

## Supporting Information

### Thermochromic and Conductive Hydrogels with Tunable Temperature Sensitivity for Dual Sensing of Temperature and Human Motion

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## **S1. Measurements and testing of structure and properties**

### *S1.1. Transparency testing of hydrogels*

Transparency of the SN-PAAM hydrogels was measured using a UV spectrometer (Lambda 365) (Platinum Elmer). The light absorption of the hydrogel samples in the wavelength range of 400 to 800 nm was used to determine the transparency. The organic hydrogel specimens were cut into strips (1 cm × 4.5 cm) and placed directly in a UV spectrometer solid test cell.

### *S1.2. FT-IR measurements of hydrogels*

An infrared spectrometer (Spectrum two) (Platinum Elmer) (RMS signal-to-noise ratio of 4 cm<sup>-1</sup>, 64 scans) was used to characterize the structure of the SN-PAAM hydrogel samples. The hydrogels did not require any treatment prior to the measurements.

### *S1.3. Rheological testing*

Dynamic rheological measurements of SN-PAAM hydrogels were performed with a torque rheometer (DHR-2, TA). The dynamic viscoelastic properties of the as-synthesized samples were tested in the range of 0.01 rad/s to 100 rad/s at a fixed strain amplitude of  $\gamma=0.1\%$ . The as-synthesized samples were swept at 25°C using a parallel plate of 25 mm diameter.

### *S1.4. Measurement of the mechanical properties of the hydrogels*

Tensile mechanical measurements were carried out using a tensile machine (6800) (INSTRON) with a stretching speed of 80 mm/min to evaluate the mechanical properties of SN-PAAM hydrogels at room temperature. Young's modulus was calculated from the slope of the stress-strain curve of 50-100% or more. Toughness was obtained by integrating the area under the stress-strain curve. The loading-unloading cycle of the hydrogel was also measured by the tensile testing machine described above. In the cyclic tensile test of a single sample, a sample of SN-PAAM hydrogel is taken

and cyclically stretched with a first tensile strain of 200%, followed immediately by a second cycle with a strain of 400%, then a third cycle with a strain of 600% and a fourth cycle with a strain of 800%, until a tensile cycle with a strain of 1,000% is completed (5 cycles per strain), and then unloaded, always using the same sample during the stretching cycle, with no dwell time between any two cycles.

#### *S1.5. Performance testing of the hydrogel sensors*

The organic hydrogel was used as a strain sensor. Copper wires were connected to both sides of the organic hydrogel sensor to act as electrodes. After applying a fixed voltage of 3 V to the sensor, the resistance of the organic hydrogel sensor was measured using a digital multimeter (2400) (Keithley). The conductivity ( $\delta$ ) of the hydrogel can be calculated using the following equation.  $\delta = L / (R \times S)$ , where  $\delta$ , L, R and S are the conductivity ( $S\ m^{-1}$ ), the length of the SN-PAAM hydrogel (m), the resistance ( $\Omega$ ) and the cross-sectional area of the hydrogel ( $m^2$ ), respectively. A stretching machine and a digital multimeter were used to test the strain-sensing properties of the conductive hydrogel samples. First, the hydrogel was fixed on the stretching machine. At the same time, the ends of the hydrogel were connected to a digital multimeter, which was used to record the curve of resistance of the hydrogel with strain. The change in relative resistance was calculated by:  $\Delta R/R_0 = (R-R_0)/R_0$ , where  $R_0$  and R are the resistance in the absence and presence of strain, respectively.

