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FORMATION AND RUPTURE OF GAS FILM OF ANTIBUBBLE

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Abstract

The formation and rupture of gas film in the process of formation, rupture and coalescence of antibubbles were investigated by high-speed photography. It was found that a gas film will appear and wrap a droplet when the droplet hit a layer of liquid film or foam before impacting the gas-liquid interface. The gas film may survive the impact on the gas-liquid interface and act as the gas film of an antibubble. A multilayer droplet will be formed when the droplet hits through several

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layer of liquid films, and a multilayer antibubble will be formed when the multilayer droplet impact a gas-liquid interface or a single layer of foam on the liquid surface. The way to generate antibubbles by liquid films will undergo the formation and rupture of gas films. The coalescence of two antibubbles, which shows a similar merging process of soap bubbles, also undergo the rupture and formation of gas films. The rupture of gas film of antibubble caused by aging and impact is also discussed.

Introduction

Antibubbles are unusual fluid objects in the liquid. An antibubble is a thin spherical gas film containing and being surrounded by a liquid, which is completely opposite to the soap bubble structure. Antibubbles, with the special structure, have many potential applications. Antibubble can be used for air filtration or cleaning because of its large gas-liquid surface area without significant volume increase. Anti-bubble can hold specific liquids for substance transport or drug preparation, etc. Although antibubble has been reported as early as 1932 (HUGHES et al. 1932) and has been formally named as antibubble in 1974 (STONG 1974), antibubble is still an area that we know little about. Few researches were conducted until this century some valuable researches on formation (GANAN-CALVO et al. 2001, TUFAILE et al. 2002, POSTEMA et al. 2005, KIM et al. 2008, POSTEMA et al. 2007), aging (DORBOLO et al. 2005, 2010, SCHEID et al. 2012, 2014), collapse (SOB'YANIN 2015, ZOU et al. 2013), stabilization (DORBOLO et al. 2003, KIM et al. 2006, POORTINGA 2011), optical properties (SUHR 2012) and control (SILPE et al. 2013, POORTINGA 2013) of antibubbles aroused great academic interest.

The formation of antibubble is the premise and an important aspect of antibubble research. Antibubbles are usually generated by impinging a droplet or liquid jet on a stationary liquid surface (SOB'YANIN 2015), forming a gas film between the droplet or liquid jet and the liquid surface. Under the action of gravity and impact force, the gas film wraps the liquid and sinks below the liquid surface. The jet breaks because of Rayleigh-Plateau instability, the gas film closes, and becomes spherical under the action of surface tension, thus forming a gas film.

This method of producing antibubbles requires that the liquid surface is very clean, even so, the survival rate of antibubbles is still very low. Preparing antibubbles requires great patience (as shown in Fig. 1). For this reason, some researchers have made some improvements to this method, such as vibrating the nozzle and creating an oscillation in the incident jet (BREWER et al. 2010), adding an electrical connection to prevent electrical potential difference due to triboelectric effects (DORBOLO et al. 2003), and rationalizing several aspects of the optimal window in parameter space for creating antibubbles (KIM et al. 2008).

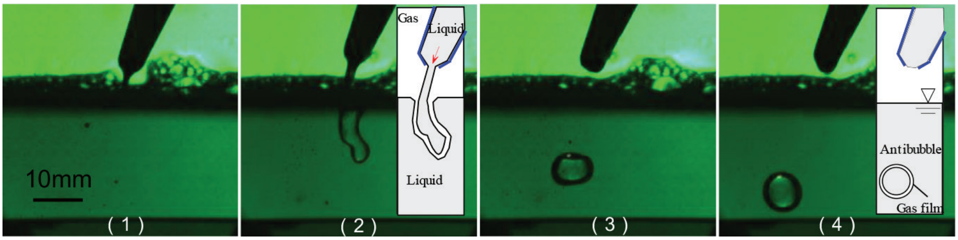

Fig. 1. Schematic diagram of experimental setup

Other researchers developed totally different methods to generate antibubbles, such as generating micron-sized antibubbles by capillary flow focusing (GANAN-CALVO et al. 2001), or generating millimeter-sized antibubbles by the coalescence between two bubbles (TUFAILE et al. 2002), or producing antibubbles by first making a particle-stabilized water-in-oil-in-water emulsion, then freeze-drying to remove both the water and the oil, and finally reconstitute the resulting powder in water (POORTINGA 2013), or oscillating contrast agent microbubbles may create a surface instability, and the re-entrant jet protrude into the gas bubble, leaving a droplet inside the bubble (POSTEMA et al. 2005, POSTEMA et al. 2007). In this paper, the membrane structure (liquid film and gas film) and its dynamic behavior in the process of antibubble formation and rupture is studied by means of high-speed photography.

