment, Seoul National University of Science and Technology, Seoul 139-743 Korea

[c]Corresponding author: Tel.: +8229706398; Fax: +8229491458; Email: jysung@seoultech.ac.kr

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ABSTRACT: *This study is devoted to investigate the dynamics of liquid driven by capillary force in a circular tube. A microscope was used to visualize the meniscus movement and the contact angle. The experiments were carried out with glycerin-water mixtures having viscosity ranging from 0.21 to 1.36 Pa·s by filling the test liquid in a borosilicate glass tube with an inner diameter of 200 µm. The wetting distances of the meniscus with time were compared with the theoretical solutions by considering the dynamic variation of contact angle. The results show that the theoretical solutions overestimate the wetting distance due to existence of transient motions in developing entrance region. The discrepancy increases with lower viscosity fluid which has higher velocity and experiences longer developing region.*

1 Introduction

Liquid penetrates into a narrow gap by capillary action at the gas-liquid interface. This is called a micro capillary flow and has practical importance in a wide range of technology such as flow through porous materials in micro heat pipes and micro coolers, and underfilling process in flip chip packaging [1].

The fundamental research for the micro capillary flow is the liquid penetration into a capillary tube. Washburn [2] derived a theoretical equation by considering the force balance between capillary force and viscous drag, neglecting gravitation and other effects. One of the critical drawbacks in the Washburn's equation is the assumption of constant contact angle at the flow meniscus. In practice, the contact angle does not always keep its equilibrium status but changes dynamically since the flow near the meniscus affects the contact line of the solid-gas-liquid interface [3]. Newman [4] suggested an equation for the dynamic contact angle in the capillary flows. He assumed the contact angle decreases exponentially as time goes on. At the earlier stage of the capillary action, the liquid experiences an acceleration force so that the transient term in the governing equation should be considered. Thus, Ichikawa and Satoda [5] and Ichikawa et al. [6] analyzed the dynamic interface motion for circular and rectangular microchannels. They proposed the dimensionless equations of the interface movement in the condition of wet solid surface. Despite of the big progress in measurement techniques for micro-scale flows, the transient behavior of dynamic contact angle in a capillary channel has not been discussed experimentally.

The present study focuses on the transient phenomena of the dynamic contact angle in micro capillary flows. With the help of flow visualization and image processing techniques, the wetting distance and meniscus velocity as well as dynamic contact angle are simultaneously measured under the flowing status. To see the effect of viscosity on these properties, glycerin-water mixtures with various volume fractions are filled into a circular micro tube. Then, the measured data are compared with the previous theoretical models in view of how the models describe the transient phenomena.

2 Theoretical Approaches

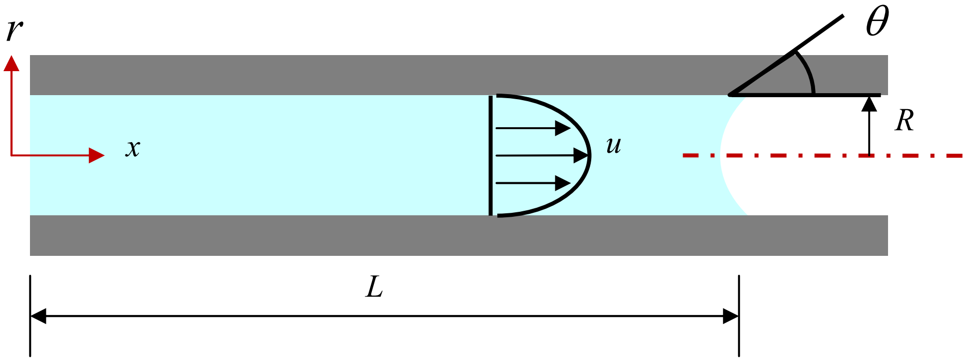


Fig. 1 Capillary-driven micro flow through a circular tube

Fig. 1 shows the schematic of the capillary-driven micro flow through a circular tube. For a Newtonian viscous liquid, if flow is laminar and incompressible, the governing equation for the unsteady flow $u(r, t)$ can be written as follows;

$$\frac{\partial u}{\partial t} = -\frac{1}{\rho}\frac{\partial p}{\partial x} + \frac{\mu}{\rho}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u}{\partial r}\right) \tag{1}$$

where μ is the fluid viscosity, ρ is the density. From the Laplace-Young equation, the driving capillary force at the meniscus is $2\sigma\cos\theta/R$ where σ is the surface tension coefficient, $\theta(t)$ is the dynamic contact angle and R is the tube radius. The pressure gradient inside the tube is assumed to be linear and the fully-developed laminar velocity profile can be used.

