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Formation of antibubbles and multilayer antibubbles

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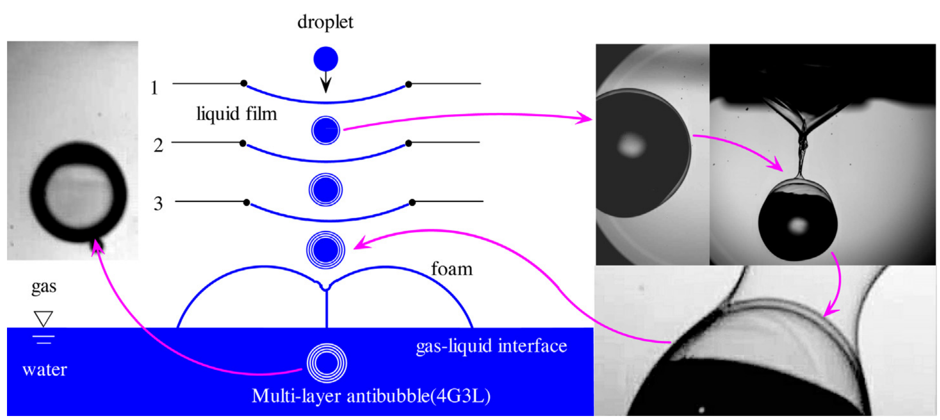
HIGHLIGHTS

- Multilayer antibubbles with 3 gas films and 2 liquid films were formed by liquid films and foams for the first time.
- The multilayer droplet on the gas-liquid interface can sink into the liquid becoming an antibubble.
- The authenticity of the existence of the multilayer antibubbles was verified.

GRAPHICAL ABSTRACT

By adding soap films between a falling droplet and a liquid pool, a multilayer antibubble can be generated. The complex hydrodynamic behavior was recorded by high-speed photography. The authenticity of the existence of the multilayer antibubble was verified.



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ABSTRACT

Soap bubbles and antibubbles are reversed-phase fluidic objects. A series of experiments were conducted in this paper to establish a link between soap bubbles and antibubbles. A new method was proposed to form antibubbles with the assistance of soap bubbles. The complex hydrodynamic behavior was recorded by high-speed photography. Multilayer antibubbles were formed by liquid films and foams for the first time. The authenticity of the existence of the multilayer antibubbles was verified.

1. Introduction

The soap bubble, as is well known to us all, is a thin spherical liquid shell with gas inside and outside. The soap bubbles floating in the air show brilliant colors in the sun because of interference of liquid film [1]. It is not so well known that a reverse phase construction may exist: the antibubble – a thin spherical gas shell containing liquid inside and surrounded by liquid outside. The antibubble floating in the water shows a thick black rim because of total reflection at the liquid – air interface [2]. Antibubble phenomenon was first reported by Hughes and Hughes [3] in 1932 and the term antibubble was coined by Stong [4] in 1974. Though several decades have

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passed since the antibubble was first reported and designated, few researches were conducted until this century on the formation [5–9], aging [10–13], collapse [14,15], stabilization [2,13,16,17], optical properties [18] and control [19,20] of antibubbles.

The formation of antibubbles is an important part and indispensable precondition of antibubble research and antibubble application (Many possible areas of their applications have been suggested [21]). An ordinary way to generate antibubbles is to gently drip or pour a small amount of the same liquid over the gas-liquid surface [14]. A gas film is formed around the liquid flow when separating the external liquid from the existing liquid. The liquid flow may break up due to the Rayleigh – Plateau instability. An antibubble will be formed from the breaking liquid flow under the effect of surface tension. This method was used by most researchers of antibubbles. Some minor improvements were also made to generate antibubbles, such as vibrating the nozzle and creating an oscillation in the incident jet [22], adding an electrical connection to prevent electrical potential difference due to triboelectric effects [2], and rationalizing several aspects of the optimal window in parameter space for creating antibubbles [8]. Other researchers developed totally different methods to generate antibubbles, such as bubbling method: to generate micron-sized antibubbles by capillary flow focusing [5], or generate millimeter-sized antibubbles by the coalescence between two bubbles [6]; and freeze-drying method: to produce 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 [20]; ultrasound method: in the ultrasonic field, 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 [7,9]. All of the above methods can not generate multilayer antibubbles. We coin the term 'multilayer antibubbles' to describe an antibubble with several gas films and several liquid films in the outer shell. Indeed there were reports of liquid onion (a droplet with several liquid films in the outer shell) [23], in which a liquid phase replaces the gas film. So the liquid onion is not antibubble. The major result of our paper is to propose a new method to form antibubbles and multilayer antibubbles. The generation of multilayer antibubbles will open up an interesting new area in the study of antibubbles.

