Supporting information

Direct Current Electric Field Induced Gradient Hydrogel Actuators with Rapid Thermo-Responsive Performance as Soft Manipulators

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Experiment section

Characterization: The morphology and size distribution of TCNCs were analyzed with transmission electron microscopy (TEM) using a JEM-2100 microscope (JEOL, Japan), where a drop of TCNCs suspension (0.01 wt %) was dropped and evaporated on a copper grid before measurement. The potentials of TCNCs were determined by ZEN3600 Zetasizer (Malvern Instruments, UK) at 25 °C. The chemical structure of the hydrogel was characterized by NICOLET 5700 Fourier transform infrared spectrometer (FTIR) (Thermo Fisher Scientific, US).

Swelling ratio: The hydrogel sample was cut into cylindrical specimen of 9 mm ×5 mm (Diameter × Height), and incubated in distilled water to reach swelling equilibrium. The swelling ratio of hydrogel actuator at 25 °C was calculated according to **Equation S1**,

$$Swelling\ Ratio = \frac{W_e}{W_d} \tag{S1}$$

where W_e is the weight of the swollen gel at 25 °C and W_d is the weight of the gel at dry state.

The deswelling kinetics of hydrogel at 40 °C was measured gravimetrically. At predetermined time intervals, the hydrogel samples were taken out from the aqueous solution. Water retention of hydrogel sample was defined as **Equation S2**,

$$Water\ Retention = \frac{W_t}{W_e} \tag{S2}$$

where W_t and W_e is the weight of hydrogel at 40 °C and the weight of hydrogel at 25 °C after swelling equilibrium.

Mechanical testing: The mechanical properties of hydrogels were carried out by a universal

material testing machine with a 100 N load cell (CMT6350, SANS, China) at room temperature. The disk-like hydrogel samples with 9 mm in diameter and 9 mm in height were compressed at a speed of 2 mm min^{-1} to obtain stress-strain curves. For tensile testing, hydrogel specimen was cut into 30 mm× 5 mm× 1 mm and stretched under a speed of 20 mm min^{-1}.

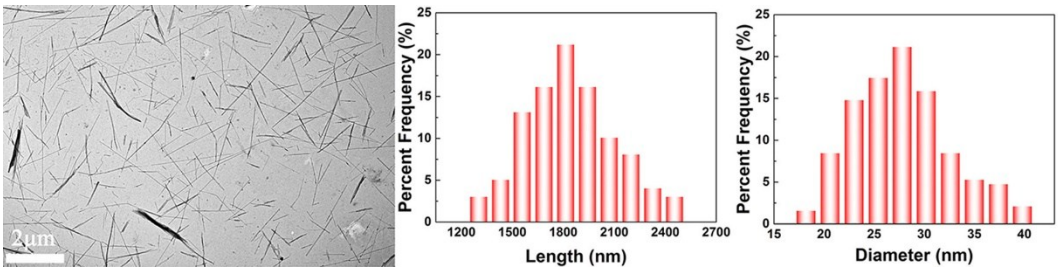

Fig. S1. TEM image (a), length (b), and diameter distribution (c) of TCNCs.

