

# Micro and Macroscopic droplet behaviors on low-surface-energy solids

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**Abstract**—Wetting phenomena are very important in industrial and chemical fields. As the scale of the system becomes small, the surface interaction becomes dominant because of the increasing surface-to-volume ratio, which, for example, affects the morphological characteristics of the flow patterns in microchannels. This means that the control of wettability leads to efficient heat transfer and chemical reactions and so on. Especially, in space, the wetting phenomena are crucial. Therefore, development of the efficient heat exchanger is important because the major heat exchange is radiation. However, there are many unresolved problems with respect to the wetting phenomena such as contact angle hysteresis and size dependency of the contact angle, not to mention the wetting phenomena under zero-gravity condition. In the present study, the wettability of micro and millimeter sized droplets on low-surface-energy solid is evaluated experimentally and theoretically. The fundamental behavior of the droplets and the gravity effect on the wetting behavior are considered. The results indicate that the gravity effect on droplet wettability can be negligible if the order of the bond number is less than  $O(10^{-3})$ .

**Keywords**—wetting phenomena, contact angle, gravity effect on wettability, droplet.

## I. Introduction

Wettability plays an important role in the printing industry, chemical and medical design and so on[1]. In particular, surface wettability affects the stability of liquid films (and the creation of dry regions) on solid surfaces and is related to flow patterns in multiphase flow[2] and heat transfer efficiency of heat exchanger [3,4], which are crucial phenomena in space environment[5,6].

Wettability is mainly determined by an interaction of liquid and solid surfaces. Contact angle of droplet is a quantitative measure of the wettability. However, the contact angle necessarily does not take a unique value. It is known as a hysteresis of the contact angle[7].

Even simple wetting behaviour such as a droplet on a solid surface is not completely understood. There are many unresolved problems with respect to the wetting phenomena because the phenomena exhibit complex behavior, not to mention the wetting phenomena under zero-gravity condition. Therefore, understanding a fundamental mechanism of the droplet wettability is very important. Recently, we have developed the wettability model[8] by considering the gravity effect and adhesion energy in the vertical and horizontal directions at the contact line. The model indicated a potential to predict the wettability of static droplets on solid surfaces[9]. In this study, the fundamental behavior of the droplets and the gravity effect on the wetting behavior are considered. Concretely speaking, the geometric variables of a droplet were measured experimentally during its natural evaporation to confirm the wettability of droplets ranging from micrometer to millimeter sizes. Then, the results are discussed on the basis of our model.

## I. Experiment

In the experiment, surface energy of liquid was measured using a DM300(Kyowa Interface Science Co., Ltd., Saitama, Japan). The liquid was purified water ( $0.0719 \text{ J m}^{-2}$ ). Three kinds of low-surface-energy materials such as silicon rubber, polyethylene(PE) and polycarbonate(PC) were mainly used. The radius, height and contact angle were measured. The droplet volumes were 0.018, 0.83, 5, 10 and 20  $\mu\text{L}$ . In this study, in order to consider the effect of solid surface energy, the critical surface tension[2,3] is evaluated using several kinds of binary mixture liquids of water and ethanol( $0.057$ ,  $0.0384$ ,  $0.0312$ ,  $0.0274$ , and  $0.0211 \text{ J m}^{-2}$ ). The setting of liquids was performed using microsyringes (outer diameters are 80 and 200  $\mu\text{m}$ ). An image of the droplet is captured using a CCD camera (Yashima Optical Co., Ltd., Tokyo, Japan). The microdroplet is measured using a microscope (Leica Microsystems, Wetzlar, Germany).

