Supporting Information

Human-Tissue-Inspired Anti-Fatigue-Fracture Hydrogel for Sensitive Wide-Range Human-Machine Interface

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Experimental Section

Materials. Acrylamide (AM), lauryl methacrylate (LMA), and sodium dodecyl benzene sulfonate (SDBS) were purchased from Aladdin Reagent Co. Sulfuric acid (H₂SO₄, 95-98%) and potassium persulfate (KPS) with the analytical grade were provided by Kelong Chemical Reagent Company (China). KPS was recrystallized from methanol/H₂O solution before use, and all other reagents were used as the received without further purification. Multiwall carbon nanotubes (MWCNTs) aqueous suspension (TNM2, the purity>95 wt%) with CNTs content of 2.1 wt% was obtained from Chengdu Organic Chemicals Co., Ltd in Chinese Academy of Sciences. The purchased MWCNTs aqueous suspension could be obtained by the ultrasonic processing with 400W for 20 min and the magnetic stirring for 5 h with the addition of 1.2 wt.% noncovalent surfactant (TNWDIS, aromatic modified polyethylene glycol ether, which could also be purchased from Chengdu Organic Chemicals Co., Ltd). Stretchable tape (VHB4905) was purchased from the company of 3M. Medical purified cotton and other chemical reagents were all purchased from Kelong Chemical Reagent Company (China).

Preparation of the CNCs aqueous suspension. The cellulose nanocrystals (CNCs) aqueous suspensions were obtained via the hydrolysis of medical absorbent cotton with sulfuric acid according to the literature.¹ Specifically, the purified cotton (10 g) was mixed with the preheated sulfuric acid solution (64 wt.%, 200 mL) at 50 ºC for 1 h. Then, the reaction was quenched by the dilution with deionized water. The obtained mixture was centrifuged at 10000 rpm for 5 min, and then the dialyzed against water was performed for one week to remove the residual sulfuric acid fully. The solid content of the obtained CNCs suspension was 0.98 wt.%.

Synthesis of HAPAM/CNCs@CNTs hydrogel. The self-healable HAPAM/CNCs@CNTs hydrophobically associating hydrogels were synthesized according to the following procedure. Taking the hydrogel with 0.8 wt.% CNCs@CNTs contents as an example. Firstly, the MWCNTs aqueous suspension (3.83 mL) was added into the prepared CNCs suspension (8.05 g), and then magnetically stirred for 6 h followed by the sonication for 10 min to obtain a stable suspension. Next, the hydrophilic monomer AM (3.0 g, 15 wt.%), the surfactant SDBS (0.6 g, 3 wt.%), and the hydrophobic monomer LMA (0.3 g, 1.5 wt.%) were dissolved in a certain amount of deionized water (4.22 mL), then stirred at least 6 h to obtain a homogeneous solution. After that, the obtained monomer solution was mixed with the as-prepared CNCs@CNTs suspension to
prepare the AM/CNCs@CNTs precursor solution. After 2 h stirring, the initiator KPS (0.017 g, 0.5 wt.% relative to the total mass of AM and LMA) was added into the mixture. Subsequently, the precursor solution was poured into home-made reaction equipment which was assembled by two PTFE plates and a hollow silicon rubber spacer (Figure S1, supporting information). Finally, the sealed reaction equipment was placed at 60 °C for 6 h for the polymerization reaction. After polymerization, the obtained plate-shaped HAPAM/CNCs@CNTs gel can be peeled off from the PTFE plate. The weight content of water in the resultant gel were 82%. Note that the size of the HAPAM/CNCs@CNTs hydrogel can be tuned by adjusting the thickness of the rubber spacer and dimensions of PTFE plates. The detailed synthesis processes of pure HAPAM, HAPAM/CNCs, and HAPAM/CNTs hydrogels are elaborated below. The total mass of all the hydrogels synthesized in this study was fixed at 20 g to achieve a reasonable comparison.