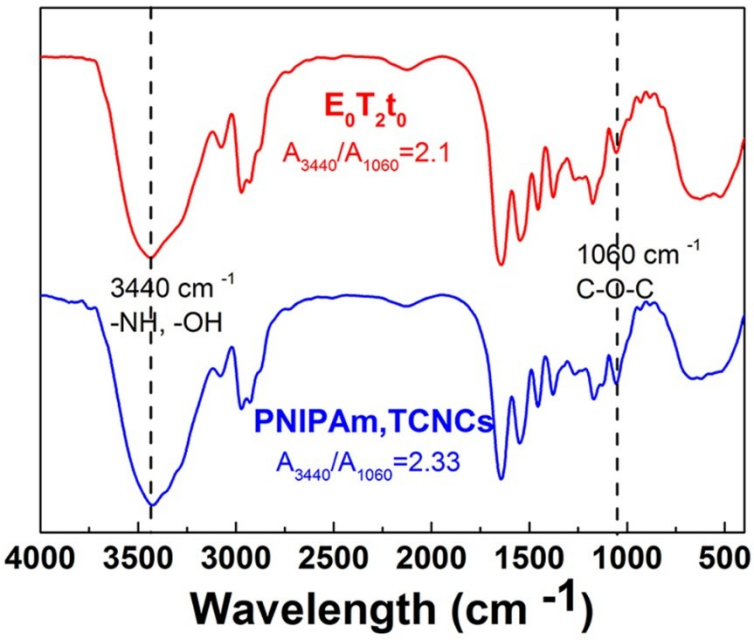

Fig. S2. FTIR spectra of the gradient hydrogel ($E_0T_2t_0$) and PNIPAM/TCNC mixture.

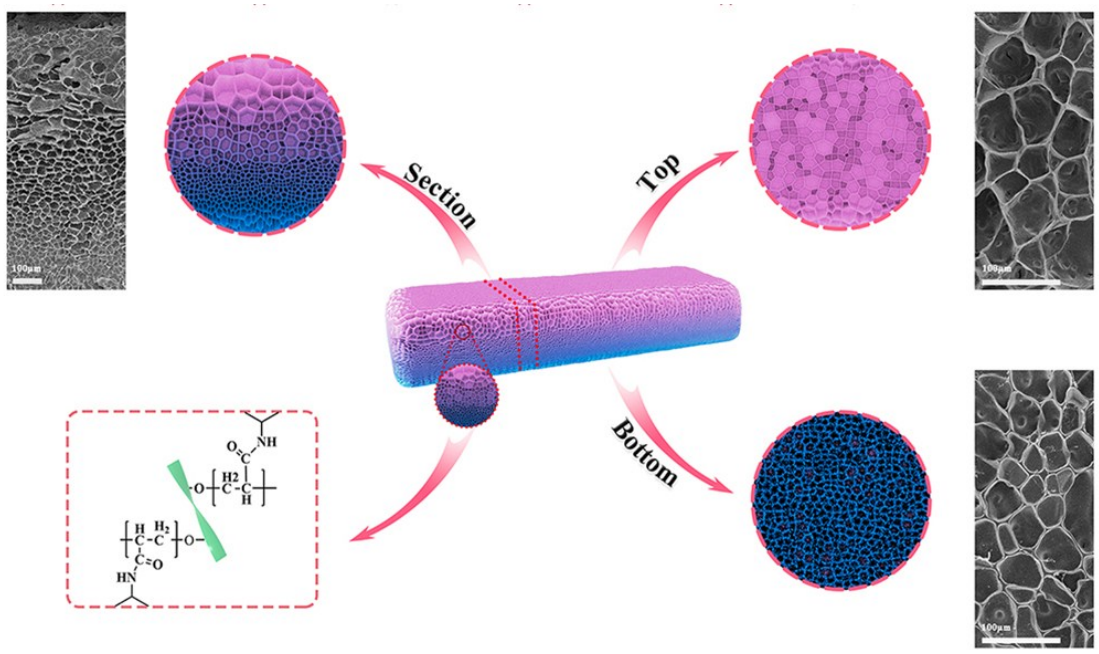

Fig. S3. Schematic illustration of the microstructure of PINIPAM/TCNC gradient hydrogel.