$$-\frac{dp}{dx} = \frac{1}{L(t)}\frac{2\sigma\cos\theta}{R} \tag{2}$$

$$u(r,t) = 2L'(t)\left(1-\frac{r^2}{R^2}\right) \tag{3}$$

where $L(t)$ is the wetting distance at time t. In most previous approaches, the transient term $\partial u/\partial t = L''$ is neglected. By substituting equations (2) and (3) for equation (1), an ordinary differential equation for the wetting distance as follows;

$$\frac{8\mu L}{\rho R^2}L' - \frac{2\sigma\cos\theta(t)}{\rho R} = 0 \tag{4}$$

If the contact angle is constant as a static contact angle θ_s, the solution for the wetting distance is the well-known Washburn equation [2]. More advanced model was proposed by Newman [4], where the contact angle decreases exponentially with time by $\cos\theta(t) = \cos\theta_s(1 - ae^{-ct})$. Here, a and c are the parameters determined experimentally.

$$\text{Washburn model}: L = \sqrt{\frac{2R\sigma\cos\theta_s}{4\mu}t} \tag{5}$$

$$\text{Newman model}: L = \sqrt{\frac{2R\sigma\cos\theta_s}{4\mu}\left(t - \frac{a}{c} + \frac{a}{c}e^{-ct}\right)} \tag{6}$$

Meanwhile, Ichikawa and Satoda [6] analyzed the interface dynamics of capillary flow in a tube considering inertial acceleration at the inlet of the tube and energy dissipation just after the meniscus. The dynamic balance of force in a horizontal tube is described as

$$(L + x)L'' + \frac{m}{2}(L')^2 + \frac{8\mu L}{\rho R^2}L' - \frac{2\sigma\cos\theta}{\rho R} = 0 \tag{7}$$

where x and m are correction factors for the inertial force and energy dissipation, respectively. Batten [7] assumed the values of $x = 7R/6$ from the inertial force in a hemispherical region of the tube inlet and $m = 3.41$ from an empirical relation. There is no analytic solution for equation (7) but it can be solved numerically using the Runge-Kutta method by considering the dynamic variation of contact angle.