Experiment

The experimental device consists of a glass tank, a dripping device, liquid films, a foam layer, a light source, lenses and a high-speed camera (as shown in Fig. 2). The dripping device can produce jets of different velocities and droplets

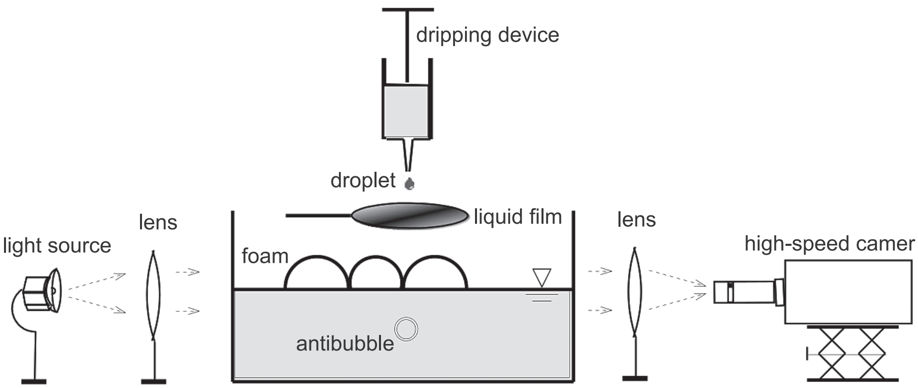

Fig. 2. Schematic diagram of experimental setup

of different sizes. A rectangular plexiglass container (220 mm × 150 mm × 170 mm) is used to hold a mixture of tap water and linear alkylbenzenesulfonate (LAS) (about 10 times the critical micellar concentration (2.2 mM)). The behavior of cavitation bubbles and antibubbles is recorded with a high-speed camera (Photron Fastcam SA-1, Photron Ltd., Japan) equipped with two long distance microscopes (Zoom 6000, Navitar, USA; LM50JCM, Kowa, Japan) respectively. The frames are illuminated with PI-LUMINOR high-light LED lamp (150 W), Cree XHP70 white high power LED, HALOGEN lamp (2600 W). In order to get a better image exposure, two lenses are used to produce parallel light for illumination in some experiments. In the experiment, the liquid in the dropper is the same as that in the rectangular glass tank. The gas in the antibubble film is the air in the environment (temperature: 20°C, pressure: 1 atm).

Results and discussion

Formation of antibubble

The preparation of antibubbles in laboratory requires harsh conditions. The dust and foam on the liquid surface affect the formation of antibubbles. It is usually necessary to use overflow devices to obtain a clean gas-liquid interface (as shown in Fig. 3). However, our experiments have found that the presence of foams on the interface sometimes helps to form antibubbles. When a single layer of foam (larger than the droplet size) is spread over the interface, the droplets or jet first penetrates the foam and then enters the water to become antibubbles (as shown in Fig. 3*b*).

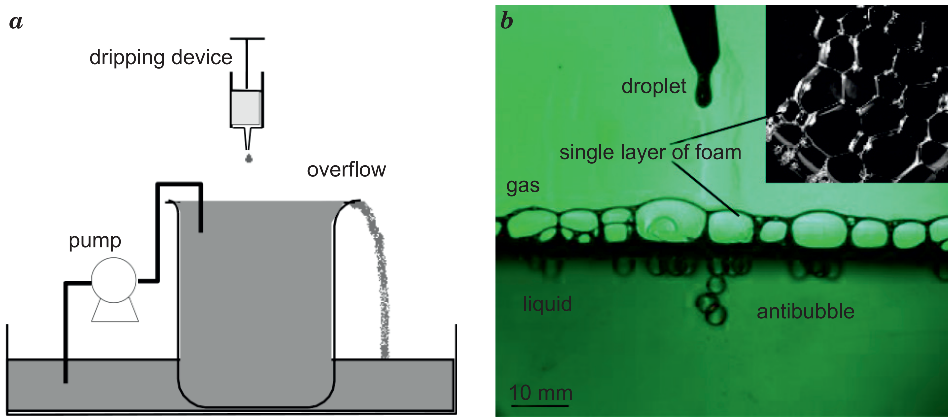

Fig. 3. Liquid surface conditions capable of producing antibubbles:
a – overflow, *b* – single layer of foam