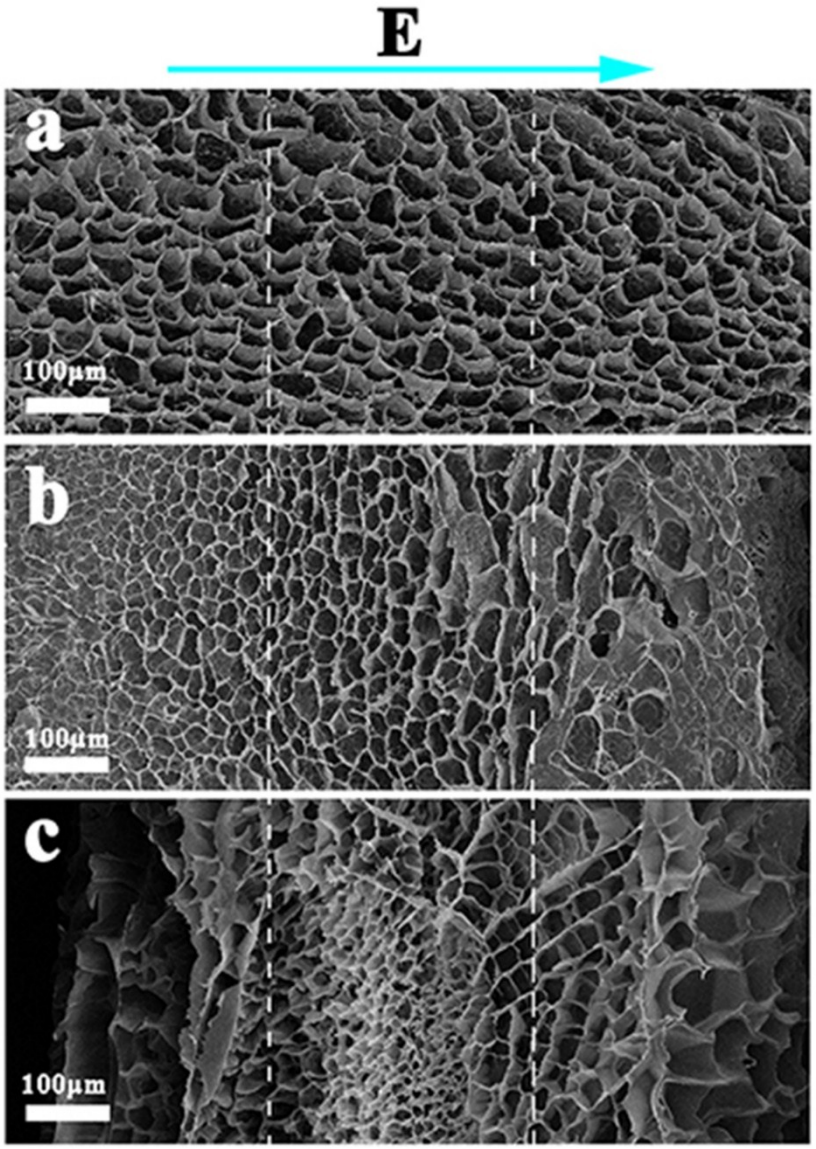

Fig. S4. SEM images of hydrogel samples: (a) $E_0T_2t_0$, (b) $E_1T_2t_{30}$, and (c) $E_2T_2t_{30}$.

