Supporting Information

A highly adhesive, self-healing and perdurable PEDOT: PSS/PAA-Fe^{3+} gel enabled by multiple non-covalent interactions for multi-functional wearable electronics

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In vitro cytotoxicity assay

Cytocompatibility of prepared hybrid hydrogels was accessed using a human skin fibroblast cell line HSF (Stem Cell Bank, Chinese Academy of science) by a non-contact method. For HSF cells culturing, Dulbecco modified eagle medium (DMEM, Gibco, NY, USA) supplemented with 10% heat-inactivated fetal bovine serum (FBS, HYCLONE) and 1% penicillin/streptomycin (P/S, HYCLONE) was used. Prior to seeding cells, gel samples were prepared directly in the 48-well culture plate. Necessary sterilization is performed under ultraviolet light for 30 min. And then, DMEM was added with a ratio of 0.2 g mL^{-1} to allow ion exchange. After immersion for 48 h, the DMEM was collected and 400 μL cell culture medium was transferred to the new 48-well plate. After that, 100 μL fresh DMEM containing 10,000 HSF cells was added. A blank sample of fresh DMEM was used as control subject. Environment of cell cultures was a humidified atmosphere of 37 °C, 95% air and 5% CO_2.

After incubating for 1, 3 and 5 days, cell viability was evaluated by cell proliferation assay and Live/Dead staining assay. Cell proliferation was examined by using Cell Counting Kit-8 (CCK-8, Dojindo, Japan) following the manufacturer's instructions. Briefly, after cell culture medium was removed, PBS solution was used to rinse the well twice, followed by adding 100 μl fresh DMEM with 10 μL CCK-8 into the plates at 37 ℃ with 5% CO_2 for 2 h. And then, 100 μL of cell medium were transferred from each well to a new 96-well plate to measure the optical absorbance at a wavelength of 450 nm on a microplate reader (Elx-800, Bio-Tek, USA). For live/dead staining assay, the LIVE/DEAD®Viability/Cytotoxicity Kit (Thermo Fisher Scientific, L-3224) was used

according to the manufacturer's instructions. Briefly, the cell culture medium was removed from the plate after a same cell culture. 100 μL of PBS with 4 μM calcein-AM and 2 μM ethidium homodimer-1 was added to stain live and dead cells, respectively. After incubating for 15 min at 37 °C, HSF cells were examined by epifluorescence microscopy (IX53, Olympus, Japan).

Preparation and characterization of gel-TENG based on PEDOT: PSS/PAA-Fe^{3+} conductive gels

Conductive gel-based TENG with sandwich structure was prepared using PEDOT: PSS/PAA-Fe^{3+} conductive organic gel and CCNTs-doped PDMS (0.5 wt.% doped) as key elastomeric substrates and encapsulant. Aluminum foil was used as another contact electrode. Finally, copper wire was performed as a connector to link TPPCN membrane and Aluminum foil to electrometer. Then it was connected with computer-controlled electrometer (Keithley 6541 system), and the corresponding output voltage and current were measured.

Preparation of PEDOT: PSS/PAA-Fe^{3+} conductive gel fibers

PEDOT: PSS/PAA-Fe^{3+} conductive hydrogel fibers were prepared by wet-spinning. AA (1 g) was added into PEDOT: PSS aqueous dispersion (10 g) to obtain the spinning solution, which was stirred constantly at 40 °C until the content of PEDOT: PSS reaching 22 $mg \cdot mL^{-1}$. After ultrasonic defoaming, it was uninterruptedly pumped through a flat needle (diameter D=0.21 mm) and vertically injected into the mixed coagulation bath of ethanol (50 wt.%) and water with Fe^{3+}ions (0.1 $mol \cdot L^{-1}$) at a speed

of 0.2~0.5 $mL \cdot h^{-1}$. The resultant fibers were also treated by EG for further improvement of conductivity and durability.

Construction and characterization of fiber-shaped supercapacitors

In this work, fiber-shaped supercapacitors (FSC) were composited by two single fibers and H_3PO_4-PVA electrolyte. Two single fibers were placed parallelly on a glass sheet with an interval of 2 mm, and two ends were fixed with silver glue. After solidification, a few drops of H_3PO_4-PVA electrolyte were added to cover the two fibers. The H_3PO_4-PVA electrolyte was prepared as follows: 5 g of PVA (M_n=74800) and 45 mL water were put into a 100 mL beaker, stirred and dissolved at 90 °C to form a gel, and then 5 g H_3PO_4-PVA was added to stir evenly. Cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) were tested, and the mass specific capacitance (C_m) of the fibrous supercapacitor was calculated using the formula: $C_m = 2It/Um$. where I is the current density, t is the discharge time, U is the discharge voltage drop, and m represents the mass of the electrolyte part (effective part) coated with H_3PO_4-PVA on the single electrode.