**Synthesis of pure HAPAM hydrogel.** The pure HAPAM hydrogel used in this study was synthesized according to the following procedure. The hydrophilic monomer AM (3 g, 15 wt.%), the surfactant SDBS (0.6 g, 3 wt.%), and the hydrophobic monomer LMA (0.3 g, 1.5 wt.%) were dissolved in 16.1 g deionized water. Then, the mixture was stirred for 6 h to obtain a uniform and homogeneous solution. Then, the initiator KPS (0.017 g) was added into the mixture. After stirring for 5 min, the mixture was placed at 60 °C with 6 h for polymerization.

**Synthesis of HAPAM/CNCs hydrogel.** The HAPAM/CNCs@CNTs hydrogel was synthesized according to the following method. 3 g AM, 0.6 g SDBS, and 0.3 g LMA were dissolved in the mixture solution of 8.05 g deionized water and 8.05 g CNCs aqueous suspension. After stirring for 6 h, 0.017 g KPS was added into the mixture. After stirring for 5 min, the mixture was placed at 60 °C with 6 h for polymerization.

**Synthesis of HAPAM/CNTs hydrogel.** The HAPAM/CNTs hydrogel can be prepared according to the following method. 3 g AM, 0.6 g SDBS, and 0.3 g LMA were dissolved in the mixture solution of 12.27 g deionized water and 3.83 g CNTs aqueous suspension. After stirring for 6 h, 0.017 g KPS was added into the mixture. After stirring for 5 min, the mixture was placed at 60 °C with 6 h for polymerization.

**Fabrication of the strain sensor and the human-motion detection.** To assemble the strain sensor, the hydrogel was cut into strips with different dimensions (40×10×2 mm for large-scale movement detection, 25×5×1 mm for facial expressions and pronunciation detection). Two
individual copper wires were wrapped around both ends of the strip sample, and then two double-sided conductive flexible copper tapes were tightly wrapped on the wires to obtain a stable signal output. Then, the strip sample was encapsulated by the stretchable VHB tape to prevent the water evaporation. Before the encapsulation, to improve the adhesion among the copper tape, VHB tape, and the gel, each surface of the gel was slightly wiped by ethanol and then dried with N₂ for 2 min via removing water from the gel surface. For the large-scale body movement detection, the sensor was fixed on the forefinger, wrist, and knee of the tester to detect their routine movements. The specific position of the sensor on the finger, wrist, and knee can be referenced from Movie S3. For the facial expression detection, the sensor was fixed on the forehead of the tester to detect the expressions of lifting the forehead and frown. Besides, the sensor was mounted on the corner of eye to detect the expressions of closing eyes. For the pronunciation detection, the sensor was tightly fixed on the Adam’s apple of the tester to collect the current change during speaking. The specific position of the sensor on the forehead, the corner of eye, and the Adam’s apple can be referenced from Movie S4 and S5.