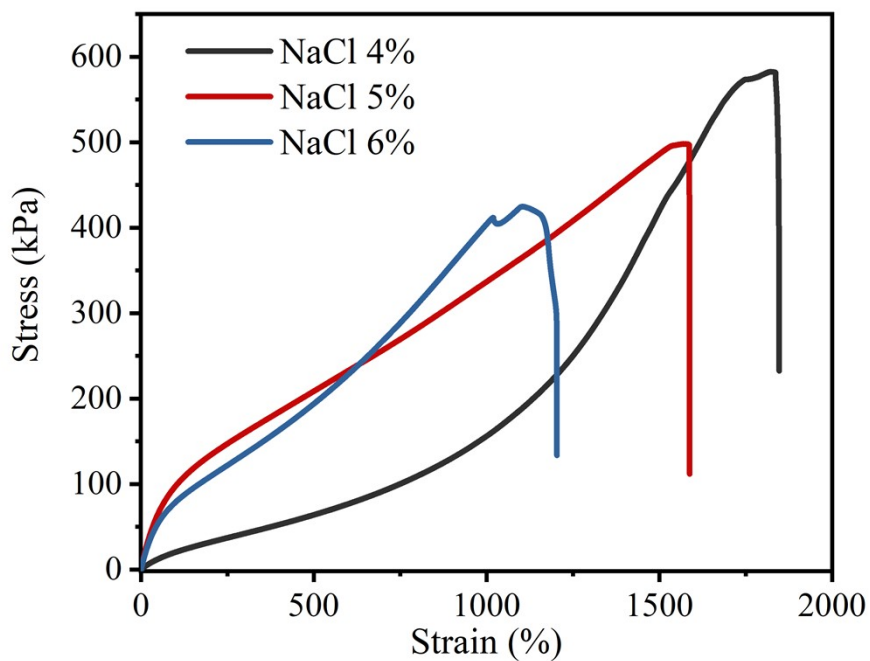
#### *S1.6. Optical microscopy analysis of the hydrogels*

Optical microscope photographs of the hydrogels were obtained by Germany Leica DM4M at different temperatures and time.

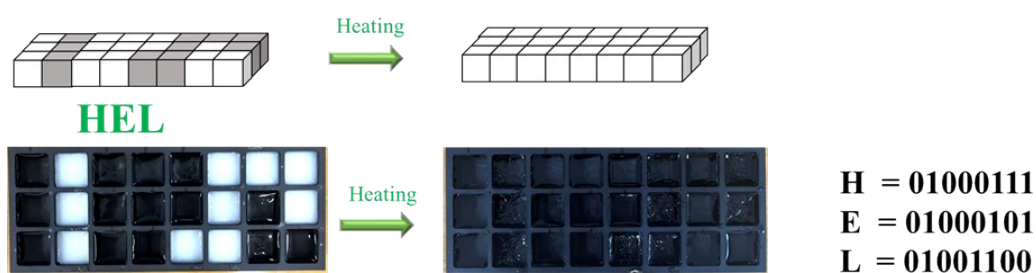
#### *S1.7. Adhesive property testing of the hydrogels*

A tensile machine (6800) (INSTRON) is used for peeling test of the hydrogels. One side of the hydrogel is adhered to various solid surfaces, and the surface is peeled off at a rate of 5 mm/min in a vertical 180° direction.