3 Experimental Setup

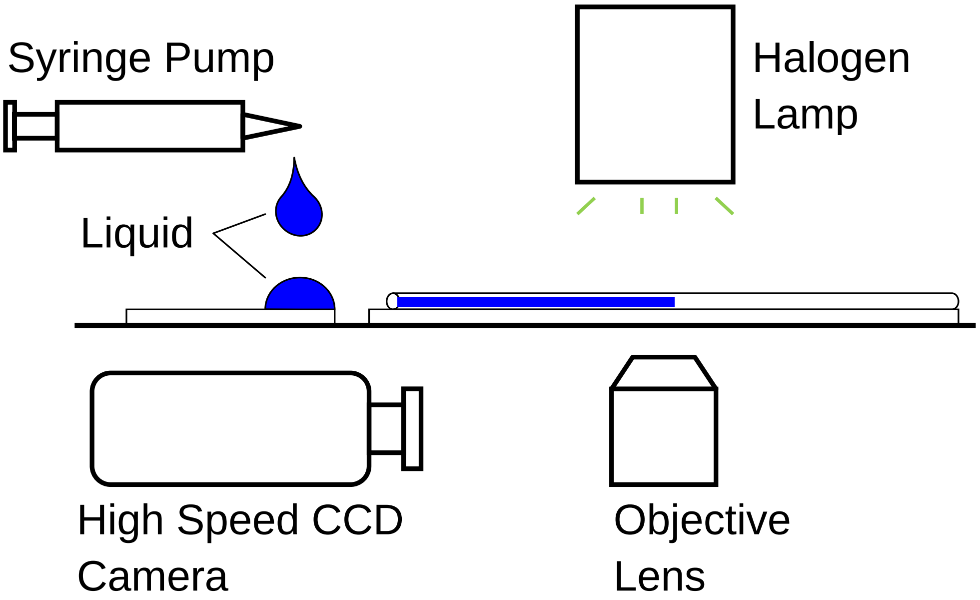


Fig. 2 Schematic of the experimental system

Fig. 2 shows the schematic of experimental setup. A drop of liquid is dispensed at the edge of a glass plate by a syringe pump and then the glass plate is attached to a capillary tube in order to fill the liquid in by only the capillary force. The motion of flow meniscus is observed using a fluorescence microscopy (Nikon, Eclipse Ti) with a 4X objective lens. Halogen lamp is used as an illuminating light source and a high-speed CCD camera (SVSI) captures the images. The capillary tube is made of borosilicate glass with an inner diameter of 200 μm. Since the contact angle is very sensitive to the surface condition, careful attention has been paid to the cleaning of the tube. In the cleaning process, the tube was kept immersed in water for 6 hours and then in a mixture of ammonium hydroxide, hydrogen peroxide and water for one hour. Again, it was immersed in water for more than 24 hours. Finally, it was dried in an electrolysis desiccator for 24 hours. By changing the volume fraction of glycerin-water mixture, the viscosity of the operating liquid is controlled. A small amount of water added to the mixture significantly changes the viscosity of the mixture. In the present study, the volume fractions of glycerin are 85.7%, 93.75%, 96.87%, 99% at room temperature 20℃, which yields the viscosity to be μ = 0.21, 0.54, 0.82, 1.36 Pa·s, respectively. The viscosity was measured by a rheometer (Brookfield DVIII+ULTRA). However, the fluid density ρ and surface tension coefficient σ are not sensitive to the small amount of added water [8][9]. Thus, ρ = 1260 kg/m^3 and σ = 0.063 N/m for all the glycerin-water mixtures. In order to measure the dynamic contact angle, the thickness T of the shadow image of the flow meniscus is obtained. If the interfacial shape is assumed to be a part of a circle, the contact angle θ can be calculated by $\theta = 2\tan^{-1}\left(0.5D/T\right) - \pi/2$ [10]. The spatial resolution for the contact angle measurement is enhanced by a high magnification microscope. The scale factor in image plane is 3.0 μm/pixel. The static equilibrium contact angle θ_s is evaluated from the image when there is no fluid motion, which is 20.07° in this experiment. The dynamic contact angle, however, varies during the meniscus movement. The measurement length of the micro tube is 0 – 10 mm, so that the CCD camera cannot capture the whole field of view at one time. Thus, the measurement length is divided into 3 sub-sections and then their data are combined successively. For each case, 5 experiments are conducted to give an averaged value.

4 Result and discussion

In Fig. 3, the movement of flow meniscus in the unsteady capillary flow is visualized for the whole measurement length in case of 85.7% glycerin-water mixture. When the meniscus lies at the entrance region, the contact angle is nearly 90° but it gradually decreases as the flow goes on. Based on the visualized images in Fig. 3, the variations of contact angle for all fluid viscosities are plotted in Fig. 4. To compare the measured contact angle with the Newman model, it is non-dimensionalized by the static contact angle such as $1 - \cos\theta / \cos\theta_s = ae^{-ct}$. Here, the symbols denote the experimental data. The solid and dotted lines are least-squares fitted data to a polynomial and exponential functions, respectively. Fig. 4 shows that the non-dimensionalized contact angle has a similar trend irrespective of the fluid viscosity but does not follow the exponential function proposed by Newman. Theoretically, the contact angle at the entrance (t = 0) is 90° since the fluid is attached to the tube at the right angle. Then, the non-dimensionalized contact angle has a value of 1. The present data indicates that it approaches to 1 at t = 0. However, the decaying rate as time goes on does not has a form of the exponential function ae^{-ct}.