Characterizations. Zeta potential (ζ potential) of CNC, CNTs, CNCs@CNTs, and the AM monomer solution, as well as the AM/CNCs@CNTs aqueous suspensions with a concentration of around 0.2 mg/mL, were measured using Zetasizer nano-ZS apparatus (Malvern, UK). Fourier transform infrared (FTIR) spectra in the region of 4000-400 cm⁻¹ were collected at a resolution of 4 cm⁻¹ with 20 scans by a Nicolet iS50 spectrometer (USA) equipped with a deuterated triglycine sulfate (DTGS) detector. Raman spectra were collected at room temperature on a Horiba LabRAM HA apparatus (Horia Co., France) with an excitation laser λ = 532 nm. The morphology of the CNCs@CNTs aqueous suspension was studied by a Dimension 3000 AFM microscope (Digital Instruments) with a soft tapping mode at 0.7 Hz. Transmission electron microscope (TEM, JEOL JEM-100CX, Japan) was used to investigate the distribution of CNTs in the gel matrix. An ultrathin section of the freeze-dried HAPAM/CNCs@CNTs hydrogel cut by a microtome was used for TEM observation. The surface morphology and microstructure of the gel were observed by scanning electron microscope (SEM, JEOL, JSM-5900LV, Japan). Before SEM experiments, the hydrogel samples were freeze-dried for 48 h. Rheological experiments of the hydrogels were performed by an AR 2000EX rheometer (TA instrument) using a parallel plate (diameter in 20 mm) on a peltier plate in a frequency sweep mode from 0.1 to 100 rad s⁻¹ at a
fixed strain of 1% at 25 °C. All the tensile and compression tests were conducted on a versatile testing machine (Instron 5567, USA) (see below for the detailed information on mechanical tests). Laser confocal microscopy (LCSM, LSM700 Carl Zeiss, German) was performed to observe the surface morphology of the original and healed sample (laser wavelength is 405 nm, the laser intensity is 5, and pinhole diameter is 1 Å). The electrical resistance was measured by a two-point probe resistance measurement system using a Keithley 2400 source meter (USA). Note that, except for the resistance of the hydrogel sample, all the resistance values displayed in this work also include the contact resistance between the copper wires and the hydrogel. The current signals of the strain sensor were measured in real-time using a two-point measurement with a Keithley 2601B source meter (USA).

**The details of mechanical tests.** All the mechanical tests were performed at room temperature using a versatile testing machine (Instron 5567, USA) with a 1 kN load cell. All the tensile tests were performed on the dumbbell-like samples (3 mm in thickness, 4 mm in width, and 50 mm in length) with a fixed stretching speed of 100 mm/min. The dumbbell-like specimen was cut from the hydrogel plate by dumbbell shape cutter. The compression test was carried out with the cylinder samples (17 mm in diameter) in a constant compressing speed of 3 mm/min. The nominal stress ($\sigma$) is calculated by dividing the force ($F$) by the initial cross-section area ($A_0$) of the sample ($\sigma=F/A_0$), and the strain ($\varepsilon$) was defined as the ratio of the gauge length ($L$) to the initial gauge length ($L_0$, 20 mm in this work) ($\varepsilon=(L-L_0)/L_0$). The elastic modulus ($E$) was calculated from the slope of the initial linear region of the stress-strain curve (0-30% strain).

The toughness ($T$) was calculated from the area under the stress-strain curve according to the following equation:

$$T = \int_{\varepsilon_0}^{\varepsilon_f} \sigma(\varepsilon)\,d\varepsilon$$

where $\varepsilon_0$ and $\varepsilon_f$ represent the initial strain and the strain at break, respectively. The cyclic tensile tests were conducted with three minutes interval between the adjacent cycles, while the cyclic compression tests were conducted with no interruption between the adjacent cycles.

**The anti-fatigue-fracture test.** The dumbbell-like hydrogel sample with a pre-crack (about 1 mm depth) was used for the anti-fatigue fracture property test. The sample was stretched and released at a speed of 500 mm/min. We sprayed the gel sample with a large amount of steams every half an hour to prevent the dehydration-induced crack propagation. Each spray process
lasts about 5 min.

**Calculations of the applied energy release rate.** To obtain the applied energy release rate for the anti-fatigue-fracture test, the dumbbell-like unnotched sample is stretched for \( N \) cycles with applied stretch \( \lambda^A \) at first. The strain energy density \( W \) of the unnotched sample under the \( N \)th cycle of applied stretch \( \lambda^A \) can be calculated as:

\[
W(\lambda^A, N) = \int_{1}^{\lambda^A} S d\lambda
\]

where \( S \) and \( \lambda \) are the nominal stress and stretch, respectively. Then, the same cyclic stretch \( \lambda^A \) is applied on the notched sample to measure the evolution of the crack length in undeformed state \( c \) with the cycle number \( N \). The applied energy release rate \( G \) in the notched sample under the \( N \)th cycle of applied stretch \( \lambda^A \) can be calculated as:

