Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information

Efficient Separation of Immiscible Oil/Water Mixture *via* Perforated Lotus Leaf

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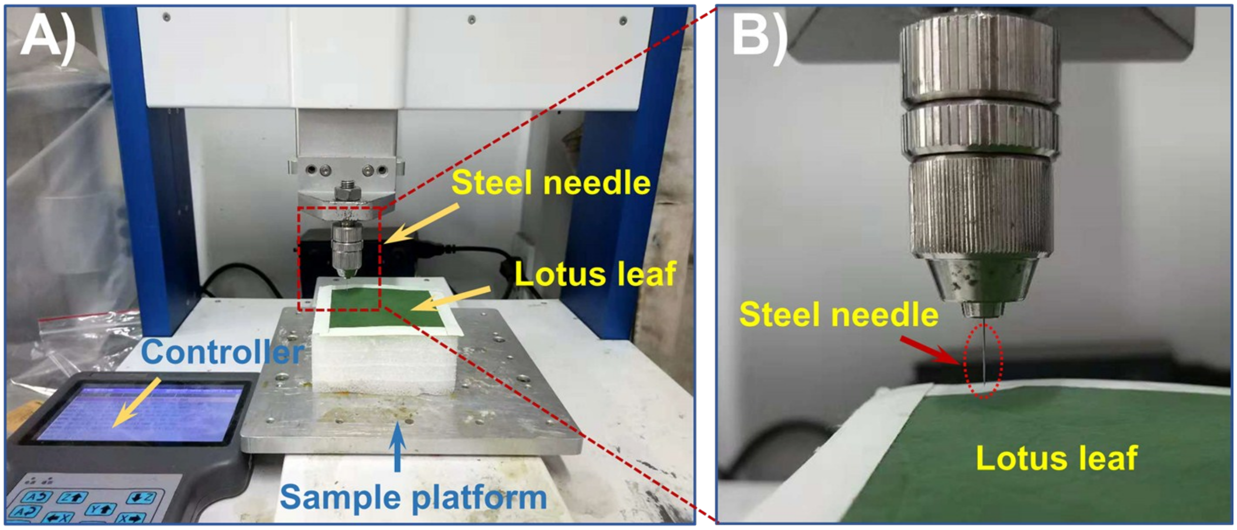

Fig. S1. A) The needle was fixed to the jet dispensing system; lotus leaf was attached to the sample platform. The perforating process was adjusted through the machine controller. B) Magnified image of the steel needle.

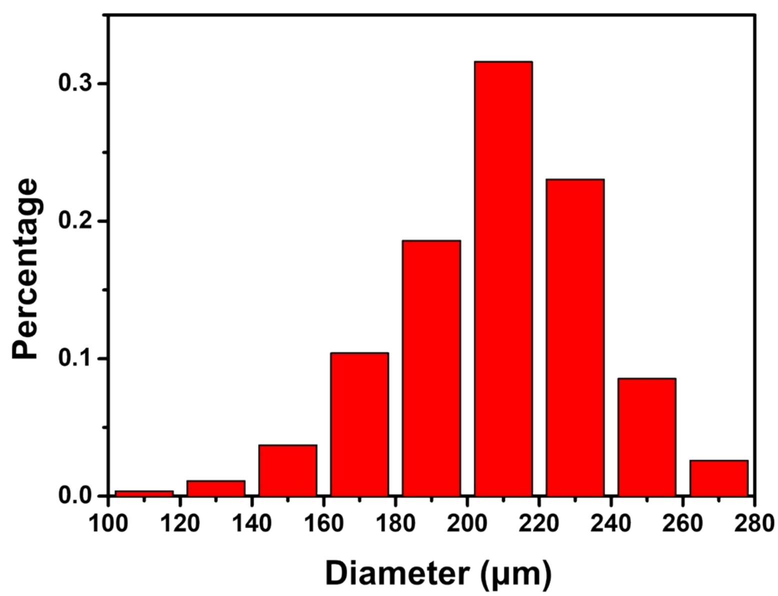

Figure S2. The distribution of perforated holes on lotus leaf. The main diameter is ~220 um, which is well corresponded to the diameter of perforate needle.