2. Experimental

The experimental setup consists of the faucet, liquid films, foams, the high-speed imaging and illumination system, opto-electric switch, fixing and adjusting devices. We inserted liquid films and/or foam between the faucet and the gas-liquid interface. Droplets falling down from the faucet will hit through the liquid films and/or foam before sinking into the water and becoming antibubbles. The faucet and liquid films were mounted to the fixing and adjusting devices. Droplets falling down from the faucet will trigger the opto-electric switch, and then the high-speed photographer (Photron Fastcam SA-1, Photron Ltd., Japan) will record the process of antibubble formation. 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 liquid mixture and laboratory temperature are maintained at about 20 °C.

3. Results and discussion

Soap bubbles and antibubbles are present in the gas phase and liquid phase respectively, the thin shell is incompressible liquid and compressible gas respectively, and the orientation of surfactant molecules (hydrophilic head and their hydrophobic tail) is also the opposite. Few relationships were discussioned between the two fluidic objects except the reversation of physical properties and the similarity of structures. In fact, soap bubble and antibubble can transform into each other under certain condition (as shown in Fig. 1(a)). When the liquid film of a soap bubble (A) collapse, droplets (B) will be formed (surface energy of soap bubble converts into the kinetic energy of the liquid). When a droplet (B) drops on the gas-liquid interface, a globule (C) will be formed (potential energy converted into kinetic energy, then into surface energy). If the kinetic energy of the falling droplet (B) is large enough, the globule (C) will sink into the liquid and transform into an antibubble (D). The antibubble (D) may collapse due to the drainage of the gas and action of van der Waals forces [12], and bubbles (E) will be formed (surface energy of antibubble convert into the kinetic energy of the fluid). Bubbles (E) float to the interface and become foams (F) (potential energy converted into surface energy). The foams (F) can break away from surface under certain forces and become soap bubble (A) again. This is indeed a cycle, though some transformation needs additional energy and the volume will become smaller.

Soap bubble and antibubble exist in different phases; however, they still can interact with each other. Soap bubble is not spherically symmetric, but cylindrically symmetric. The lower part of the liquid shell is thicker than the upper because of gravity [1]. Imagine, if the lower part of the liquid shell infinite thickening, a soap bubble (A) will become foam (F). For the liquid film, there is no essential difference between them. Antibubble is not spherically symmetric, but cylindrically symmetric. The upper part of the gas shell is thicker than the lower because of buoyancy [2]. Imagine, if the upper part of the gas shell infinite thickening, an antibubble (D) will become a globule (C). For the gas film, there is no essential difference between them. So, the interaction at the contact point between foam (F) and globule (C) is actually the interaction between soap bubble (A) and antibubble (D). It is found that there is a multilayer structure at the contact point (1G1L, i.e. one gas film one liquid film, as shown in Fig. 1(b)). We introduce soap bubble (A) or foam (F) to the transformation process of droplet (B) to globule (C) (as shown the dotted curve with arrow in Fig. 1(a)). With the help of multilayer structure, we can generate antibubble and multilayer antibubble very easily.

3.1. Foam

When we generate antibubbles with traditional method (pouring liquid over the gas-liquid interface), it is required that no foam should appear on the surface [8]. As a result, overflow is used to keep the surface clean. A foam layer was produced on the surface deliberately in our experiment to generate antibubbles (as shown in Fig. 2(e); the diameter of the droplet is 0.8 mm–1.6 mm; the impact velocity of droplet is about 1.1 m/s.). As far as we know, this is the first report on the formation of antibubbles by foams. The formation process was recorded by high-speed photography.

