

Supplementary Information

Stretchable, healable, adhesive, transparent, anti-drying and anti-freezing organohydrogels toward multi-functional sensors and information platforms

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Fabrication of organohydrogel sensor and electrical measurements

The organohydrogel sensor was fabricated by connecting two adhesive copper electrodes with conductive wires to both sides of an organohydrogel strip. To evaluate the performance of as-prepared organohydrogel as a strain sensor, the relative resistance changes under different strain states were investigated by an electrochemical workstation (Chenhua, CHI760E). The relative change of the resistance ($\Delta R/R_0$) was calculated on the basis of the monitored current-voltage curves at different strains: $\Delta R/R_0 = (R - R_0)/R_0$, where R_0 and R are the resistances without and with applied strain, respectively. The gauge factor (GF) was defined as $GF = (\Delta R/R_0)/\varepsilon$, where ε is the applied strain. Meanwhile, the sensitivity of the organohydrogel for monitoring temperature

variations can be calculated from the slope of linearly fitted curve ($S = \Delta R/R_0/\Delta T$).¹ Moreover, in order to protect the skin, the organohydrogel sensor is first attached to the commercial VHB tapes and then they are integrated onto human joints for detecting various human motions.

Mechanical property and transmittance measurements

The tensile tests of organohydrogels were carried out using a microcomputer-controlled electronic universal testing machine (JM-140PT, Hangzhou, China). The sample was made into a strip shape with 40 mm × 9 mm × 2 mm (effective length = 20 ± 1 mm), and the strain rate was fixed at 50 mm min⁻¹. The transmittance of the organohydrogel was tested by using a UV-vis spectrophotometer (Mapada P4, Shanghai, China) from 700 to 350 nm. The sample was cut into a strip shape with thickness of 1 mm, and then was attached to the inside of the quartz cell.

Anti-freezing property and long-term moisture-retention property measurements

For anti-freezing property of the organohydrogel, the temperatures of freezing and ice melting were characterized by using differential scanning calorimetry (DSC, TA Q2000). The sample was cooled from 20 to -90 °C at a rate of -5 °C min⁻¹ and then heated to 20 °C. The long-term moisture-retention performance of the organohydrogel was obtained by the weight change of sample at room temperature.

Self-healing property measurement

To test the healing ability, an organohydrogel sample was cut into half using a scalpel and half portion of the organohydrogel was dyed purple. Subsequently, the purple organohydrogel was kept in contact with the complementary half of the organohydrogel, and they were then placed under normal environment (20 °C) for healing the damage. Similarly, during the self-healing process at low temperature, the contacted organohydrogel at the fresh surface was placed in the

refrigerator of $-50\text{ }^{\circ}\text{C}$ for healing the damage.

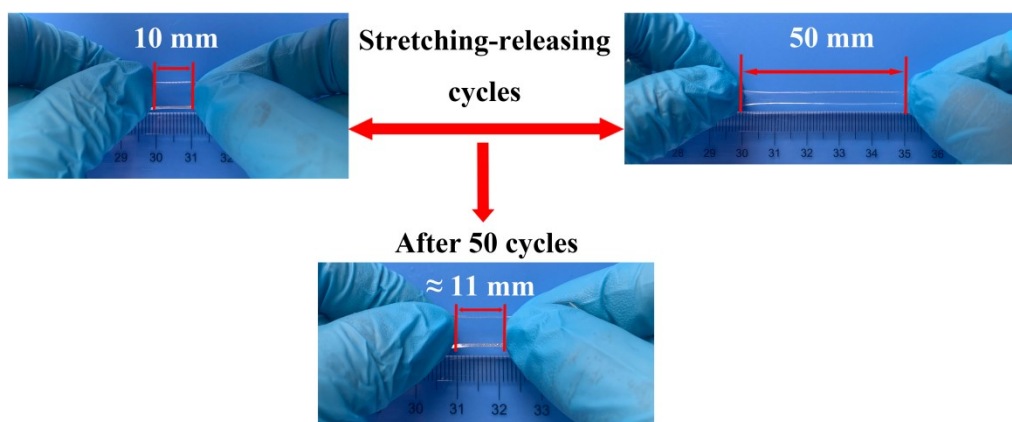


Fig. S1. The excellent fatigue resistance of organohydrogels during 50 consecutive stretching-releasing cycles.