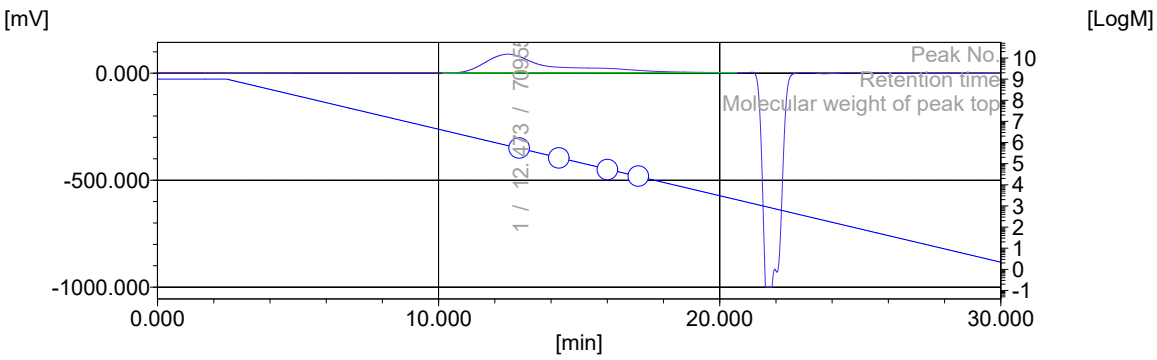

Fig. S1. A GPC curve of PAA.

Tab. S1. Relative peak value

	Time / min	M_n	M_w	Mz	Polydisperrsity(M_w/M_n)
1	12.413	94702	555848	957820	5.869

