Rupture of an air bubble on the solid surfaces

Shingo ISHIHARA*, Yoshiyuki TAGAWA* and Masaharu KAMEDA*

* Department of Mechanical Systems Engineering, Tokyo University of Agriculture and Technology, Naka-cho 2-24-16, Koganei, Tokyo 184-8588, Japan
E-mail: s158163w@st.go.tuat.ac.jp

Received: 28 March 2018; Accepted: 17 July 2018

Abstract
We investigate behavior of bouncing and rupturing air bubbles on solid surfaces experimentally. We focus our attention to how the hydrophilicity of the solid surface alters the rupture process. We observe motion of the single bubble of a fixed diameter on several flat glass plates using high-speed cameras. In this experiment, we use two kinds of plate whose contact angles are different from each other. The bubble rises and bounces on the glass surface several times without touching the plate. It is found that, on the weakly hydrophilic glass plate whose contact angle is 65 degree, the liquid film between the bubble and the solid surface ruptures within about a hundred milliseconds after bouncing. The rupture starts at a single site. In contrast, on a highly hydrophilic glass whose contact angle is 7.6 degree, rupture time becomes much longer, i.e. more than 30 minutes. The rupture starts at several sites simultaneously. Note that in the both cases bounce times, bounce intervals, and bounce distance, are quite similar. Quite large difference in onset of rupture indicates that the existence of surface nanobubbles on the weakly hydrophilic glass enhances the rupture of liquid film. In the case of highly hydrophilic glass, the penetration of air into the liquid film after the rupture exhibits the pattern similar to viscous fingering.

Keywords: Bubble, Rupture, Hydrophilicity, Thin liquid film, Interference fringe

1. Introduction
Improvement of water quality is indispensable for the current life. Flotation is a water treatment process that clarifies wastewaters by the removal of suspended matter such as sludge. Injected bubbles adhere to the suspended matter and rise towards float to the liquid surface of the water due to their buoyancy. Finally, the floating matters removed by a skimming device. The behavior of the hydrophilic substance included in suspended matter has a large influence on the selectivity of the flotation process (Koide and Yamada 1990).

A key physical process that controls the bubbles adhering to the suspended matter is the rupture dynamics of the thin film between a bubble and a solid surface. Various researches about the bubble’s bounce to rupture process have been reported. Krasowska et al. (2009) studied the solid surface roughness dependence of the liquid thin film formation experimentally, and showed that the rougher surface becomes, the earlier the film forms. As for the mechanism of drainage dynamics through bounce to rupture process, Hendrix et al. (2012) observed these processes by high-speed photography. They inspected the time variation of liquid film thickness and formation, and they analysed that the central portion of a droplet surface of the film continues to thin, then the film ruptured from this central portion. Zawara et al. (2007) observed the bounce process on the solid surfaces with various hydrophilic surfaces. They reported that the bounce behaviors of the bubble on mica (θ=0°). The contact angle θ is the angle formed by the solid surface and the tangent of the water droplet.) and fluorite (θ=40°) are quite similar. Albadaiw et al. (2014) calculated the air bubble’s collision and bounce process by using the Volume of Fluid (VOF) method, and showed their numerical results agreed well with an experimental result by Tsao et al. (1997). However, they did not calculate the rupture process of the liquid film which occurs after bouncing.

Compared with the bouncing dynamics, less attention was paid to the rupture process, which is important at the final stage of an adherent process. Kadoura et al. (2013) reported that a thin water film sprayed onto a surface typically ruptured near the center of the substrate, directly under the nozzle. But they did not argue about the the time history of film
thickness. Schulze et al. (2001) observed the interference fringe of a liquid film between the hydrophobic surface and the portion of the air which was injected by the capillary tube, and they inspected the rupture process. On the hydrophobic surface, the rupture started at a single site, and the time scale was $O(1)$ ms. Stockelhuber (2003) used the same set up as Schulze et al. (2001), and they observed the rupture of liquid film between the hydrophilic surface charged by Al$^{3+}$ and the air injected by the capillary tube. On the hydrophobic surface, the rupture started at several sites simultaneously, and the time scale was $O(1)$ s. However, he did not argue the no charged surface and the no compulsion air condition.

