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Supplementary Information

Reversible thermo-responsive transitions between aerogel-based porous liquids and solid-liquid-vapor triple-state gels

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Supplementary Note 1. Materials.

Superhydrophobic silica aerogel particles (SSAMs) purchased from IBIH Technology Co., LTD. (analytical pure), tap density 0.0629 g/cm^3; gelatin (used in microbiology, gel strength ~ 250 g Bloom), xanthan gum (USP/NF), methylcellulose (MC, 40000 mPa·s) and sodium phosphate dibasic (AR, 99 %) were purchased from Aladdin Limited; Potassium phosphate monobasic (AR, 99.5 %) purchased from Fuchen Chemical reagent factory; Sodium chloride (AR, 99.5 %) and potassium chloride (AR, 99.5 %) were purchased from Sinopharm Group Chemical reagent Company; Deionized water was used in all experiments.

Supplementary Note 2. Materials Preparation

Preparation of UC-PL and UC-TSG. Gelatin (2 g) was dissolved in 17.94 g of deionized water with vigorous stirring at 500 rpm for 30 min (60 °C), finally the gelatin solution was obtained. Then xanthan gum (0.06 g) was added to the gelatin solution and stirred for 30 minutes at room temperature to prepare gelatin/XG aqueous solution (UC-L). SSAMs (1.23 g, corresponding to 19.6 mL) was then added into 19.6 mL of UC-L and stirred at 2500 rpm for two hours to obtain gelation-porous liquid (UC-PL). UC-TSG was obtained by placing UC-PL at 4 – 25 °C for 20 minutes to obtain UC-TSG.

Preparation of LC-PL and LC-TSG. Sodium chloride (20 g), potassium chloride (0.5 g), disodium hydrogen phosphate (3.6 g), and dipotassium hydrogen phosphate (0.6 g) are dissolved in distilled water until the total volume reaches 500ml to prepare the 5 wt.% PBS solution. Then methyl cellulose (MC, 0.2 g) was added to the 19.8 g of PBS solution and stirred at 500 rpm for 60 min (90 °C). Subsequently, the upper mixture solution was stirred in the ice water bath for 2 hours to obtain PBS/MC aqueous solution (LC-L). SSAMs (1.23 g, corresponding to 19.6 mL) was then added to 19.6

mL of LC-L and stirred at 2500 rpm for two hours to obtain LC-PL. Finally, LC-TSG was obtained by placing at 40 °C for 10 min.

Calculation details

Supplementary Note 3. Calculation details for the porosity volume of the porous fluid

$$P = 1 - \frac{\rho}{\rho_0} \tag{S1}$$

$$\rho_0 = \rho_{water} \times \omega_{water} + \rho_{sio_2} \times \omega_{sio_2} \tag{S2}$$

The parameter P refers to the porosity volume of the PL, ρ is the measured density of the PL, ρ_{water} is the density of the polymer aqueous solution (1.02 g·cm^{-3}), ω_{water} is the mass fraction of the polymer aqueous solution (94.16 wt%), ρ_{sio_2} is the intrinsic density of silica (2.2 g·cm^{-3}), and ω_{sio_2} is the mass fraction of SSAMs in PL (5.84 wt%).

