

Gas marbles: ultra-long-lasting and ultra-robust bubbles formed by particle stabilization

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Abstract Bubbles and foams are ubiquitous in daily life and industrial processes. Studying their dynamic behaviors is of key importance for foam manufacturing processes in food packaging, cosmetics and pharmaceuticals. Bare bubbles are inherently fragile and transient; enhancing their robustness and shelf lives is an ongoing challenge. Their rupture can be attributed to liquid evaporation, thin film drainage and the nuclei of environmental dust. Inspired by particle-stabilized interfaces in Pickering emulsions, armored bubbles and liquid marble, bubbles are protected by an enclosed particle-entrapping liquid thin film, and the resultant soft object is termed gas marble. The gas marble exhibits mechanical strength orders of magnitude higher than that of soap bubbles when subjected to overpressure and underpressure, owing to the compact particle monolayer straddling the surface liquid film. By using a water-absorbent glycerol solution, the resulting gas marble can persist for 465 d in normal atmospheric settings. This particle-stabilizing approach not only has practical implications for foam manufacturing processes but also can inspire the new design and fabrication of functional biomaterials and biomedicines.

Keywords bubble, particles, interfaces, armored bubble, liquid marble, gas marble, Pickering emulsion

1 Stability of soap bubbles

Soap bubbles are small gas pockets enclosed by liquid films in an air environment. They are commonly seen in

children's play and artistic performances. They are also building blocks that constitute foams, which are ubiquitous in our daily life and industrial processes ranging from foods, cosmetics and medicines to mining. Studying the behaviors of bubbles is crucial for these foam manufacturing industrial processes; their varied behaviors also attract the attention of researchers [1,2]. For instance, bubble films tend to minimize their surface area under given boundary conditions; thus, they are representative experimental models of minimal surfaces for verifying complex mathematical problems involving minimal optimization [3,4].

Soap bubbles are thought to be fragile and transient, and their rupture is related to viscous surface tension as well as Marangoni and nuclei effects depending on the composition of the bubble shell and the surrounding environment [1,3]. Without any stabilizers, the bursting of bare bubbles is primarily caused by the gravity-induced drainage of the liquid film, the thickness h of which follows the dynamics [5] $h = h_0 \exp(-t/\tau)$, where τ is the characteristic time of drainage scaling as $\eta/\rho g R$, h_0 denotes the initial film thickness, η is the liquid viscosity, ρ is the liquid density, and R is the bubble radius, respectively. When the bubble surface is thinned to a critical value, normally on the order of tens of nanometers, long-range van der Waals interactions accelerate the thinning process, and the bursting of bubbles consequently occurs [5]. Increasing the liquid viscosity of the liquid film can prevent film drainage and prolong the lifetime of bubbles [6]. However, bare viscous bubbles are still transient and can only last seconds. By adding surfactants to the bubble shell, surfactant molecules can induce the Marangoni effect on the surface or even immobilize surface boundaries [7], which significantly prevents film drainage and can promote bubble life to minutes. Even so,

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surfactant-stabilized bubbles eventually rupture due to liquid evaporation and/or the nucleation of holes caused by dust in the surrounding environment. In a dustless, vibration-free environment with saturated vapor atmospheres to suppress the nuclei and to prevent the evaporation of liquid, a bare viscous bubble can reach a lifetime as long as 2 years [5]. However, in a normal environment, overcoming film drainage, evaporation, and nuclei effects and achieving a long bubble lifetime are challenging tasks [8].

2 Particulate interfaces: particle-covered droplets in liquid, particle-covered bubbles in liquid and particle-covered droplets in air

