Electronic Supplementary Information
for
Liquid Marble Coalescence via Vertical Collision

*Jing Jin, Chin Hong Ooi, Dzung Viet Dao and Nam-Trung Nguyen**

Queensland Micro- and Nanotechnology Centre, Griffith University, Brisbane, Queensland 4111, Australia.

*To whom correspondence should be addressed. E-mail: nam-trung.nguyen@griffith.edu.au

Supplementary optical microscope images of the multilayer coating of a liquid marble

(a) (b)

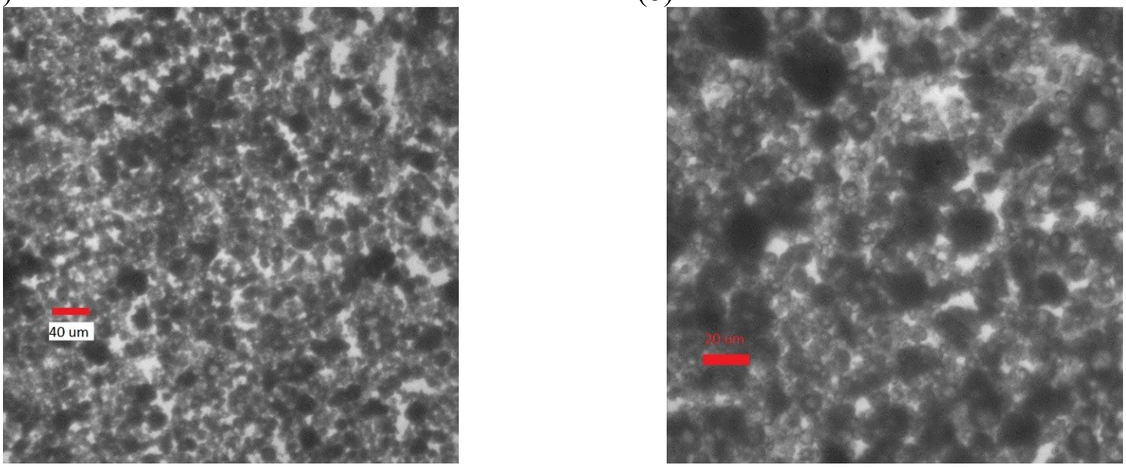

Fig. S1 Top-view microscope images of a water marble coated by 1 μm PTFE particles. (a) 4X microscope image; (b) 10X microscope image.

The rolling method is widely used in the manual fabrication of liquid marbles. It is simple and easy to handle. After rolling, the liquid droplet may be coated by multiple particle layers. It should have a random aggregate of particles on the marble surface. We make it roll for enough time to ensure the liquid core is covered as fully as possible. According to the measurement of microscope images above, we estimate the coverage of multilayer particles on the droplet surface at 90%.

Supplementary images of reversible deformation of liquid marbles caused by DEP force