Supplementary Figures

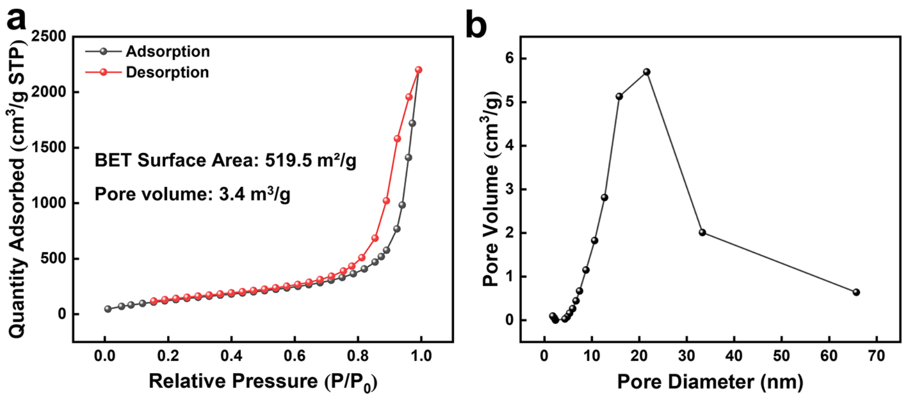

Figure S1. The BET analysis of SSAMs. (a) Nitrogen adsorption and desorption isotherms; (b) pore size distribution curve.

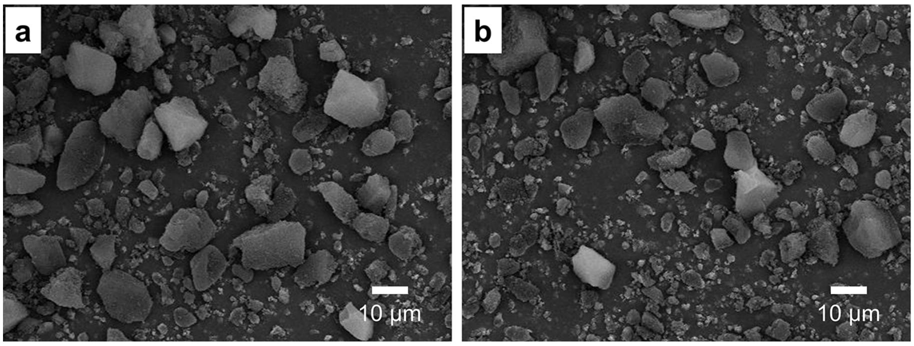

Figure S2. The SEM image of SSAMs.

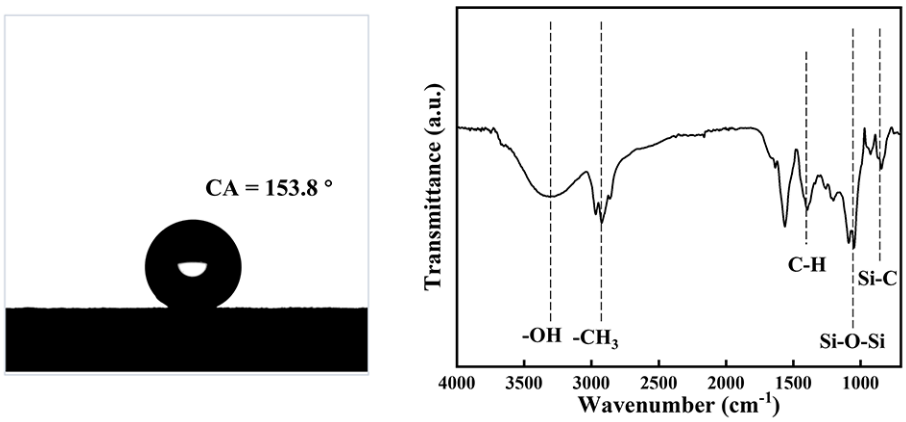

Figure S3. The contact angle and FT-IR spectrum of SSAMs.

Figure S4. Optical photos of UC-PL without adding XG during 2 hours standing test.