The total reflection caused by the gas-liquid interface on both sides of the gas film makes the antibubble shine in the reflected light and show a thick black edge on the outer edge in the transmission light. This is the most obvious difference between ordinary bubbles and antibubbles underwater (as shown in Fig. 4*a*, *b*). When the droplet drops into a honeycomb monolayer foam on the liquid surface, if the impact point is near the intersection line of three soap bubbles (plateau boundary) or near the interface of two soap bubbles (common liquid membrane),

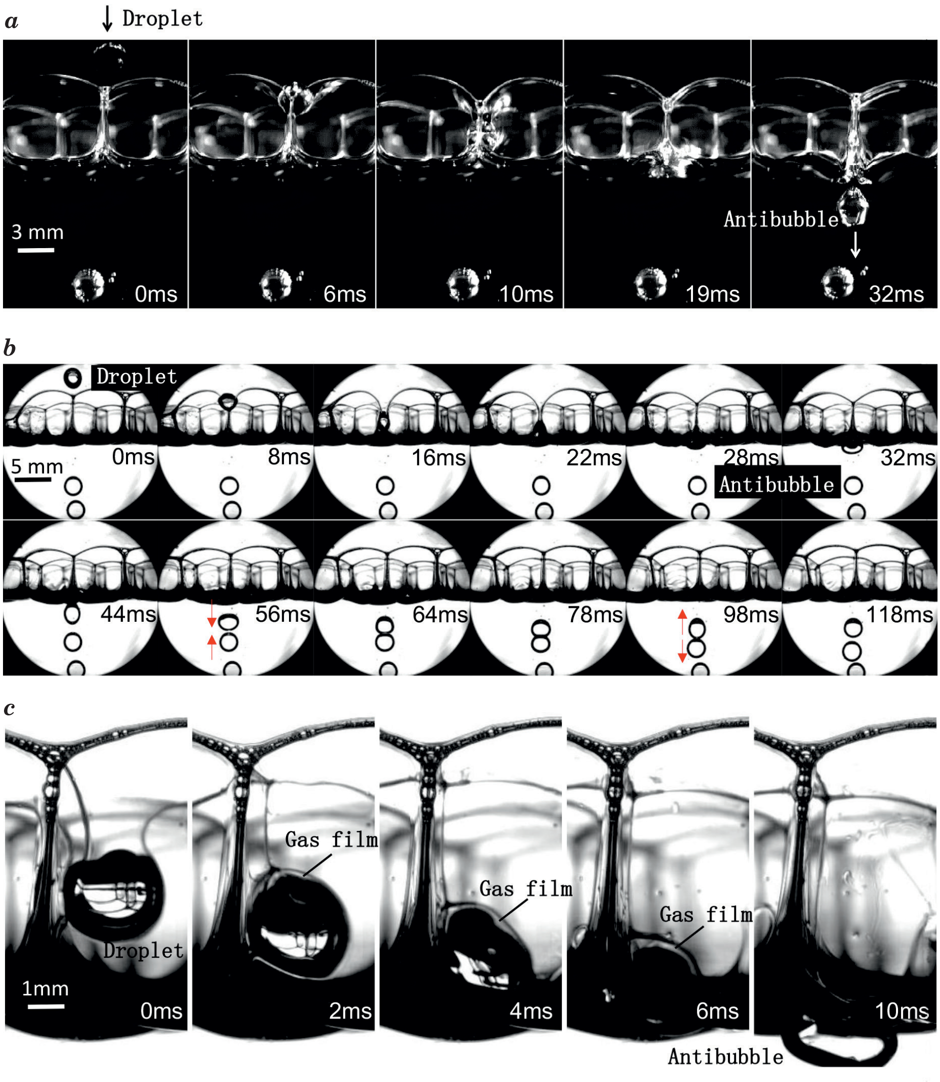

Fig. 4. Antibubble formation by hitting through a foam layer:
a – reflected light, *b* – transmitted light, *c* – detailed layout

because of the Gibbs-Marangoni effect, the surfactant molecules on the liquid film surface have the ability of self-repair to resist external disturbance, and the liquid film will deform without breaking. Because the hydrophobic end of surfactant in the liquid film of soap bubbles and interface of falling droplets is point towards the gas side, the plateau boundary or common liquid film of soap bubbles is separated by droplets, and there is a layer of gas film between the droplets and the liquid film (as shown in Fig. 4*c*). When the droplets with a layer of gas film and a layer of liquid film fall to the liquid surface, the liquid film and the liquid in the container are united, while the gas film still exists. Under the action of gravity and inertia, the droplets with gas film sink into the water and become antibubbles. The deformability of the foam layer causes the droplet to form a gas film before strongly impacting the liquid surface, which is the key to increase the production rate of the antibubbles. It is found in the experiment that when droplets impact near the dome ($\overset{\downarrow}{\frown}$) of the soap bubble, droplets will pass through the liquid film of the soap bubble, and then fall into the water. No antibubbles can be formed. However, if we reverse the bending direction of the liquid film ($\underset{\smile}{\downarrow}$), in most cases, an antibubble will be formed (as shown in Fig. 5).

