

Visualized study on the interaction between single bubbles and curved solid surface in flotation separation process

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ABSTRACT

The present study has been devoted to bubble–curved solid surface interaction in water, which is critical to the separation of suspended particles by air flotation. For this purpose, two particular stages of the interaction (collision and attachment) have been examined visually using high-speed photography in a laboratory-scale flotation column. The effects of the surface material and surfactant concentration on these two stages have been also studied quantitatively. The considered solid materials are the cleaned glass as hydrophilic surface and Teflon as hydrophobic surface. The experimental results show that the presence of surfactant significantly affects the collision and rebound process of a gas bubble, while there is no obvious effect of the surface material on the rebound process. An increase in surfactant concentration has been observed to suppress the rebound number and maximal distance of the bubble from the surface. Moreover, the three-phase contact time of the bubble is a strong function of the surfactant concentration and surface hydrophobicity as well as of the bubble diameter. Another important finding is that the bubble attachment is only observed at the hydrophobic Teflon surface below the surfactant CMC (critical micelle concentration). Results of this study are relevant for deep understanding of the attachment mechanism and to determine the proper conditions for a selective flotation process.

Key words | attachment, curved solid surface, flotation bubbles, high-speed photography, three-phase contact time

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INTRODUCTION

Flotation is a surfactant-based separation process widely used in environmental and municipal industrial applications such as wastewater treatment, water purification, de-inking of wastepaper and plastic recycling. In the process, gas is generally introduced in the form of small bubbles which act as carriers transporting suspended particles to the liquid surface, where particles are removed (Malysa *et al.* 2005; Finch *et al.* 2008). To create small bubbles or modify interfacial properties, surfactants are commonly added into the system (Lima *et al.* 2008; Watcharasing *et al.* 2009; Wiley *et al.* 2009; Lu *et al.* 2010; Li *et al.* 2013). Obviously, the interaction between gas bubbles and particles is critical to the formation of stable bubble–particle aggregates, which is a fundamental step required for achieving higher efficiency in flotation separation.

According to Derjaguin & Dukhin (1961), bubble–particle interaction can be divided into three processes:

collision, attachment and detachment. With approach of bubbles and particles to the contact distance, collision can occur, which is determined mainly by the hydrodynamics of the liquid phase. As the particle and bubble come closer, the influence of intermolecular and interfacial forces is decisive for the attachment and detachment process. For a stable bubble–particle attachment, a liquid film formed between the bubble and particle surface needs to be ruptured, and a liquid/gas/solid three-phase contact (TPC) line must be formed (Albijanac *et al.* 2010; Firouzi *et al.* 2011; Niecikowska *et al.* 2012). So far, bubble–particle collision has been well studied and many models have been well established (Basařová *et al.* 2010; Parkinson & Ralston 2011). In contrast, the attachment process has not been fully explored yet due to its fast period and complex physicochemical mechanism. Recently, some studies have appeared on the attachment of a rising air bubble to solid

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surfaces (Malysa *et al.* 2005; Krasowska *et al.* 2009; Zenit & Legendre 2009; Fujasová-Zedníková *et al.* 2010; Parkinson & Ralston 2011; Niecikowska *et al.* 2012). However, most of those studies focused on interaction of bubble-flat solid surfaces. Although some of these studies observed directly the collision and attachment, how to quantitatively characterize the bubble-solid surface interaction was still questionable.

The aim of the present work is to investigate the interaction of a single air bubble with a curved surface experimentally. Because the bubble may slide away from the curved surface instead of staying below the surface if the attachment cannot occur, the curved surface arrangement could reveal easily whether or not the TPC is formed compared to the horizontal surfaces. Therefore, the emphasis of the present study is on the effects of surfactant and solid material on the two particular stages of bubble (collision and attachment) at the curved solid surfaces. The effects of the bubble size on the bubble collision and attachment are also examined.

EXPERIMENTAL

Experimental setup

Figure 1 shows schematically the present experimental setup. A single bubble was injected through stainless steel capillaries with different diameters, installed at the bottom of a square plexiglas column. Gas flow rate was regulated to produce single bubbles (ca. 0.8–1.8 mm in diameter) by controlling the rotational speed of a peristaltic pump in cooperation with a rotameter. The real particle was replaced by an immersed curved surface (80 mm in diameter) located 35 cm above the capillary tip, where the bubble was found to

reach the dynamic steady state after testing. The bubble motion was monitored and recorded using a high speed camera (MS 75K, Canada). The illumination system consisting of a halogen light of 2,000 W and a diffuser permitted the light to disperse into the column uniformly to avoid reflection from the bubble surface.