In this study, we clarify the effect of hydrophilicity of the solid surface on the rupture of the liquid film using high-speed photography. The motion of liquid film including the onset of rupture is observed in detail by a long time interferometric imaging. Difference in rupture due to surface hydrophilicity is discussed in the relation with nanobubbles.

2. Experimental setup and procedure

A schematic of the experimental set up is presented in Fig. 1. A rectangular acrylic tank is filled with ultrapurified water. A plain microscope slide made from water-white glass plate settled in the tank as a solid surface. We use two kinds of glass plate (Matsunami Glass Industry S2112, roughness = 1 nm; Muto Pure Chemicals 511611, roughness < 1 nm (rms)). We establish the fixed base as approximately 20 mm each side using the adhesive tape (Scotch SPU19, 3M) on the glass plate (Fig. 2). Horizontal level of the glass plate is adjusted by a goniometric stage equipped below the tank. A bubble is released from a fine needle (inner diameter 0.40 mm) connected to a syringe pump. The volume flow rate is 12 µL/s. The diameter of a bubble is 2.8 mm. The bubble rises upwards due to its buoyancy, then bounces on the glass which is settled 25 mm upward the needle. During the bouncing process of bubbles, a thin liquid film exists between the bubble and the plate. After bouncing, the liquid film ruptures.

We observe the bounce to the rupture process by high-speed cameras. Two high-speed cameras (FASTCAM SA-X, Photron) are used. The spatial resolution is 1,024 ×848 pixels, and frame rate is 15,000 fps. One is for interference fringe [Fig. 3(a)] generated by using a monochromatic light (wavelength 630 nm, UHP-Mic-LED-630, Prizmatix co.) to observe the deformation of liquid film precisely, and the other is for the side view [Fig. 3(b)] which reveals the bounce process. Two cameras are synchronized by a delay generator (Model575 Pulse/Delay Generator, BNC). A co-axial lens (12x Co-axial Ultra Zoom Lens, Navitar) and an objective lens (M-PLAN PRO 5x, Mitsutoyo) are attached to the camera for fringe. A telephoto lens (AF-s MICRO NIKKOR 105mm 1:1.28G ED, Nikon) is attached to the camera for side view observation. Using desktop capture software, the whole rupture process is observed.

The contact angle of the glass surface is measured by a contact angle gauge (DMs-400, Kyowa Interface Science). The contact angles are 65.4° (Matsunami S2112, hereinafter called “weakly hydrophilic glass”) and 7.6° (Muto 511611, hereinafter called “highly hydrophilic glass”) (Fig. 4), which means the two glasses have different nature in hydrophilicity.

Fig. 1 Schematic drawing of the setup. The bubble is injected from a submerged needle placed 25 mm below the plate. The bubble rises upward and bounces on the glass surface several times without touching the plate until the liquid film ruptures.
Fig. 2 Fixed base on the plate for long time scale observation. A bubble is fixed by the base.

Fig. 3 Typical images from the high-speed cameras. (a) A snapshot of the interference fringes. (b) Side view showing the bubble image taken at the same time as the interference pattern is obtained.

Fig. 4 Water droplet on the glass plates. (a) Weakly hydrophilic glass (Matsunami S2112), contact angle is 65.4° (b) Highly hydrophilic glass (Muto 511611), contact angle is 7.6°.

3. Results and discussion
3.1 Bounce and rupture processes

In this experiment, a bubble released from the needle rises upwards due to its buoyancy, then bounces on the glass surface several times without touching the plate. In the bounce process, the liquid film exists between the glass plate and the bubble. The deformation of the film is observed from the interference fringes in detail.

Figure 5(a) shows the time variation of the side view of bubbles bouncing on the weakly hydrophilic glass and Fig. 5(b) shows that on the highly hydrophilic glass. Figure 5 indicates that bounce interval and distance is quite similar on the glasses irrespective of degree of hydrophilicity. It indicates that the properties of solid surface do not affect the bouncing behavior of the bubble while it does affect drainage of the liquid film after the bouncing process.