In the experiment, a metal frame is used to support the liquid film. Because of gravity, the arc surface of the liquid film is concave. When the droplet passed through the liquid film, the droplet was wrapped by the liquid film, and there was a layer of gas film between the liquid film and the droplet (as shown in Fig. 5*a*). Reflected light was used to illuminate the surface of droplets. It can be clearly seen that the pressure wave caused by the lifting of the liquid film causes the fluctuation of the upper part of the droplet. A depression is formed at the top of the droplet and a micro bubble is wrapped into the droplet. This phenomenon is very common in the experiments of droplet penetrating the liquid film. If the droplet penetrates through two layers of liquid film, the droplet will be wrapped by two layers of liquid film and two layers of gas film (as shown in Fig. 5*b*). If the droplet penetrates through three layers of liquid film, three layers of liquid film and three layers of gas film will be wrapped outside the droplet (as shown in Fig. 5*c*).

It can be seen from Figure 6 that when the droplet impacts the liquid film, the liquid film is gradually stretched as the droplet falls. The surface tension of the liquid film tends to minimize the surface energy, which causes the liquid film to shrink toward the center and form a saddle shape. With the acceleration of contraction, the liquid film in the neck finally contacts each other and produces a liquid jet in the upper and lower directions of the neck. The liquid film in the metal frame and the liquid film encapsulating the droplets are closed separately. The liquid column between the two liquid films splits into micro droplets under the action of surface tension.

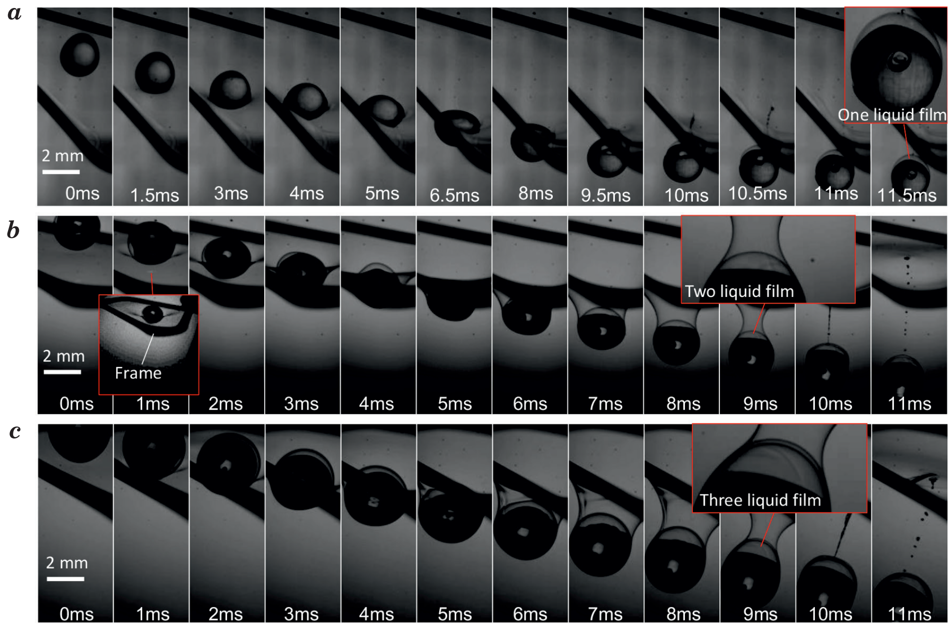

Fig. 5. Antibubble formation by hitting through a liquid film:
a – single layer liquid film, reflected light, *b* – double layer liquid film, transmitted light,
c – three layer liquid film, transmitted light