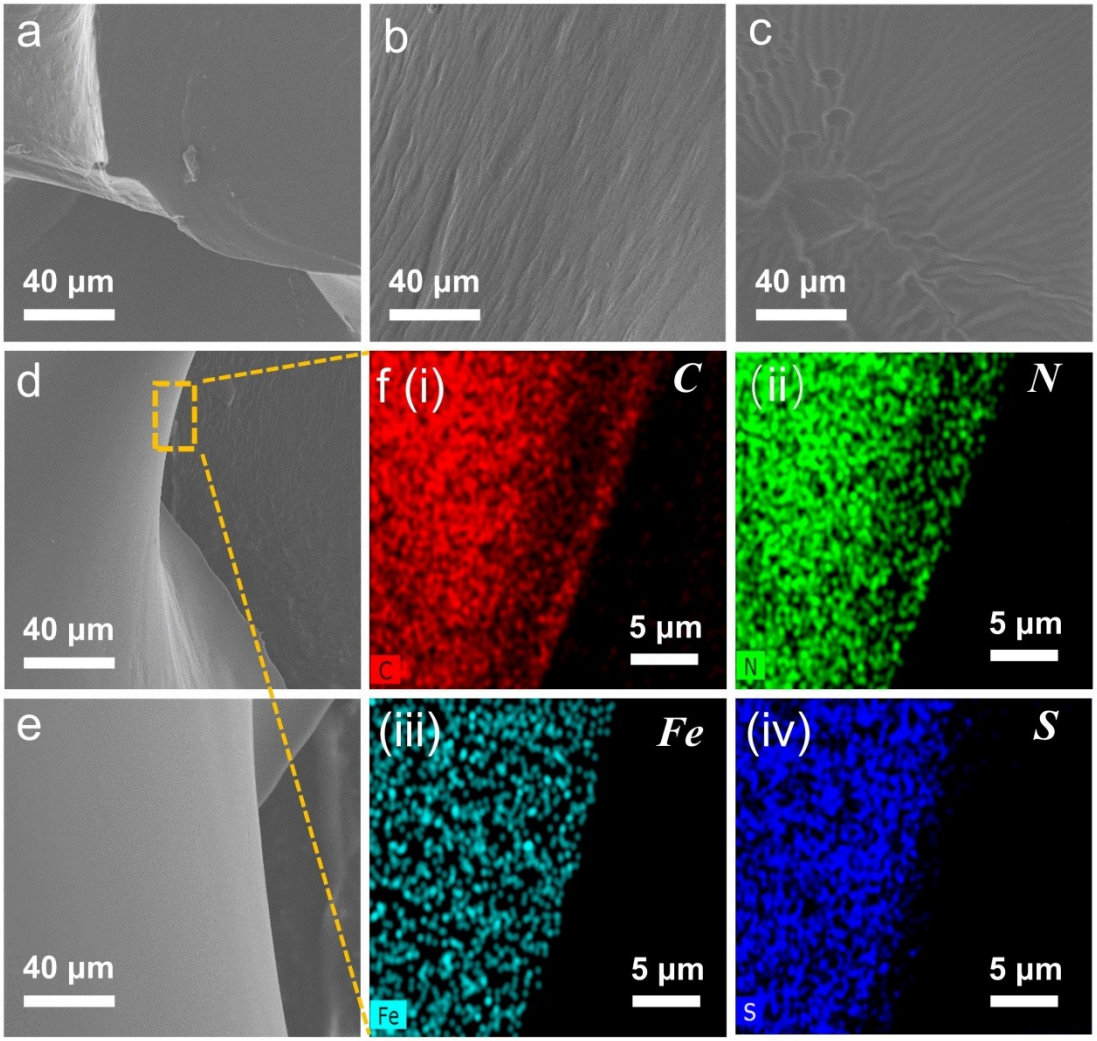

Fig. S2. SEM images and EDS mapping of PAA and PEDOT: PSS/PAA-Fe^{3+} organic gels with different x values. (a) pure PAA, (b) x=1, (c) x=2, (d) x=3, (e) x=5, (f) mapping of x=3.