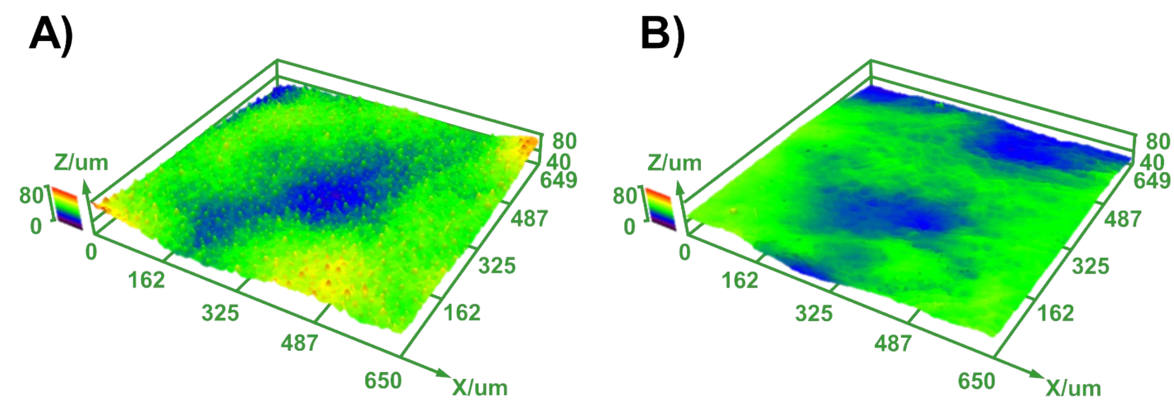

Fig. S3. The laser confocal microscope images of the upside and downside surfaces of lotus leaf. A) The upside surface of lotus leaf is full of hierarchical rough structures. B) Compared to upside surface, the downside surface is relatively smooth.

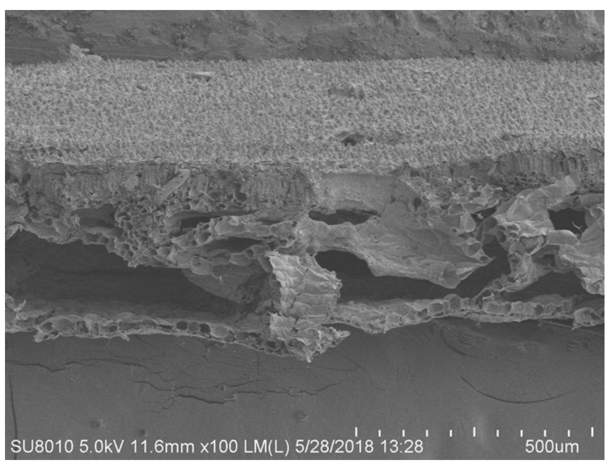

Fig. S4. SEM image of cross-section of lotus leaf.