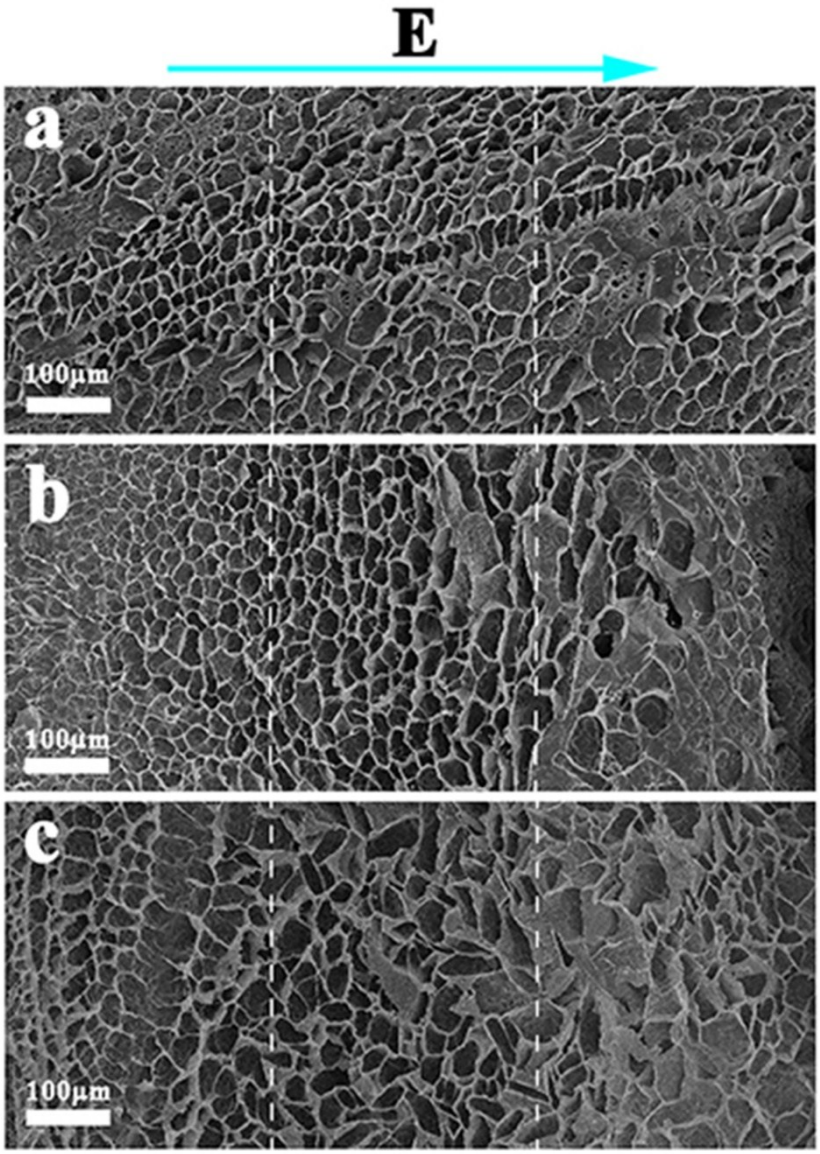

Fig. S5. SEM images of hydrogel samples: (a) $E_0T_0t_0$, (b) $E_1T_2t_{30}$, and (c) $E_1T_5t_{30}$.

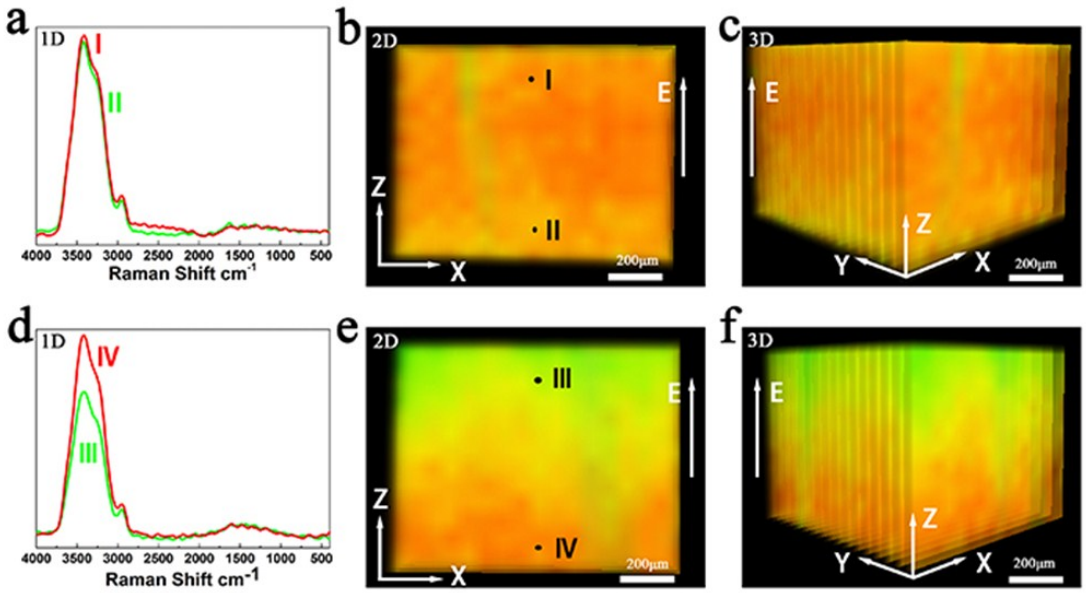

Fig. S6. (a, d) Raman spectra of the –OH/-NH stretching intensities (3000−3400 cm^{-1}) at I, II, III, and IV, (b, e) reconstructed 2D images of the -OH/-NH stretching intensities distribution on section, (c, f) 3D Raman images of $E_0T_2t_0$ and $E_1T_2t_{30}$, respectively, which were derived from multivariate curve resolution (MCR) model with OMNIC software.

