Supplementary Information (SI) for New Journal of Chemistry.

Supporting Information

Tannic acid-iron dual-catalyst-induced polyacrylamide/sericin hydrogel for strain and pressure sensing

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Experimental section

1. Characterization

The surface morphology of the SS/PAM/Borax/TA-Fe^{3+} hydrogel was observed using an in-situ emission scanning electron microscope (Quatro S, Semmerfeld, China) at an acceleration voltage of 20 kV. Fourier transform infrared spectroscopy of the SS/PAM/Borax/TA-Fe^{3+} hydrogel was recorded using an IRPRESTIGE-21 spectrometer (Shimadzu). Rheological measurements were performed using an Anton Paar MCR302 rheometer equipped with a CP20-1 cone-plate tool. X-ray photoelectron spectroscopy (XPS) analysis was conducted using a Thermo Fisher Scientific K-Alpha XPS system. Mechanical properties were tested using an LD24.202 universal testing machine. All electrochemical characterisation, including conductivity and IT measurements, was performed on a CHI760E electrochemical workstation.

2. Detailed test methods

Mechanical properties

Mechanical property testing was conducted using LD24.202 universal testing machine equipped with a 200N load cell. For tensile testing, dumbbell-shaped hydrogel specimens (20 mm × 4 mm × 2 mm) were used to evaluate mechanical properties at a rate of 100 mm/min. In compression measurements, cylindrical SS/PAM/Borax/TA-Fe^{3+} hydrogel specimens (12.5 mm diameter × 8 mm thickness) underwent compressive strain at a rate of 40 mm/min.

Adhesion and Self-Healing Properties

The adhesion strength of the SS/PAM/Borax/TA-Fe^{3+} hydrogel was determined using the lap shear method. All rectangular samples (2 mm thick) were sandwiched between two substrates (iron sheet, copper sheet, plastic, cardboard, wood, and pig skin as test models). Then, the samples were stretched at a constant speed of 10 mm/min using a tensile testing machine. Three samples were taken from each group for adhesion testing.

Electrical properties

The electrical conductivity of the SS/PAM/Borax/TA-Fe^{3+} hydrogel was measured using an electrochemical detector (Chi760e, Shanghai Chenhua Instrument Co., Ltd., China). The relative resistance change rate is the rate of change in resistance (ΔR) relative to the initial resistance (R0) during deformation at a given strain. The relative resistance change ($\Delta R/R_0$) of the SS/PAM/Borax/TA-Fe^{3+} hydrogel samples was recorded using an electrochemical workstation and a multimeter. By performing linear fitting of the relative resistance change curve, the sensitivity factor (GF) of the SS/PAM/Borax/TA-Fe^{3+} hydrogel sensor was obtained, reflecting its strain sensitivity.

$$\sigma = L/RS \times 10^3$$

where σ represents conductivity (S/cm), L (cm) represents the distance between the two probes, R (Ω) and S (cm^2) represent the resistance and cross-sectional area of the hydrogel sample, respectively.

$$\Delta R/R_0 = (R - R_0)/R_0 \times 100\%$$

Here, R_0 denotes the initial resistance, and R denotes the resistance at a specific strain at a given time.

$$GF=(\Delta R/R_0)/\varepsilon \times 100\%$$

Here, R_0 denotes the initial resistance, and R represents the resistance at a given time under a specific strain.

For pressure sensing characteristics, the variation of current with compressive strain was measured using an electrochemical workstation (Chi760e, Shanghai Chenhua Instrument Co., Ltd., China). A real-time current-time (I-t) curve was acquired by connecting the SS/PAM/Borax/TA-Fe^{3+} hydrogel to a complete circuit and applying a constant voltage (2V).

$$\Delta I/ I_0 = (I - I_0)/ I_0$$

where I_0 denotes the initial current without applied compressive strain, and I denotes the real-time current after applying compressive strain.

$$\Delta S = (\Delta I / I_0)/ \Delta P$$

where I represents the change in current, I_0 is the initial current, and ΔP is the change in pressure.