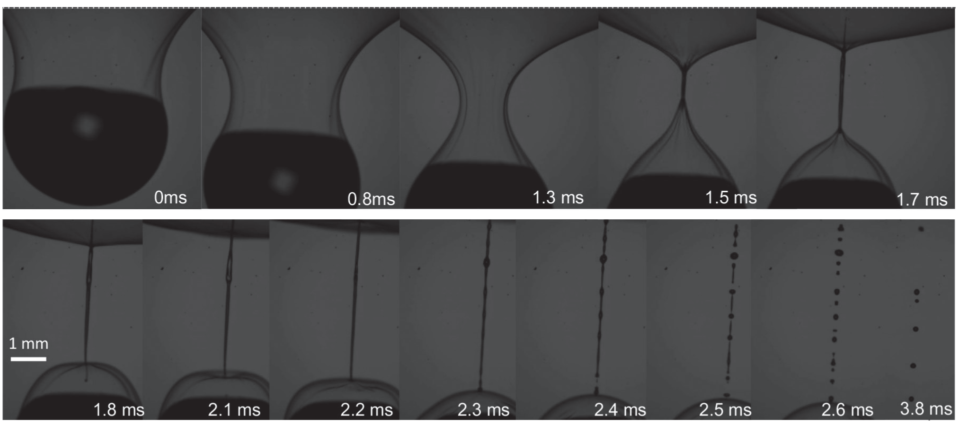

Fig. 6. The process of droplet packing by liquid film

Under the action of surfactants, ordinary droplets or droplets coated with one or more layers of liquid film and gas film usually form a layer of gas film between the outermost layer of the droplet and the liquid surface (as shown in Fig. 7*b* and *c*). The above-mentioned gas film holds the droplet floating on the liquid surface, forming a floating droplet or multilayer floating droplet. However, under the pressure of gravity and lifting force, the gas in the film will drain rapidly, and the film will become thinner until it breaks due to van der Waals force. After the film breaks down, the liquid film on the outermost layer of the droplet merges with the liquid in the container. Mechanically unstable droplets will undergo a series of deformation until the next stable state. For multi-layer floating droplets (as shown in Fig. 7*a*, liquid film and gas film can be clearly seen), when the gas film breaks down, the droplets gradually narrow in the horizontal direction, making the droplets nearly conical. Subsequently, as the droplet width increases gradually, the top of the droplet begins to drop rapidly, and a surface wave extending outward is formed on the surrounding liquid surface. As the droplet sinks, it can be clearly seen that the circular intersection line between the top of the droplet and the liquid surface gradually reduces to a point and disappears , while releasing surface waves to propagate outward (as shown in Fig. 7*a*, *b*). For ordinary floating droplets (as shown in Fig. 7*c*), when the film breaks down, the droplet diameter decreases horizontally and forms a vertical boundary while the height remains unchanged. The boundary gradually shrinks towards the droplet axis, and at the same time, obvious surface waves are formed on the droplet surface. Subsequently, the droplets evolved from a cylindrical shape to a conical shape, and then, while the upper diameter of the cone remained unchanged, the bottom contracted rapidly to form a small diameter cylinder, which then sank into the water. During the whole deformation process of the droplet, surface waves continue to release and expand to the surrounding liquid surface (as shown in Fig. 7*c*).

The multi-layer floating droplets will sink into the water to form antibubbles after the breakdown of the gas film, while the ordinary floating droplets

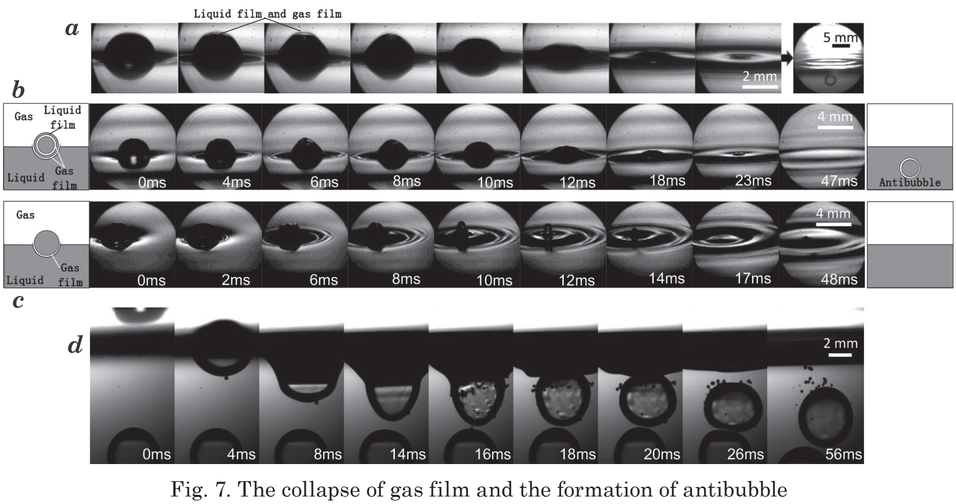

Fig. 7. The collapse of gas film and the formation of antibubble
(*a*) and (*b*) multilayer floating droplet (on the surface) (*c*) ordinary floating droplet
(*d*) multilayer floating droplet (under the surface)