\[
G(\lambda^A, N) = 2k(\lambda^A) \cdot c(N) \cdot W(\lambda^A, N)
\]

where \( k \) is a slowly varying function of the applied stretch expressed as \( k = 3/\sqrt{\lambda^A} \), \( c \) is the current crack length at the undeformed state, and \( W \) is the strain energy density measured in the unnotched sample. In this work, for HAPAM/CNCs@CNTs hydrogel, the loading curve and unloading curve at the 2000th cycle are already highly overlapped (see Figure S8). Thus, the stretching curves of the 2000th cycle were used to calculate the \( G \) values.

**The electrical self-healing efficiency.** The electrical self-healing efficiency (\( S \)) of the hydrogel is defined as:

\[
S = \left(1 - \frac{R_h}{R_0}\right) \times 100\%
\]

where \( R_h \) and \( R_0 \) represent the resistance of the healed sample and the original sample, respectively, in each healing cycle. It points out that this equation is only valid when \( R_h < 2R_0 \).

**Experiments involving human subjects (live subject statement).** All the sensing experiments in Figure 5 and Figures S25-S28 were performed in compliance with the relevant laws and the approval of the Scientific Committee of Polymer Research Institute of Sichuan University. The informed consent of all participants involved in the sensor experiments was obtained.
**Table S1.** Comparisons of the reported hydrogel-based sensors