It is found in our experiment that a multilayer structure (1G1L, i.e. one gas film plus one liquid film, as shown in Fig. 2(b)) is formed between the droplet and the foam when the droplet passes through the foam layer. The liquid film of foam bubbles is stretched, and the foam bubbles deformed slightly. The multilayer structure is well preserved without breaking up during the impact in most cases. Antibubbles are most likely to be formed when the droplets drop on the interface of two foam bubbles or on the plateau border.

Fig. 2(a–d) shows the schematic diagram of the process of a droplet passing through foams and generating an antibubble. Fig. 3 shows the high-speed photos of this process (the diameter of the droplet is 3.3 mm). It is found that the droplet is pushed toward the interface of two foam bubbles or plateau border by the foam bubbles when the droplet hits the foam (the impact velocity of the droplet is about 0.9 m/s, as shown point A in Fig. 3(d)) because foam bubbles tend to minimize the surface energy [24]. When the droplet

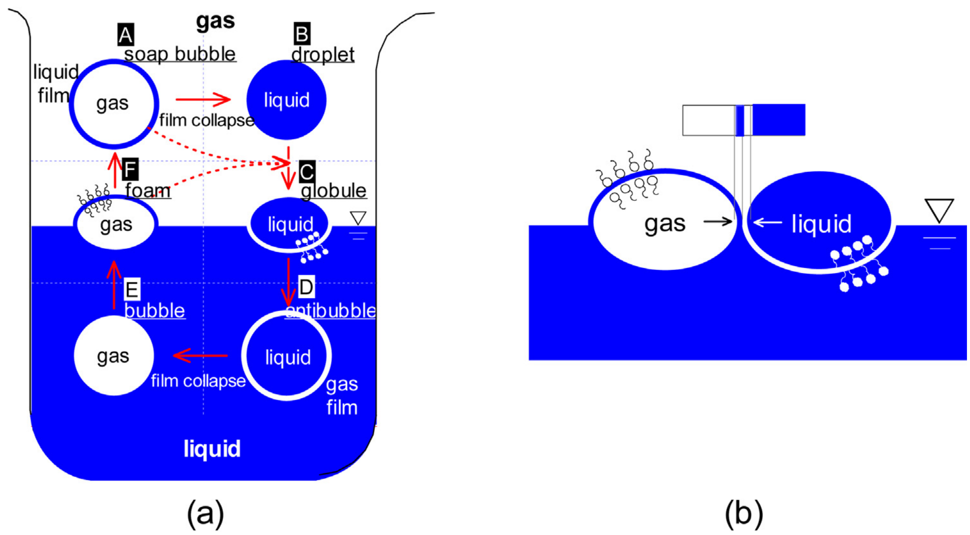

Fig. 1. (a) Cycle of fluidic objects. (b) Interaction between soap bubble and antibubble.

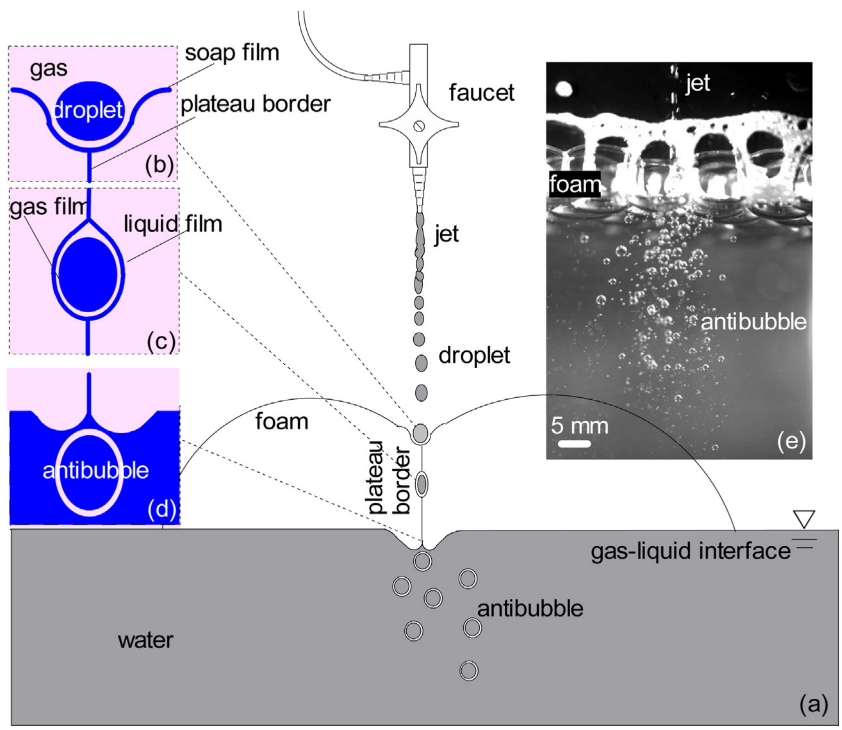

Fig. 2. Schematic diagram of antibubble formation (foam method).