Since Ramsden [9] and Pickering [10] found a century ago that particles are surface reactive and can therefore adsorb onto interfaces, particles have been intensively used as particulate agents to stabilize multiphase interfaces similar to surfactant molecules (Fig. 1). For instance, particles can occupy the interface of immiscible liquids, stabilizing either water-in-oil or oil-in-water emulsions [11–15], as shown in Figs. 1(a)–(c). These particle-stabilized emulsions are called Pickering emulsions [16], named after S.U. Pickering, who discovered them in 1907 [10]. In addition, particles can adsorb on the liquid–air interface and thus serve as stabilizers for air bubbles in liquid, forming particle-covered bubbles in a liquid environment, which are also known as “armored bubbles” [17–23] (Figs. 1(d–g)). This can also coat liquid droplets in air, forming particle-covered droplets termed “liquid marbles” [24–30]. Particles can even hang onto interfaces with ultralow interfacial tensions, such as in the interface of aqueous two-phase systems. The two immiscible aqueous phases are formed by phase separation in an aqueous solution dissolved with polymers, biomolecules and salts [31–33]. Recent works have revealed that protein particles [34] and fibrils [35,36] can effectively stabilize aqueous-in-aqueous emulsions.

The accumulation of particles on the interface is driven by the minimization of the total surface energy of the system [13,41,42]. For instance, the spherical particle straddled at the immiscible interface has an adsorption energy of $\Delta G = \pi a^2 \gamma (1 - |\cos\theta|)^2$, where a denotes the radius of the colloidal particle, γ indicates the interfacial tension, and θ is the contact angle at the interface [41]. For particles with nonspherical shapes, such as rod-like particles and disk-like particles, the adsorption energies at the fluid surface are even larger than those with spherical particles of the same volume, thereby strengthening their attachment to fluid interfaces [39,41,43]. For most particle-laden interfaces, the adsorption energy of particles is several orders higher than the thermal energy

kT , with k and T being the Boltzmann constant and the temperature, respectively. As a result, the adsorption of particles at interfaces is irreversible, which differs from surfactant molecules that constantly adsorb and desorb. More interestingly, Janus particles that have two distinct surface regions with opposite chemical compositions and wetting properties are considerably more effective than homogeneous particles in stabilizing multiphase interfaces [44–47].

3 Gas marbles: a recently discovered particle-covered bubble in air

Particulate materials, such as fat globules and protein aggregates, have been applied extensively for stabilizing foams in the food industry [2,41]. Most studies have focused on the collective behaviors of foams against coalescence and flocculation, which are crucial for the quality and shelf life of foam-based food products. In comparison, research on the individual behavior of particle-stabilized bubbles has been inadequate. Recently, a compact monolayer of microparticles has been demonstrated to straddle on air/liquid/air interfaces and stabilize a single bubble, forming a new soft object called a “gas marble” [8,48,49], as shown in Fig. 2. A gas marble consists of gas coated by a layer of particles that entrap a liquid thin film exposed to the atmosphere, as shown in Figs. 2(a,b). Although the states of their constituent phases are different, the appearance of a gas marble is similar to that of a liquid marble. Importantly, the delimiting particle armor in liquid marble straddles at the single-layer liquid–gas interface, while that in gas marble straddles at the bilayer liquid–gas interfaces, as shown in Fig. 2(a).

Compared to soap bubbles, a gas marble with particle-entrapping liquid film has significantly higher robustness [48]. The mechanical stability of a gas marble can be characterized by measuring the pressure difference $\Delta P = P_b - P_{\text{atm}}$ that causes bursting, where P_b and P_{atm} denote the inner pressure of the gas marble and the atmospheric pressure, respectively. There are two scenarios in which a gas marble can rupture: overpressure $\Delta P^+ > 0$ when the gas marble undergoes an inflation test, and underpressure $\Delta P^- < 0$ while a gas marble is in the deflation process. It has been found that particulate bubbles can sustain both overpressures and underpressures with amplitudes ~ 10 times greater than the Laplace pressure, $\Delta P_{\text{cap}} = 4\gamma/R_b$ (a gas marble has two liquid/air interfaces), which suggests that the particle monolayer at the thin liquid film dramatically improves the stability of bubbles [48]. The outstanding mechanical strength is attributed to the strong cohesive nature of the particle-assembled shell on the bubble surface. More interestingly, the normalized pressures $\Delta P^+/\Delta P_{\text{cap}}$ and $\Delta P^-/\Delta P_{\text{cap}}$ of a gas marble are