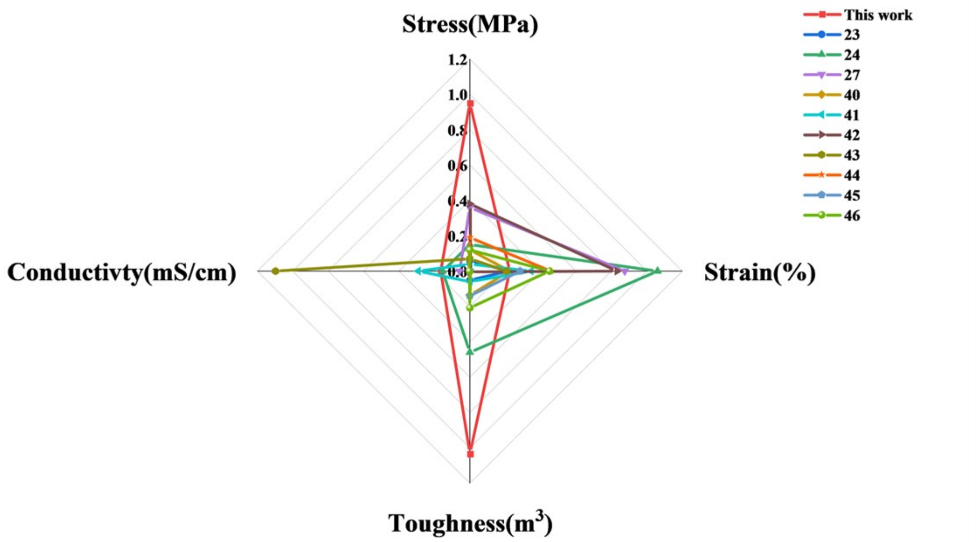

Fig. S1 The stress, strain, toughness and conductivity of hydrogels (9wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax) were compared with some silk-based hydrogels reported in the literature.

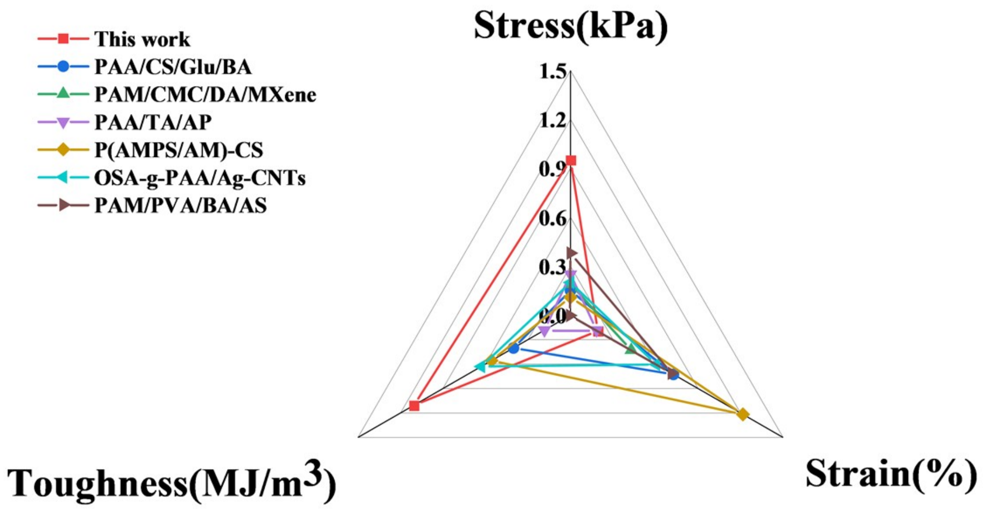

Fig. S2 Comparison of the stress, strain and toughness of hydrogels (9wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax) with those of several adhesive hydrogels reported in the literatures.