Materials

An anionic surfactant, sodium dodecyl sulfate (SDS), was employed in this study. It was commercial grade and used as received without further purification. Fresh deionized water (18.2 Ω) with temperature of 20 °C was employed for solution preparation. The concentrations of SDS used were 3.0×10^{-5} , 9.0×10^{-5} , 5.0×10^{-4} , 8.5×10^{-3} and 1.0×10^{-2} mol/L. Glass and Teflon cylindrical surfaces purchased commercially were used as the curved solid surfaces. They were detected to have low roughness and, further, their contact angles measured by the droplet image method were 38° and 107°, indicating that the glass and Teflon surfaces were hydrophilic and hydrophobic, respectively.

Experimental and analytical methods

For tracking the bubble collision and attachment process, images with $1,280 \times 1,024$ pixels were recorded by the high speed camera at 1,000 fps, and then analyzed off-line using the AVI View software to obtain bubble position and geometric parameters. Since the bubble generated was generally near-spherical in shape, the equivalent diameter (D_{eq}) was calculated as follows (Li *et al.* 2013):

$$D_{eq} = (d_v d_h^2)^{1/3} \quad (1)$$

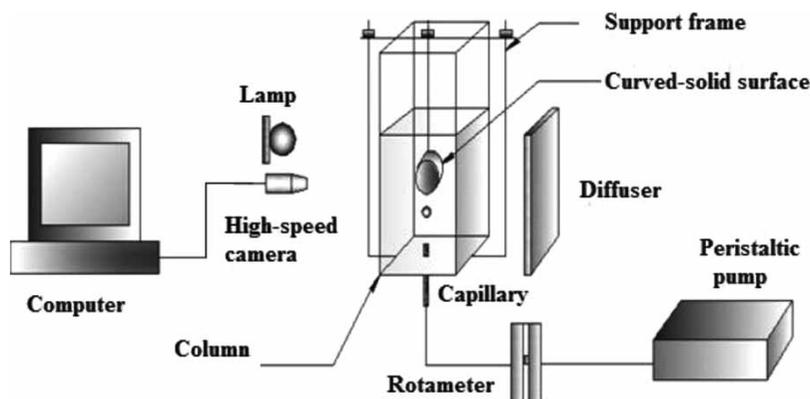


Figure 1 | Sketch of the experimental setup.

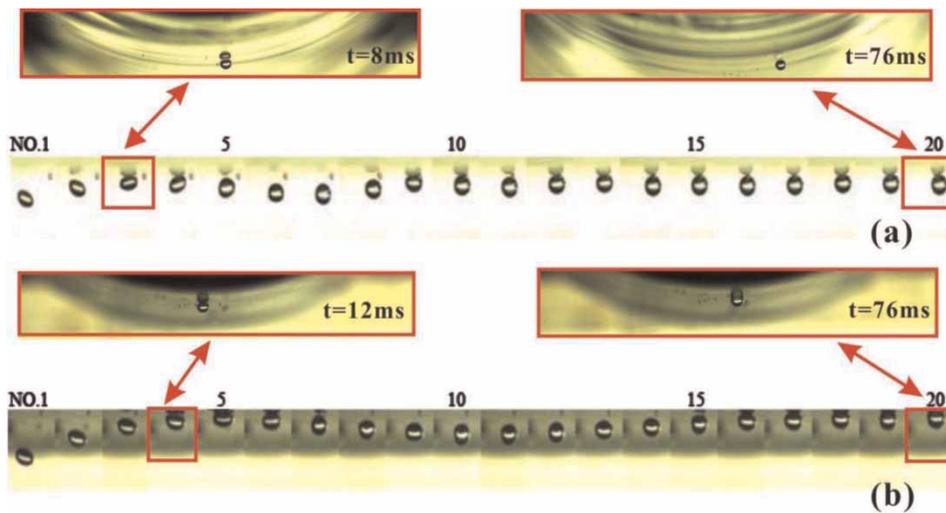


Figure 2 | Sequential images of bubble collision against (a) curved glass surface, (b) curved Teflon surface in pure water. Bubble size is about 1.5 mm.

where d_v and d_h represent the long and short axes of the bubble, respectively. The instantaneous bubble velocity (U) was determined by differentiating the center-of-mass position from each sequential frame. The impact velocity (U_{im}) was defined as velocity corresponding to the impact moment when the distance between the center of mass of the bubble and the surface is equal to d_h .

