

Supplementary Information

Fabrication of stable monolayer liquid marbles with reduced particle coverage and locomotion on hydrophilic surface

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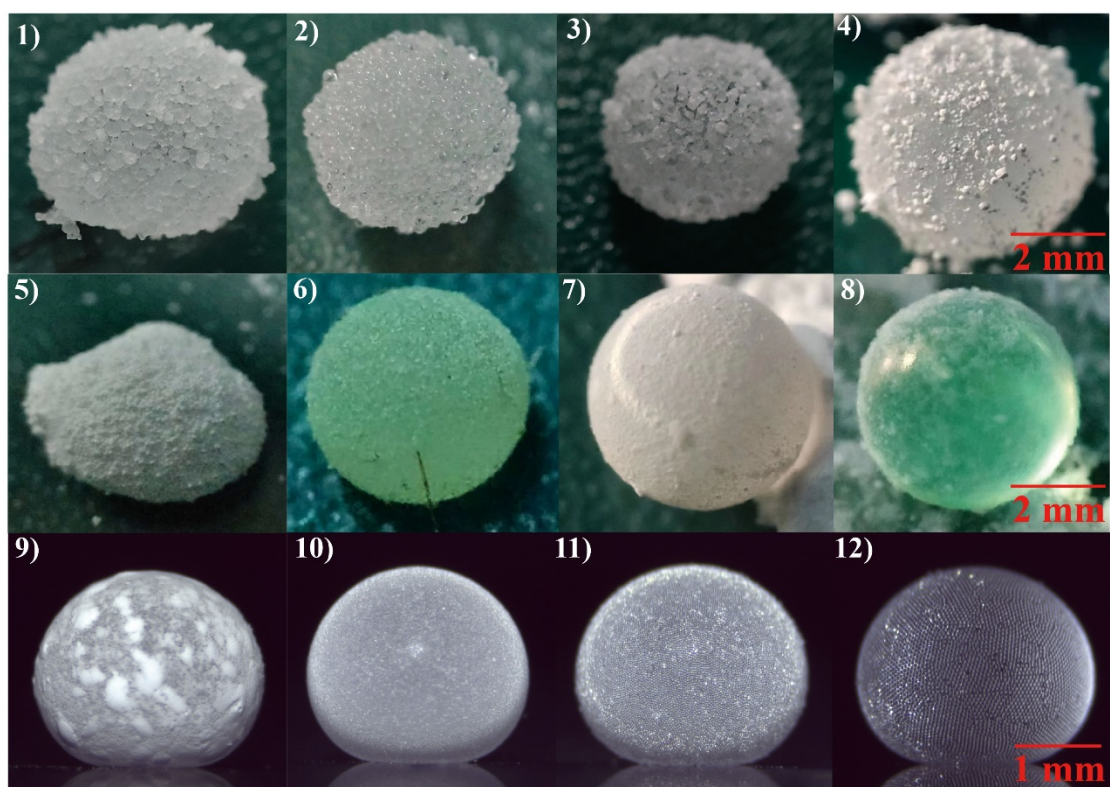
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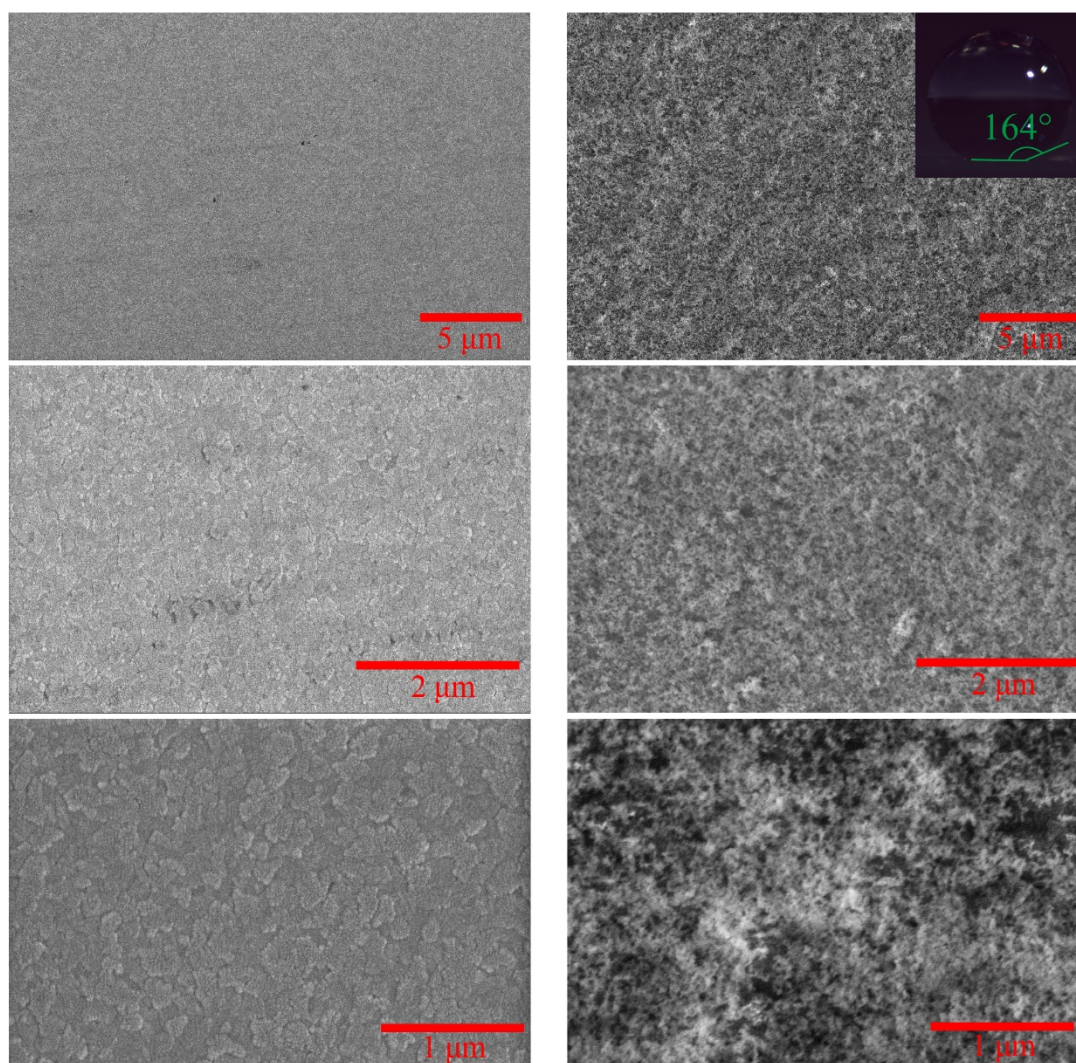
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Supplementary Figure 1. Various liquid marbles coated with different shell particles. 1) 300- μm PE, 2) 300- μm PMMA, 3) 300- μm PP, 4) 1- μm PTFE, 5) 200- μm PVC, 6) $\sim\mu\text{m}$ Lycopodium, 7) 25- μm PTFE, 8) $\sim\text{nm}$ SiO_2 , 9) $\sim\mu\text{m}$ PMSQ, 10) 10- μm PS, 11) 30- μm PS, and 12) 50- μm PS, respectively (Scale bar: panels 1-8) 2 mm, panels 9-12) 1mm).