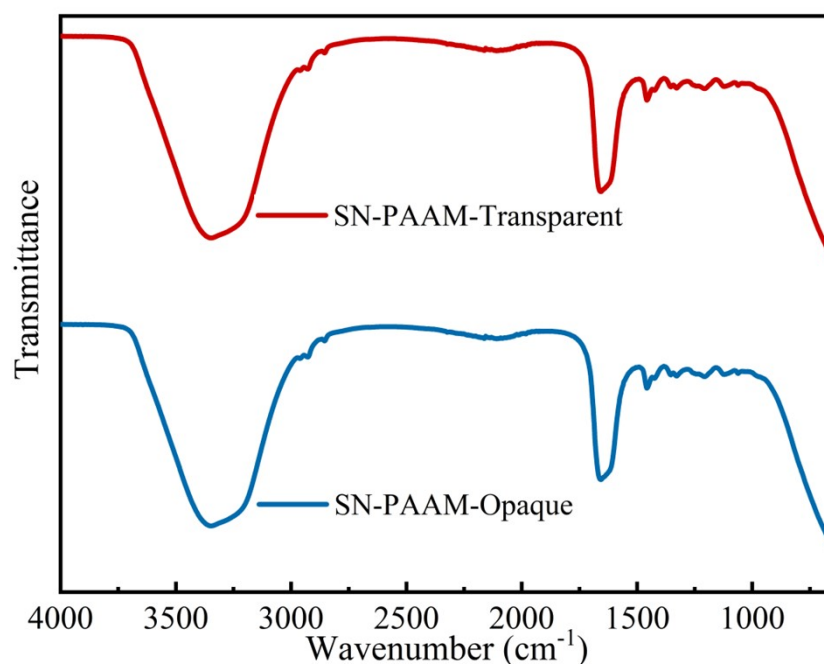
## S2. Supporting Figures



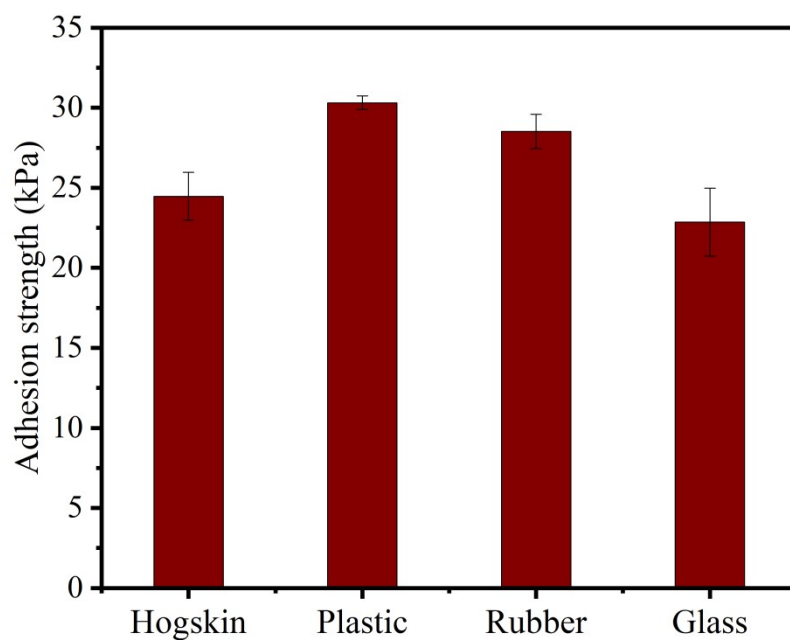
**Figure S1.** Tensile stress-strain curves and corresponding toughness and modulus of elasticity of SN-PAAM hydrogels with different NaCl contents (4%, 5%, and 6%).



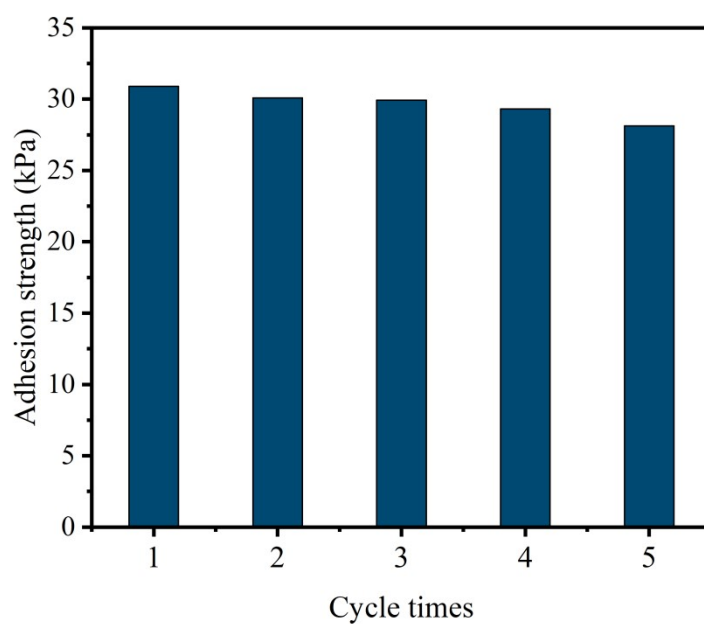
**Figure S2.** Binary encoding of hydrogels: schematic diagram of binary encoding of hydrogels and pictures of binary encoding of hydrogels.