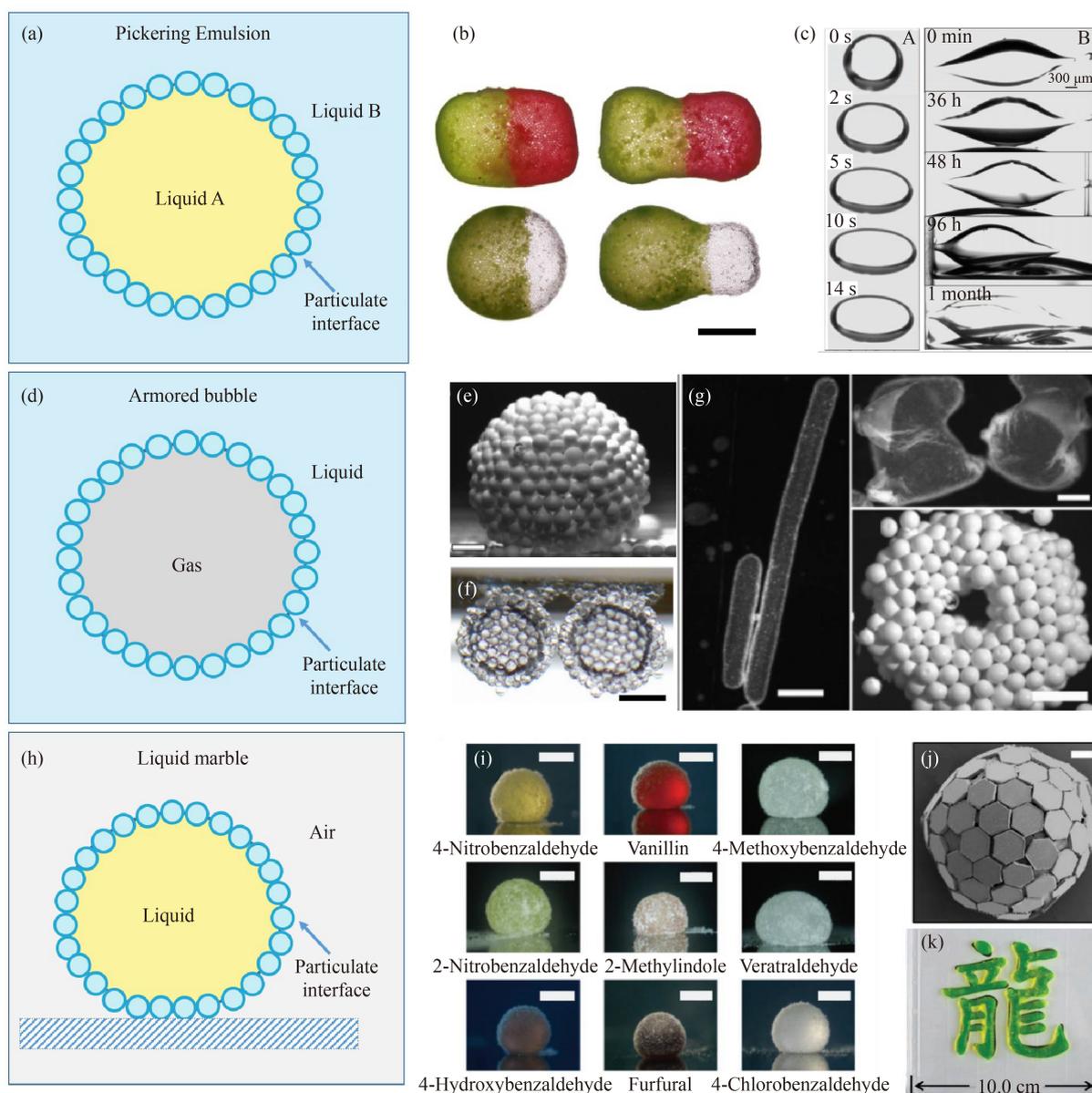


Fig. 1 (a) Schematics of a Pickering emulsion. (b) Asymmetric Janus Pickering emulsions through particle jamming of coalesced emulsions. The scale bar is 500 μm . Reproduced with permission from Ref. [15], copyright 2014, Springer Nature. (c) The deformation and stability of Pickering emulsions in an electric field. The scale bar is 300 μm . Reproduced with permission from Ref. [11], copyright 2013, The American Association for the Advancement of Science. (d) Schematics of an armored bubble. (e) Optic images of a spherical armored bubble. The scale bar is 400 μm . Reproduced with permission from Ref. [18], copyright 2006, American Chemical Society. (f) Two floating armored bubbles do not coalesce due to particle stabilization. The scale bar is 200 μm . Reproduced with permission from Ref. [37], copyright 2020 Elsevier. (g) Nonspherical armored bubbles with various shapes [18]. The scale bar is 200 μm . (h) Schematics of a liquid marble. (i) Photographs of liquid marbles encapsulating various chemical solutions. The scale bar is 2 mm. Reproduced with permission from Ref. [38], copyright 2019, Wiley-VCH. (j) SEM image of a dried polyhedral liquid marble stabilized by hexagonal fluorinated PET plates. The scale bar is 200 μm . Reproduced with permission from Ref. [39], copyright 2019, Wiley-VCH. (k) Complex particle-stabilized liquid/air surfaces forming a complex structure representing a Chinese dragon symbol. The scale bar is 10 cm. Reproduced with permission from Ref. [40], copyright 2018, Wiley-VCH.

much larger than those of liquid marbles and armored bubbles [23,50], as shown in Fig. 2(c). The significant differences in underpressures between gas marbles and liquid marbles come from the capacity of gas marbles to resist fluid loading up to 10 times the Laplace pressure of corresponding bare bubbles, whereas liquid marbles do not possess any strength for such a solicitation mode [48].

4 Gas marbles represent ultra-long-lasting bubbles in the atmospheric environment

Gas marbles are more robust than bubbles, liquid marbles and armored bubbles underwater [48]. Do they have a longer life than bubbles with no particles? A recent work