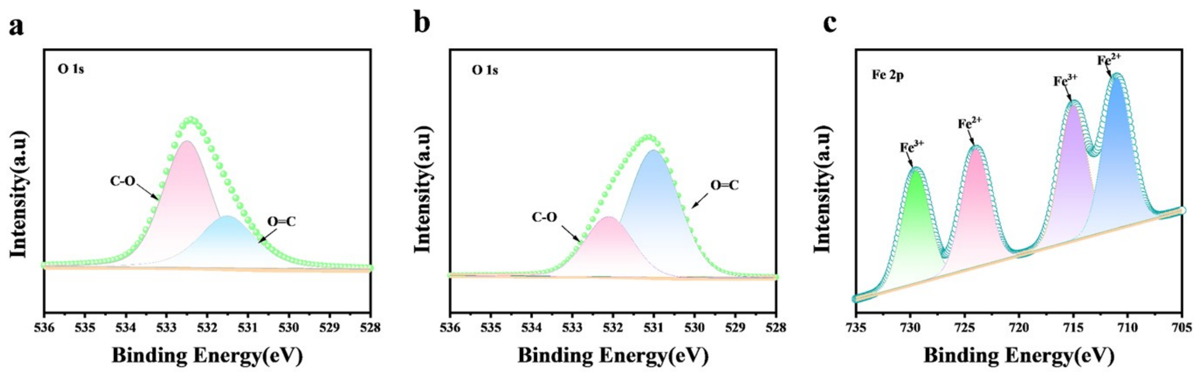

Fig. S3 XPS of hydrogels (9wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax). (a,b) XPS signals of TA-free and TA-containing O 1s regions. (c) XPS signals of the Fe 2p region containing TA.