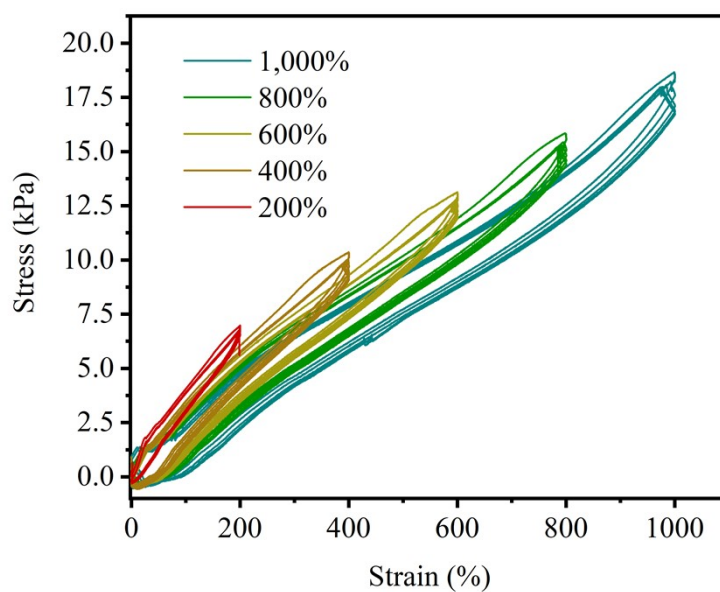
**Figure S3.** FT-IR spectra of SN-PAAM hydrogels in transparent and opaque states.



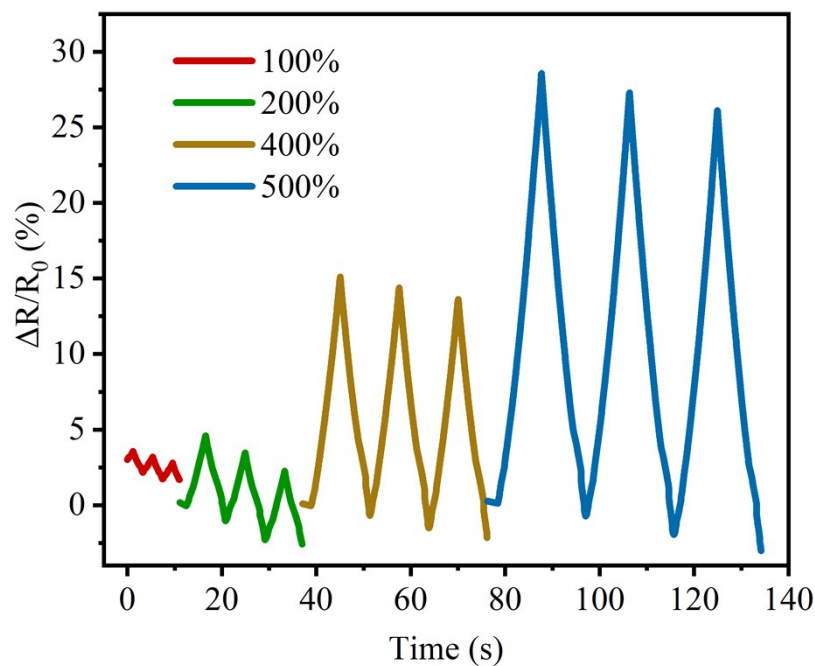
**Figure S4.** Adhesion strength of the SN-PAAM hydrogel on different surfaces.