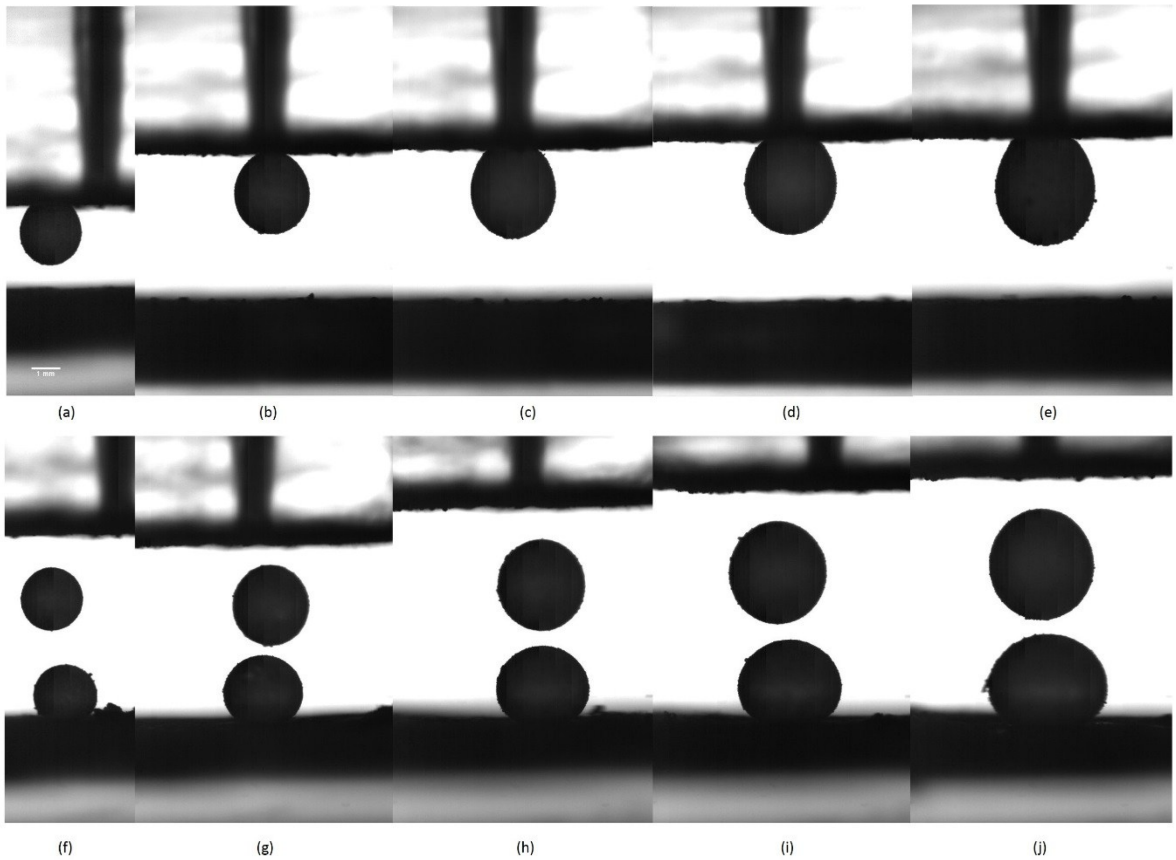

Fig. S2 Images of liquid marble deformation caused by the dielectrophoretic picking and recovery after releasing. (a)(f) 5 μL water marbles; (b)(g) 10 μL water marbles; (c)(h) 15 μL water marbles; (d)(i) 20 μL water marbles; (e)(j) 25 μL water marbles.

It is obviously that the liquid marble was deformed when it was picked up by dielectrophoretic force. But the deformation is absolutely limited and would not cause irreversible damage after releasing from the PMMA holder.

Supplementary image sequence of the crack formation process on the particle shell