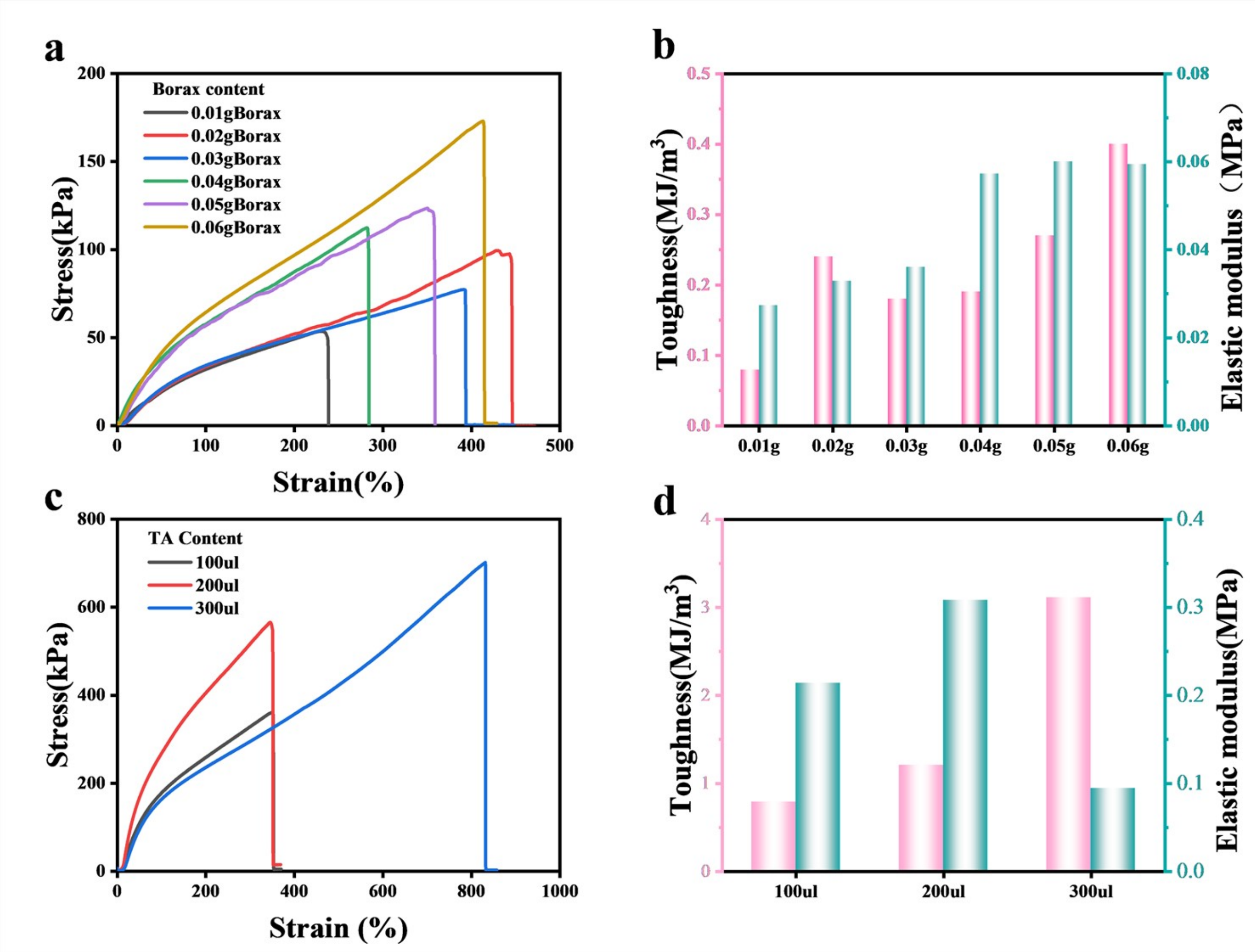

Fig. S4 Mechanical properties of hydrogels (9 wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax). (a) Tensile stress-strain curves of SS/PAM/Borax/TA-Fe^{3+} hydrogels at different borax concentrations, along with (b) the corresponding toughness and Elastic modulus. (c) Tensile stress-strain curves of SS/PAM/Borax/TA-Fe^{3+} hydrogels at different TA contents, along with (d) the corresponding toughness and Elastic modulus.