<table>
<thead>
<tr>
<th>Refs.</th>
<th>Materials</th>
<th>Self-healing</th>
<th>Gauge factor (GF)</th>
<th>Conductivity (S/m)</th>
<th>Tensile stress (MPa)</th>
<th>Stretchability</th>
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<tr>
<td>This work</td>
<td>HAPAM/CNCs@CNTs</td>
<td>Yes</td>
<td>1.65 (0-600% strain) 3.06 (600-1400% strain) 7.63 (1400-2000% strain)</td>
<td>0.826</td>
<td>0.31</td>
<td>2900%</td>
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<td>Adv. Sci., 2017, 4, 1600190</td>
<td>PVA/Borax/SWCNTs PVA/Borax/Graphene PVA/Borax/Silver nanowire</td>
<td>Yes</td>
<td>1.51 at 1000% strain 0.92 at 1000% strain 2.25 at 1000% strain</td>
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<td>--</td>
<td>1000%</td>
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<td>ACS Appl. Mater. Interfaces, 2018, 10, 20897</td>
<td>PAAm/dopamine hydrochloride/KCl</td>
<td>Yes</td>
<td>0.125 at 100% strain 0.693 at 1000% strain</td>
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<td>0.009</td>
<td>960%</td>
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<td>ACS Appl. Mater. Interfaces, 2018, 10, 14045</td>
<td>PNIPAM/PAni</td>
<td>No</td>
<td>3.9 (0-120% strain)</td>
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<td>0.4</td>
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<td>J. Mater. Chem. C, 2018, 6, 9200</td>
<td>PAA/PAni</td>
<td>No</td>
<td>2.0 at 350% strain</td>
<td>5.12</td>
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<td>J. Mater. Chem. B, 2019, 7, 24</td>
<td>PVA/PSBMA</td>
<td>No</td>
<td>1.5 (0-300% strain)</td>
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<td>Carbon, 136 (2018), 63</td>
<td>PAA/rGO/ Fe^{3+}</td>
<td>Yes</td>
<td>0.31 (100% strain) 1.32 (500% strain)</td>
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<td>0.4</td>
<td>650%</td>
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<td>ACS Appl. Mater. Interfaces, 2017, 9, 26429</td>
<td>PAAM/k-carrageenan/KCl</td>
<td>Yes</td>
<td>0.23 at 100% strain 0.63 at 1000% strain</td>
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<td>0.5</td>
<td>2000%</td>
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<tr>
<td>Mater. Horiz., 2019, 6, 326</td>
<td>SA/PAAm/NaCl</td>
<td>Yes</td>
<td>2.0 (0-200% strain) 2.7 (200-1800% strain)</td>
<td>0.023</td>
<td>0.65</td>
<td>2000%</td>
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<td>Journal, Year, Volume, Pages</td>
<td>Polymer Composition</td>
<td>Conductivity</td>
<td>Breakdown Voltage</td>
<td>Strain</td>
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<tr>
<td>Chem. Mater., 2018, 30, 8062</td>
<td>P(AAm-co-HEMA)/PAni</td>
<td>No</td>
<td>5.7 at 0.3% strain 1.48 (40-300% strain)</td>
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<td>Adv. Mater., 2016, 28, 8037</td>
<td>PAAm/PANI/Chitosan</td>
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<td>5.00</td>
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<td>J. Mater. Chem. C, 2018, 6, 13316</td>
<td>PVA/PAANa</td>
<td>No</td>
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<td>2.65</td>
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<td>Adv. Funct. Mater. 2017, 27, 1703852</td>
<td>PVA/PDA/FSWCNTs</td>
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<td>Adv. Funct. Mater. 2019, 29, 1806220</td>
<td>PVA/HPC</td>
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<td>Adv. Mater., 2017, 29, 1604827</td>
<td>PAAm/PDMS/LiCl</td>
<td>No</td>
<td>0.84 at 40% strain</td>
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<td>ACS Appl. Mater. Interfaces, 2018, 10, 13685</td>
<td>PNaSS/PMPTC/PAni</td>
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<td>ACS Appl. Mater. Interfaces, 2017, 9, 25559</td>
<td>PVA/PVP/CNCs-Fe³⁺</td>
<td>Yes</td>
<td>0.478 (0-200% strain)</td>
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<td>Adv. Mater., 2017, 29, 1700533</td>
<td>PAA/PPy/Chitosan</td>
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<td>Adv. Mater., 2017, 29, 1700321</td>
<td>ACC/PAA/alginate</td>
<td>Yes</td>
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<td>0.006</td>
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<td>Small, 2018, 1801711</td>
<td>PMMA/PAA/CaCl₂</td>
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<td>J. Mater. Chem. C, 2018, 6, 4183</td>
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<td>Adv. Funct. Mater., 2018, 1802576</td>
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<td>Adv. Mater., 2016, 28, 10244</td>
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<tr>
<td></td>
<td>Skins$^{2-4}$</td>
<td>Muscle fiber$^5$</td>
<td>Spinal cord and brain$^{2-4}$</td>
<td>Blood Vessel$^4$</td>
<td>Our hydrogel</td>
<td></td>
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<td>--------------------------------------</td>
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<tr>
<td><strong>Young’s modulus (KPa)</strong></td>
<td>10-100</td>
<td>6-500</td>
<td>0.1-10</td>
<td>~40</td>
<td>10-25</td>
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<td><strong>Elongation at break (%)</strong></td>
<td>40-80</td>
<td>10-30</td>
<td>5-45</td>
<td>~50</td>
<td>2900</td>
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**Table S2.** Comparisons of the mechanical properties of soft human tissues with our hydrogel.
Supporting Figures

Figure S1. Schematic illustration of the preparation process of the HAPAM/CNCs@CNTs conductive hydrogel plate. (a) PTFT plate and the hollow silicon rubber spacer; (b) casting polymer precursor solution on the plate; (c) sealed with another PTFE plate, and then placed at 60 °C with 6 h for polymerization; (d) the resultant HAPAM/CNCs@CNTs hydrogel plate can be easily peeled off from PTFT plates.
We can observe that the Zeta potential of the neat CNTs dispersion is -29.4 mV, which shows relatively poor suspension stability, leading to the easy precipitation in aqueous solution. With the addition of CNCs, the Zeta potential of CNCs@CNTs decreases to -40.3 mV, giving a good dispersibility and stability. Then, after mixing with the AM monomer solution, the Zeta potential of AM/CNCs@CNTs further drops to -43.5 mV due to the existence of surfactant SDBS, which is also helpful for dispersing CNTs.6

Figure S2. (a) Digital photographs and Zeta potential values of CNCs, CNTs, CNCs@CNTs, AM, and AM/CNCs@CNTs aqueous suspensions.
Figure S3. Photographs of the AM/CNCs@CNTs precursor solution before and after placing for three months.