Every measurement was conducted at least five times and average values were then calculated to reduce evaluation uncertainty. Before each test, the column was emptied and carefully cleaned. All tests were carried out under consistent conditions, such as at room temperature of 20 ± 1 °C.

RESULTS AND DISCUSSION

Bubble collision and rebound at curved solid surfaces

Figure 2 presents sequential images of a bubble colliding with the curved glass and Teflon surface in pure water. The time interval of two consecutive images was 4 ms. As seen, the bubble approached to a sufficient distance, and then collision occurred. When the bubble deformation reached maximum, the bubble was almost motionless (e.g. 3rd, 9th, 13th and 18th images in Figure 2(a) and 5th and 18th images in Figure 2(b)). The bubble then visibly rebounded from the surface. Several rebounds could be observed from both the glass surface and Teflon surface, suggesting that there were similar phenomena, called the

‘impact-rebound-slide’ cycle, for the bubble colliding with the hydrophilic and hydrophobic surfaces.

Moreover, as shown in the entire images (e.g. $t = 8$ and 76 ms), such collision process was found to be accompanied by a tangential shift of the bubble on the curved surface due to the present curved surface arrangement, which was different from other previous research on flat surfaces (Malysa *et al.* 2005; Krasowska *et al.* 2009; Fugasová-Zedníková *et al.* 2010; Parkinson & Ralston 2011; Niecikowska *et al.* 2012).

Figure 3 illustrates the variations in bubble velocity during collisions with the curved glass surface in different solutions. Here 10 ms prior to colliding was set as the initial moment ($t = 0$). In pure water the bubble approached the surface with an approach velocity of ca. 0.305 m/s. On collision

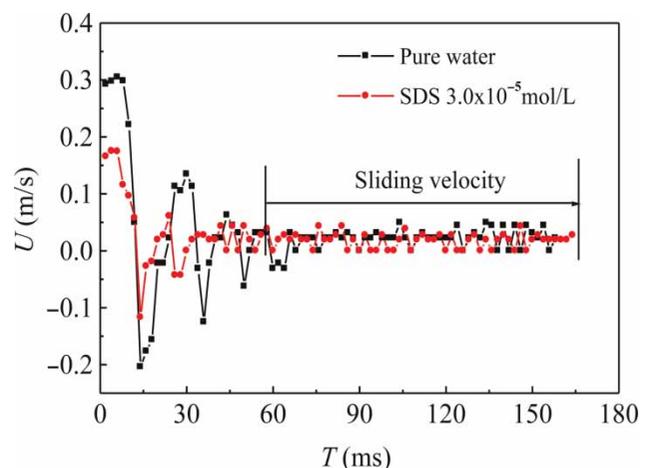


Figure 3 | Variations in local velocity during bubble colliding against curved glass surface.

with the curved surface, the bubble velocity decreased rapidly and then the bubble moved backwards, reaching a velocity up to 0.20 m/s. Afterwards, the bubble started its second approach to the surface with a lower velocity (ca. 0.13 m/s). After about four cycles, the bubble slid away from the curved glass surface with a sliding velocity of about 0.035 m/s. In the case of 3.0×10^{-5} mol/L SDS solution, the bubble approach velocity was about 0.174 m/s due to the so-called Marangoni effect and the first and second rebound velocity were about 0.118 and 0.04 m/s, respectively. After three cycles, the bubble slid away from the curved glass surface with a sliding velocity of about 0.02 m/s. The velocity variation tendency in SDS solution was the same as that in pure water, but approach velocity and second impact velocity were reduced evidently. According to Li et al. (2012), as the bubble rises in water contaminated by surfactant, the surfactant moves off the bubble front and accumulates in the bubble rear, which induces a surface tension gradient and further causes the so-called Marangoni tangential shear stress on the bubble surface. The shear stress can make the bubble surface stiff. Therefore, the drag force acting on the

bubble due to the reduced mobility increases, resulting in a decrease in bubble rise velocity.