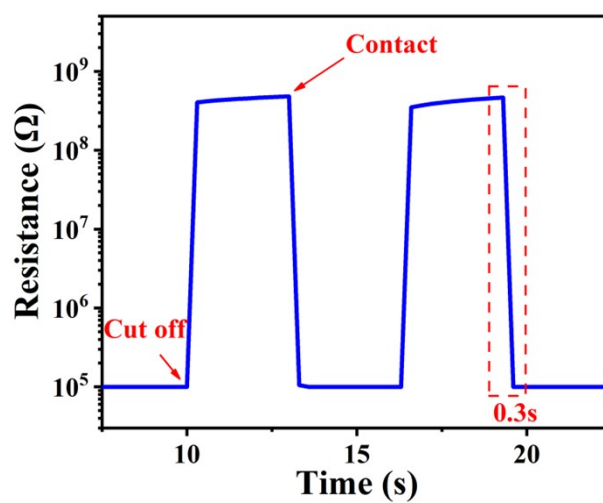


Fig. S2. Time evolution of the healable process for the organohydrogel by the real-time resistance measurements.

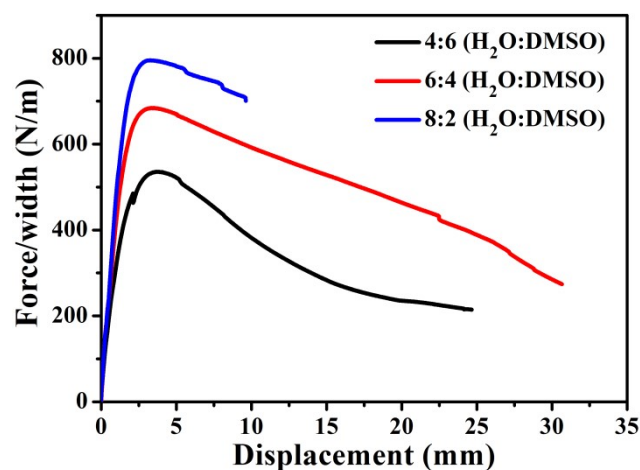


Fig. S3. Peeling strength of different solvent component ratios based organohydrogels

onto glass surface.

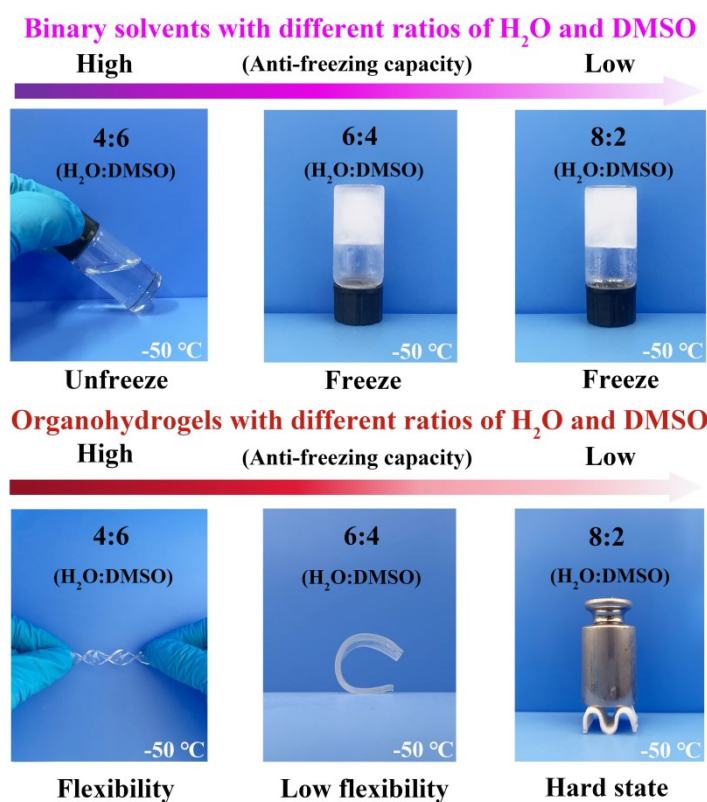


Fig. S4. The anti-freezing abilities of binary solvents and organohydrogels

with different ratios of H₂O and DMSO.

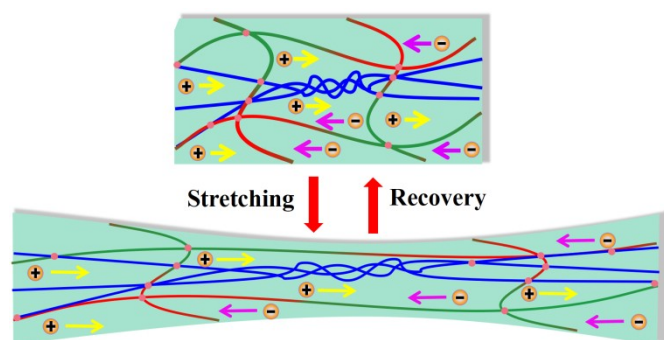


Fig. S5. Schematic illustration of the changes of organohydrogel structure according to external stretching deformation.