Photographs of the AM/CNCs@CNTs aqueous suspension demonstrate its outstanding stability. We can observe that the amphiphilic CNCs effectively disperse CNTs in water without aggregation or precipitation even after placing for three months. The excellent stability of the AM/CNCs@CNTs precursor solution ensures the well-dispersion of CNCs@CNTs in the gel matrix, which is essential for the formation of the continuous conductive pathways in the gel.
Hydrogels are viscoelastic materials, which exhibit the capacity of storing and dissipation energy. The amount of energy stored and dissipated within the gel under the oscillatory stress is manifested by the storage modulus (G’) and the loss modulus (G’’), respectively. The G’ and G’’ values of HAPAM, HAPAM/CNTs, and HAPAM/CNCs@CNTs hydrogels are shown in Figure S4. The wide linear viscoelastic region reveals their dynamically cross-linked gel states during the dynamic frequency sweep tests, and further confirmed by the fact that the G’ is higher than that of the G’’ in each case.7-8 Besides, the G’ values of HAPAM/CNCs@CNTs hydrogel is higher than those of HAPAM/CNTs and pure HAPAM hydrogels, revealing the reinforcement effect of the partially crystallized fiber-like CNCs@CNTs in the gel matrix.
Figure S5. Typical XRD patterns of CNCs, CNTs, and CNCs@CNTs thin films (prepared from vacuum-assisted filtration).

XRD analysis was carried out to characterize the crystalline structure of the obtained CNCs, CNTs, and CNCs@CNTs nanohybrids. For neat CNCs, the diffraction peaks at 2θ angles of 14.9°, 16.5°, 22.6°, and 34.5° are assigned to the typical reflection planes of cellulose I crystals, 110, 110, 200, and 004, respectively.⁹ For CNTs, the peaks at around 25.6° and 43.0° are attributed to the 002 and 100 of the CNT’s crystal structure, respectively.¹⁰ In the XRD pattern of the CNCs@CNTs, both the characteristic peaks of CNCs and CNTs can be clearly observed, indicating the crystalline structure of CNCs@CNTs.
Figure S6. Typical Raman spectra of CNCs, CNTs, and HAPAM/CNCs@CNTs hydrogels.

In Raman spectra of CNCs, the peak at 2895 cm\(^{-1}\) and the bands of 1050-1150 cm\(^{-1}\) are ascribed to the stretching vibration of C–H and the skeletal vibration of C–O–C pyranose rings, respectively.\(^{11}\) For CNTs, the peak around 1348 cm\(^{-1}\), 1585 cm\(^{-1}\), 2692 cm\(^{-1}\), and 2932 cm\(^{-1}\) are assigned to the D, G, 2D, and D+G bands, respectively.\(^{12}\) In Raman spectra of the HAPAM/CNCs@CNTs hydrogel, characteristic bands of CNCs and CNTs are both observed, which indicates the successful synthesis of the HAPAM/CNCs@CNTs hydrogel. Furthermore, we randomly selected five points on the surface of the HAPAM/CNCs@CNTs hydrogel for the Raman experiment and obtained five similar results. This result reveals the good distribution of CNCs@CNTs in the gel matrix.
The characteristic band at 1060 cm$^{-1}$ assigned to the skeletal vibration of the C–O–C pyranose ring in CNCs (Figure S7b) appears in HAPAM/CNCs and HAPAM/CNCs@CNTs hydrogels, which confirms the successful synthesis of the HAPAM/CNCs@CNTs hydrogel. Besides, a new shoulder peak around 1728 cm$^{-1}$ appears after the addition of CNCs and CNCs@CNTs. This new peaks probably can be attributed to the newly formed hydrogen bonding between C=O groups of HAPAM and the –OH groups of CNCs.