will merge with the surrounding liquid after the breakdown of the gas film. The process of gas film breakdown and anti-bubble formation under liquid surface was photographed by high-speed photography under transmission light (as shown in Fig. 7*d*). When multi-film droplets fall on the liquid surface, there are thick black edges in the outer ring of the droplet and the area intersecting with the liquid surface due to the existence of gas film and the total reflection of light caused by it (as shown in Fig. 7*d*, τ = 8 ms). When the gas film breaks down (as shown in Fig. 7*d*, τ = 16 ms), the light transmitted from the droplet center appears very mottled due to the disturbance of the fluid and the change of the inner film thickness caused by the disturbance. At this moment, the black edge around the droplet still exists, indicating that the droplet is still encapsulated by the gas film. As the droplet sinks under the action of gravity and inertia force, an antibubble can be clearly observed below the liquid surface (as shown in Fig. 7*d*, τ = 56 ms).

The phenomenon of droplets bouncing on the liquid surface sometimes occurs when the gas film of ordinary floating droplets breaks down (as shown in Fig. 8). Every time the droplet bounces, it will become smaller. This is because when the gas film breaks down and the floating droplet deforms to the stage of thin cylinder, the lower part continues to shrink until it breaks, making the upper liquid form an independent smaller droplet. The smaller droplet falls to the surface again under the action of gravity, forming a new floating droplet. This newly formed floating droplet can undergo the same process to form another smaller droplet that falls onto the surface of the liquid. In this way, droplets can bounce up to four or five times on the surface of the liquid. The energy of these deformations comes from the surface energy released by film breakdown and droplet reduction.

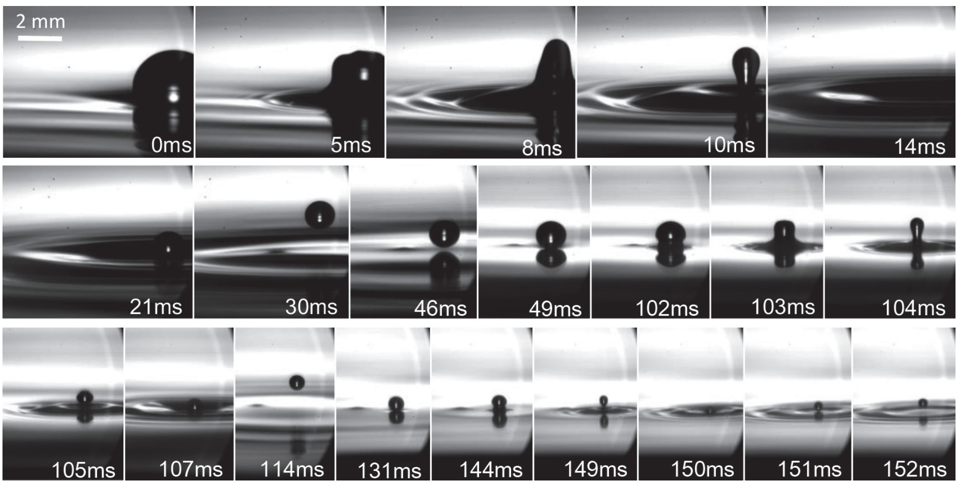

Fig. 8. Bouncing droplets on the liquid surface

Rupture of antibubble

Figure 4*a*, *b*, *c* and Figure 7*a*, *b*, *d* show the formation process of antibubbles. When droplets pass through several layers of liquid film and then fall on the liquid surface or fall on a single layer of foam, multi-layer antibubbles enveloped by several layers of gas film and several layers of liquid film may form under water. Because the gas density is smaller than that of water, the gas in the lower part of the anti-bubble film will gather up to the upper part, so the lower part of the film is very thin and the upper part is very thick. When the droplets are wrapped by several gas films, the black edges in the outer ring of the anti-bubble is much thicker at the top than at the bottom (as shown in Fig. 9). When one gas film breaks down in a multi-layer antibubble (whether inner or outer), the number of gas film layers decreases, but it does not lead to the disappearance of the whole antibubble (as shown in Fig. 10). The inner gas film of a multi-layer antibubble bursts at $\tau = 4$ ms. Many bubbles formed by gas film breakdown float up in the spherical droplets and coalesce at the top of the liquid inside the antibubble.