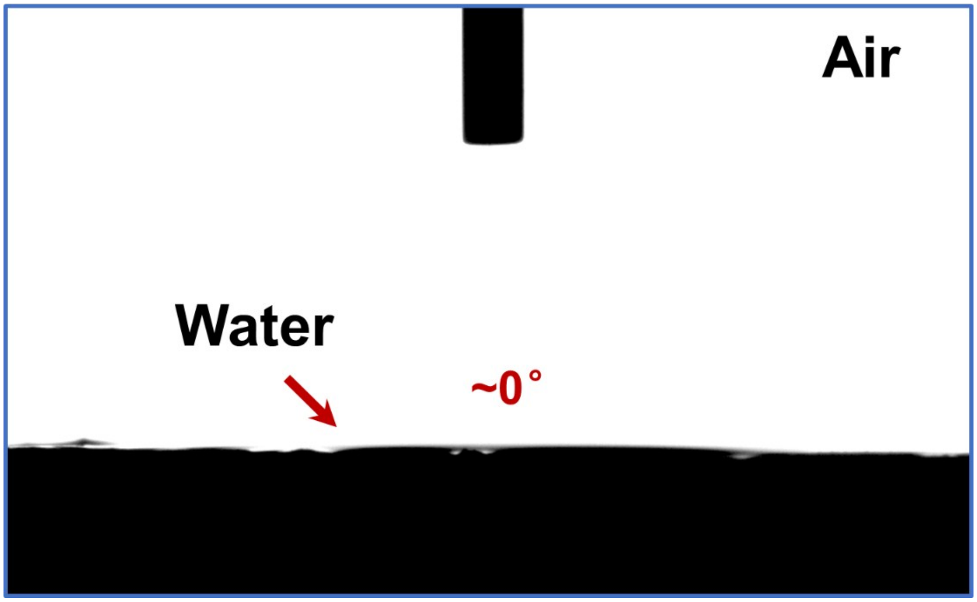

Fig. S5. The water contact angle(WCA) on the pre-wetted downside surface of lotus leaf. The pre-wetted downside surface is superhydrophilic, demonstrating the enhanced wettibility after pre-wetting process.

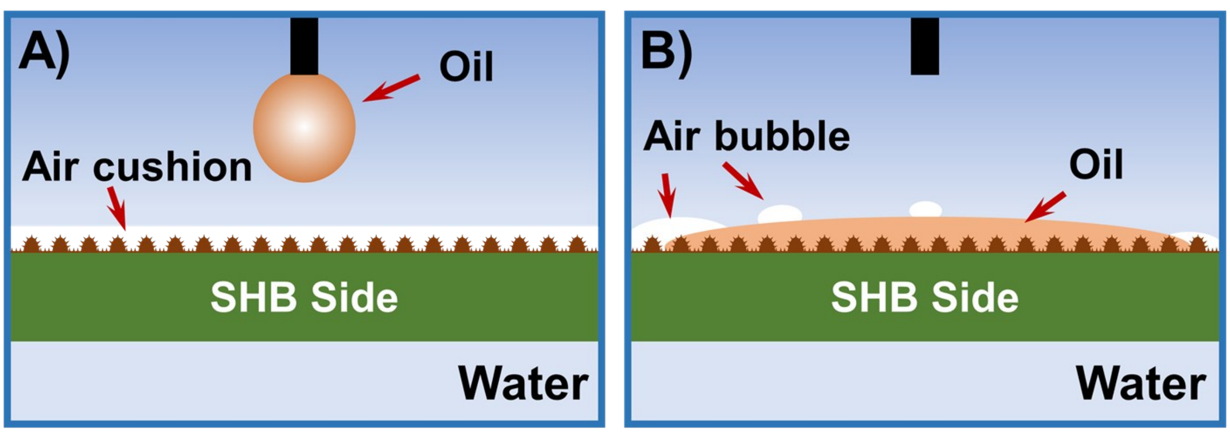

Fig. S6. The generation of bubbles during the oil wetting process on SHB Side of Lotus. A) The micro-nano structures on SHB side can trap air tightly and form a thin layer of air cushion. B) Dripping an oil droplet on SHB side, the oil can wet the substrate and the air cushion will be replaced by oil. Then the air film gather in several bubbles.

