

Preparation of a Janus copper mesh *via* nanoparticle interface self-assembly for
unidirectional water transportation

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Experimental Section: Materials and method

2.1 Materials

SDS-gel was synthesized based on our previous work¹. Hydrophobic colloidal particles (900 nm polystyrene (PS) and 1500 nm SiO₂, bought from Tianjin BaseLine Co., Ltd., China. Dilute nitric acid (HNO₃, wt=20%), sodium hydroxide (NaOH), ammonium ((NH₄)₂S₂O₈), and anhydrous ethanol were purchased from Shanghai Energy Biochemical Co., Ltd., China. The copper mesh (#150 mesh) was purchased from Anping Hangying Wire Mesh Co., Ltd., China. Double-sided tape was purchased from Shunsheng Co., Ltd., China. The square silicon wafer was purchased from Asatech Co., Ltd., China. Ultrapure water was used in all the experiments.

2.2 Preparation of superhydrophilic copper mesh by chemical oxidation

First, the original copper mesh was cleaned ultrasonically with anhydrous ethanol for 10 min and etched ultrasonically with nitric acid (wt.20%) for 5 min. The copper mesh was washed with deionized water and dried naturally. Secondly, the cleaned copper mesh was put into a mixed solution of 30 mL 0.2 mol/L (NH₄)₂S₂O₈ solution and 30 mL 5 mol/L NaOH solution and reacted for 15 min to grow the Cu(OH)₂ nanowires on the acid-etched surface, and finally rinsed with deionized water and dried naturally overnight. As a result, a superhydrophilic copper mesh was obtained.

2.3 Single-faced preparation of the hydrophobic coating on the superhydrophilic copper mesh

For design of nanoparticle layer, the nanoparticle layer was used to endow the one side of the copper mesh with superhydrophobic property. In this work, SiO₂ and PS nanoparticles were selected as building blocks. The PS nanoparticles were designed to act as adhesion layer to anchor the SiO₂ nanoparticle layer after fully melting. Therefore, the size of PS nanoparticle will not affect the performance of as-prepared Janus mesh. The SiO₂ nanoparticles was used to act as hydrophobic layer. An ideal hydrophobic surface relies on a hierarchical structure (micro/nano scale) and a low surface energy. Hence, the hydrophobic SiO₂ nanoparticles with proper size is important for the construction of hierarchical structure on the copper mesh. In this work, 1500 nm SiO₂ nanoparticles was used to construct this structure.

First, the 900 nm hydrophobic PS particles were distributed ultrasonically at 90 Hz in anhydrous ethanol for 5 min. The copper mesh was attached to the surface of the silicon wafer with double-sided tape. Secondly, the petri dish was filled with deionized water, and the PS suspension was gently added to the water with a dropper until it nearly overspread the water surface. The gel held with the wide-tipped tweezers was placed into one side of the petri dish and quickly removed after a few seconds. In the meantime, the colloidal particles self-assembled at the liquid-air interface. The copper-mesh-attached silicon wafer held with pointed tweezers was then dipped into the water moved below the particle layer, and removed from the bottom up so that the self-assembly monolayer was transferred to the surface of the copper mesh. The water attached to the tweezers was absorbed by the tissue paper, and the as-prepared sample was placed flat in a dry and windless place and dried naturally overnight. Subsequently, the glass pane was placed in the oven, and the copper mesh removed from the wafer was placed onto the glass pane when the temperature reached 150 °C and heated for 3 min. Finally, the copper mesh was attached to the cleaned wafer again, and the 1500 nm SiO₂ hydrophobic particles were used to repeat the above self-assembly process. After drying naturally overnight, the silicon wafer attached to the copper mesh was placed in the oven and heated at 150 °C for 3 min to obtain the Janus copper mesh.

2.4 Characterizations

The surface topography of all the samples is observed by a Scanning Electronic Microscope (SEM, HITACHI, SU8220), and the chemical components of all the samples are detected by the Energy Dispersive Spectrometer (EDS, Bruker). The dynamic and static contact angles of a water droplet on all the samples are measured by the optical contact angle measuring instrument (Dataphysics, OCA 15). The dynamic behavior of water droplets on the as-prepared Janus mesh was recorded by a mobile phone (iPhone 13).