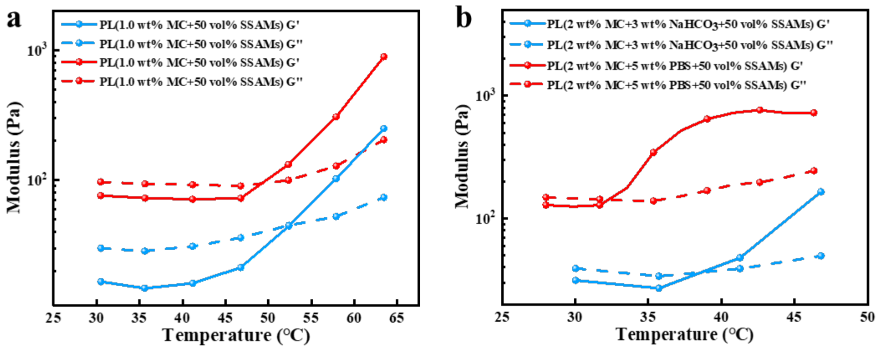

Figure S5. The storage modulus (G′) and loss modulus (G″) of PLs (the deformation value set at 1.5%). (a) PLs were fabricated by mixing equal volumes of SSAMs and a MC aqueous solution at concentrations of 1.0 wt% and 2.0 wt%, respectively. (b) PLs were fabricated by mixing equal volumes of SSAMs and MC aqueous solution (2.0 wt%) with the addition of PBS (5.0 wt%) and $NaHCO_3$ (3.0 wt%), respectively.

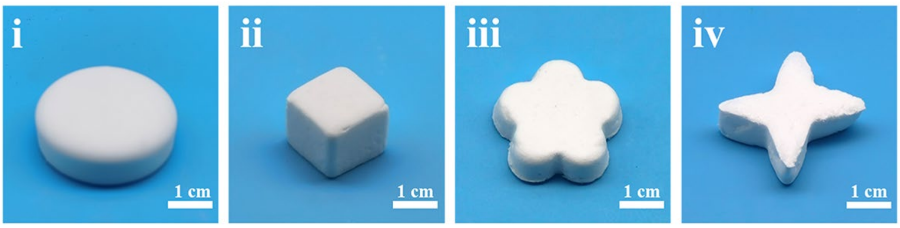

Figure S6. Optical photos of various shapes of TSGs. (i) and (ii) are UC-TSG; (iii) and (iv) are LC-TSG.

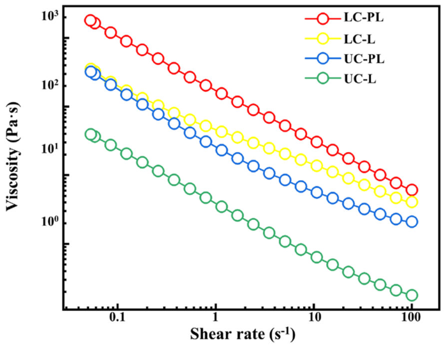

Figure S7. The curve of shear viscosity with the shear rate. The rheological tests of gelatin-directed fluids and MC-directed fluids were carried out at 30 °C and 28 °C, respectively.

Figure S8. Contact angle of *n*-hexane on UC-TSG (ca. 0°).

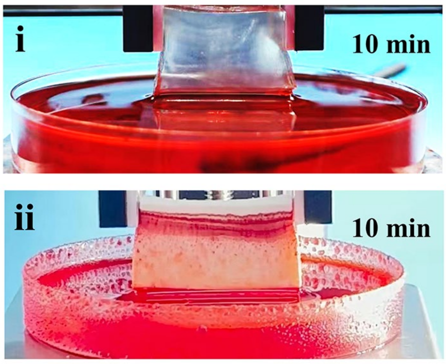

Figure S9. Optical photographs of (i) UC-G and (ii) UC-TSG for adsorbing *n*-hexane dyed with Sudan Red III after 10 min.

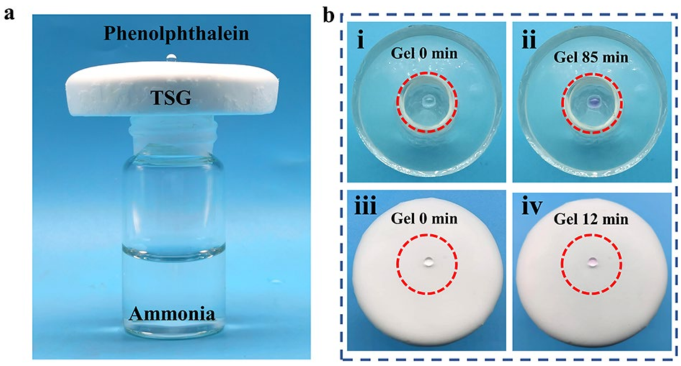

Figure S10. (a) The device for NH_3 transfer determining. (b) The comparison for NH_3 passing through UC-G (I and ii) and UC-TSG (iii and iv). A 73 mins faster NH_3 transfer was observed on UC-TSG than UC-G as NH_3 changes the color of the phenolphthalein indicator in a shorter time when passing through UC-TSG.