falls into the interface of two foam bubbles (or plateau border) completely, two (or three) curved plateau borders are formed due to the existence of gas film. The droplet surrounded by an air film and a liquid film moves downwards under the gravity. The included angle (on the droplet side) of plateau border (on the lowermost point) is greater than 120° and the lowermost point moves downwards. The included angle (on the droplet side) of plateau border (on the uppermost point) is less than 120° and the uppermost point moves downwards. Hence the droplet and the two layers move downwards keeping the multilayer structure. When the droplet falls to the bottom of the interface of two foam bubbles (or plateau border) (the velocity of the droplet is about 0.5 m/s, as shown point B in Fig. 3(d)), the liquid film is opened, and the droplet surrounded by a gas film falls into the liquid. An antibubble is formed.

3.2. Liquid film

Liquid layers also help to generate antibubbles. A droplet passes through a liquid film before dropping to the gas-liquid interface (as shown in Fig. 4(A)). A special fluidic object is formed: the multilayer droplet – a droplet with several gas films and several liquid films in the outer shell. The formation process was recorded by high-speed photography (as shown in Fig. 5(A)). A gas film is formed between the droplet and the liquid film when the droplet falls on the liquid

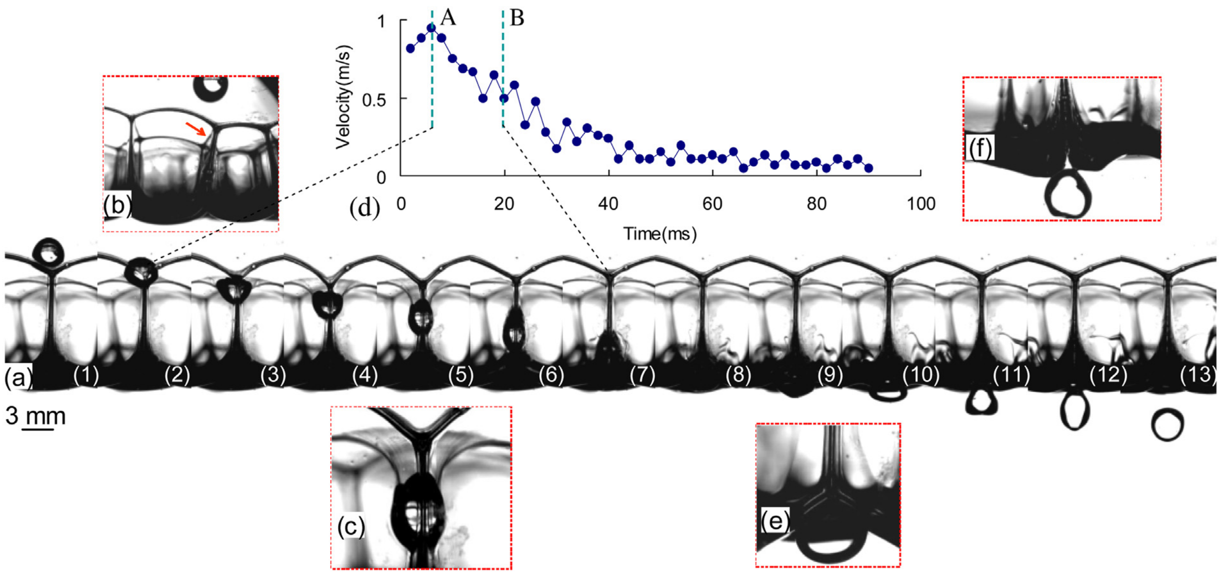

Fig. 3. (a) A droplet passes through foams and generates an antibubble. (b) The droplet hits the interface (public liquid film) of two soap bubbles. (c) The soap bubbles deform and encase the droplet. (d) Droplet falling velocity during the process of antibubble formation. (e) (f) The droplet coated with gas film sank into the water.