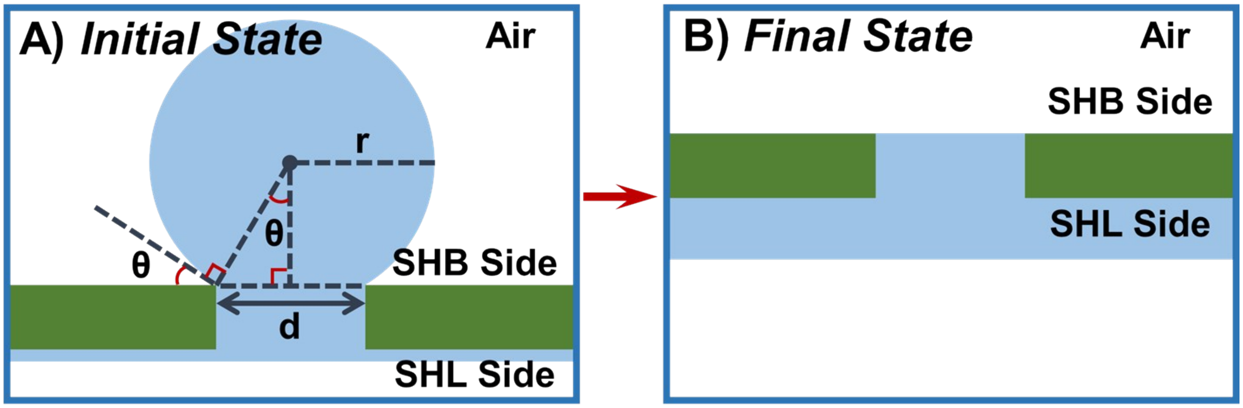

Fig. S7. Illustration of permeation process of water droplet on perforated lotus leaf from energy point-view.

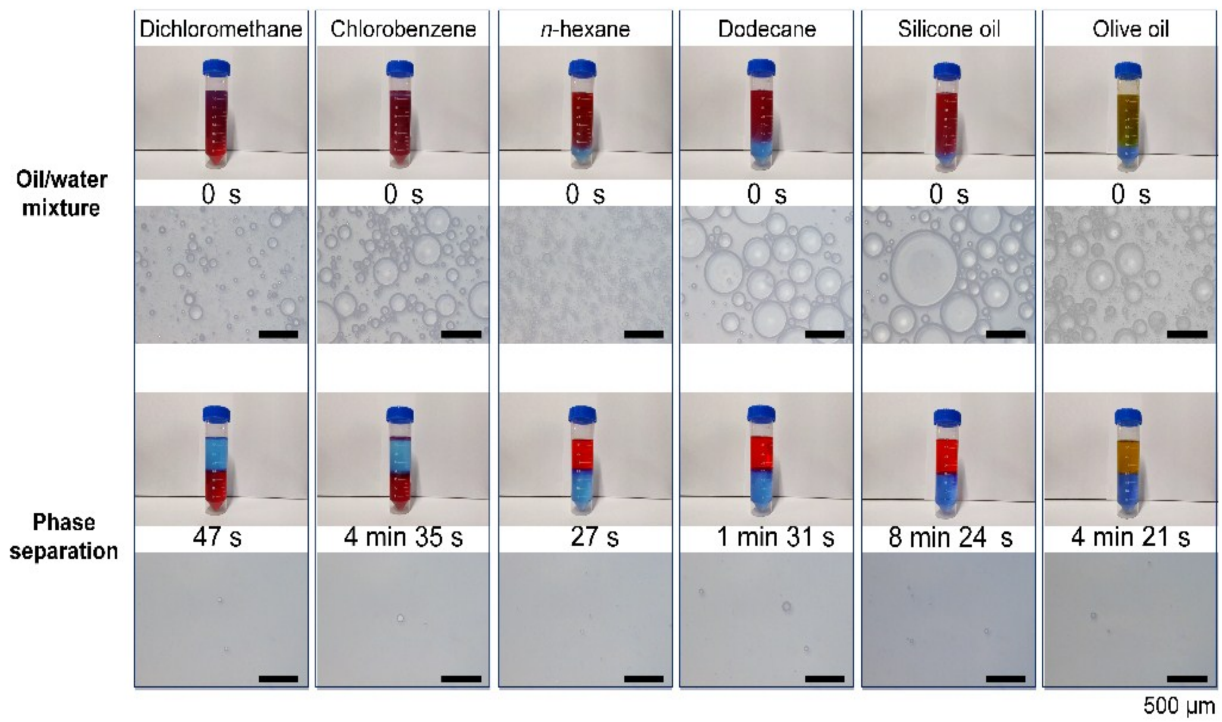

Fig. S8. Spontaneous phase separation of immiscible oil/water mixture.