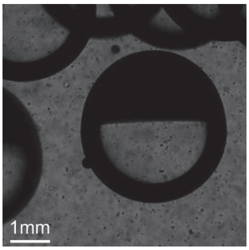

Fig. 9. Multilayer antibubbles

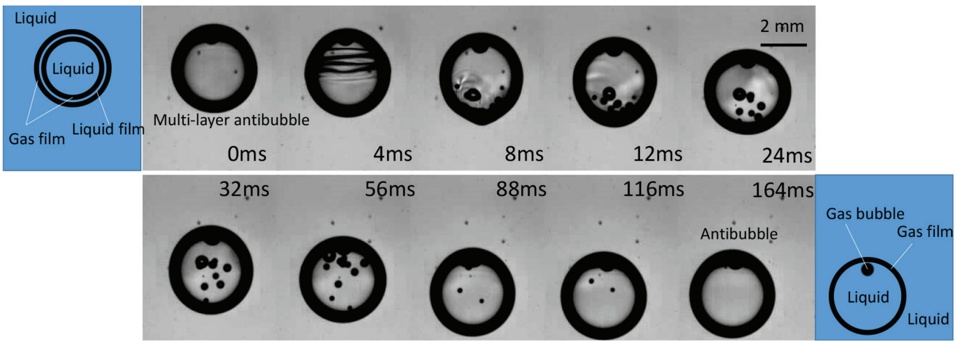

Fig. 10. The collapse of gas film of a multilayer antibubble

In order to explore the details of antibubble rupture, a single-gas-film antibubble breakdown process was photographed by a high-speed camera in a framing rate of 4000 fps and an exposure time of 1/178 000 s (as shown in Fig. 11). Because of the buoyancy of a small amount of gas in the antibubble gas film, the antibubble attached to the liquid surface (there is a layer of gas film between the antibubble and the liquid surface) slightly lifts the liquid surface to form a bump (τ = 0 ms). When the antibubble film breaks down at the lower right, the rest of the film remains intact, so the black edge caused by total reflection still exists. Due to total reflection, we cannot see the starting point of film breakage, but the surface wave generated by the breakdown has been formed and propagates to the upper left (τ = 0.50 ms). When the film disappears by one third, the black edge caused by total reflection in the lower right part disappears, and the outer edge of the damaged film and the small bubbles formed by fragmentation can be clearly seen (τ = 1.25 ms). After the antibubble collapses, a gas bubble is formed at the intersection with the liquid surface and attached below the liquid surface (τ = 4.75 ms).