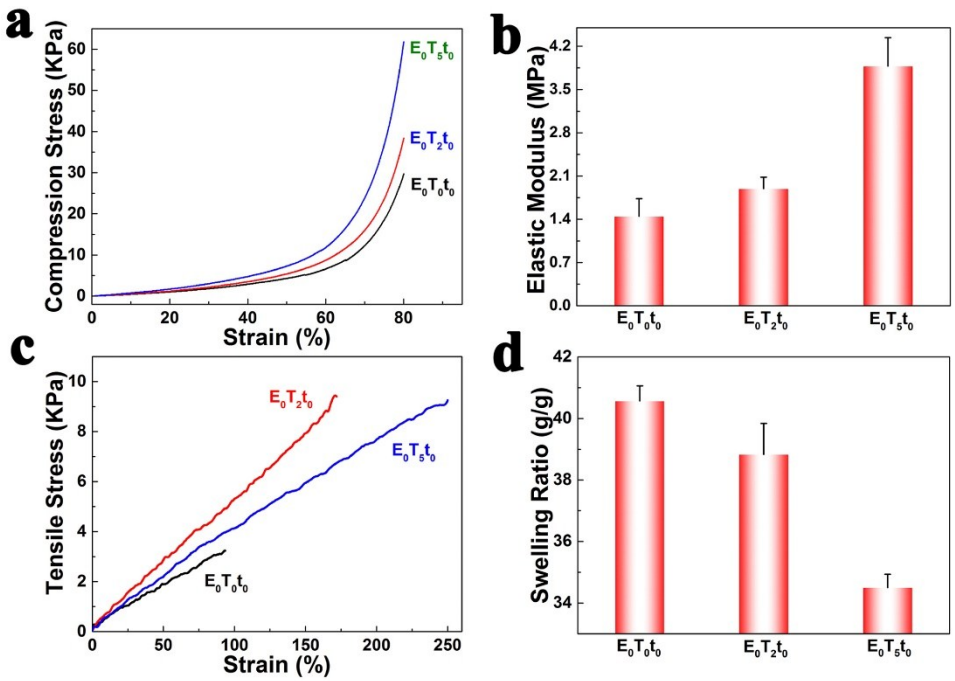

Fig. S7. (a) Compressive stress-strain curves, (b) related elastic modulus, (c) tensile stress-strain curves, and (d) equilibrium swelling ratios of hydrogel samples.

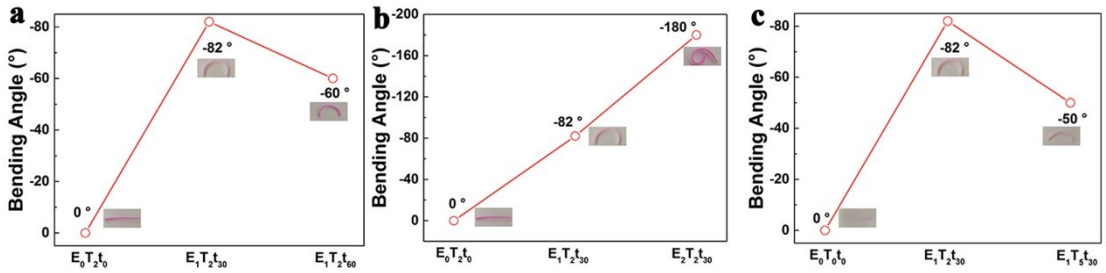

Fig. S8. Effect of different preparation conditions on bending angle of gradient hydrogel after equilibrium swelling in 25 °C water: (a) inducted time, (b) intensity of electrical field, and (c) TCNCs contents.