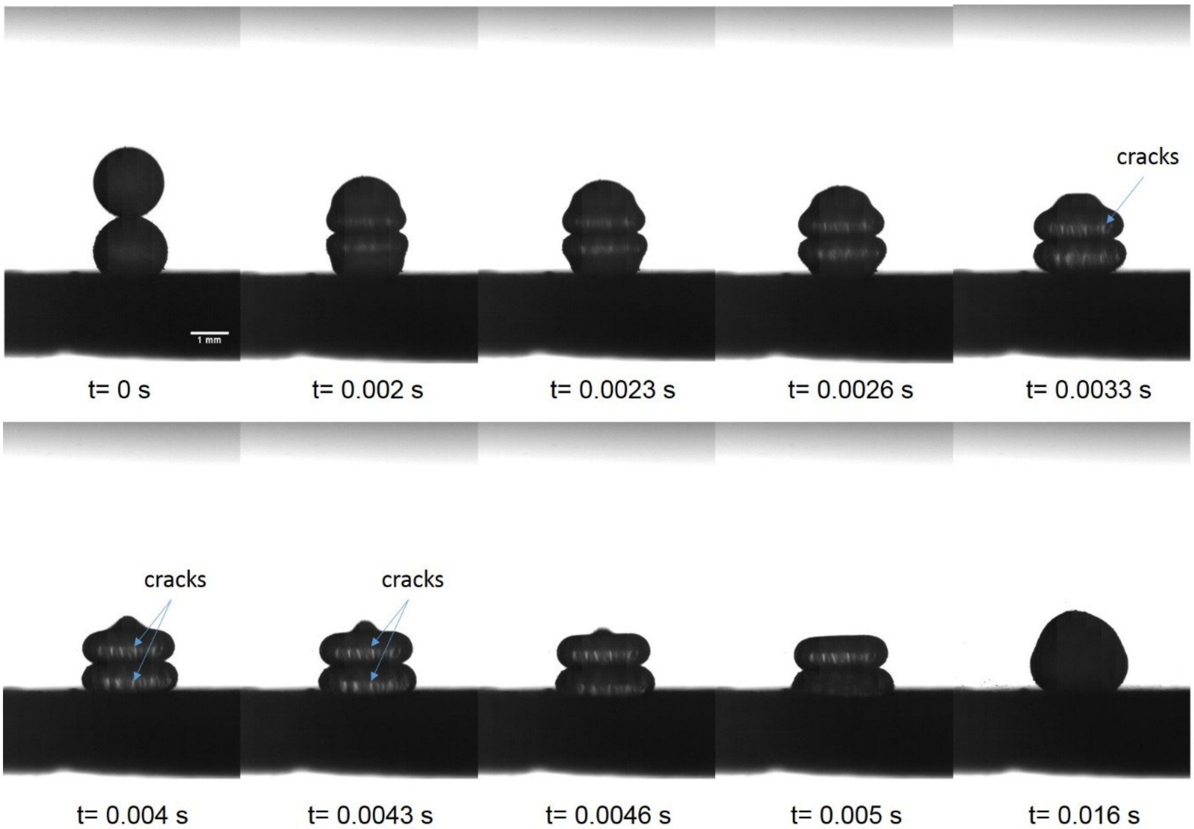

Fig. S3 Image sequence of the formation process of cracks on the particle shell.

There were some cracks formed on the particle shells, which effectively proves the direct liquid-liquid contact between two coalescing LMs.

Supplementary discussion on the dynamic elasticity of liquid marbles

To understand the dynamic elasticity of liquid marbles with different volumes during collision, we did a simple experiment to compare them. We fabricated the PTFE marble with various volumes of 5, 10, 15, 20 and 25 μL. We subsequently transported the liquid marble with a chemical spoon to a certain height (10mm) above a microscope glass slide and let it fall down freely towards the rigid glass slide (Fig. S4). A high-speed camera was used to record the side view of the whole collision process. The recorded images were analysed in ImageJ and the value of the first rebound height was measured. By comparing different rebound heights of various liquid marbles, we deduced the relative sizes of dynamic elasticity of liquid marbles with volumes of 5, 10, 15, 20 and 25 μL respectively.

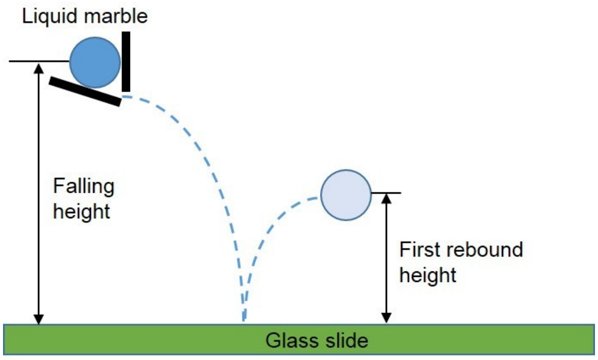

Fig. S4 Schematic of the experiment for the dynamic elasticity of liquid marbles.

Marble volume/μL	Falling height/mm	First rebound height/mm
5	10.2	4.6
10	10.2	4.0
15	10.5	Rupture
20	10.5	Rupture
25	10.3	Rupture

Table S1 The experimental data for relative sizes of the dynamic elasticity of liquid marbles.

From the measurement values of the first rebound height in Table S1, we can clearly see that the 5 μL liquid marble has the best dynamic elasticity during vertical collision with first rebound height to 4.6 mm. As the marble volume increases, the dynamic elasticity decreases gradually. When the marble volume increases to 15, 20 and 25 μL, the integrity of these liquid marbles is lost easily after colliding with the rigid substrate vertically even with a small impact velocity.

Supplementary discussion on the shear rate between two colliding liquid Marbles

The shearing effect plays an important role in the off-centre vertical collision of liquid marble different offset ratios. The shear rate for the fluid flowing between two liquid marbles is defined as:

$$\dot{\gamma} = \frac{v'}{r},$$

where $\dot{\gamma}$ is the shear rate, measured in s^{-1}, v' is the impact velocity of upper liquid marble just before collision along the shear interface that is perpendicular to the connecting line between two centres of mass, measured in $m \cdot s^{-1}$ and r is the radius of liquid marble, measured in m.

Following are the schematic graphs depicting the geometrical relation of two equally-sized liquid marbles with different offset ratios when they contact with each other. We can easily find the velocity vector along the shear surface based on geometrical calculation and then derive the shear rate of two colliding liquid marbles, as listed in the table below.