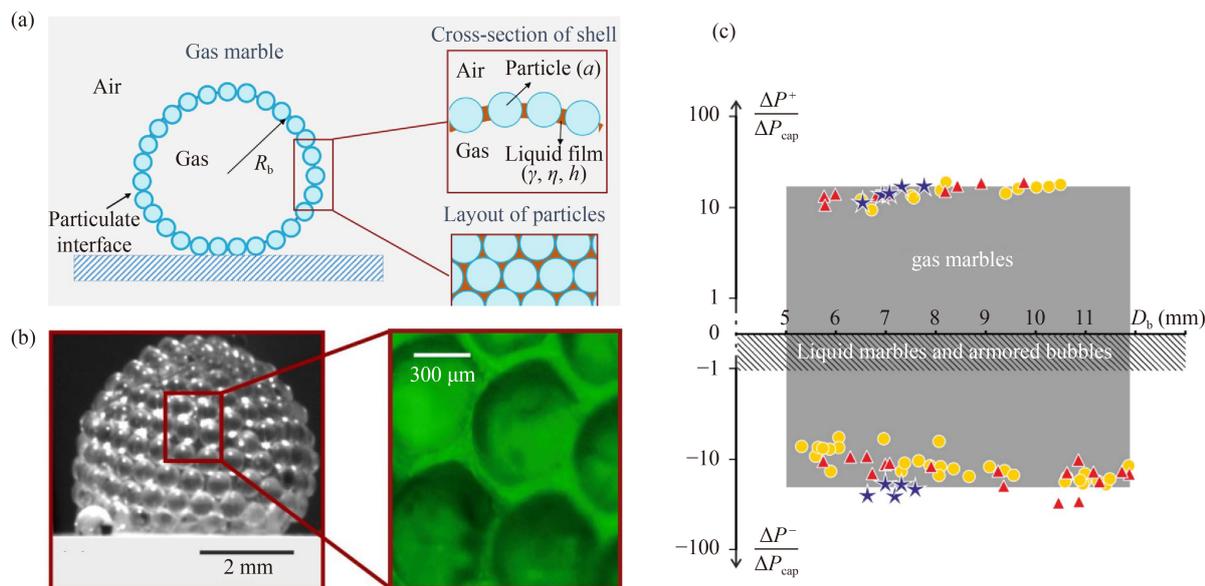


Fig. 2 (a) Schematic of a gas marble. Insert illustrating the cross-section of the gas marble shell and the layout of particles on the marble surface. (b) Optical image of a gas marble. The fluorescent picture demonstrates the enlargement of the particle layout. (c) Comparison of mechanical stability among gas marbles, liquid marbles and armored marbles at different sizes of bubbles and drops (D_b). Both the critical overpressures (ΔP^+) and underpressures (ΔP^-) are normalized by capillary pressure (ΔP_{cap}) to make a fair comparison. Reproduced with permission from Ref. [45], copyright 2017, American Physical Society.

demonstrated that particle-stabilized bubbles can maintain their integrity for more than 1 year in a standard atmosphere [8]. The ultra-long-lasting bubble is a gas marble featuring a particle-entrapping thin film of glycol aqueous solution. Its long life is attributed to the conduction of film drainage, liquid evaporation and gas diffusion, which accounts for the otherwise transient and fragile nature of bare bubbles. First, the particle shell can slow the drainage of the film through wetting forces. These partial-wetting particles adsorb on the two liquid interfaces, forming a monolayer through cohesive attractions. This monolayer traps the liquid by capillarity, makes the liquid passages constricted and tortuous, and thus significantly hinders the overall drainage within the thin film [41]. Second, the particles on the film can reduce the area of the surface across which gas diffuses, thus making the particulate film less permeable to gas than their pure liquid counterparts. The low gas permeability of the film slows the aging of gas marbles since evaporation is inhibited [49]. For instance, a normal water soap bubble ($R_b = 3.7$ mm) could burst within 1 min because of evaporation. When coated with the particles, the lifetime of a water bubble with an identical radius can be prolonged to 9 min, as shown in Fig. 3(a). The phenomenon wherein particulate film reduces evaporation also exists in liquid marbles. It has also been suggested that the higher the surface coverage is, the lower the evaporation rate [51]. Last but not least, to further slow the evaporation and make the bubble longer lasting, a mixture of water and glycerol can be formulated to generate a gas marble (Fig. 3). Glycerol has a strong affinity for water molecules due to its rich hydroxyl

groups. Thus, the glycerol within the particulate film of bubble can absorb water molecules contained in air, which compensates for water evaporation and enhances bubble stability (Fig. 3(a)). In a gas marble, adsorption and evaporation of water can be balanced, and the resultant marble can be further stabilized by optimizing the glycerol mass ratio and relative humidity, as summarized in the phase diagram shown in Fig. 3(b) [8].

5 Perspectives of gas marbles

Recent progress in particle-stabilized bubble with ultra-robustness and ultra-long-lasting life not only extends our understanding of particulate-stabilized interfaces but could also have important implications for applications. Gas marbles can inspire the design and fabrication of novel materials. For instance, highly robust bubbles could inspire new strategies for foam stabilization, which is crucial for developing foam-based materials or products [1,2,23]. By inhibiting the coalescence of foams on the level of a single bubble, well-controlled aerated materials can also be designed. Additionally, particulate-stabilized bubbles with high mechanical strength and long life can be exploited as gas storage materials. By designing the film composition and inhibiting the gas permittivity, valuable or polluted gases can be encapsulated inside the bubbles with insignificant gas diffusion/exchange with the environment.

Gas marbles can also be utilized as a new type of confined microreactor. For instance, we can use gas