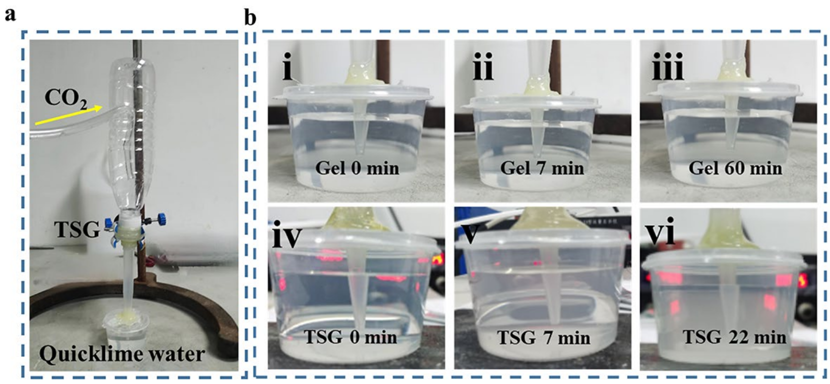

Figure S11. (a) CO_2 transfer detector. (b) The comparison for CO_2 passing through UC-G (I, ii and iii) and UC-TSG iv, v and vi). At least 53 mins faster CO_2 transfer was observed on UC-TSG than UC-G as CO_2 clouding quicklime water in a shorter time when passing through UC-TSG.

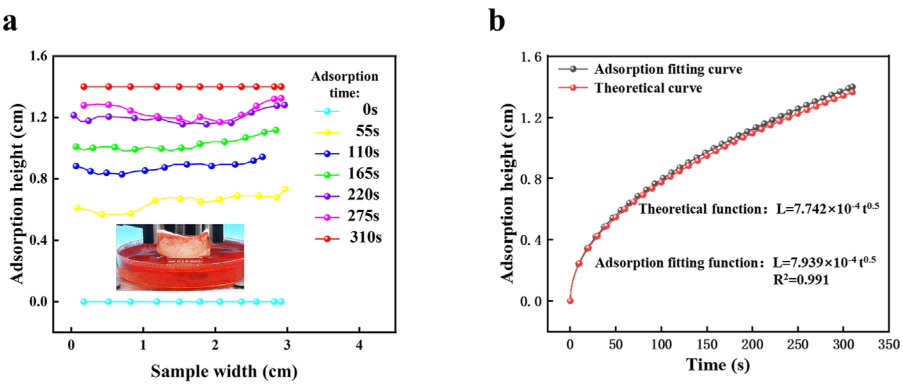

Figure S12. (a) The adsorption height of dyed *n*-hexane by LC-TSG within 310 s (inset is the photographs of LC-TSG in *n*-hexane after 310 s). (b) The comparison between the theoretical and actual adsorption equations.

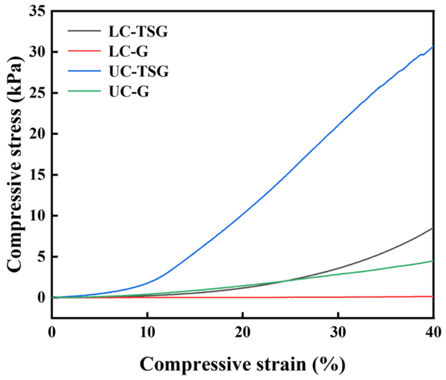

Figure S13. Compressive stress-strain curves of TSGs and hydrogels.