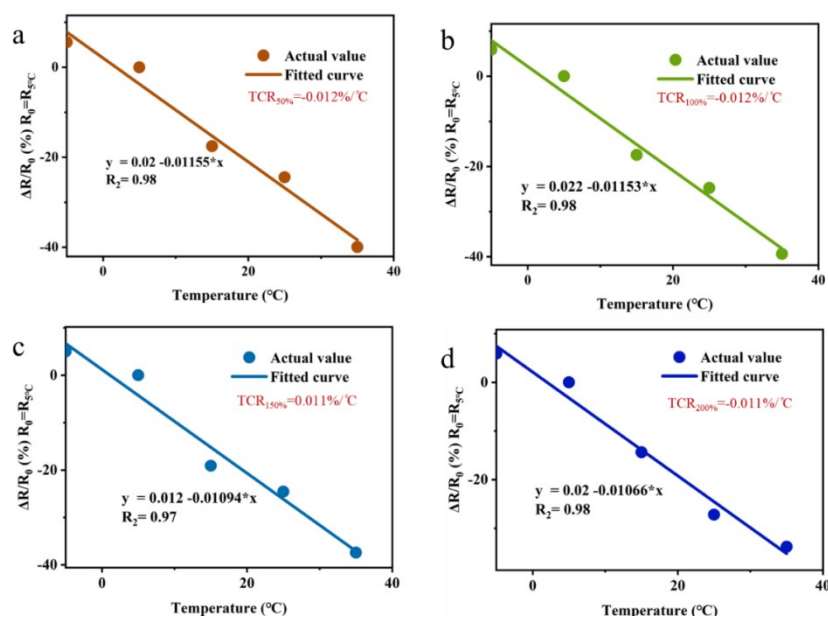
**Figure S5.** Repeatable adhesion behavior of the SN-PAAM hydrogel on plastics.



**Figure S6.** Five loading-unloading tests of the hydrogels at different strains (200%, 400%, 600%, 800% and 1,000%) with no standstill interval.



**Figure S7.** Electrical sensing performance of the hydrogels. Relative resistance variation of the organo-hydrogel sensor at medium strains (100%-500%).



**Figure S8.** Relative resistance as a function of temperature from  $-5^{\circ}\text{C}$  to  $35^{\circ}\text{C}$ .  $R_0$  is the resistance at  $20^{\circ}\text{C}$ , and the hydrogel with a NaCl content of 4% is used as a temperature sensor. a, b, c, and d show the temperature coefficient of resistance under strains of 50 %, 100 %, 150 %, and 200 %, respectively.

### S3. Supporting Tables

**Table S1.** Performances and functions of our hydrogel compared with some previously reported multifunctional hydrogels. (“Yes” stands for having the function; “No” stands for NOT having the function).

Ref.	Mechanical properties	Dual response	Application
<b>Our work</b>	Tensile strength: ~581 kPa; Elongation: ~1,836 %	Yes	Strain and temperature
[15]	Tensile strength: ~700 kPa; Elongation: ~1,956 %	No	Strain
[19]	-	No	Strain
[16]	Tensile strength: ~1,255 kPa; Elongation: ~1,600 %	No	Strain
[34]	Tensile strength: ~2,200 kPa; Elongation: ~522 %	Yes	Strain and UV light
[37]	Tensile strength: ~1,000 kPa; Elongation: ~900 %	No	Strain
[38]	Tensile strength: ~3.25 kPa; Elongation: ~1,919 %	No	Strain
[39]	Tensile strength: ~152 kPa; Elongation: ~3,000 %	No	UV light

**Table S2.** Feeding amount for preparation of SN-PAAM hydrogels.

	SDS (g)	NaCl (g)	AM (g)	LMA (μL)	KPS (g)	H <sub>2</sub> O (mL)
<b>1</b>	1	0	2	65	0.07	20
<b>2</b>	1	0.3	2	65	0.07	20
<b>3</b>	1	0.6	2	65	0.07	20
<b>4</b>	1	0.9	2	65	0.07	20
<b>5</b>	1	1.2	2	65	0.07	20