Supplementary Material for

Capillary rise method for effective surface tension of monolayer nanoparticle-covered liquid marbles

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1. Preparation of the xerogel SiO₂ coating.

The superhydrophobic SiO₂ coating with weak binding forces was prepared by a sol-gel method as described elsewhere^{1,2} and summarized below.

A silica sol was synthesized with tetraethyl orthosilicate (TEOS) as the precursor, in ethanol (EtOH). The molar ratio was TEOS/EtOH/NH₃/H₂O at 1/38/1.08/3.06 (all water was from the 25% ammonia solution, and the volume ratio was TEOS/EtOH/ammonia solution at 5/50/1.8). First, EtOH was mixed with TEOS with stirring for ~15 min. Then, ammonia solution was added and stirred for 1 h. The mixed solution was subjected to aging at room temperature for 7 d to form a SiO₂ sol, after which it was suitable for coating. Hexamethyl disilazane (HMDS) was then added into the sol with stirring for ~2 h for alkylation (molar ratio, TEOS/HMDS at 1/0.6 and volume ratio, TEOS/HMDS at 5/2.82). The alkylated SiO₂ sol was subjected to aging for 1 d, after which it was ready for use.

Clean glasses were dip-coated using the alkylated SiO_2 sol, with air drying for 2-10 min. Finally, superhydrophobic SiO_2 coatings were generated. Such a coating consists of hydrophobic SiO_2 nanoparticles (NPs) and features very weak binding forces, for which the NPs can be transferred to water surface just by making the coating and water contact with each other.





Fig. S1. (a) Photograph of a coated glass after droplet impinging at an angle, demonstrating the property of particle transfer. Arrows denote areas contacted with the impinging droplets. The color differences between the denoted areas and the rest resulted from the variation in coating thickness due to detachment of a layer of nanoparticles. (b) AFM image of the boundary area of a denoted patch in (a), showing a clear step of ~20 nm which is in accordance with the particle size. (c) and (d) SEM images of the uncontacted and contacted areas, which are basically the same. (e) FTIR spectrum of the superhydrophobic SiO₂ coating, which consisted of monodispersed and connected SiO₂ particles, as sketched in (f), with abundant methyl groups.

(1) Li, X. G.; Xue, Y. H.; Lv, P. Y.; Lin, H.; Du, F.; Hu, Y. Y.; Shen, J.; Duan, H. L. Liquid plasticine: controlled deformation and recovery of droplet with interfacial nanoparticle jamming. *Soft Matter.* **2016**, *12*, 1655-1662.

(2) Li, X. G.; Shi, H. X.; Wang, Y. Q.; Wang, R. X.; Huang, S.; Huang, J. C.; Geng, X. G.; Zang, D. Y. Liquid shaping based on liquid pancakes. *Adv. Mater. Interfaces.* 2018, *5*, 1701139.