Figure S8. Typical stress $S$ versus stretch $\lambda$ curves for HAPAM/CNCs@CNTs hydrogel at applied stretch (a) $\lambda^a = 2.5$ and (b) $\lambda^a = 5.0$.  

Figure S7. (a) FTIR spectra of HAPAM, HAPAM/CNCs, and HAPAM/CNCs@CNTs hydrogels; (b) FTIR spectra of CNCs.
Figure S9. The ultra-depth-of-field microscope images of the notch before and after 2000 stretching cycles ($\lambda=5.0$).

Figure S10. (a) Representative load-unloading tension curves (10 cycles) under a tension of 1000%. (b) Representative load-unloading compression curves (10 cycles) under 90% strain.

Figure S11. The HAPAM/CNCs@CNTs hydrogel can basically recover to its original height in 10 mins after suffering 90% compression strain.
Figure S12. The HAPAM/CNCs@CNTs hydrogel is strong enough to withstand the 2 Kg load.

Figure S13. (a) Photograph of the surgical blade; (b) the excellent cutting resistance of the HAPAM/CNCs@CNTs hydrogel to a surgical blade at a compression strain of about 90%.
Figure S14. The HAPAM/CNCs@CNTs hydrogel can easily withstand large deformations, such as curling, knotting, and stretching, and it can easily recover from these large deformations.

Figure S15. Photographs of the self-healed HAPAM/CNCs@CNTs hydrogel before and after stretching.
Figure S16. LCSM image of the hydrogel sample before cutting.

Figure S17. Tensile properties of the HAPAM/CNCs@CNTs hydrogel after self-healing with different (a) healing cycles, (b) times, and (c) temperatures.

Figure S18. Repeated electrical conductivity recovery performance in successive three self-healing cycles.
In this experiment, we measured the resistance of the original hydrogel sample at first. After that, we cut the sample into two parts, and then gently brought them into contact to allow them self-healing. Afterward, we measured the resistance of the sample every 12 hours in the next two days. Note that, the sample was sealed with a PP bag to minimize the water evaporation. This process was repeated for three times to prove the reliability of the electrical healing ability of our hydrogel. In this experiment, three samples were used to obtain an accurate result.

Figure S19. Photographs of the HAPAM/CNCs@CNTs hydrogel as a self-healing conductor to light an LED bulb during the healing process. (a) undamaged; (b) completely cutting; (c) after healing for 12 h.

Figure S20. Illustration of the fabricated strain sensor based on HAPAM/CNCs@CNTs hydrogel.
Figure S21. Relative resistance change of the sensor as a function of the applied strain. (a) the HAPAM/CNTs hydrogel. (b) The pure HAPAM hydrogel.

Figure S22. Current changes of the self-healed (a) and the damaged (b) sensors under different small tensile strains.
Figure S23. Current changes of the original, the self-healed, and the damaged sensors under 100% (a, c) and 200% (b, d) tensile strains.
**Figure S24.** Response times of the strain sensor towards 1%, 10%, and 20% strains.

**Figure S25.** Current response of the original strain sensor for the wrist movement detection.
Figure S26. Current responses of SH (a-c), D (d-f), and DS20000 (g-i) sensors for the finger bending, the wrist bending, and the walking (90 s) detection.
Figure S27. Current responses of SH (a-c), D (d-f), and DS20000 (g-i) sensors in the facial expression detection.
Figure S28. Current responses of SH (a-c), D (d-f), and DS20000 (g-i) sensors in the pronunciation detection.

References
2. X. Chen, Small Methods, 2017, 1600029.