Figure 6 shows the time variation of the interference fringes when using the weakly hydrophilic glass plate. In Fig. 6, 0 ms denotes the time at which the rupture site appears after finishing the bounce process. As shown in Fig. 6, the rupture starts at a single site, and the whole region of illuminated area is covered by air after O(100) ms.

Figure 7 shows the time variation of the interference fringes when using the highly hydrophilic glass plate. The time displayed in Fig. 7 denotes the time after the first bounce of the bubble is observed. As shown in Fig. 7, the drainage of the liquid film proceeds slowly. When the time after 30 min, the thickness of liquid film become uniformity in the range of a half wavelength of the monochromatic light (315 nm). When the time reaches 60 min, the rupture starts at several sites simultaneously. The process of gas penetration into the liquid film is very slow. The air covers the entire region of observed area at O(100) min.

The rupture of an aqueous wetting films can be distinguished between the two mechanisms; a bubble nucleation at the rupture site caused by very small gas bubbles adhering at the solid surface, and a capillary wave when using the capillary tube and charging (Stockelhuber 2003). In this case, we do not use the capillary tube, and the glass plate is not to be
charged by the ion. So, the reason that induce the liquid film to rupture is the nucleation.

Our experimental result suggests that the surface nanobubbles exist even if the surface is weakly hydrophilic as well as mechanically flat within 1 nm. Although the nanobubbles are typically observed on hydrophobic surfaces whose contact angle is larger than 90 degrees (Borkent et al. 2010; Suna et al. 2016), the hydrophobicity is not a necessary condition for the surface nanobubbles to be stable. Several researches (Weijts and Lohse 2013; Liu and Zhang 2013; Lohse and Zhang 2015; Chan et al. 2015) indicate that the three-phase contact line pinning, which results from the intrinsic nanoscale physical roughness or chemical heterogeneities of substrates, leads to stable surface nanobubbles. The weakly hydrophilic surface in our experiment is a silane-treated glass, that is, a hydrophobic film is partially formed on its surface. This chemical property may stabilize nanobubbles in the range of several hours.

On the other hand, it is improbable that nanobubbles adhere at such good wettable surfaces (contact angle <10°). In this case that takes long time to rupture on a hydrophilic surface, there is a possibility that the surface is contaminated with some hydrophobic spots which induce the nucleation. It is conceivable that the derivation of the hydrophobic spots is very small amount of organic substance in ultrapurified water, the dust in the air adhering the surface until putting in water after washing the glass by ultrapurified water, the dust in the air descending to the container, or the glass grinding dust adhering the surface which could not be removed by washing process.

Compared with the result of Stockelhuber’s (2003), the rupture process on the highly hydrophilic surface (S511611, θ=7.6°) starting from several sites simultaneously is similar to his study. And the rupture process on the weakly hydrophobic surface (S2112, θ=65.4°) that starts from a single site is similar to his study using the hydrophobic surface. The rupture process has similar with the results of Stockelhuber’s (2003) in the view of the appearing pattern of the rupture site, but in detail, there are several differences. In his study, the rupture started from a single site on the hydrophobic surface, but the site location was in the middle of the radius of the liquid film between the bubble and the surface. On the other hand, in this study, the rupture site on the surface (S2112, θ=65.4°) is located along the circumference of the liquid film shown in Fig. 6. When using the capillary tube and compressing the air straight down uniformly, the liquid film is drained while maintaining the uniform thickness. The bubble rises upwards due to its buoyancy, the liquid film forms the dimple shape whose outer area is thinner than inner area. Hendrix et al. (2012) observed the dimple formation of the liquid film through the collisions between millimeter-size bubbles in water against a glass plate by high-speed photography. The thinnest point exists on the rim of the liquid film, then ruptures from it. In the case of the rupture on the surface (S511611, θ=7.6°), the dimple shape gradually deforms to the uniform shape because the drainage of the liquid film proceeds slowly whose time scale is thousands of times longer than that on the weakly hydrophilic glass (S2112, θ=65.4°).