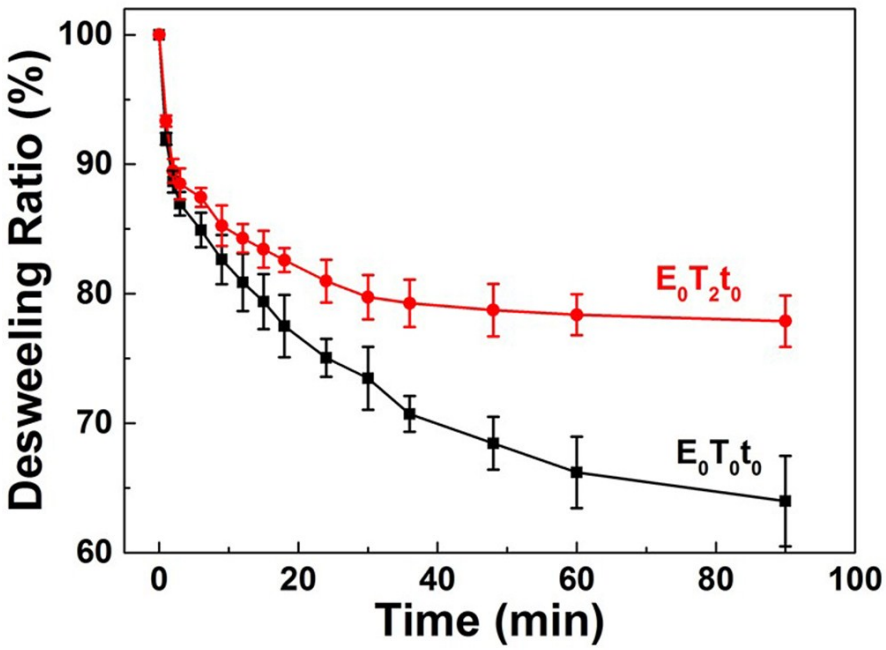

Fig. S9. Deswelling kinetic curves of $E_0T_0t_0$ and $E_0T_2t_0$ in water at 40 °C.

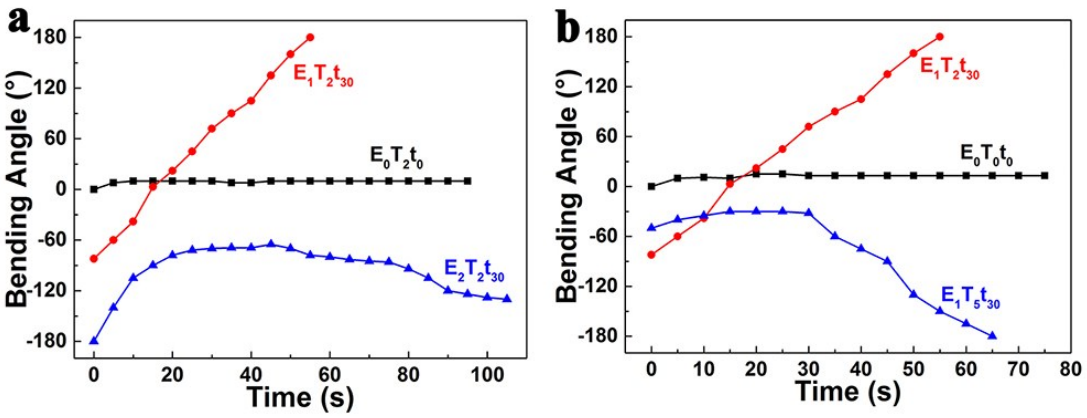

Fig. S10. Effects of preparation conditions on temperature-sensitive bending of gradient hydrogels at 40 °C: (a) intensity of electrical field and (b) TCNCs contents.