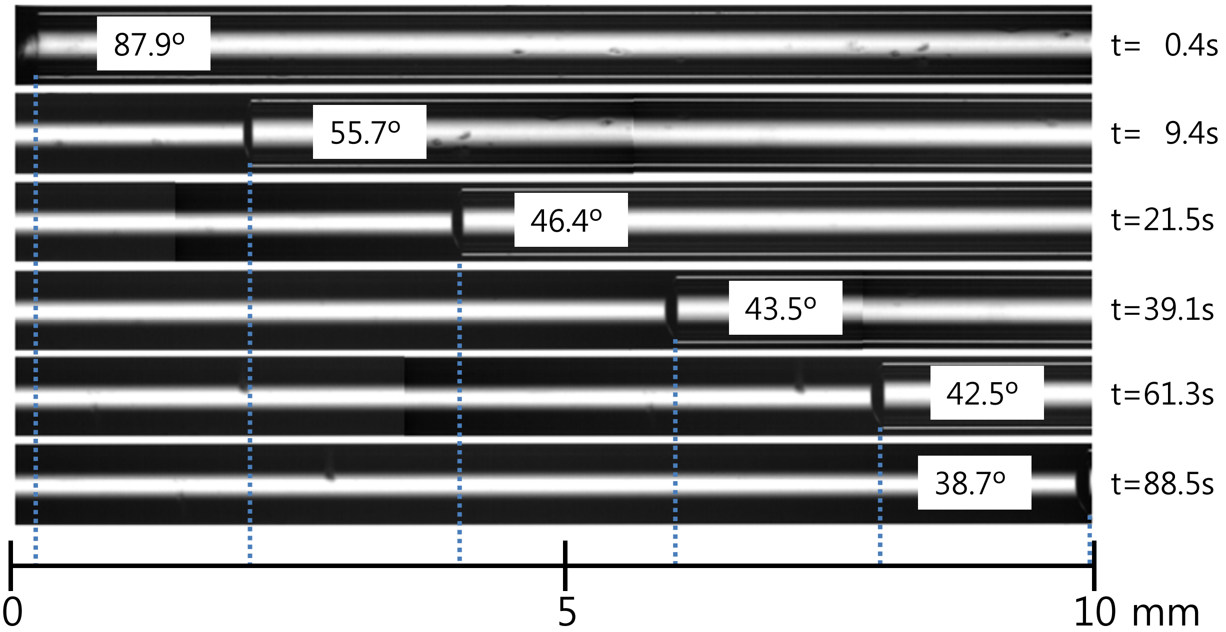


Fig. 3 Movement of flow meniscus and dynamic variation of contact angle in the unsteady capillary flow in case of 99% glycerin-water mixture

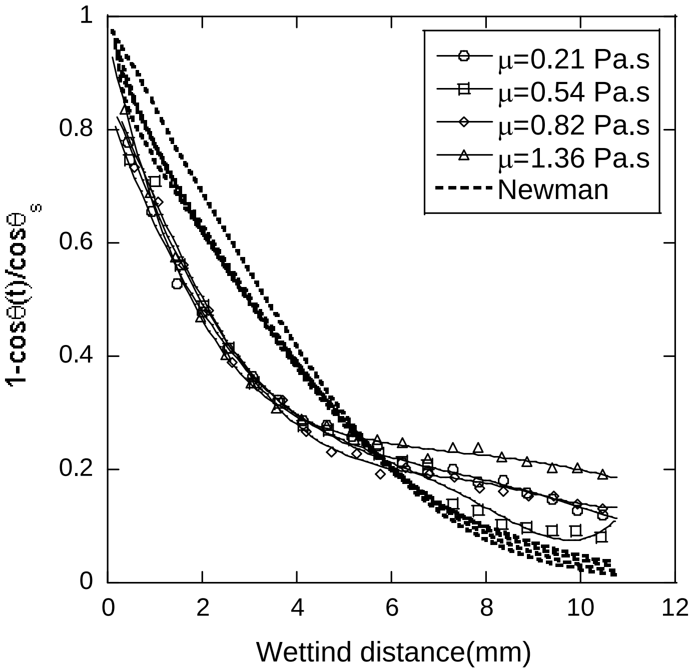
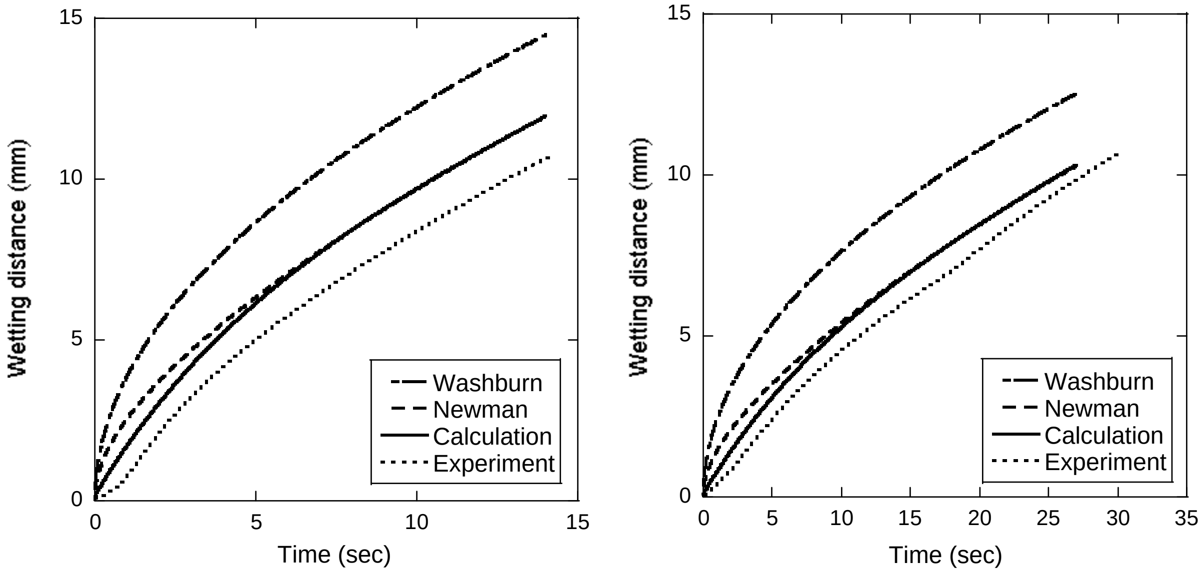