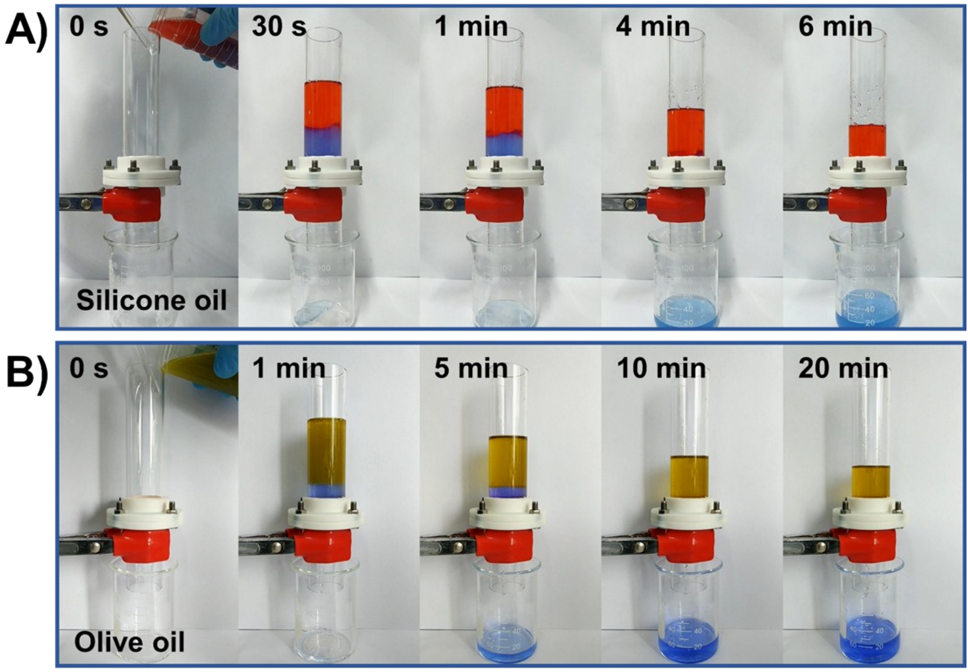

Fig. S9. Optical images of tank oil/water separation processes. A) The separation of silicone oil/water mixture. B) The separation of olive oil/water mixture.