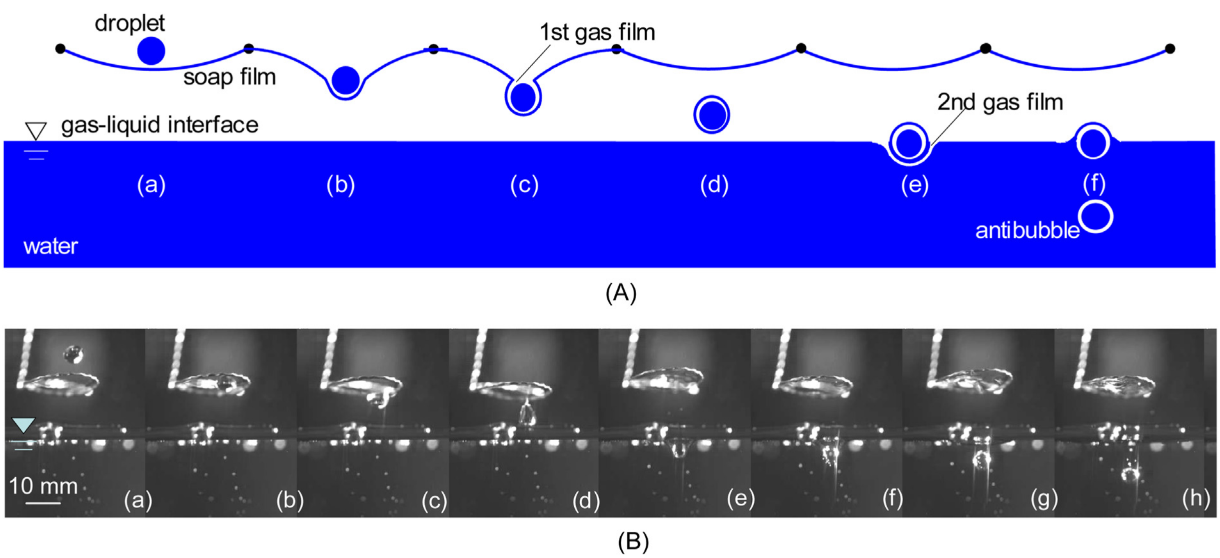

Fig. 4. Schematic diagram and photos of antibubble formation (liquid film method).

film. The liquid film stretches and wraps the droplet completely under the surface tension. The multilayer droplet (1G1L) falls on the gas-liquid interface, and another gas film is formed between the multilayer droplet and the interface. When the second gas film breakups, the liquid film of the multilayer mixes into water, and the droplet surrounded by a gas film sinks into the liquid becoming an antibubble (as shown in Fig. 4(B); the diameter of the droplet is 0.6 mm; the impact velocity of the droplet is about 1.3 m/s, the distance between the liquid film and gas-liquid interface is 15 mm).

Fig. 5(A) shows the process of multilayer droplet formation (the diameter of the droplet is 4.6 mm; the impact velocity of the droplet is about 1.6 m/s.). When the droplet impacts the liquid layer, the liquid layer is stretched and the stretching waves spread outwards. The gas film between the droplet and the liquid film is too thin to be observed, but we can perceive its existence during its breaking up (as shown in Fig. 5(A–b)). The surface of the droplet fluctuates because of the impact with liquid film (as shown in Fig. 5(A–c)). The fluctuation is so large sometimes that a small gas bubble can be involved in the droplet. The neck of liquid film gradually shrinks because of surface tension [1] (as shown in Fig. 5(A-a(9, 10)). The peripheral walls of the neck eventually bond together and scatter into small droplets because of Rayleigh – Plateau instability (as shown in Fig. 5(A-a(11, 12), d, e and f)). The liquid film and gas film is closed. The upper part of the gas film is thicker than the lower in the beginning, but it will turn into uniform thickness over time (as shown in Fig. 5(A-g, h, i)).