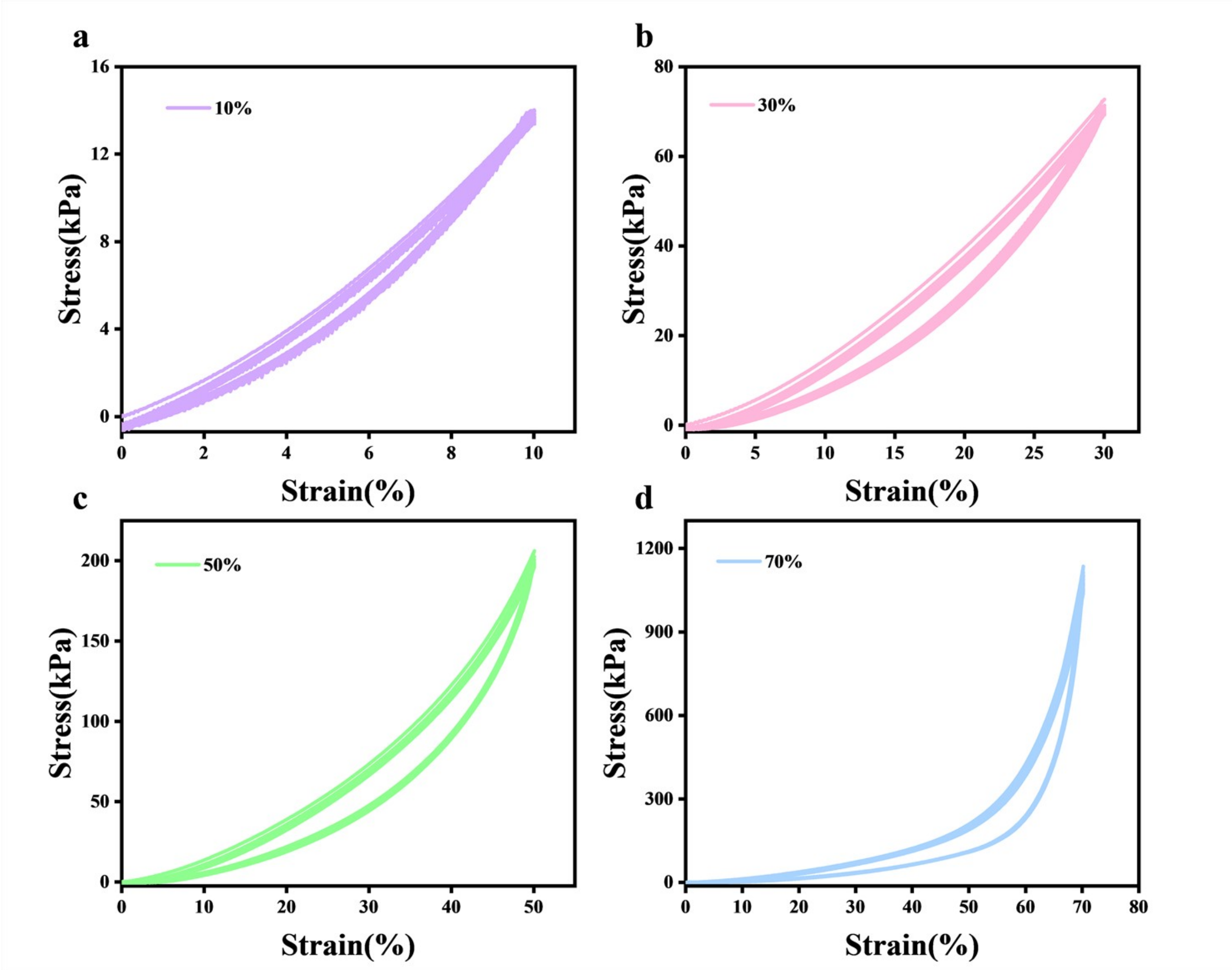

Fig. S5 Pressure properties of hydrogels (9 wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax).underwent 10 load-unload cycles at compression levels of (a)10%, (b)30%, (c)50%, and (d)70%.

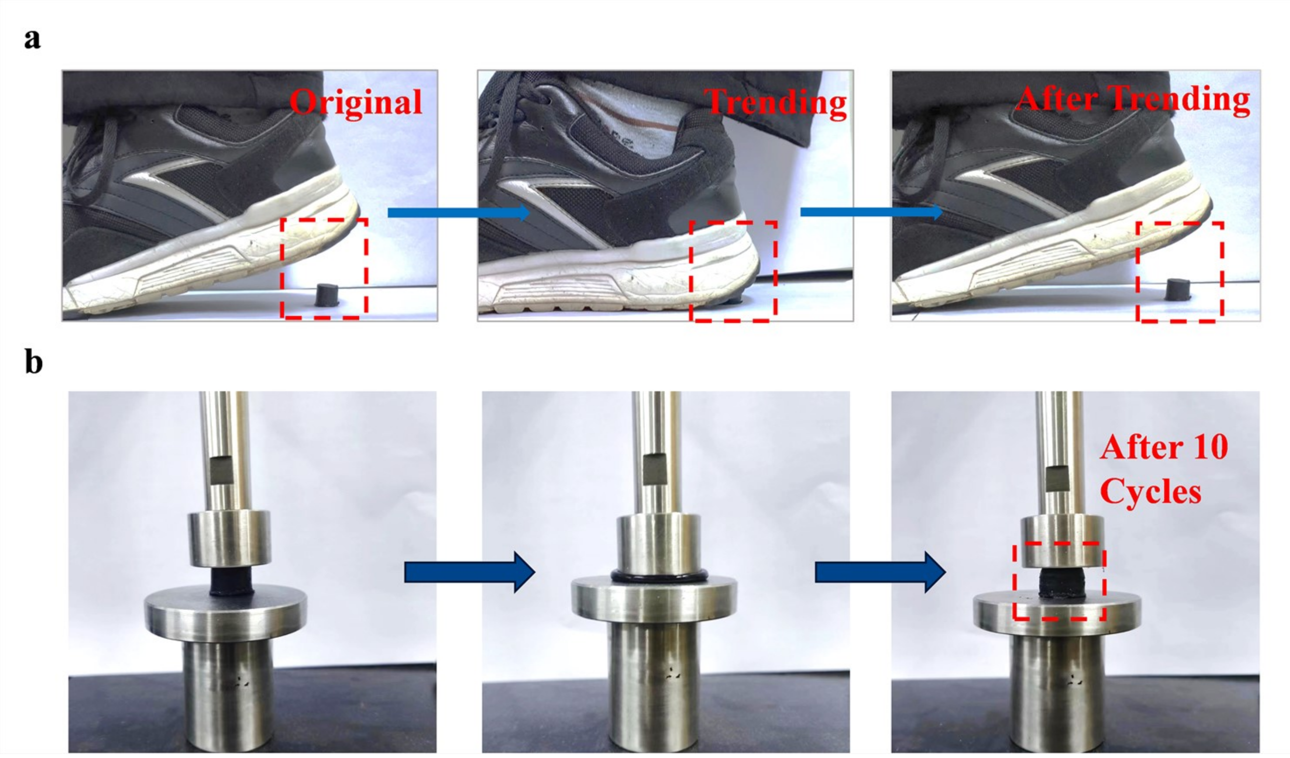

Fig. S6 Compressed images of hydrogels (9 wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax). (a) Process of stepping on the hydrogel; (b) Images of the hydrogel before and after 80% compression.