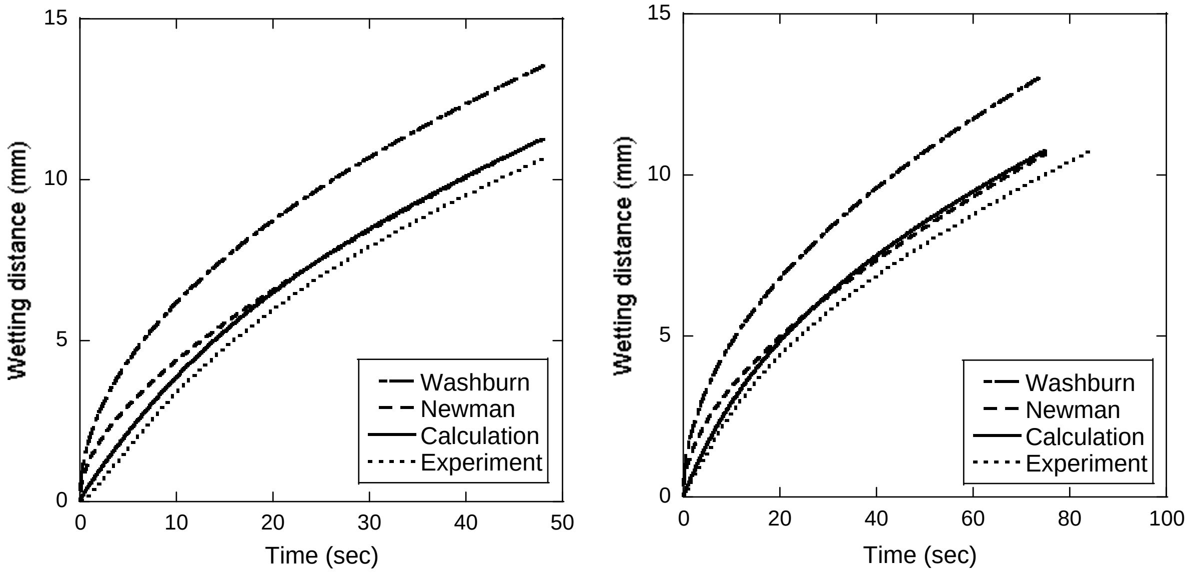
Fig. 4 Transient behavior of the non-dimensionalized contact angle for liquids with various viscosities