Fig. 5(B and C) shows the transformation of multilayer droplets into antibubbles. A gas film is formed between a multilayer droplet and the gas-liquid interface. The multilayer droplet floats stably on the interface after fluctuating up and down several times. Most gas accumulates in the upper part of gas film because of the high pressure from the lower part. The lower part of the multilayer droplet is dark because of the total refection at the liquid – gas interface (2G1L) before the second gas film breaks up. The gas film between the multilayer droplet and the interface (2nd gas film) will break up because of aging or disturbance. The lower part of the multilayer droplet will become pervious to light when the second gas film breaks up (as shown in Fig. 5 C(2)) (the thick black outer edge still

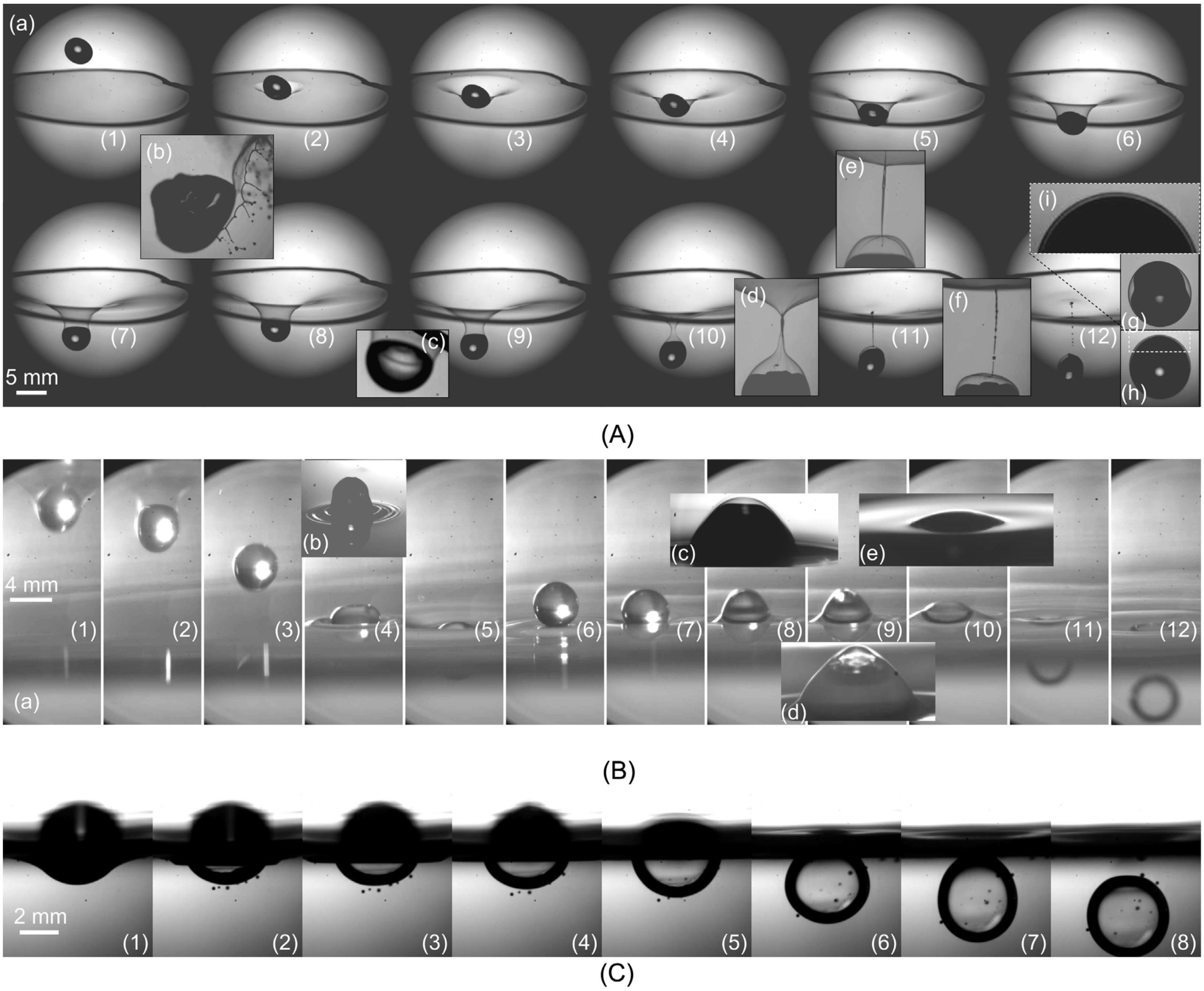

Fig. 5. (A) Formation of droplets coated with a layer of gas film and a layer of liquid film (multilayer droplets, 1G1L). (B) The transformation of multilayer droplets to antibubble (over the surface). (C) The transformation of multilayer droplets to antibubbles (below the surface).

exists due to the first gas film). Without the lift (surface tension) of second air film, the fluidic object sinks into water becoming an antibubble.