Experimental Section: Numerical Simulation

1. Models

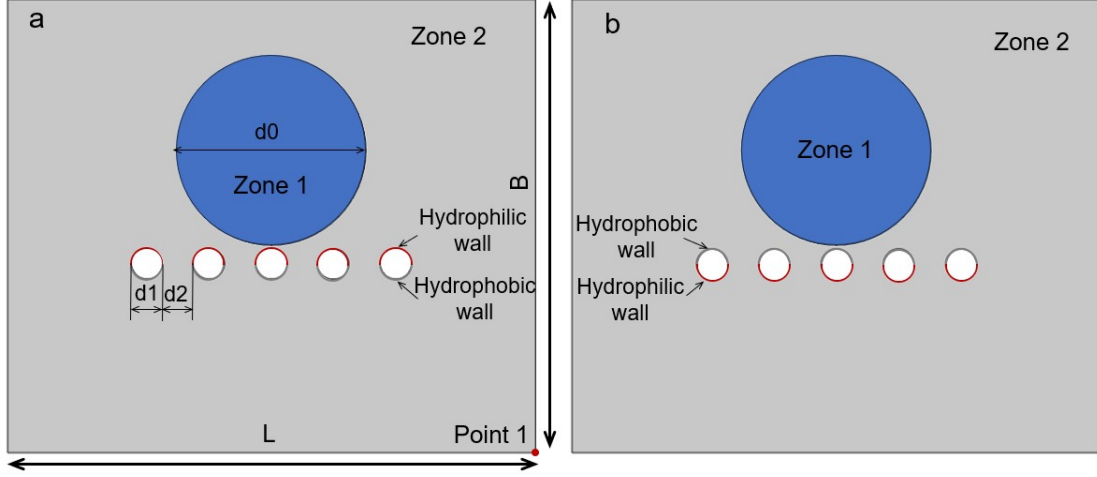


Figure S1. Simulation models of the dynamic behaviors of a water droplet on the Janus mesh: (a) The hydrophilic side facing the water droplet; (b) the hydrophobic side facing the water droplet

The simulation models and computation domains are shown in Figure S1. The initial value of Zone 1 was set as water phase and the diameter of Zone 1 was set as 0.5 mm (d_0). The initial value of Zone 2 was set as air phase. The red semicircular arc was set as the hydrophilic walls with a contact angle of 120° . The gray semicircular arc was set as the hydrophobic walls with a contact angle of 5° . These two semicircular arcs form a circle with a diameter of $80 \mu\text{m}$ (d_1), which was used to simulate the wire of the Janus mesh. The gap between the two circles is set as $85 \mu\text{m}$ (d_2), which simulates the pores of the Janus mesh. The whole computation domain is a rectangle with a size of $1.4 \text{ mm} \times 1.2 \text{ mm}$.

2. Governing Equations

The flow field is formulated by the continuity and the Navier-Stokes equations:

$$\rho \frac{\partial \mathbf{u}}{\partial t} + \rho(\mathbf{u} \cdot \nabla)\mathbf{u} = \nabla \cdot [-p\mathbf{I} + \mu(\nabla\mathbf{u} + (\nabla\mathbf{u})^T)] + F_{st} + \rho\mathbf{g} \quad (1)$$

$$\nabla \cdot \mathbf{u} = 0 \quad (2)$$

Where \mathbf{u} represents the velocity, \mathbf{I} denotes the identity matrix, ρ denotes the density, μ denotes the dynamic viscosity, p is the pressure, F_{st} represents the surface tension force exerted on the interface of two phases.

The phase field model was used to track the diffusive interface between water and

air. A dimensionless phase-field variable $\phi(x)$ is used to characterize two phases (water and air), where x is the position vector. $\phi_a = \phi(x) = 1$ represents water phase; $\phi_w = \phi(x) = -1$ represents the air phase; the diffusive interface between water and air is represented by the region with $-1 < \phi(x) < 1$.

The evolution of phase-field variables with time is produced by solving the Cahn-Hilliard equations, as follows

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = \nabla \cdot \frac{\gamma \lambda}{\varepsilon^2} \nabla \psi \quad (3)$$

$$\psi = -\nabla \cdot \varepsilon^2 \nabla \phi + (\phi^2 - 1)\phi \quad (4)$$

Where \mathbf{u} represents the velocity, γ represents the mobility parameter, and t is the time. λ is referred to as the mixing energy density, ε represents the interface thickness parameter, and ψ represents the phase-field help variable. The interfacial tension parameter (σ), mixing energy density and interface thickness parameter meet the following relation:

$$\sigma = \frac{2\sqrt{2}\lambda}{3\varepsilon} \quad (5)$$

3. Boundary conditions

All the boundary walls were set as non-slip boundaries. The contact angle of the boundary walls of the rectangle area was set as 90° . Point 1 was set as the pressure point constraint with a constant pressure of 0 Pa.