Figure 4 shows the normalized maximum rebound distance (S_{\max}/D_{eq}) after the first collision. The data reported here were the average and standard deviation of at least five measurements for each data point. The standard deviation was presented in the form of error bars. It can be found that the maximum rebound distance of the bubble decreased obviously with increasing SDS concentrations for different solid materials (Figures 4(a) and 4(b)), suggesting that the rebound process was strongly affected even at the smallest surfactant concentration. Another important finding was that the solid material seemed to have little effect on rebound distance, especially in SDS solutions (Figures 4(c) and 4(d)). For example, in SDS solution of 3.0×10^{-5} mol/L, the normalized maximum rebound distance of bubble at glass and Teflon surfaces was 0.3218 ± 0.0074 and 0.3176 ± 0.0095 for $Re = 95$ and 99, respectively, and 0.1289 ± 0.0079 and 0.1321 ± 0.0074 for $Re = 66$ and 65, respectively, in SDS solution of 5.0×10^{-4} mol/L. Apparently, no significant difference was found between the

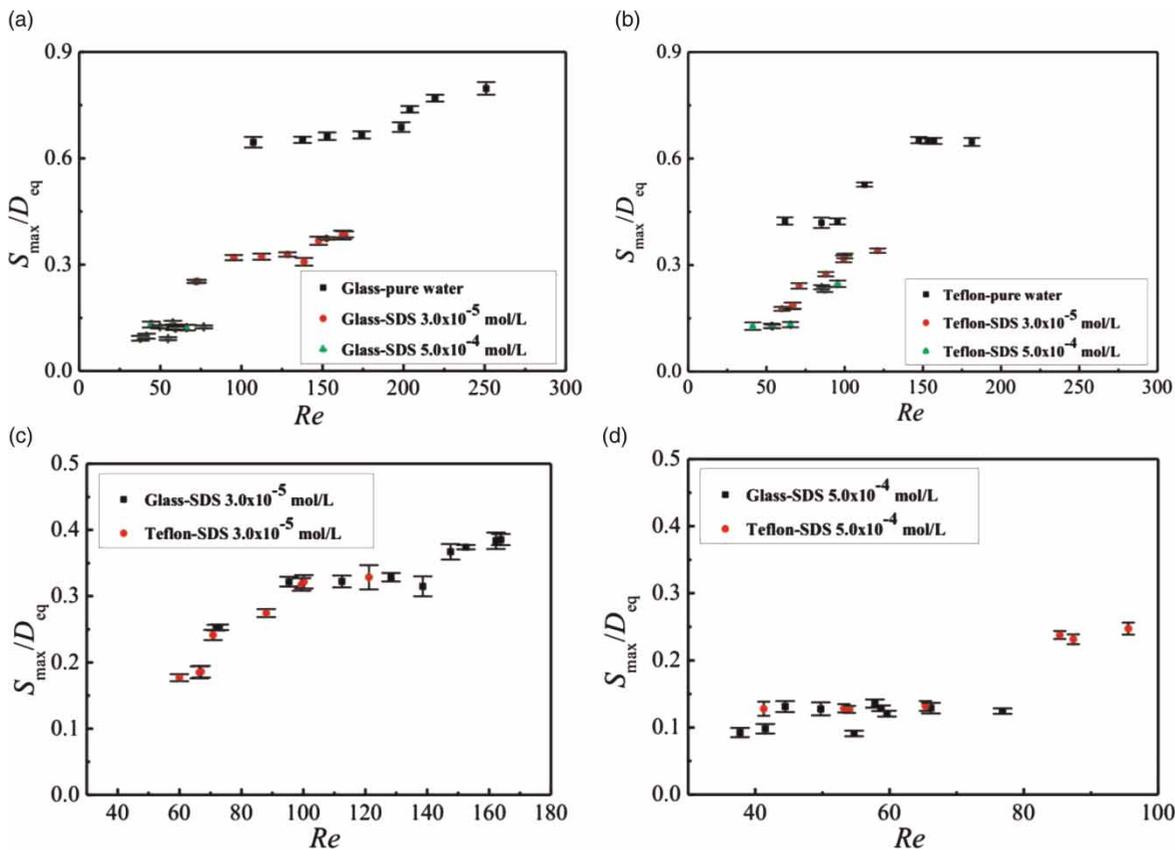


Figure 4 | Normalized maximum rebound distance after first collision vs Reynolds number ($Re = U_{\text{im}}D_{\text{eq}}/\nu$).

present hydrophilic and hydrophobic surfaces used. This phenomenon may be due to the existence of a liquid intervening film between the bubble and the solid surface, resulting in indirect contact of the bubble with the solid surface (Fujasová-Zedníková *et al.* 2010).