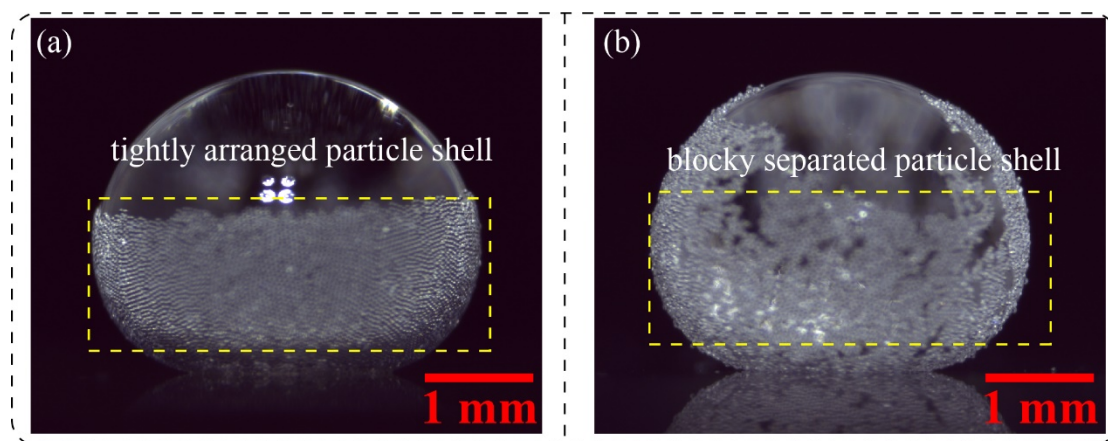


before treatment

after treatment

Supplementary Figure 2. Micrographs of a glass slide before and after hydrophobic treatment.

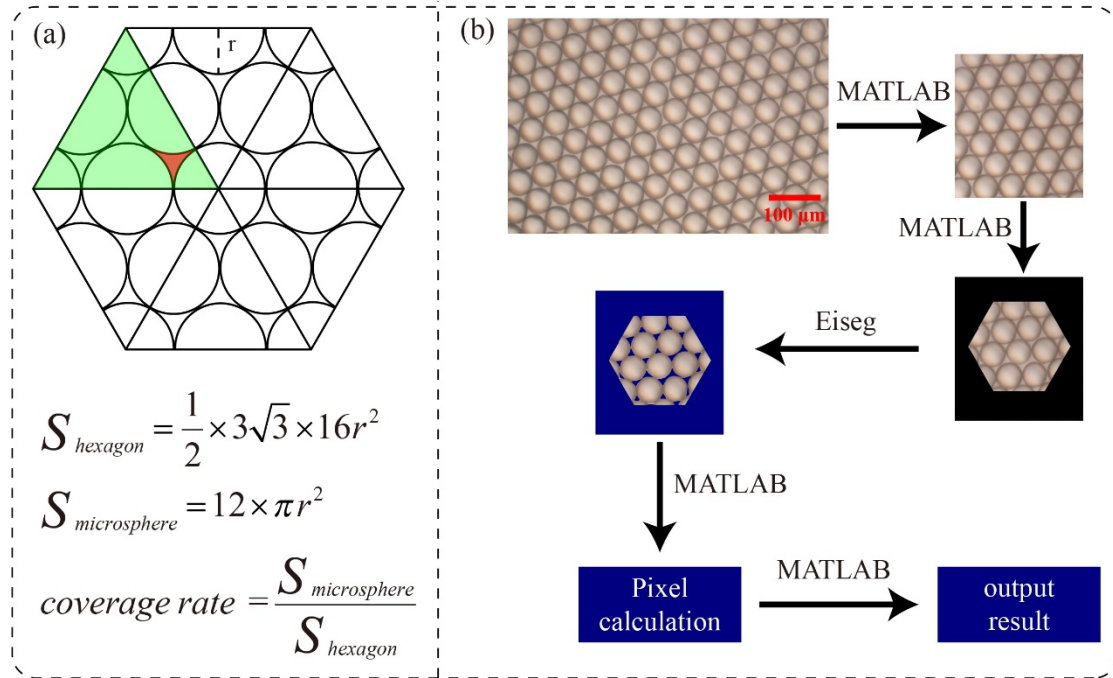
Supplementary Note 1. Characterization of the effectiveness of hydrophobic treatment



Supplementary Figure 3. The distribution trend of treated particles in different batches. (a) Effective hydrophobic treatment. **(b)** Ineffective hydrophobic treatment.

