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Supporting Information

Programmable Unidirectional Liquid Transport on Peristome-Mimetic Surfaces Under Liquid Environments

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Supporting Information Includes:

One PDF file with Experimental Section, 7 Supporting Figures, and Video file with 3

Supporting Movies.

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Movie S1-S3

Movie S1. Schematic demonstration of the fabrication process of the peristomemimetic surface.

Movie S2. Schematic demonstration of the possible mechanisms of oil transport on

the solid PDMS surface swelled and without swelled by oil in water/oil/organogel system.

Movie S3. Unidirectional transportation dynamics of liquids on peristome-mimetic surfaces.

Experimental Section

Materials and methods. Pitcher plants, *Nepenthes alata*, are purchased from XCCT Corporation, Guangzhou, China. Optical images of the pitcher from juvenility to the maturation stage and a cross-sectional view of the peristome surface are captured by a 105 mm macro lens using a digital camera (D750, Nikon, Japan). The photopolymerized resin, VeroClear-RGD 810, is purchased from Stratasys Ltd., USA. The PDMS pre-polymer, curing agent and PVA are purchased from Sigma-Aldrich, USA. Ethanol, hexadecane and silicone oil 20 are purchased from Sinopharm, Beijing, China. Water is acquired from Milli-Q with a resistance of 18.2 MΩ.

3D Printed Mold. 3Ds Max 2018 Software (Autodesk) is used to draw the surface morphology of inverted peristome-mimetic models. 3D printer (Formlabs, USA) is used to print the inverted peristome-mimetic mold at a resolution of 25 μ m. In the process of printing, we take advantage of a slippery surface with ultra-low adhesive energy to overcome the unavoidable vertical adhesion due to the inhibition of the direct contact between the cured resin and the solid surface.¹ After 3D printing, the printed mold is developed in ethanol for 5 min to remove the uncured resin. A post-curing process is performed with LEDs emitting 405 nm light for 20 min at room temperature to enhance the mechanical property of the printed mold. The other experimental setups such as the fixture are also printed by the 3D printer (Formlabs, USA).

The fabrication of Peristome-mimetic PDMS surface. The peristome-mimetic surfaces are achieved by replicating the morphology of 3D-printed molds. PDMS polymers are used as replicas. The PDMS prepolymer and curing agent are mixed with a ratio of 10:1, stirred mechanically for 15 min and then poured into a dish with the 3D-printed molds mount on the bottom surface. After removing the bubbles in the dish by vacuuming, the PDMS-coated template is heated at 80 °C for 3h in a vacuum oven. After the demolding from the 3D-printed molds, the as-prepared PDMS replica is fully swollen by hexadecane to obtain an organogel. Before conducting the experiments, using ethanol to remove hexadecane in the organogel surface, and then removing the bubbles on the surface of the Peristome-mimetic PDMS organogel surface in an underwater environment by vacuuming for 15 minutes.

The fabrication of Peristome-mimetic PVA hydrogel surface. The peristomemimetic surfaces are achieved by replicating the morphology of 3D printed molds. PVA is used as a replica. PVA powders of 20 g amount are first dissolved in a mixed solvent consisting of 45 mL of water and 135 mL of DMSO at 90 °C. Then the mixture is heated and stirred at 95 °C for 3 h. After the as-prepared PVA solution is cooled to 70 °C, we immediately poured it into the mold. After removing the bubbles by vacuuming and cooling completely, transferring the mold covered by PVA into a refrigerator at -20 °C for 6 h. Next, peel off the peristome-mimetic PVA replica from the mold and then submerge the replica into water for 8 hours changing the water every two hours. The PVA hydrogel is finally prepared.

The fabrication of Peristome-mimetic heart-shaped resin surface. The peristome-mimetic surfaces are fabricated directly through DLP 3D printing methods, which are based on the sequential layer-by-layer deposition of photocurable resin. 3Ds Max 2018 Software (Autodesk) is used to draw the heart-shaped surface morphology. The 3D printing process is conducted by a homemade 3D printer at a resolution of 25 μ m. After 3D printing, printed parts are developed in ethanol for 5 min to remove the uncured resin. To enhance the mechanical properties, a post-curing process is performed in a tank with 20 multidirectional LEDs emitting 405 nm light for 15 min at room temperature. The heart-shaped resin surface is finally prepared.

Characterization. High-speed cameras (FASTCAM Mini UX 100, Photron, Japan) are used to record the spreading dynamics of drops. Analysis software (FASTCAM Mini UX 100, Photron) is used to analyze the spreading distances and time of the hexadecane and water on the peristome-mimetic organogel and hydrogel surface, respectively. SEM images of the peristome-mimetic surface are obtained using a field-emission scanning electron microscope at 10 kV by a Hitachi S-4800. Contact angles are measured with a contact angle measurement device (OCA20, DataPhysics) with water and hexadecane droplets of 3 μ L, 2.5 μ L, respectively. Each reported contact angle is an average of at least five independent measurements. High-resolution 3D X-ray microscopy images and computed tomography are taken by a MicroXCT-200, X-radio. Individual X-ray exposure slices reconstruct the 3D copy of the sample in under-liquid environments.



Fig. S1. Scanning electron microscopic (SEM) images of the peristome surface. (a) and (e) Inner scanning electron microscope view of the peristome surface at different growth cycles. (b) – (d) and (f) – (h), High-magnification SEM image of the peristome surface showing two-order microgrooves and periodic duck-billed microcavities scattering along the second-order microgroove.



Fig. S2. Stereoscopic microscope images of the artificial surface. (a) - (c) The peristome mimetic PDMS surface without immersed in oil, immersed in oil, immersed in oil and then washed with ethanol, respectively. An obvious liquid bridge can be observed in $\mathbf{b}_{3, 4}$. (d) and (e) The peristome mimetic PVA surface dried in air and the wetted peristome mimetic PVA surface. There is a microgroove distribution with a periodicity similar to the natural *Nepenthes* peristome, and the microgrooves consist of periodic microcavities.



Fig. S3. Surface wettability of the peristome mimetic surface. (a) and **(b)** The peristome mimetic PVA hydrogel surface that placed in the air, and submerged in oil. They both show superhydrophilicity. **(c)** and **(d)** The peristome mimetic PDMS surface that placed in the air, and submerged in water. The former exhibits superoleophilicity. Whereas, the peristome mimetic surface that submerged in water exhibits oleophilicity.



Fig. S4. Uni-directional transportation dynamics on the outside of a peristomemimetic three-dimensional *Mobius* strip-shaped surface for the oil on the surface of PDMS from top (a) and side views (b).



Fig. S5. Uni-directional transportation dynamics on the inside of a peristomemimetic three-dimensional *Mobius* strip-shaped surface for the oil on the surface of PDMS from top (a) and side views (b).



Fig. S6. Swelling of the peristome-mimetic PDMS surface. (a) Length change of PDMS surface during the process of being immersed in n-hexadecane.



Fig. S7. Time sequences of the unidirectional transporting distances of the oil on the

surface of PDMS.

Notes and references

1. L. Wu, Z. Dong, H. Du, C. Li, N. Fang and Y. Song, *Research*, **2018**, 4795604.