The behavior occurring during the rebound process can be explained by the energy balance of the bubble. Before touching the solid surface, the bubble decelerated and deformed obviously from its initial shape, indicating that part of the initial kinetic energy is dissipated due to viscosity of the liquid film and part is transformed into bubble surface energy (Zenit & Legendre 2009). Afterwards, part of the surface energy is again transformed into kinetic energy and the bubble rebounds. Obviously, it is the loss of energy mainly due to viscous dissipation that causes the subsequent decreased bubble rebound and impact velocities. Further, the energy loss of the bubble seems to depend on the solution concentration. This is likely because the viscous dissipation is related directly to the interface mobility. As mentioned above, the Marangoni effect induced by uneven distribution of surfactants on the bubble surface may dampen bubble deformation and reduce bubble mobility.

Bubble attachment to curved solid surfaces

As seen in Figure 2, a significant difference appeared during the attachment process between the bubble and different curved surfaces. The bubble slid away from the curved hydrophilic glass surface after the disappearance of bouncing movement. The TPC was not formed after much longer times than those shown in Figure 2(a), implying that no bubble attachment occurred in this case. A similar finding was also reported by several previous studies. Sñol & González-Cinca (2010) and Niecikowska *et al.* (2012) observed that the bubble remained suspended below the flat hydrophilic surfaces after the visible bubble movement. Although the size of the present solid surface looks much larger than that of the bubble, visual results shown in Figure 2 can illustrate clearly a panorama of the curved surface and bubble sliding motion. Therefore, it is rational to consider the present surface as a curved surface.

In the case of hydrophobic surfaces, however, an unstable liquid film was formed between the bubble and the Teflon surface. This film was then squeezed and ruptured, resulting in TPC formation and, further, the bubble attachment to the solid (18th image in Figure 2(b)). According to Krasowska *et al.* (2009) and Niecikowska *et al.* (2012), the stability of the wetting film and the kinetics of the TPC formation were governed by the interplay of surface

electrical charge and surface hydrophobicity. The wetting film on hydrophilic surfaces was stable even in the presence of the electrostatic attractive forces, but was unstable and ruptured at the hydrophobized surfaces due to so-called long-range hydrophobic attractive forces via a nucleation mechanism, in which nanometer or submicrometer-sized bubbles pre-attached on the hydrophobic surface due to surface roughness, can cause rupture of the wetting film. No such submicrometer-sized bubbles were observed in the present study. This hypothesis needs to be further and widely verified in future studies.

To quantify the bubble attachment to curved Teflon surfaces, the effect of SDS concentration and bubble size on TPC time are shown in Figure 5. The time of the TPC formation, t_{TPC} , was defined as the period from the moment the bubble stopped bouncing to the moment of the TPC formation here. It can be seen that the TPC time decreased with increasing SDS concentration for various sized bubbles examined here, suggesting that the presence of surfactants can facilitate the bubble attachment to the hydrophobic surfaces. In addition, the TPC time was also found to decrease as the bubble size decreased, which means that smaller bubbles are easier for attachment and flotation than larger bubbles.

These results are similar to previous work by Niecikowska *et al.* (2012), who presented the TPC time at a flat hydrophobized titania surface as a function of bubble size and salt concentration. To gain a deeper insight into these results, a simple model of TPC time can be given by the Taylor drainage theory (Seyyed Najafi *et al.* 2008)

$$t_{\text{TPC}} = \frac{6\pi\mu R_b^2}{F} \ln\left(\frac{h_{\text{cr}}}{h_0}\right) \quad (2)$$

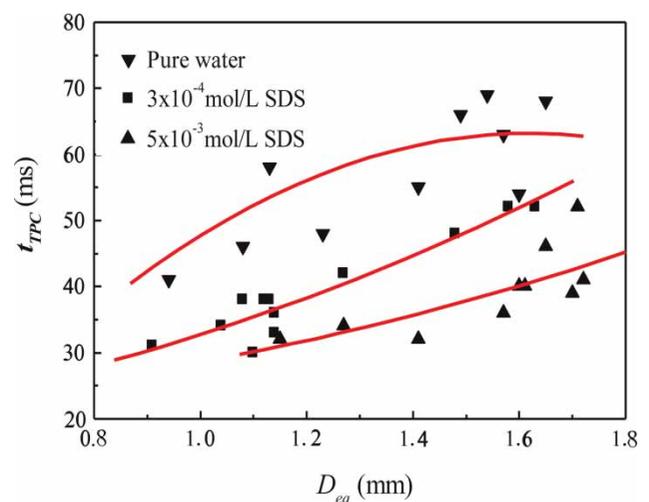


Figure 5 | Effect of SDS concentration and bubble size on the TPC time.