## II. Theory

The gravity effect on the droplet wetting behavior was discussed using the following analytical equation[8, 9].

$$\frac{\rho_l g h V}{2} = \pi R^2 \sigma_{lg} (1 - \cos \theta) - \pi R h \sigma_{lg} \sin \theta. \quad (1)$$

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This equation is derived by considering the gravity potential, adhesion forces in the horizontal and vertical direction at the contact line. In this equation,  $\sigma_{lg}$  is the surface energy of liquid.  $\rho_l$  is the density of the liquid ( $\text{kg m}^{-3}$ ),  $g$  is the gravitational acceleration ( $\text{m s}^{-2}$ ),  $R$  and  $h$  are the droplet radius and height(m), respectively.  $V$  and  $\cos\theta$  are the volume of droplet ( $\text{m}^3$ ) and the contact angle (-), respectively. Here, in order to solve Eq. (1), (i) the droplet volume and (ii) the contact angle must be determined. With respect to these problems, we evaluate their values on the basis of experimental data as below.

With respect to the droplet volume, the relationship between the droplet radius and height is considered as shown in Fig. 1. The liquid is purified water and the solid materials are silicone rubber(black circle), PE(white circle) and PC(white triangle). The solid lines are evaluated by the analytical relation as follows;

$$V_0 = \pi h \left( \frac{h^2}{6} + \frac{R^2}{2} \right). \quad (2)$$

This relation is derived assuming spherical cap shape of droplet. The experimental data in Fig. 1 deviate from the analytical lines as the droplet volume increases because of gravity effect. Even so, the droplet volume can be roughly evaluated by Eq. (2). Therefore, the value of the droplet volume in Eq. (1) is evaluated using Eq. (2) as an approximation ( $V \approx V_0$ ).

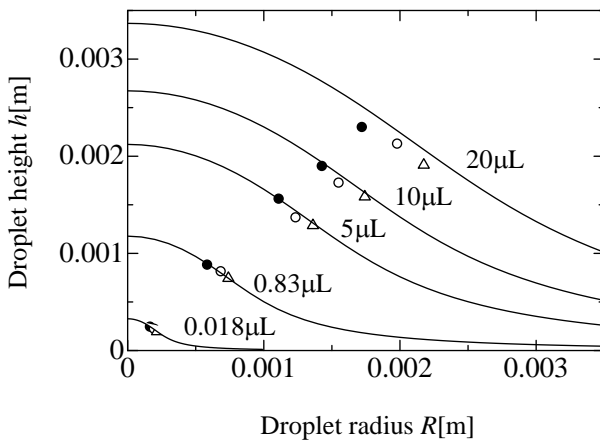


Figure 1: The relationship between the droplet radius and height. The liquid is purified water and the solids are silicone rubber(black circle), PE(white circle) and PC(white triangle).

Then, with respect to the contact angle, the following relation is used[10].

$$\cos \theta = -1 + 2 \frac{\sigma_c}{\sigma_{lg}}, \quad (3)$$

where the  $\sigma_c$  represents the critical surface tension which characterizes surface energy of solid. The value of the critical surface tension is estimated using experimental data.

### III. Result and discussion

Figure 2 shows the relationship between the droplet radius and the contact angle of purified water under the natural evaporation. As an example, the results of the droplet volumes of 0.018, 0.83 and  $10\mu\text{L}$  are shown. From the result of the silicone rubber, initial contact angles at the static condition in all cases are almost same.

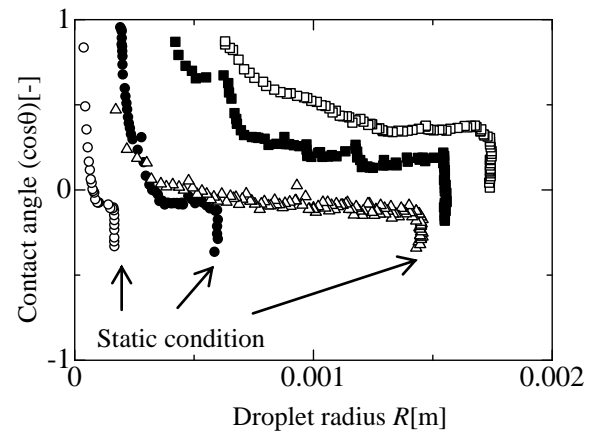


Figure 2: Relationship between the droplet radius and the contact angle; white circle( $0.018\mu\text{L}$ ), black circle( $0.83\mu\text{L}$ ) and white triangle( $10\mu\text{L}$ ) are silicone rubber, black( $10\mu\text{L}$ ) and white( $10\mu\text{L}$ ) squares are PE and PC, respectively.