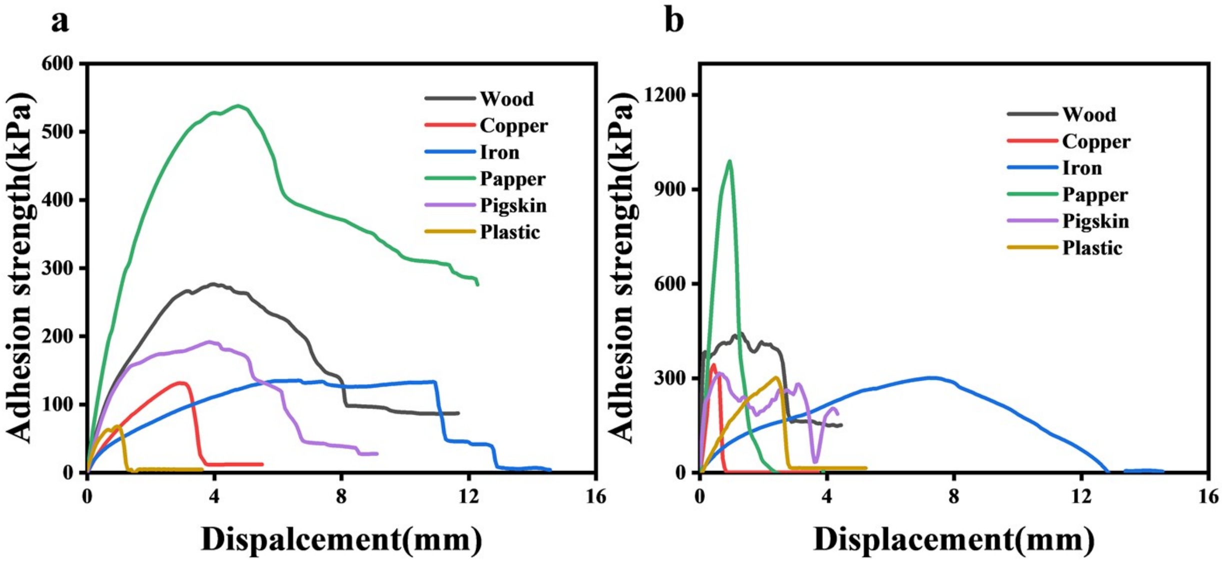

Fig. S7 Adhesive tensile properties of hydrogels (9 wt% SS, 45 wt% AM, 0.5 wt % TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax). Strength of adhesion of hydrogels to various substrates after (a) instantaneous and (b) 24h adhesion to various substrates.