3.3. Multilayer antibubble

Combining the above two methods, we can generate multilayer antibubbles (as shown in Fig. 6). The droplet passes through several liquid films and becomes a multilayer droplet coated by several gas films and several liquid films. Then the multilayer droplet passes through a layer of foams and becomes a multilayer antibubble. Fig. 7(A–C) shows the high-speed images of the formation of multilayer droplets (1G1L, 2G2L, 3G3L). When the multilayer droplet sinks to the liquid and becomes a multilayer antibubble, we can not observe the layers directly because of the total reflection at the liquid–air interface. A proper way to ascertain the numbers of layers is to count how many times the gas films will break up (as shown in Fig. 8). The multilayer antibubble (the diameter of the droplet is 3.9 mm) in Fig. 8(1) has three gas films and two liquid films. The outer gas film (the average thickness of the gas film is 35 μm) breaks up in Fig. 8(2) and forms a small bubble that floats up. The multilayer antibubble in Fig. 8(3–5) has two gas films and one liquid film. The outer gas film (the average thickness of the gas film is 8.3 μm) breaks up in Fig. 8(6) and forms the second small bubble that floats up. The multilayer antibubble in Fig. 8(7–10) is an ordinary antibubble (1G0L). The only gas film (the average thickness of the gas film is 9.6 μm) breaks up in Fig. 8(11) and forms

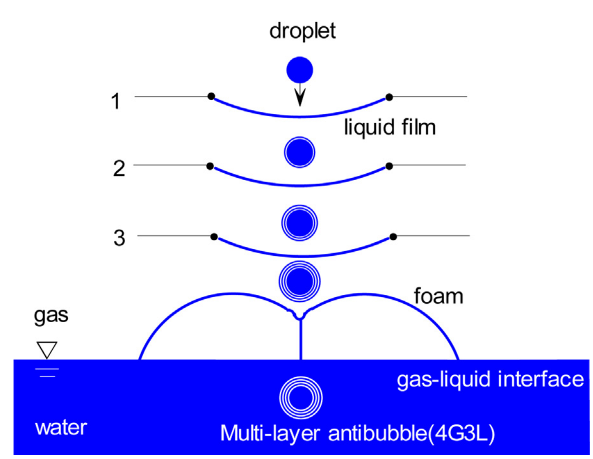

Fig. 6. Schematic diagram of multilayer antibubble (4G3L) formation.