As the liquid evaporates, the contact angle recedes from the initial contact angle but radius remains constant for all three sizes. After reaching a certain contact angle, the contact line recedes and the droplet radius decreases. Then, the contact angle decreases. For the results of PE and PC, similar droplet behaviors are observed. Here, we analyzed the droplet behavior focusing on the static conditions on the basis of our model. Figure 3 shows the relationship between the droplet radius and the height of the initial droplets on the silicon rubber. The analytical lines are obtained using Eqs. (1), (2) and (3). The dashed and bold solid lines represent the relationships between  $R$  and  $h$  under 0 and  $2g_0$  gravity conditions where  $g_0$  is  $9.8 [\text{m s}^{-2}]$ . In the evaluation of the change in the gravity condition, we assume that the contact angle takes almost constant value and can be expressed by Eq. (3) even if the gravity changes. From the result, as the droplet volume becomes small, the experimental data approaches the dashed line. With respect to the analytical solutions, the three analytical lines gradually merge as the droplet size becomes

small. Figure 4 and 5 show the relationships between the gravity and the droplet radius, and the gravity and the droplet height, respectively. In both figures, the horizontal axis represents the acceleration of gravity. In the present analysis,

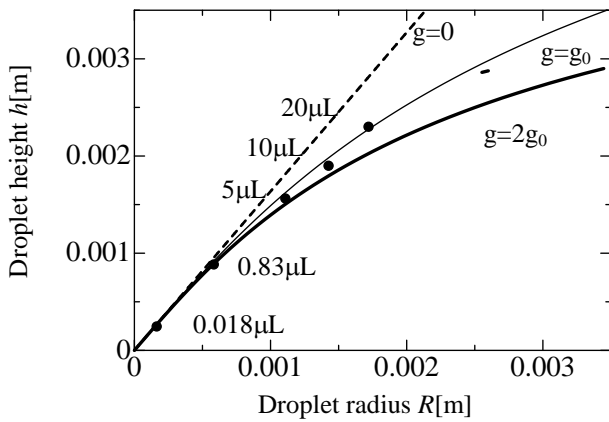


Figure 3: Comparison of the experimental data with the theory under some gravity conditions.

the gravity is evaluated by a factor  $N$  as  $g=N \times g_0$  where  $g_0=9.8 \text{ m s}^{-2}$ . Thus, the number of  $N$  is mainly used in this figure. The white circle, black circle, white triangle, black triangle and white square are the analytical results of different droplet volumes of  $0.018\mu\text{L}$ ,  $0.83\mu\text{L}$ ,  $5\mu\text{L}$ ,  $10\mu\text{L}$  and  $20\mu\text{L}$ , respectively. When the droplet is deposited on the solid surface under each gravity condition, the droplet radius and height will exhibit a tendency as shown in these figures. In Fig. 4, the droplet radius almost takes constant for  $0.018\mu\text{L}$  and  $0.83\mu\text{L}$  cases. The droplet height also takes constant. On the other hand, in the cases of  $5\mu\text{L}$ ,  $10\mu\text{L}$  and  $20\mu\text{L}$ , the droplet radii gradually increase as the gravity increases. In these cases, the droplet heights decrease as the gravity increases. Here, the bond numbers of droplets are  $0.386(20\mu\text{L})$ ,  $0.243(10\mu\text{L})$ ,  $0.153(5\mu\text{L})$ ,  $0.046(0.83\mu\text{L})$  and  $0.0036(0.018\mu\text{L})$ , respectively. Therefore, the gravity effect on the wetting phenomena is not necessarily negligible even if the bond number is less than unity. In addition, the results indicate that the gravity effect will be negligible if the order of the bond number is, at least, less than  $O(10^{-3})$ . Moreover, from these results, the droplet behavior under zero-gravity condition may be able to predict to a certain extent. Concretely speaking, in the case of  $g=g_0$ , the contact angle of droplet which is initially deposited on the solid surface can be evaluated using Eq. (3) even if the gravity effect is non-negligible. Thus, under zero-gravity condition, the contact angle of droplet may be able to predict using same equation. Moreover, considering the results of Figs. 2 to 5, the trajectories between the droplet radius and the contact angle for large droplet as shown in Fig. 2 may shift in the right direction if the gravity increases. On the other hand, the trajectories may shift in the left direction if the gravity decreases.