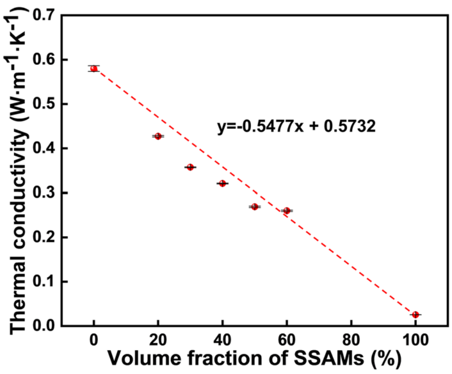

Figure S14. Approximate linear relationship between thermal conductivity (*k*) of gelatin-directed gels and volume fraction of SSAMs ($\phi_{aerogel}$).

The relationship between the thermal conduction (k) of SSAMs-based gels and volume fraction of SSAMs (*ϕaerogel*) is based on following equation:

$$k = k_{aerogel} \times \phi_{aerogel} + k_{hydrogel} \times (1 - \phi_{aerogel}) \quad \text{(S3)}$$

where $k_{aerogel}$ is the thermal conductivity of SSAMs (0.0254 $W \cdot m^{-1} \cdot K^{-1}$), $k_{hydrogel}$ is thermal conductivity of UC-G (0.5798 $W \cdot m^{-1} \cdot K^{-1}$).

So,

$$k = -0.5344\phi_{aerogel} + 0.5798 \quad \text{(S4)}$$

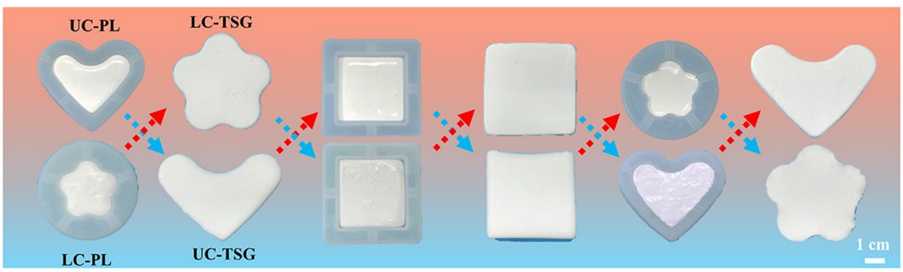

Figure S15. TSG's plastic-like processing capabilities.

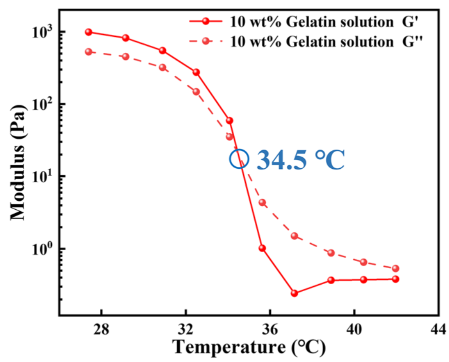

Figure S16. The UCST of gelatin solution measure by the intersection point of energy storage modulus (G ') and loss modulus (G").

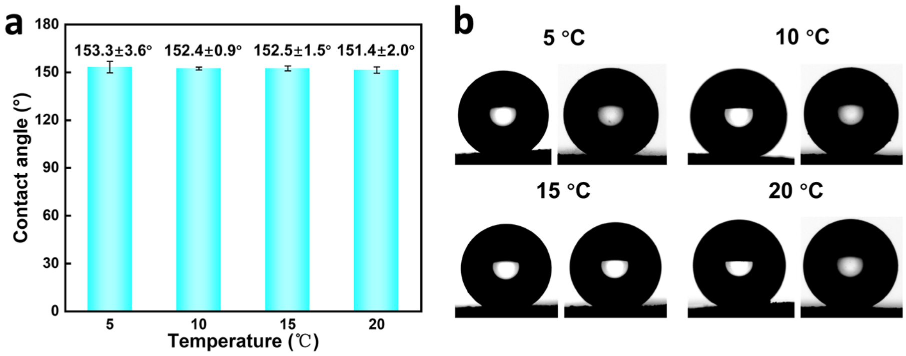

Figure S17. The contact angle changes of UC-TSG at various temperature. (a) The contact angles of UC-TSGs at various temperature. (b) Photos recorded by contact angle measuring instrument.