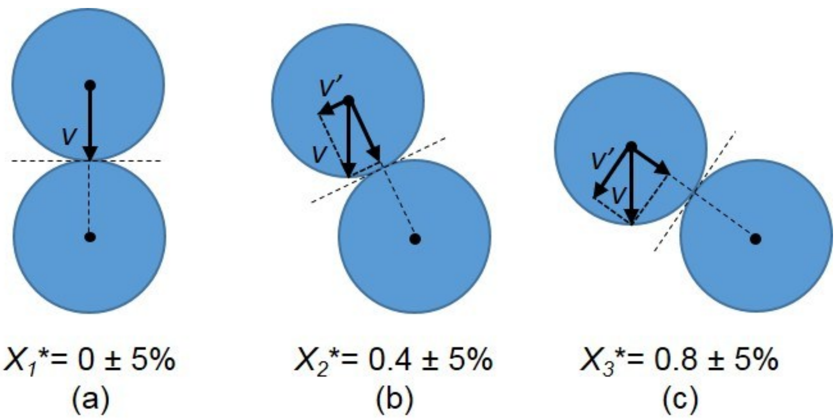

Fig. S5 Schematic of the geometrical relation of two equally-sized liquid marbles with different offset ratios.

Table. S2 The value of shear rate (s^{-1}) for all the experimental groups.

Marble volume/μL		5			10			15			20			25		
Diameter/mm		2.15			2.66			3.01			3.40			3.61		
Offset ratio		0	0.4	0.8	0	0.4	0.8	0	0.4	0.8	0	0.4	0.8	0	0.4	0.8
Impact velocity /m·s^{-1}	0.333	0	124	248	0	100	200	0	89	177	0	78	157	0	74	148
	0.386	0	144	287	0	116	232	0	103	205	0	91	182	0	86	171
	0.438	0	163	326	0	132	263	0	116	233	0	103	206	0	97	194

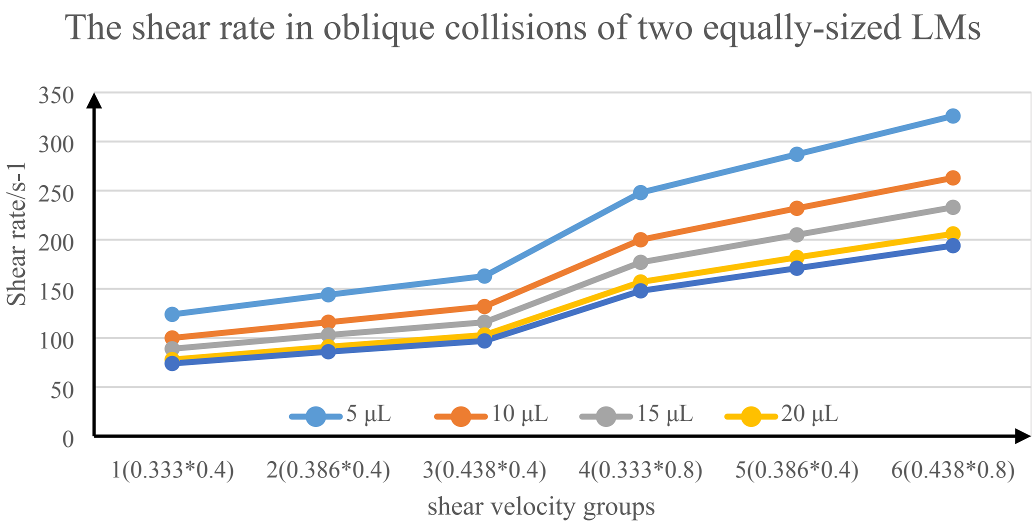

Fig. S6 The values of shear rate in oblique collisions of two equally-sized LMs.

For small-volume liquid marbles, the shear stress has a huge effect on the rupture of the interface between two colliding marbles. The shear stress promotes the particle motion on the surface and thus shears the particle layer. As the diameter of liquid marbles increased, the shear rate decreases significantly so that the shear stress did not affect the oblique collision outcome seriously. If the offset ratio increases from 0.4 to 0.8, the shear rate grows almost twice. However, due to the short contact time and the small compression force toward the centre of mass of the bottom liquid marble, the shearing effect did not dominate the collision process in larger offset ratio vertical collision.