As shown in Supplementary Figure 3, the distribution of particles after effective and ineffective treatment on the surface of liquid marbles differs a lot. After liquid injection, effectively treated particles will tightly pack and move towards the bottom of the droplet. Ineffectively treated particles, on the other hand, will lump together and crack during liquid injection.

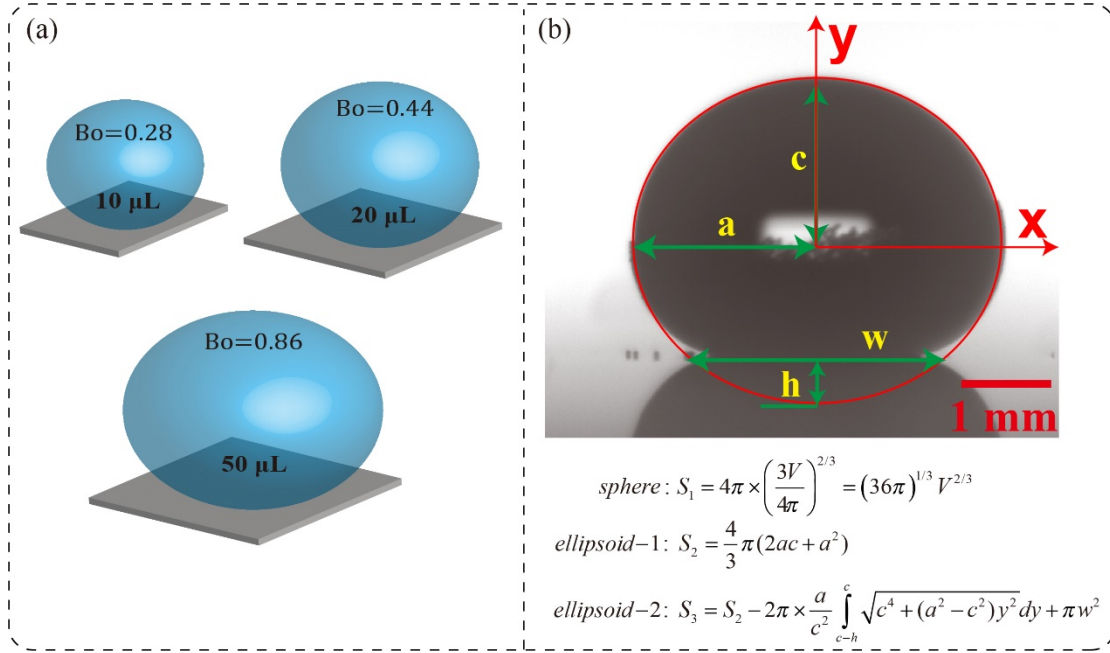
Supplementary Note 2. Particulate image recognition



Supplementary Figure 4. Theoretical coverage rate calculation and the detailed image processing procedures. (a) The hexagonal arrangement structure of microspheres and theoretical calculation equations of the corresponding microsphere area, where the red area represents the gap between the microspheres and r is the radius of the microsphere used. (b) The image recognition procedure for determining the bottom microsphere area.

The detailed processes of image recognition are as follows. First, cut the original image into appropriate size and add a positive hexagonal mask using MATLAB. Then employ Eiseg tool to label the microspheres in the processed image to obtain a comparison-enhanced image. Finally, use MATLAB to calculate the ratio of pixels occupied by the microspheres in the hexagonal area of the image and output the results. In this calculation model, the algorithm yielded a result of $S_{microsphere}/S_{hexagon} = 90.69\%$ when calculating the perfect hexagonal arrangement of microspheres. The results obtained from the image recognition method aforementioned were 90.56%, with a difference of only 1.3%.