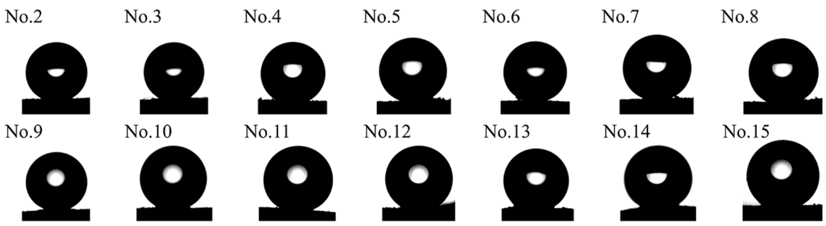

Figure S18. Contact angle of water on UC-TSG during 15 sol-gel transitions.

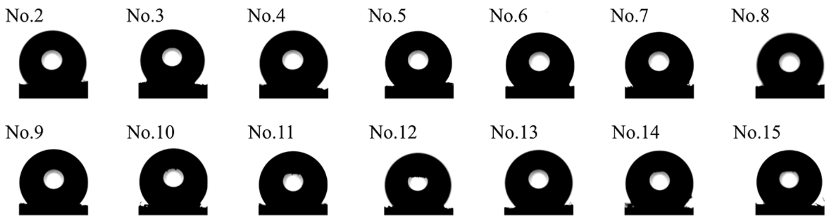

Figure S19. Contact angle of water on LC-TSG during 15 sol-gel transitions.

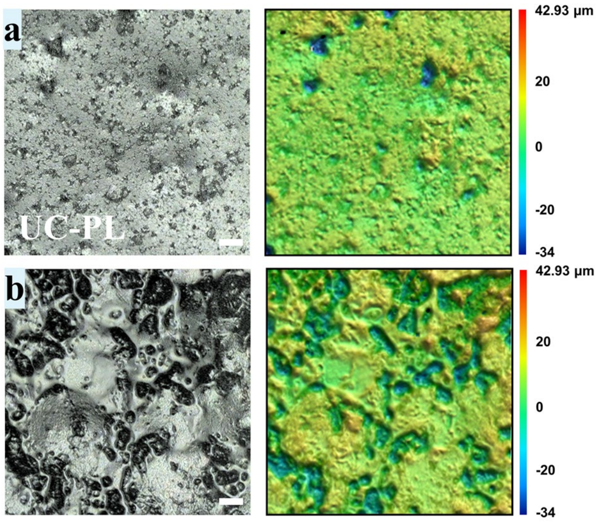

Figure S20. The 3D confocal microscopy image and surface height fluctuation analysis of UC-PL (a) and LC-PL (b) (Scale bar 20 μm).

Figure S21. (a) Photograph of a water droplet on UC-PL. (b) Photograph of a water droplet on UC-PL using a contact angle meter. (c) Schematic for a water droplet on UC-PL.

Figure S22. (a) Photograph of a water droplet on LC-PL. (b) Photograph of a water droplet on LC-PL using a contact angle meter. (c) Schematic for a water droplet on LC-PL.

Supplementary Tables

Table S1. The adsorption height of *n*-hexane dyed with Sudan Red III for gelatin-directed gels with various volume fraction of SSAMs.

Volume fraction of SSAMs in gelatin-directed gels (vol%)	Adsorption time (s)	Adsorption height of dyed *n*-hexane (mm)
10	710	2
20	710	2
30	710	13
50	710	20

Table S2. Thermal conductivity of TSG and its corresponding PLs.

Sample	Thermal conductivity [W/(m·K)]
UC-TSG	0.2677
UC-PL	0.2843
LC-TSG	0.2577
LC-PL	0.2390