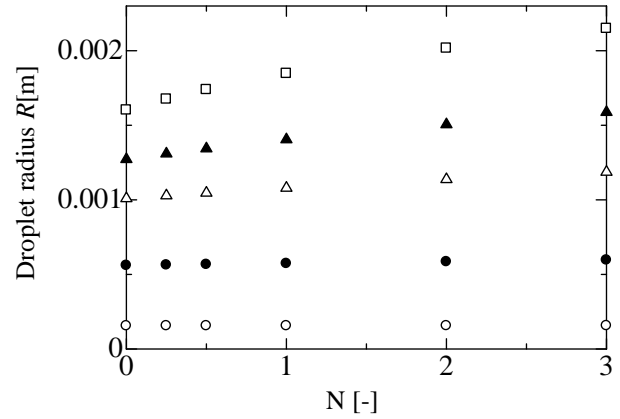


Figure 4: Relationship between the gravity and the droplet radius:  $g=N \times g_0(g_0=9.8\text{ms}^{-2})$ ,  $0.018\mu\text{L}$ (white circle),  $0.83\mu\text{L}$ (black circle),  $5\mu\text{L}$ (white triangle),  $10\mu\text{L}$ (black triangle) and  $20\mu\text{L}$ (white square).

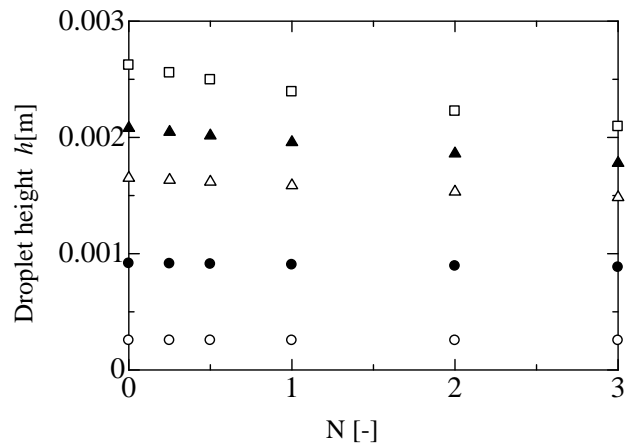


Figure 5: Relationship between the gravity and the droplet height:  $g=N \times g_0(g_0=9.8\text{ms}^{-2})$ ,  $0.018\mu\text{L}$ (white circle),  $0.83\mu\text{L}$ (black circle),  $5\mu\text{L}$ (white triangle),  $10\mu\text{L}$ (black triangle) and  $20\mu\text{L}$ (white square).

#### iv. Conclusion

In the present study, the wettability of micro and millimeter seized droplets on low-surface-energy solid is evaluated experimentally and theoretically. The fundamental behavior of the droplets and the gravity effect on the wetting behavior are considered. The results reveal that the gravity effect on the wetting phenomena is not necessarily negligible even if the bond number is less than unity. The gravity effect will be negligible if the order of the bond number is, at least, less than  $O(10^{-3})$ . From the present study, the droplet wettability under zero-gravity condition is predict to a certain extent. However, in order to understand detail droplet behavior, further

investigation of contact angle behavior under various gravity conditions will be needed.

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