Supplementary Note 3. Geometric features of expanded liquid marbles

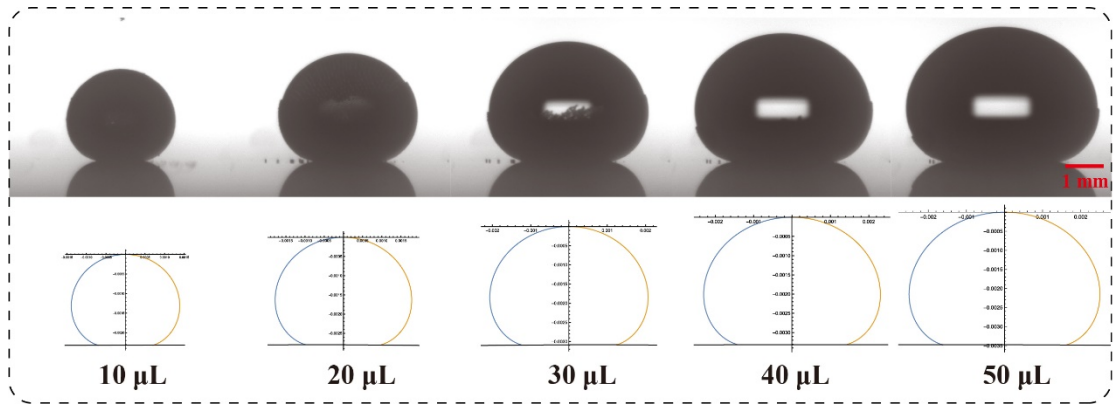


Supplementary Figure 5. Morphology changes and surface area calculation of liquid marbles.

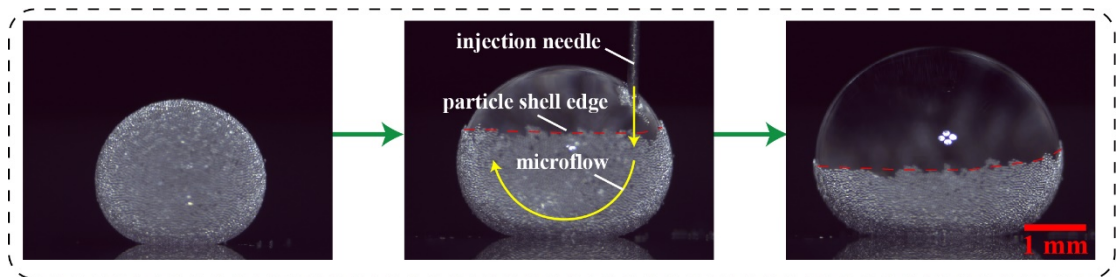
(a) The bond number of liquid marbles with different volumes. (b) Approximate ellipsoids for fitting a sessile liquid marble and three geometrical methods to calculate its surface area: ideal spherical model S_1 , ideal ellipsoidal model S_2 , and deformed ellipsoidal model S_3 .

$Bo = \frac{\rho_m g R_m^2}{\gamma_m} = \left(\frac{R_m}{l_c}\right)^2$, where R_m is the radius of a liquid marble, γ_m is the effective surface tension between the marble and air, ρ_m is the density of liquid, g is the gravitational constant.

Here $l_c = \left(\frac{\gamma_w}{\rho_w g}\right)^{1/2} = 2.7 \text{ mm}$ is the capillary length of pure water, which is approximately equal to the diameter of a $10\text{-}\mu\text{L}$ liquid marble. γ_w and ρ_w are the surface tension and the density of water, respectively.

