Supporting Information for:

Mechanical compression to characterize the robustness of liquid marbles

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(1) Controlling the liquid droplet size by changing the diameter of the needle

We form the droplets by dripping the liquid phase out of a needle at a low flow rate Q. Upon droplet formation, the gravitational force together with the inertial force will drive the droplet extension while the surface tension will prevent this. The ratio of the inertial force over surface tension can be characterized by the Weber number $\rho u^2 d/\gamma$, where ρ and γ are the density and surface tension of the liquid respectively, d is the diameter of the needle, u is the velocity of the liquid which is estimated by $4Q/\pi d^2$. In our experiment, the Weber number is less than 10^{-3} , indicating that the inertial force is negligible compared with the surface tension. Therefore, upon droplet detachment, it is the gravitational force that balances the surface tension: $\rho Vg \sim \pi d\gamma$, where V is the volume of the droplet. The resultant droplet breakup from the needle should have a volume: $V \sim \pi d\gamma / \rho g$. From this formula, we find that the volume can be controlled by varying the diameter of the needle. We generate the droplets using needles of different diameter (300 µm, 550 µm and 900 µm). Indeed, the measured volume of the droplets agrees relatively well with the size of the needle, as shown in Figure S1.



Figure S1. Plot of the measured droplet volume *versus* the predicted volume for aqueous drops containing 2 wt.% sodium alginate.

(2) Microscopy of the surface of liquid water marbles coated with different particles

To demonstrate the size and shape of particles, we have taken optical microscope images of 35 μ m PTFE particles, 200 μ m PTFE particles and 6 μ m silica particles on the surface of water marbles. Also, we have taken an SEM image showing the morphology of powdered Aerosil R812 silica nanoparticles.

(a) PTFE micro-particles- 35 μm



(c) Silica micro-particles-Tospearl 2000B*





(d) Silica nanoparticles- Aerosil R812





Figure S2. Optical microscope images of (a) 35 μ m PTFE particles, (b) 200 μ m PTFE particles and (c) 4-6 μ m silica particles on the surface of water marbles. (d) SEM image showing the morphology of powdered Aerosil R812 silica nanoparticles.

(3) Influence of viscosity of liquid core on P_c

Experiments were carried out to investigate the influence of the viscosity of the liquid on the robustness of liquid marbles. We use different concentrations of sodium alginate to increase the viscosity of water. The results indicate that the droplets with a higher viscosity have a larger critical pressure, as shown in Figure S4. We therefore use 2 wt.% sodium alginate solution to fabricate our water marbles as they are not easily destroyed or ruptured during the manipulation/transportation of them on the substrate.



Figure S3. Influence of viscosity of the liquid on the critical pressure of liquid marbles. The liquid marbles are fabricated by pure water (1 mPa s), 1 wt.% sodium alginate solution (79.8 mPa s), 2 wt.% sodium alginate solution (576 mPa s); all of the droplets are coated with 35 μ m PTFE particles, $R_0 = 1.28$ mm.

(4) Influence of the compression speed on P_c

Experiments were carried out to test the influence of compression speed on the critical pressure, as shown in Figure S3. The results indicate that P_c increases slightly upon increasing the compression speed within the range we have tested (from 15 μ m/s to 75 μ m/s). However, P_c remains almost constant when the compression speed is below 50 μ m/s. Therefore, we think that the compression speed of 45 μ m/s is sufficiently low to test the critical pressure of liquid marbles.



Figure S4. Influence of the compression speed on the critical pressure of liquid marbles. The liquid marbles are composed of pure water droplets ($R_0 = 1.28$ mm) coated with 35 µm PTFE particles.

(5) A movie demonstrating the squeezing test of a liquid marble using our technique

A video demonstrating the squeezing test of a liquid marble using our technique is shown in "Movie_squeezing test". The liquid marble is formed by coating 2 wt.% aqueous sodium alginate droplets ($R_0 = 1.56$ mm) with PTFE particles of diameter 35 μ m.