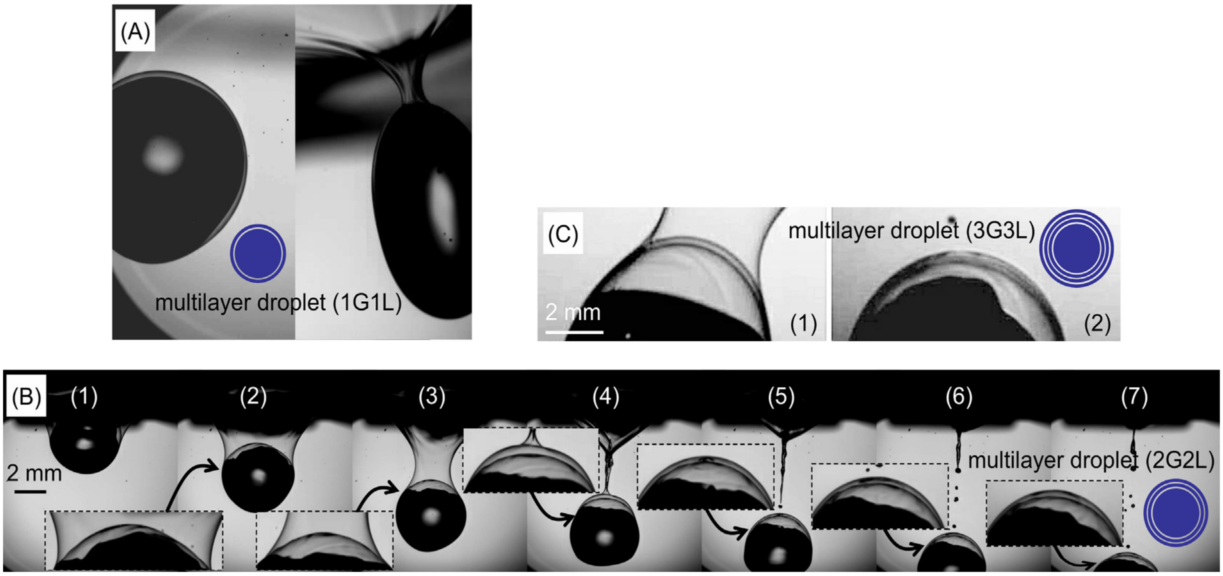

Fig. 7. High-speed images of multilayer droplets (A) 1 gas film and 1 liquid film. (B) 2 gas films and 2 liquid films. (C) 3 gas films and 3 liquid films.

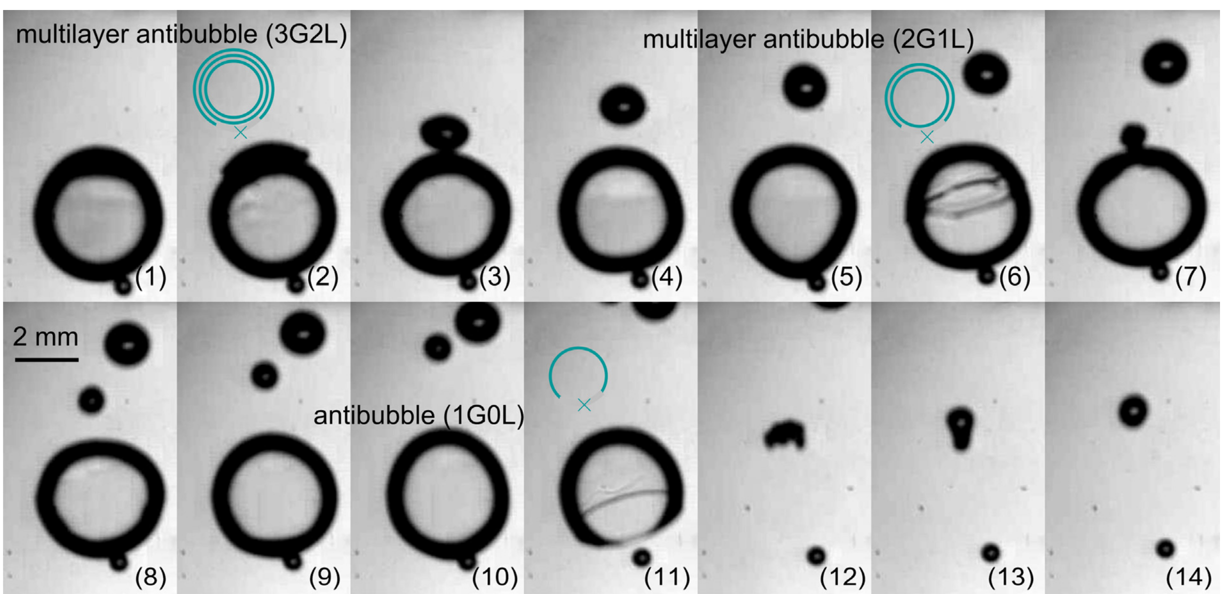

Fig. 8. The collapse of multilayer antibubbles.

the third small bubble that floats up. The gas films of multilayer antibubbles do not necessarily break up from outside. It was also found that the inner gas film may break up first some times.

4. Conclusions

This paper focused on dynamic process of antibubble formation. A series of experiments were carried out on the soap bubbles and antibubbles. New methods of generating antibubbles were found. The physical process was observed detailedly by high-speed photography. Multilayer antibubbles were formed by liquid films and foams for the first time. Regardless of the type of surfactant used, antibubbles can be easily formed by foams as long as there is a stable foam layer on the liquid surface. The authenticity of the existence of the multilayer antibubbles was verified.

Acknowledgments

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