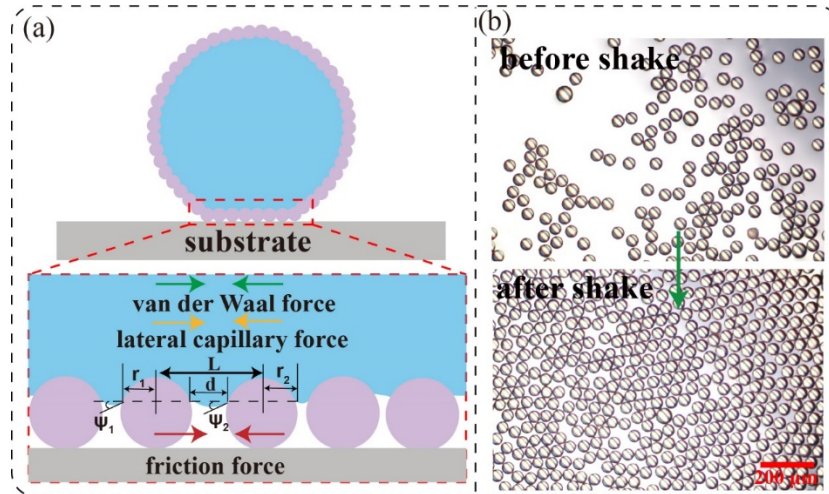


Supplementary Figure 6. True profiles of liquid marbles at each volume and estimated profiles obtained by solving the Young-Laplace equation.



Supplementary Figure 7. The effect of asymmetric microflow generated by liquid injection on the edge development of the monolayer particle shell.

Supplementary Note 4. Interaction between the underneath coating particles



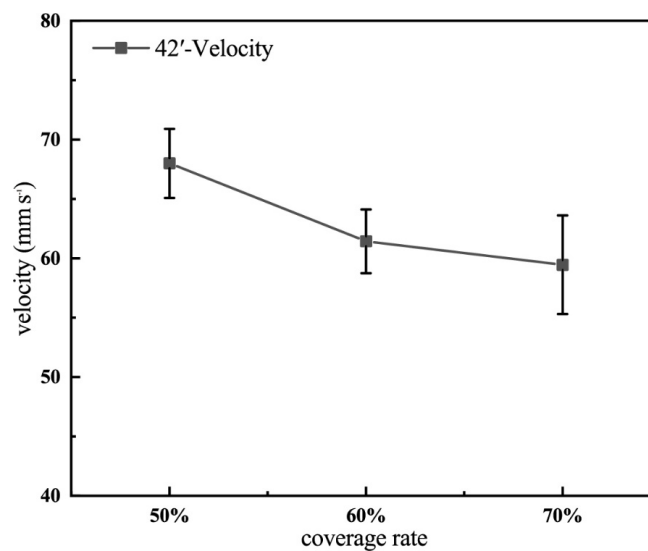
Supplementary Figure 8. Distribution characteristics of bottom particles. (a) Schematic of the close interaction of underneath coating particles. (b) The detailed distribution of bottom microspheres before and after gentle shaking.

van der Waals force:
$$F_{\text{vdw}} = \frac{Ar}{12d^2}$$

lateral capillary force:
$$F_C = 2\pi\gamma_w Q_1 Q_2 / L$$

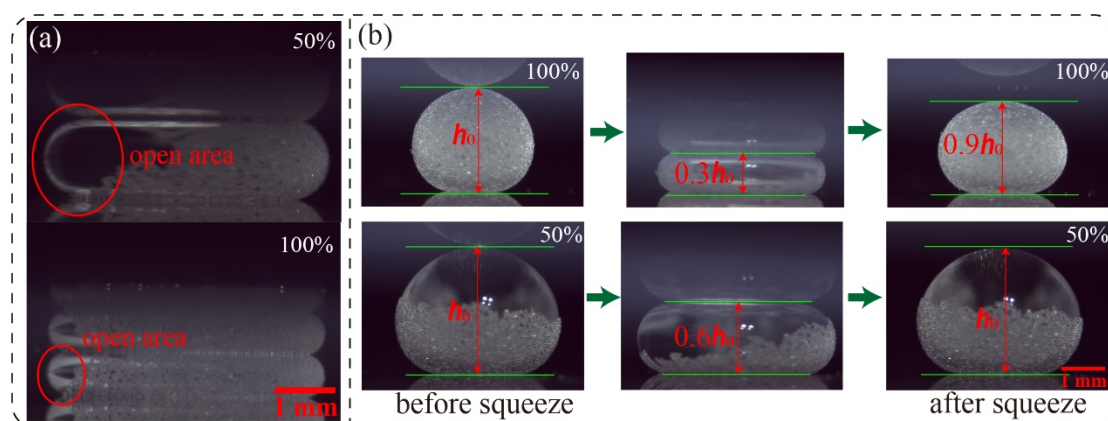
friction force:
$$F_f = \mu \frac{G}{n}$$