Fig. 5 shows the wetting distance of the flow meniscus for time. The measured data for all fluid viscosities are compared with those obtained by theoretical approaches. In the Washburn model, only the equilibrium contact angle is considered. In the Newman model, the exponential function obtained from Fig. 4 is used for the calculation of dynamic contact angle. Note that in both models, the transient term in the governing equation is neglected ($\partial u / \partial t = 0$). On the other hand, in the present calculation, the correction factors for the inertial force and energy dissipation are considered as well as the transient term. In addition, the dynamic contact angle fitted based on the polynomial function in Fig. 4 is used for the calculation. On the whole, it takes longer time for the meniscus to move a given distance as the fluid viscosity increases. However, the measured wetting distance deviate from the theoretically estimated ones. The Washburn model overestimates the wetting distance because the contact angle is assumed to have the lowest value by the equilibrium contact angle, which give rise to largest capillary force. The Newman model reflected the dynamic contact angle but it also slightly overestimates due to the neglected inertial effect at the entrance region. The present calculation estimates it more realistically than these two models. Nevertheless, it still deviates from the experimental data, especially for low viscosity fluid. The main reason seems to result from the assumption that the velocity profile is always parabolic as is in laminar pipe flow. At the entrance, there exists a developing region in velocity profile since the contact angle is nearly 90° and the velocity is uniform across the cross-section. The entrance length is longer for the lower viscosity fluid.



(a) viscosity = 0.21 Pa·s

(b) viscosity = 0.54 Pa·s



(c) viscosity = 0.82 Pa·s

(d) viscosity = 1.36 Pa·s

Fig. 5 Wetting distance of the flow meniscus for time. The measured data are compared with the theoretical solutions for liquids with various viscosities

Figure 6 shows the variation of meniscus velocity for various fluid viscosities, which are compared with the numerical solution for equation (7). The meniscus velocity is evaluated from the temporal behavior of the wetting distance. The velocity is very high at the entrance region and then it decreases gradually. As the viscosity is lower, there found higher velocity and higher decreasing rate. Since the variation of the velocity profile is not considered in the numerical solution, the results shows higher velocity than the measured ones at the entrance region. The deviation becomes more significant at lower viscosity fluid, which can explain why the calculation data in Fig. 5 overestimate the measured data. As a consequence, the developing velocity profile needs be considered in further study to estimate more accurately by the numerical solution.

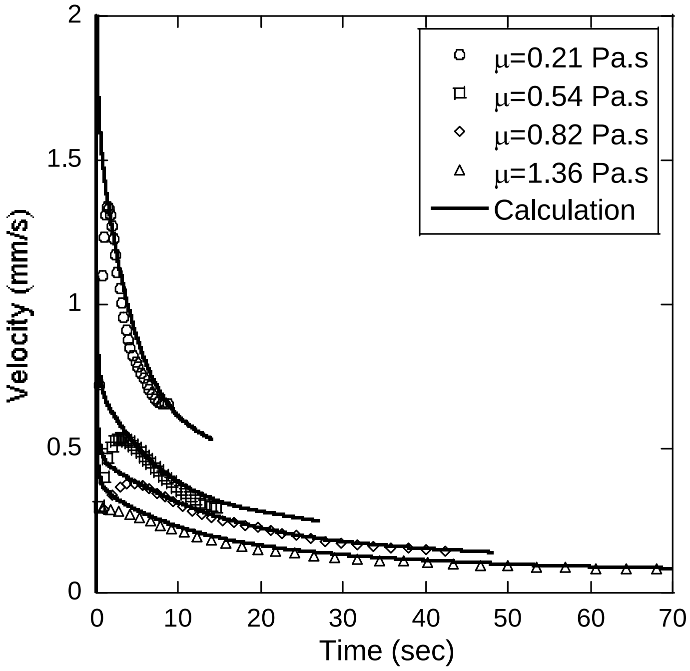


Fig. 6 Variation of meniscus velocity with time for liquids with various viscosities

5 Conclusions

The present study investigated the dynamic behavior of the micro capillary flow in a circular tube. The wetting distance, dynamic contact angle and meniscus velocity have been measured for liquids with different viscosity using a microscope. To reflect the transient phenomena near the tube inlet, flow images are acquired with a high magnification. Then, the experimental data are compared with the analytical models. The dynamic contact angle obtained by an in-situ measurement technique decreases as the meniscus marches but its variation does not follows the exponential decay as is proposed by Newman. When estimating the wetting distance by some analytical models, it is certain that the dynamic variation of contact angle significantly affect the results. The influence of dynamic contact angle increases in lower viscosity fluid. For a more realistic estimation, an unsteady governing equation which considers the entrance effect and energy dissipation as well as the dynamic contact angle is solved numerically. However, due to the existence of developing region in velocity profile near the entrance, the numerical solution still overestimates the wetting distance, especially in lower viscosity fluid.

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