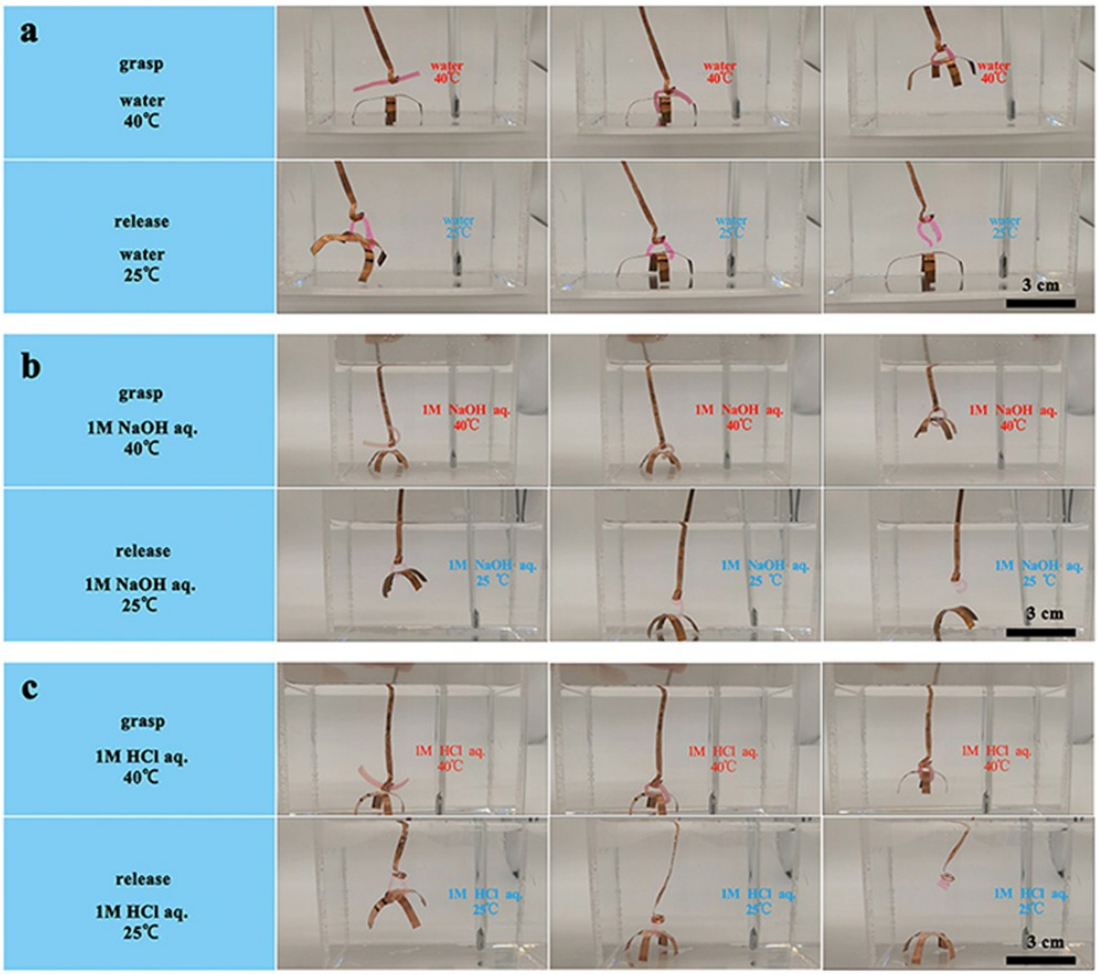

Fig. S11. Application of PNIPAM/TCNC gradient hydrogel actuator as a manipulator to grab and release objects in water (a), 1M NaOH (b), and 1M HCl (c) aqueous solutions.

Table S1. Summarization for the bending and recovery velocity of hydrogel actuators.

Samples	Bending V (°/s)	Recovery V (°/s)	Bending T (°C)	Recovery T (°C)	Ref.
PNIPAM/PAAM-PTCA BH	3.3	0.06	45	25	1
PNIPAM/PDMAPMA BH	2.3	0.02	60	25	2
PNIPAM/P(AAc-co-AAM) BH	3.3	0.03-0.04	40	15	3
PNIPAM/PDMAEMA BH	2.2	0.27	45	15	4
PNIPAM/GO BH	0.6	0.1	40	20	5
PNIPAM /PAM/Clay BH	0.8	0.19	42	24	6
Al-alginate/PNIPAM BH	3.5	0.2	30	20	7
PNIPAM/Clay BH	0.6	0.2	40	25	8
PNIPAM/GO BH	1.7	0.1	25	20	9
PNIPAM/GO GH	0.8-3.8	0.1	50	25	10
PNIPAm/Laponite GH	0.3-1.2	0.2	50	25	11
PNIPAM/TCNCs GH	4.8	1.4	40	25	--

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