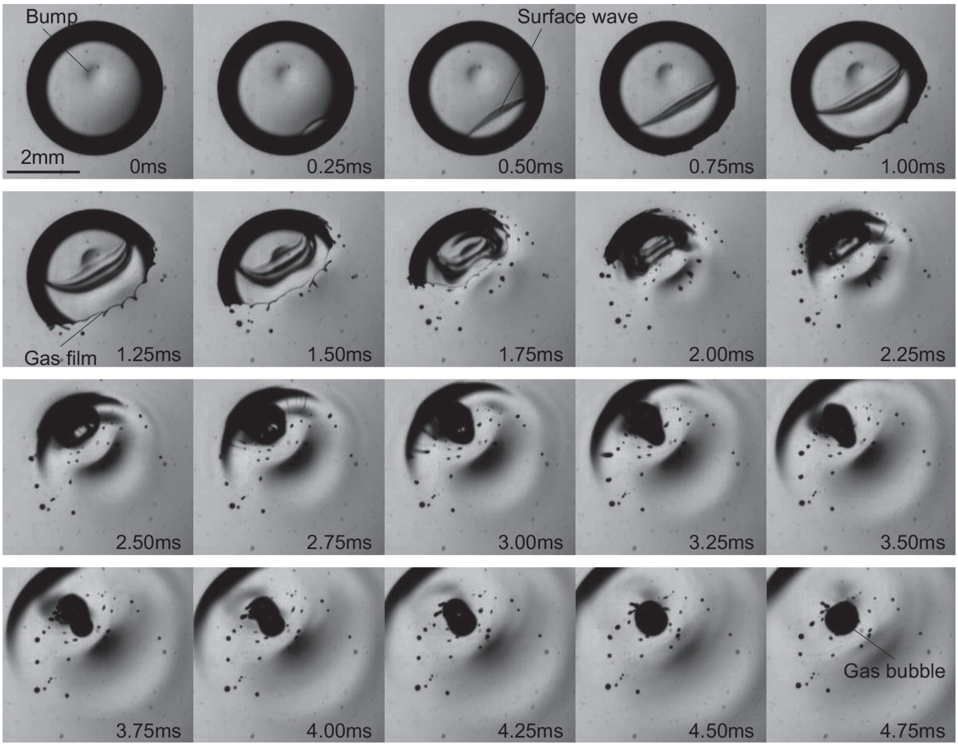

Fig. 11. The collapse of an antibubble

Coalescence of antibubbles

Due to the action of surfactants, two antibubbles usually bounce off when they collide with each other (Fig. 4*b*, τ = 56 ms – 118 ms; Fig. 7*d*, τ = 18 ms – 56 ms). However, if the film breaks when two antibubbles collide, the two antibubbles may merge with each other (as shown in Fig. 12). It is well known that there are two gas-liquid surfaces in the gas film of an antibubble. When two antibubbles collide, if the outer gas-liquid surface of the two gas films ruptures and merges, the two antibubbles will share an outer gas-liquid surface while retaining the original inner gas-liquid surface, forming an "8" shape structure. In this state, a shared gas film is formed between two droplets. If the shared gas film breaks down, in other words, the inner gas-liquid surface merges, then two droplets wrapped in the gas film merge, and two antibubbles merge into an antibubble. The coalescence of two antibubbles shows a similar merging process of soap bubbles. The present work is, to the best of the authors' knowledge, the first analysis about the coalescence of antibubbles.

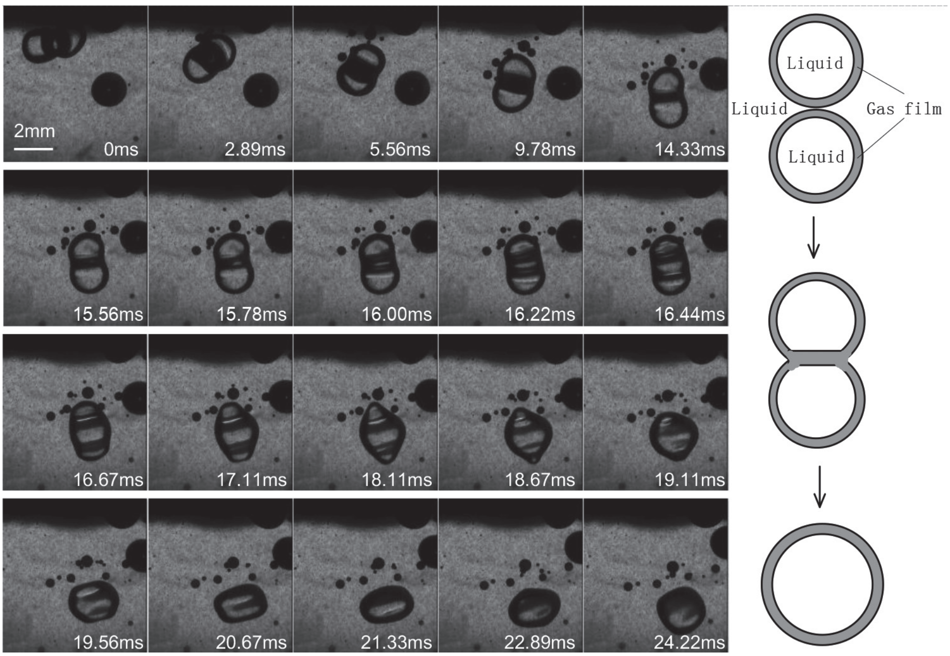

Fig. 12. The coalescence of antibubbles

Conclusion

The formation, rupture and coalescence of antibubbles are experimentally investigated by high-speed photography. The research focuses on the interaction between antibubble and film (gas film and liquid film) in these physical processes. It is found that if droplets or jets interacts with a liquid film or liquid membrane structure (foam) before impinging on the liquid surface, a gas film structure coated with liquid film will be formed. Then the impact on the liquid surface will greatly increase the survival rate of the antibubbles. This fluid structure encapsulated by gas film and liquid film impinges on the liquid surface, which will greatly improve the survival rate of antibubble. When droplets pass through several layers of liquid film and then fall on the liquid surface or fall on a single layer of foam, multi-layer antibubbles enveloped by several layers of gas film and several layers of liquid film may form under water. Because of the potential energy of the membrane structure, the rupture of films will produce new fluid structures. When two antibubbles interact, the rupture and coalescence of gas films can be decomposed into the process of the rupture and coalescence of two gas-liquid surfaces and different fluid structures can be formed.

Acknowledgments

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