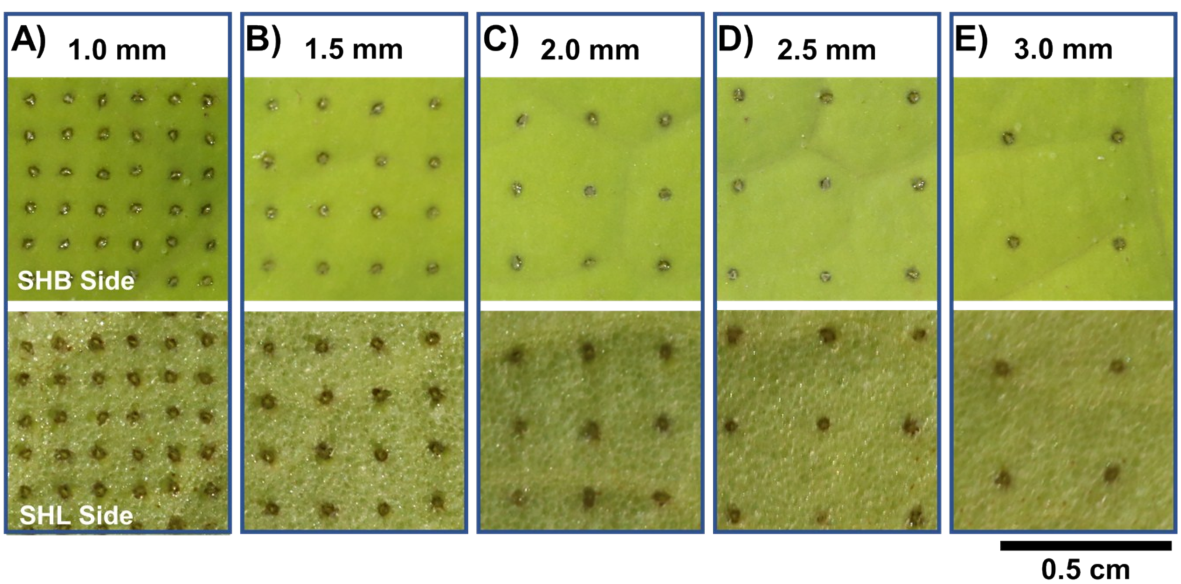

Fig. S10. Optical images of the perforated lotus leaf with different micro-hole spacing.

Experimental Section

Materials: Fresh lotus leaf, particularly the lotus leaf floating on water. (picked up from lotus pond of Tsinghua University, China).

Preparation of perforated lotus leaf: The fresh lotus leaf was cleaned with distilled water. Then the lotus leaf was attached to the sample platform of a jet dispensing system. An acupuncture needle with the diameter of ~240 μm was attached to the machine bit. Regular micro-hole arrays on the lotus leaf can be easily fabricated through the perforating process manipulated by the machine controller. The punching process can be clearly viewed in supporting **Movie S7**.

Preparation of feed solutions: The feed solution can be easily prepared through mixing 20 mL selected oil or organic liquid and 20 mL deionized water. Magnetic stirring apparatus (S10-3, Shanghai Sile, China) was used to stir the feed solution for about 10 min (600 r/min) to make sure the intensive mixing of the feed solution. These procedures are also included in **Movie S7**. The prepared feed solution was used immediately.

Oil/water Separation: The perforated lotus leaf was fixed between two PTFE fixtures, which were connected with two glass tubes (26 mm in internal diameter, 30 mm in external diameter). During whole separation process, the hydrophilic side of lotus leaf was prewetted by a small amount of water in advance, then, the mixture of water and oil or organic solvent (40 mL, V_{water}: V_{oil} = 1:1) was quickly poured into the designed separating device and the separation process was driven by the gravity. To clealy show oil and water, the oil or organic solvent was dyed by oil red O and showed red colour, the water was dyed by methane blue and showed blue colour.

Recycling test: Silicone oil/water mixture was used in the recycling test, of which process is same as single oil/water separation test. After each oil/water separation, new feed solution was added in the separation device directly.

Instrumentation and Characterization: The surface morphology of the as-prepared perforated lotus leaf was characterized a desktop scanning electro-microscope (Phenom Pro, Phenom World, Netherlands). The SEM image of the cross-section of lotus leaf was obtained with scanning electron microscope (SU 8010, Hitachi, Japan). The behaviours of the water/oil droplet on the perforated lotus leaf were recorded by using a high-speed camera (i-SPEED 3, Olympus, Japan). The water and oil contact angles were measured using a video-based contact angle measuring device (OCA 40, Data-physics, Germany) at ambient temperature (the volume of each tested liquid drop was 4 μL). The observation of phase separation in **Figure S8** was carried through an optical microscope (Olympus BX53). The oil concentration of the collected water after the separation process was measured by the infrared spectrometer oil content analyser (OIL480, China). The residual oil was extracted by 20 mL CCl_4 from 10 mL collected water before analysis. Use the extract liquor as the test liquid to test the oil concentration, then the oil analysis software can convert the oil concentration of the extract liquid into the oil concentration of collected water.