4. Mesh and Solving Method

The simulation was carried out by COMSOL Multiphysics. A free triangle mesh was used to divide the model. The maximum cell grid and minimum cell grid are set as 0.000804 mm and 0.000024 mm, respectively. The segregated solver was used in the model.

Additional Figures

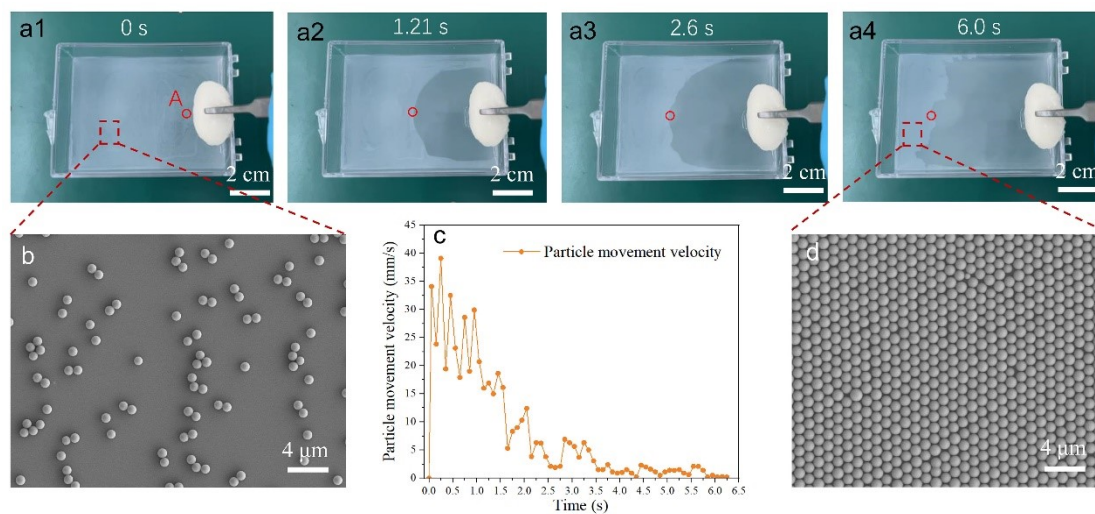


Figure S2. (a1-a4) Snapshots of SDS hydrogel-induced surface tension-induced colloidal nanoparticle self-assembly by taking 900 nm polystyrene particles as an example. (b) SEM image of loose colloidal particle layer formed at the water/air interface before the SDS-gel was inserted in water. (c) The Variation of particle movement velocity (point A) over time at an interval of 0.1s. (d) SEM image of the formed close-packed colloidal particle layer at the water/air interface.