where μ is the liquid viscosity, h_o is the initial distance between the bubble of radius R_b and the solid surface, h_{cr} denotes the critical thickness at which the liquid film will rupture spontaneously, and F presents the net driving force acting on the bubble. Obviously, the TPC time is proportional to the square of the bubble radius. Therefore, a reduction in bubble size will result in a shorter TPC time. As reported by Li *et al.* (2012, 2013), the presence of surfactants could reduce the bubble size, leading to a shorter attachment time. It is also important to note that the critical thickness required in Equation (2) is dependent on the interfacial properties and particle hydrophobicity and, thus, is affected by adsorption of flotation chemicals. According to an empirical power model ($h_{cr} = 23.3[\sigma(1 - \cos \theta_a)]^{0.16}$) developed by Schulze (1989), h_{cr} is proportional to the surface tension (σ) and contact angle (θ_a). Therefore, the presence of surfactants can reduce the surface tension and contact angle, resulting in a lower critical thickness and a shorter TPC time.

It was worth noting that the effect of surfactants on the attachment discussed above was only at low concentration. Quite a different situation can be found at high solution concentration. This is illustrated in Figure 6. The TPC time can be seen to decrease with increasing SDS concentration up to 5.0×10^{-4} mol/L, and then to increase dramatically, implying that the stability of the liquid films increased when the SDS concentration exceeded 5.0×10^{-4} mol/L, making attachment more kinetically unfavorable. Especially, in SDS solutions above the CMC (critical micelle concentration), which is about 8.5×10^{-3} mol/L, the bubble attachment to the curved Teflon surface did not happen at all, instead sliding away from the curved Teflon surface. It needs to be underlined that the reproducibility of this

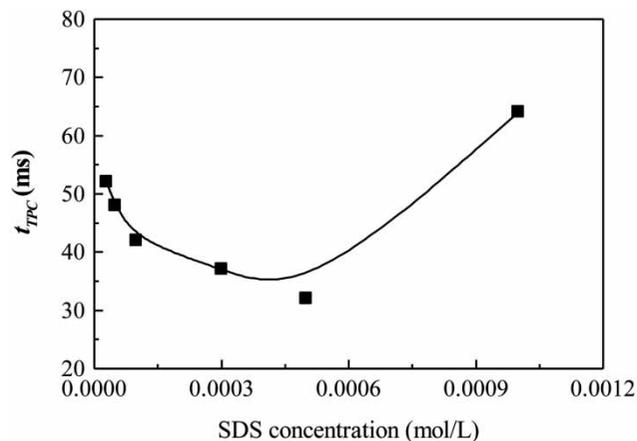


Figure 6 | Effect of SDS concentration on the TPC time at the curved Teflon surface.

interesting finding was good. This might be attributed to the formation of a foam film instead of the wetting film separating the colliding bubble from the surface at very high surfactant concentrations. It is well known that stability of the foam films increases with surfactant concentration, resulting in the decreased hydrophobic attractive forces. Thus, the TPC time was prolonged at high surfactant concentrations. Of course, it is necessary to carry out more experiments concerning effects of surfactant type on bubble attachment at high concentration. Despite this limitation, it is still clear that such results are helpful to choose the proper surfactant concentrations for selective flotation process.

CONCLUSION

The effects of surfactant and solid material on two particular stages of bubble-curved surface interaction have been studied and visualized using high-speed photography. Experimental results show that the presence of surfactants has a significant effect on the collision and rebound process of single bubbles. However, no significant effect of the surface material on the collision process can be observed. During the bubble collision with glass and Teflon surfaces, several rebounds, so-called impact-rebound-slide cycles, can be observed and the impact and rebound velocity can also be found to decrease with rebound number due to energy dissipation in pure water and in SDS solutions. Moreover, compared to pure water, the addition of SDS can reduce the bubble rebound number and rebound distance. Further, the bubble rebound distance decreases with increasing SDS concentration.

Another important result is that the bubble attachment process is dependent on the solid material and the SDS concentration as well as bubble diameter. The formation of three-phase contact of the bubble at the curved surface is found only in the case of the curved Teflon (hydrophobic) surface, not in the hydrophilic glass surface. An additional interesting finding is that the TPC time decreases with increasing SDS concentration up to 5.0×10^{-4} mol/L, and then increases dramatically. When SDS concentration is above the CMC, the bubble attachment cannot be found. Although the present study is restricted only to one type of hydrophobic surface and one type of surfactant, these results are still useful to deeply understand the attachment mechanism and to optimize operating conditions in the air flotation process.

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