![Image of bubble behavior](image_url)

**Fig. 5** The bounce behavior of the bubble on the glass. (a) Bounce on the weakly hydrophilic glass (θ =65.4°), (b) Bounce on the highly hydrophilic glass (θ =7.6°).
Fig. 6 The time variation of the interference fringe on the weakly hydrophilic glass ($\theta=65.4^\circ$). 0 ms denotes the time at which the rupture site appears after finishing the bounce process.

Fig. 7 The time variation of the interference fringe on the highly hydrophilic glass ($\theta=7.6^\circ$).

### 3.2 Gas penetration after the film rupture on highly hydrophilic surface

Figure 8 shows the close-up images of the drainage of the liquid film on the highly hydrophilic glass ($\theta=7.6^\circ$). The formation of air region exhibits finger pattern. The area of the contact surface is measured shown in Fig. 9. Table 1 shows the measurement result of the drainage speed on the weakly hydrophilic glass ($\theta=65.4^\circ$) calculated from the time-series of the images from 2 ms to 140 ms. Table 2 shows the measurement result of the drainage speed on the highly hydrophilic glass ($\theta=7.6^\circ$) from each photography with the time is 60 min to 100 min, which is before combining other sites. The drainage area speed on the highly hydrophilic glass ($\theta=7.6^\circ$) is approximately $19 \text{ mm}^2/\text{s}$. On the other hand, the drainage area speed on the weakly hydrophilic glass ($\theta=65.4^\circ$) is approximately $1.0 \times 10^{-5} \text{ mm}^2/\text{s}$. From the very low speed of drainage of the liquid film and formation of fingering pattern shown in the rupture on the highly hydrophilic surface, there is a possibility of occurring viscous fingering. Viscous fingering is the formation of patterns in a morphologically unstable interface between two different viscous fluids such as air and oil. It usually occurs in in a porous medium (Homsy 1987) or in a Hele-Shaw cell (Saffman 1986). It occurs when a less viscous fluid is injected displacing a more viscous one. Saffman (1986) reported that the dependence of the capillary number on the formation of viscous fingering. Bensimon (1986) argued the stability of viscous fingering, and the existence of a nonlinear instability which threshold decreases exponentially with increasing velocity. It is future work of investigation whether such instability occurs in such
a nanomillimeter-thickness of the film.

Fig. 8 The time variation of the interference fringe on the weakly hydrophilic glass. The drainage formation looks like finger pattern.

Fig. 9 Measurement of the contact surface area. Using analysis software, the area variations are measured.

Table 1 The area of contact surface on the weakly hydrophilic glass.

| Time [ms] | Area [mm²] |
|-----------|------------|
| 2         | 0.007      |
| 20        | 0.113      |
| 60        | 0.907      |
| 100       | 1.721      |
| 140       | 2.662      |

Table 2 The area of contact surface on the highly hydrophilic glass.

| Time [min] | Area [mm²] |
|------------|------------|
| 60         | 0.005      |
| 70         | 0.011      |
| 80         | 0.015      |
| 90         | 0.019      |
| 100        | 0.030      |

4. Conclusions

We observe the rupture process of an air bubble on the glass surface with different hydrophilicity. The interference fringes generated using the monochromatic LED light are observed to determine the rupture sites on the surface. When the weakly hydrophilic surface whose contact angle is 65.4 degrees, the rupture starts from a single site, and the time scale of the rupture is \( O(100) \) ms. On the other hand, when the highly hydrophilic surface whose contact angle is 7.6 degrees, the rupture starts at several sites simultaneously, and the time scale is of the rupture is \( O(100) \) min.

Our observation implies that the surface nanobubbles exist even if the surface is weakly hydrophilic as well as
mechanically flat within 1 nm. Differences in chemical composition on the surface may select whether the nanobubbles are held or not.

From the very low speed of drainage of the liquid film and formation of fingering pattern shown in the rupture on the highly hydrophilic surface, there is a possibility of occurring the viscous fingering.

This observation suggests that in the practical flotation process, the selectivity of hydrophilic/hydrophobic matters proceeds without the serious interference because the time scale of the liquid film’s rupture to the matter’s surface with weak hydrophilicity is about $2 \times 10^6$ times faster than that with high hydrophilicity.

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