where A and d are a modified effective Hamaker constant and the separation distances, respectively. $Q_i = r_i \sin \psi_i$ is the 'capillary charge' of the particle, $i = 1, 2$, here r_i and ψ_i are the radius of the contact lines and the meniscus slope angles of two microspheres, respectively. μ , G and n are the static friction coefficient of the PS microsphere used, the gravity of droplet core and the number of bottom microspheres, respectively.



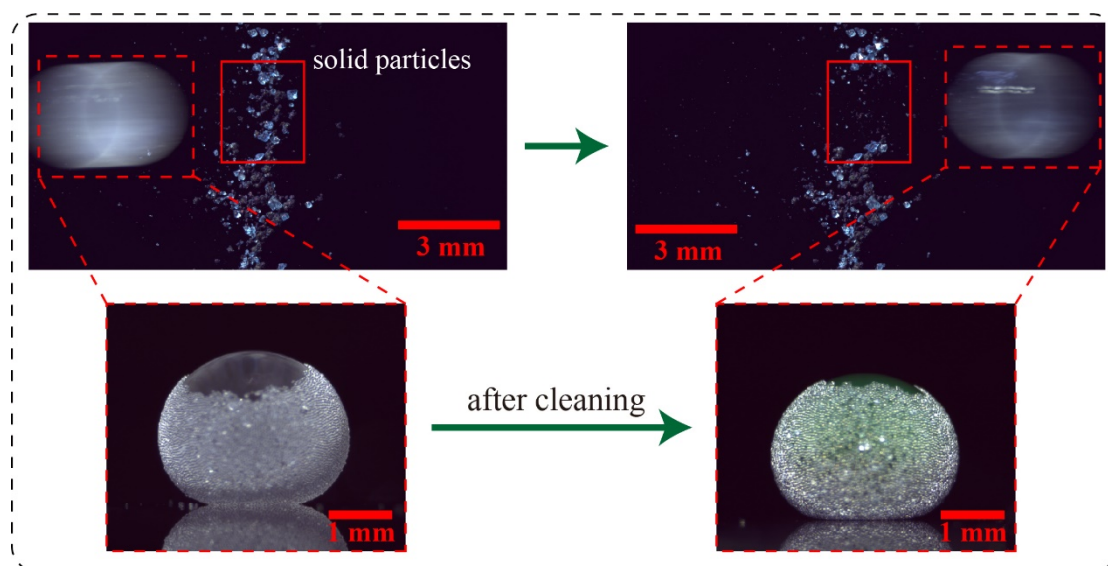
Supplementary Figure 9. The corresponding rupture velocity for 50%-, 60%- and 70%-coverage liquid marbles at an activation angle of 42'.

Supplementary Note 5. Elasticity of opened liquid marbles



Supplementary Figure 10. Squeezing breakage principle and rebound features of liquid marbles with openings. (a) The squeezing process for an opened liquid marble and a fully covered liquid marble. (b) The comparison of elasticity between an opened liquid marble and a fully covered liquid marble.

As illustrated in Supplementary Figure 10(a), the breakage of the 50%-coverage liquid marble is mainly caused by the uneven development of the particle shell, while the 100%-coverage liquid marble s is due to the cracks created during the squeezing process's expanded surface area. As illustrated in Supplementary Figure 10(b), the 100% coverage liquid marble shows visible deformation after compression, and the 50%-coverage liquid marble returns to its original shape.



Supplementary Figure 11. Snapshots of the cleaning process of an opened liquid marble on a hydrophilic surface.