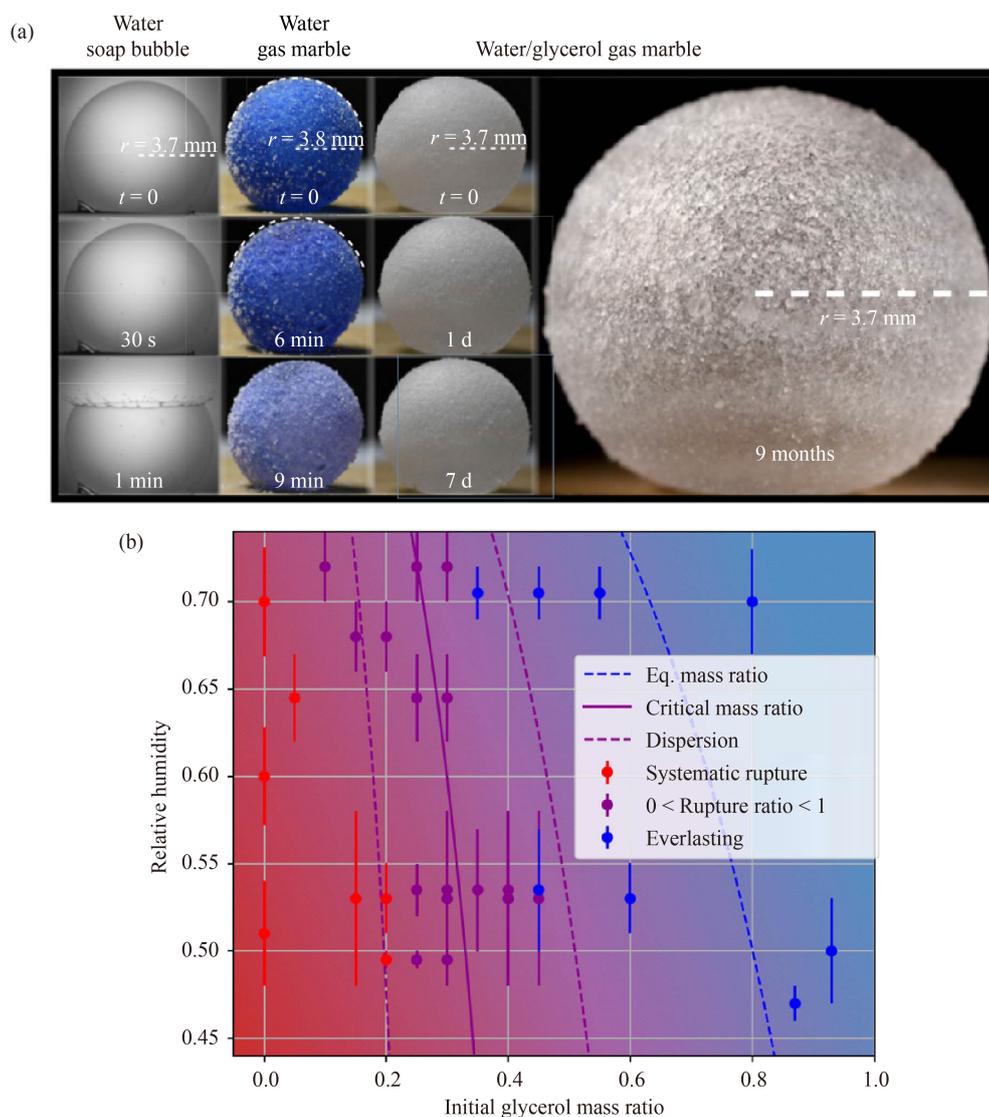


Fig. 3 (a) Morphology and lifetimes of different marbles: soap water bubble, water gas marble and water/glycerol gas marble. The water/glycerol gas marble has the longest lifetime, which maintains its morphology after 9 months. (b) Phase diagram of different regimes of gas marble depending on the initial glycerol mass ratio and the relative humidity. Reproduced with permission from Ref. [8], copyright 2022, American Physical Society.

marbles for miniaturized reactions between interior and exterior gases. By tuning the film composition, we can control the permittivity of gas marbles and explore the reaction dynamics of the two gases. In addition, the thin film of gas marbles can be the confinement where miniaturized liquid/gas reactions with high efficiency take place. The thin film of gas marbles possesses an ultrahigh surface-to-volume ratio: $S/V \sim 1/h$, where h is the thickness of the film. With its unique properties, this new soft object, the gas marble, could open new possibilities both for a fundamental understanding of particle-laden interfaces as well as for the development of novel bubble-based materials and novel microreactors.

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