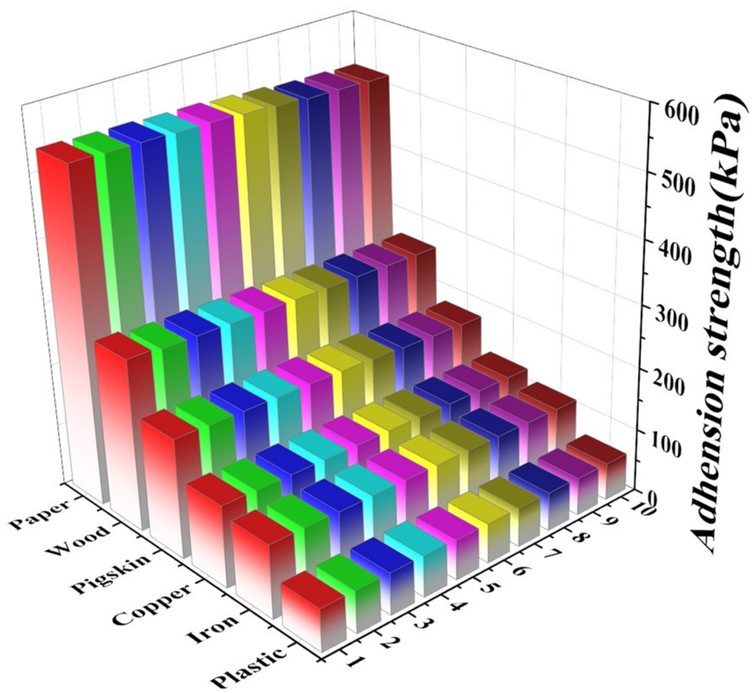

Fig. S8 Adhesion curves after 10 cycles for the hydrogel (9 wt% SS, 45 wt% AM, 0.5 wt% TA, 0.2 wt% $FeCl_3$ and 2 wt% borax) on various substrates.

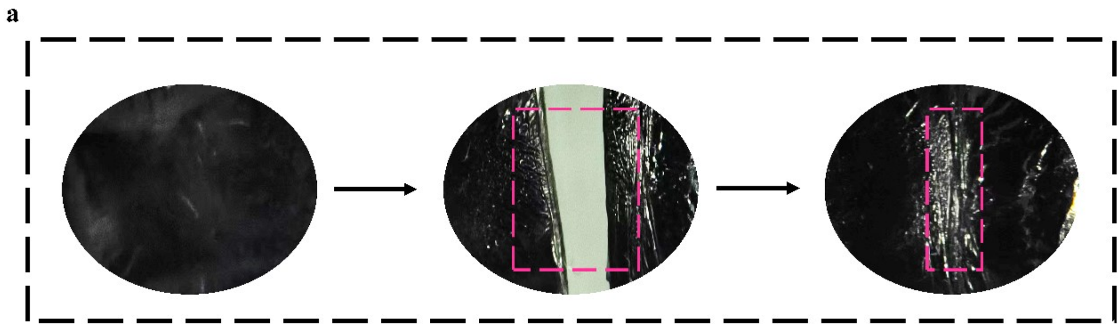

Fig. S9 Macroscopic self-healing of hydrogels (9 wt% SS, 45 wt% AM, 0.5 wt% TA, 0.2 wt% FeCl3 and 2 wt% Borax).

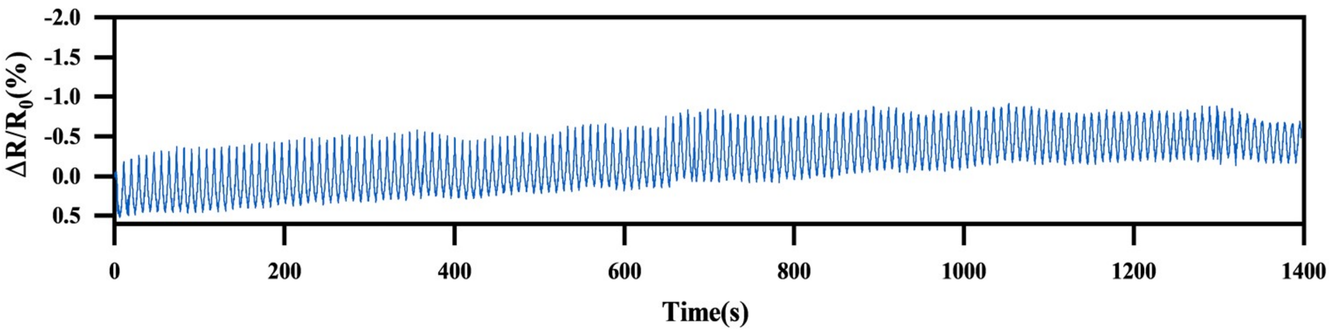

Fig. S10 The stability of the hydrogel (9 wt% SS, 45 wt% AM, 0.5 wt% TA, 0.2 wt% $FeCl_3$ and 2 wt% Borax) after loading-unloading application of bending motion.