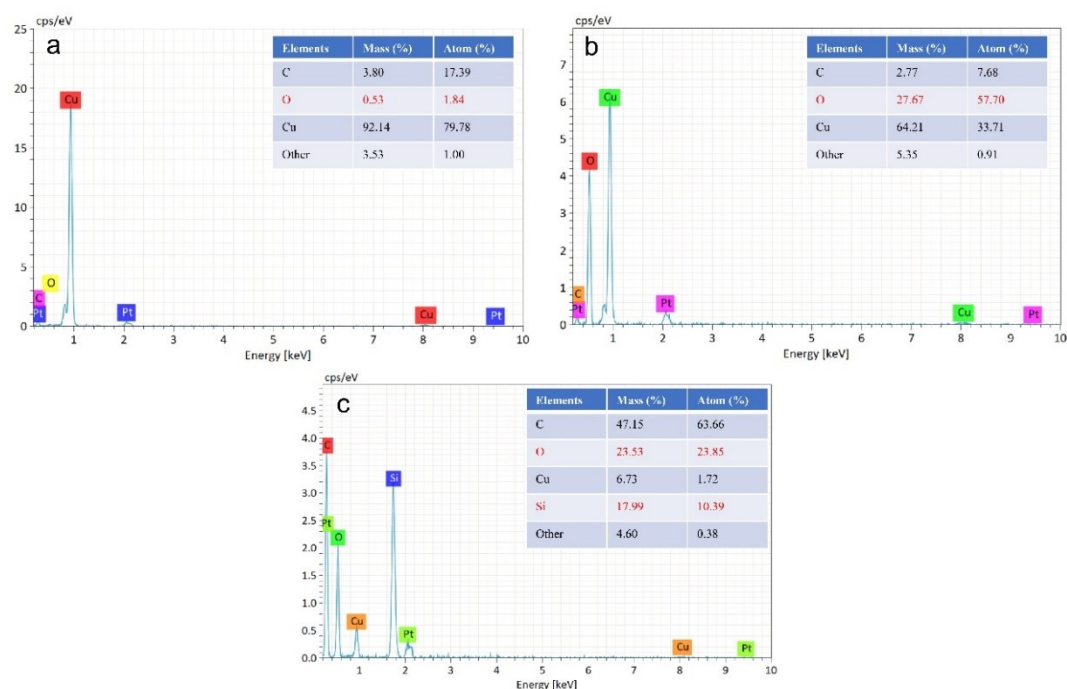


Figure S3. EDS spectrum of (a) pristine copper mesh (b) the hydrophilic side and (c) the hydrophobic side of the as-prepared Janus mesh.

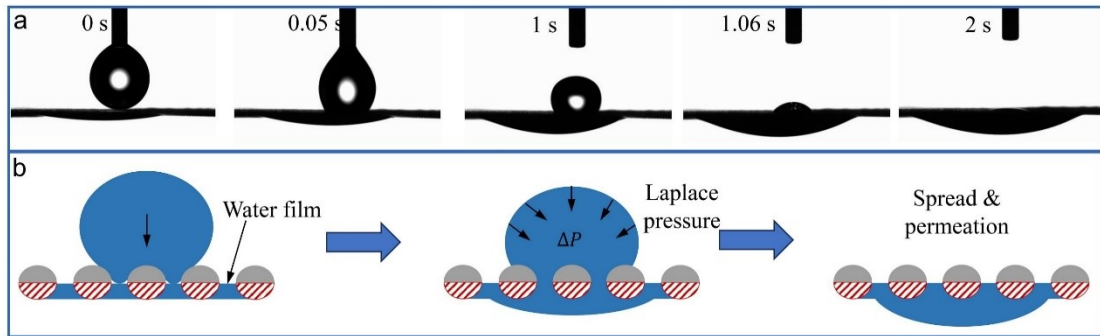


Figure S4. (a) Experimental Snapshots and (b) the corresponding illustration of the second water droplet permeating through the Janus mesh after the first water droplet permeating through the Janus mesh.

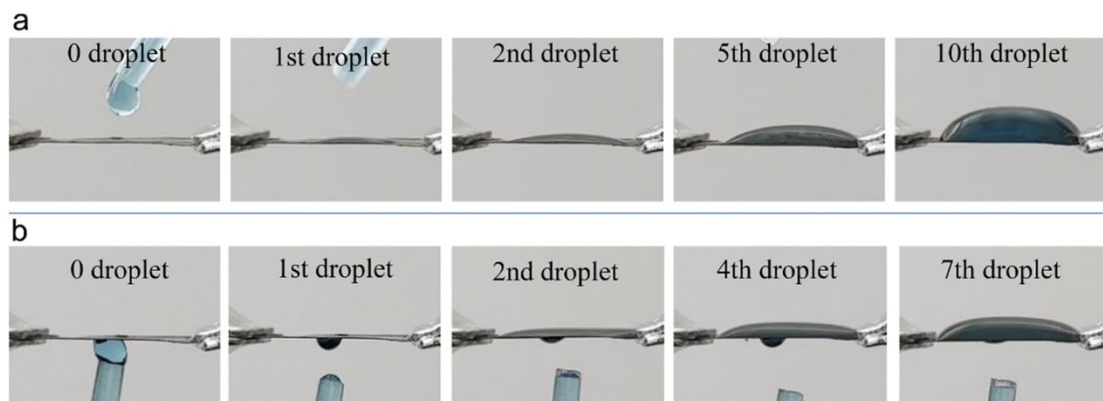


Figure S5. (a) Snapshots of water droplets dripping on the hydrophilic side of the Janus copper mesh drop-by-drop; (b) snapshots of the anti-gravity water directional transportation process (the water was dyed with methyl blue).

References

1. Li, X.; Chen, L.; Ma, Y.; Weng, D.; Li, Z.; Song, L.; Zhang, X.; Yu, G.; Wang, J., *Advanced Functional Materials* 2022, 32 (45).