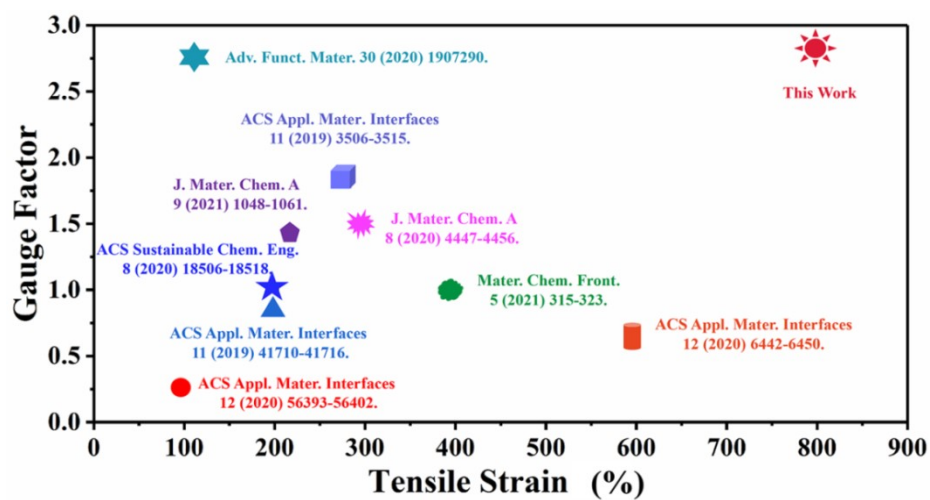


Fig. S6. A scatter diagram showing the gauge factor of some reported strain sensors.

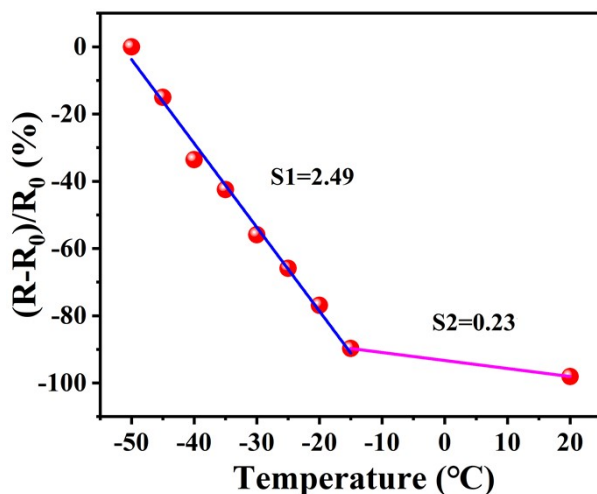


Fig. S7. Relative resistance-thermal response curve of the organohydrogel sensor.

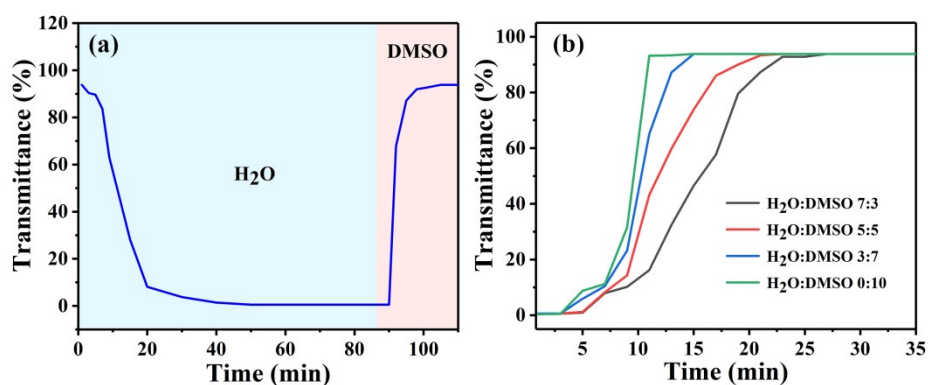


Fig. S8. (a) Transmittance changes of the organohydrogel in water and DMSO. (b) Transmittance changes of the organohydrogel in mixed solutions of DMSO and water at diverse ratios.

In the organohydrogel system, the salt ions of sodium citrate (Na_3Cit) can induce gelatin chains to form hydrophobic interaction regions.² When as-prepared organohydrogel was immersed in water, the DMSO molecules inside the organohydrogel were replaced by the solvent exchange.³⁻⁵ As consequently, the hydrophobic interaction regions in the organohydrogel may be separated from the water and aggregated gradually, which led to the transparent

organohydrogel turned completely opaque white within 1.5 h (Fig. S8a). However, the water-treated organohydrogel changed from opaque white to transparent in the DMSO. This change can be explained by the theory in which the aggregated hydrophobic regions were re-dissolved in the DMSO solution during the penetration of DMSO molecules into the organohydrogel.⁶ In addition, the transparent changes of the organohydrogel in a mixed solution of DMSO and water at different ratios were investigated and the results as shown in Fig. S8b. The time required for the organohydrogel to return to a transparent state decreased gradually as the content of DMSO increased.

References

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