Fig. S11 Strain(100%) and compression(20kPa) sensing tests on a hydrogel (9 wt% SS, 45 wt% AM, 0.5 wt% TA, 0.2 wt% $FeCl_3$ and 2 wt% borax) after 1, 5 and 9 days of storage at room temperature.

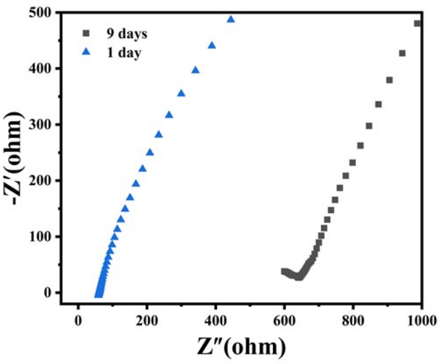

Fig. S12 Impedance spectra of the hydrogel (9 wt% SS, 45 wt% AM, 0.5 wt% TA, 0.2 wt% $FeCl_3$ and 2 wt% borax) after storage at room temperature for 1 day and 9 days.

Table S1. Effect of Borax content in hydrogels on tensile stress-strain.

SS:AM(aq)	Borax	SS	AM	0.48 wt% $FeCl_3$(aq)	5 wt% TA(aq)
1:5	0.01 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.02 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.03 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.04 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.05 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.06 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.08 g	0.26 g	1.3 g	3 ml	100 ul

Table S2. Effect of SS to AM ratio in hydrogels on tensile stress-strain.

SS:AM(aq)	Borax	SS	AM	0.48 wt% $FeCl_3$(aq)	5 wt% TA(aq)
1:1	0.05 g	0.26 g	0.26 g	3 ml	100 ul
1:3	0.05 g	0.26 g	0.78 g	3 ml	100 ul
1:5	0.05 g	0.26 g	1.3 g	3 ml	100 ul
1:7	0.05 g	0.26 g	1.82 g	3 ml	100 ul
2:3	0.05 g	0.52 g	0.78 g	3 ml	100 ul
2:5	0.05 g	0.52 g	1.3 g	3 ml	100 ul
2:7	0.05 g	0.52 g	1.82 g	3 ml	100 ul
3:4	0.05 g	0.78 g	1.04 g	3 ml	100 ul

Table S3. Effect of TA content in hydrogels on tensile stress-strain.

SS:AM(aq)	Borax	SS	AM	0.48 wt% $FeCl_3$(aq)	5 wt% TA(aq)
1:5	0.05 g	0.26 g	1.3 g	3 ml	100 ul
1:5	0.05 g	0.26 g	1.3 g	3 ml	200 ul
1:5	0.05 g	0.26 g	1.3 g	3 ml	300 ul

Table S4. Effect of ferric chloride content in hydrogels on tensile stress-strain.

SS:AM(aq)	Borax	SS	AM	0.48 wt% $FeCl_3$(aq)	5wt%TA(aq)
1:5	0.05 g	0.26 g	1.3 g	1 ml	300 ul
1:5	0.05 g	0.26 g	1.3 g	2 ml	300 ul
1:5	0.05 g	0.26 g	1.3 g	3 ml	300 ul

Table S5. Comparison of hydrogel sensors with other hydrogels based on silk-based proteins.

Materials	Stress(Mpa)	Strain(%)	Toughness(MJ/m^3)	Conductivity(mS/cm)	Ref
PAM/SS/Borax/TA-Fe^{3+}	0.95	450	2.58	4.8	This work
PAM/SS/NaCl	0.36	1750	-	1.7	27
PAM/SS/Pedot:PSS	0.15	2120	1.15	4.5	24
PAM/SS/PA/Al^{3+}	0.06	430	0.13	-	23
PVA/SA/SS@CNTs	0.12	450	0.34	-	40
PAM/TA/SF/MXene	0.04	692	0.15	8.5	41
PAM/SF	0.38	1663	0.01	-	42
PAA/GMA/SF	0.07	415	-	32	43
PAA/Gly/SF/TA-Fe3O4@MXene	0.19	946	<1.2	0.2	44
BF/SF/Borax/$CaCl_2$	<0.09	567	0.35	-	45
RSF/PDMA/NaCl	0.12	900	0.52	0.07	46