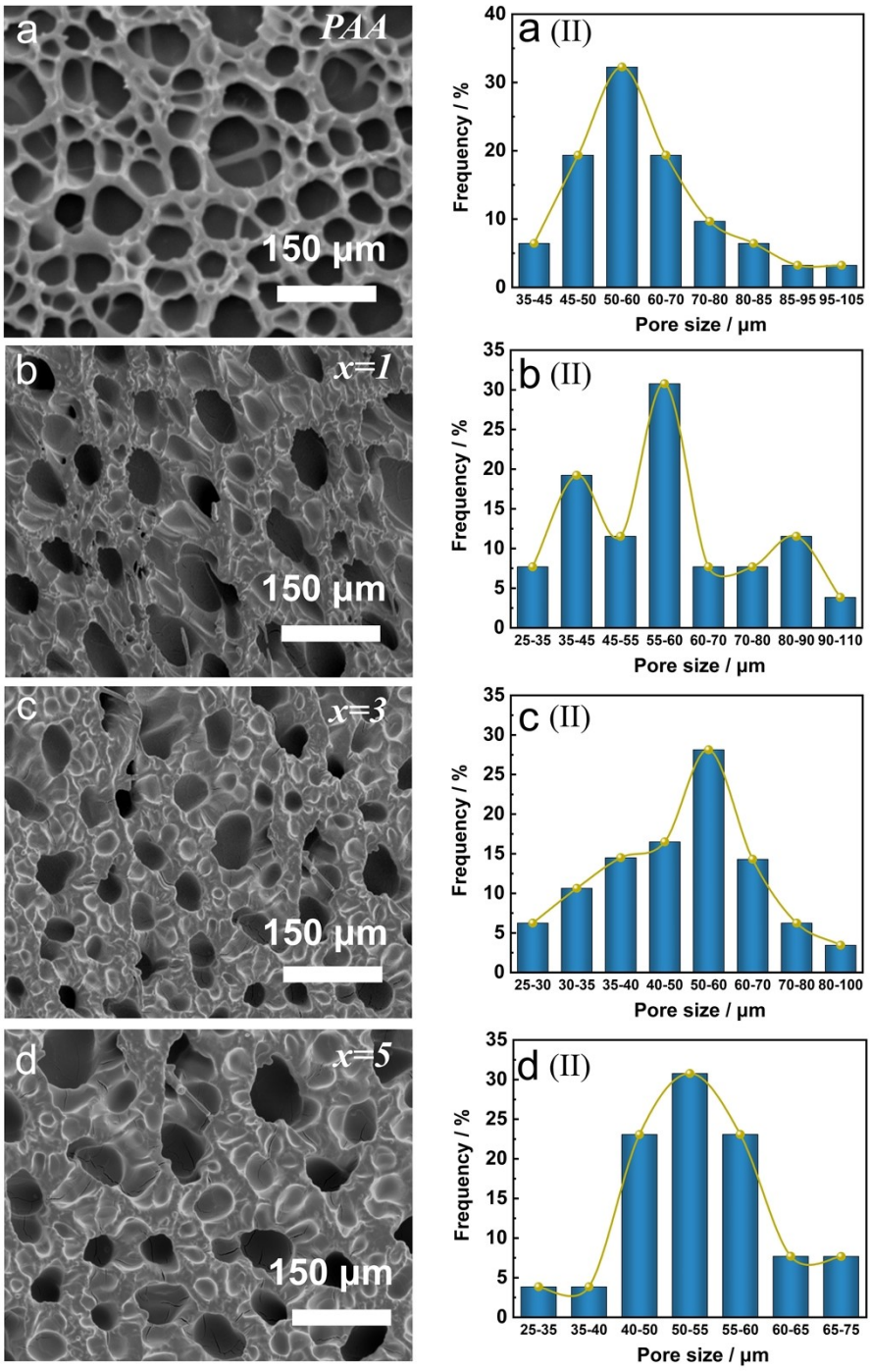

Fig. S3. SEM images and pore size of PAA and PEDOT: PSS/PAA-Fe^{3+} gels with different x values after freeze-drying. (a) pure PAA, (b) x=1, (c) x=3, (d) x=5.

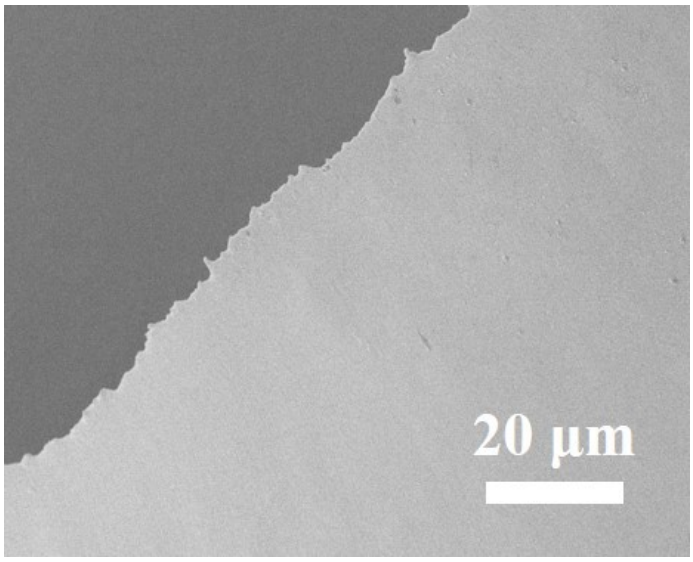

Fig. S4. A SEM image of PEDOT: PSS/PAA-Fe^{3+} conductive gel with EG treatment.

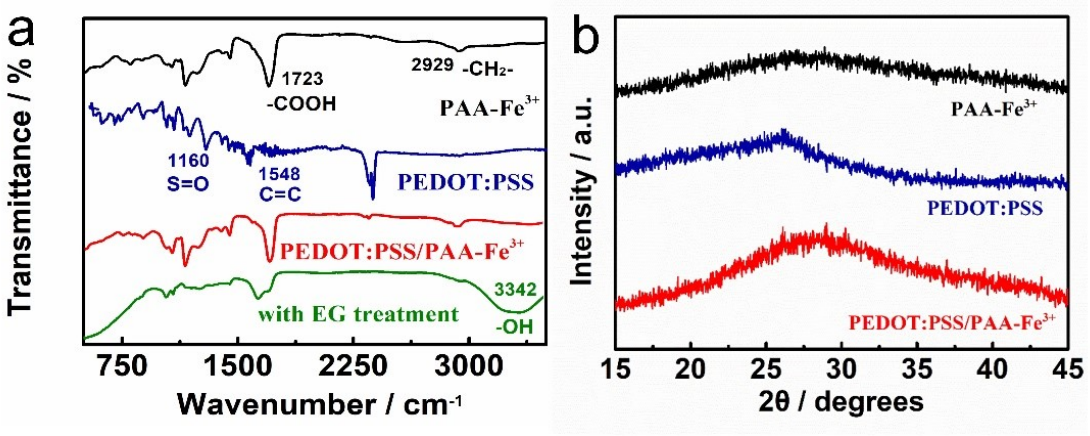

Fig. S5. FT-IR spectra (a) and XRD curves (b) of pure PAA-Fe^{3+}, PEDOT: PSS and PEDOT: PSS/PAA-Fe^{3+}gels.

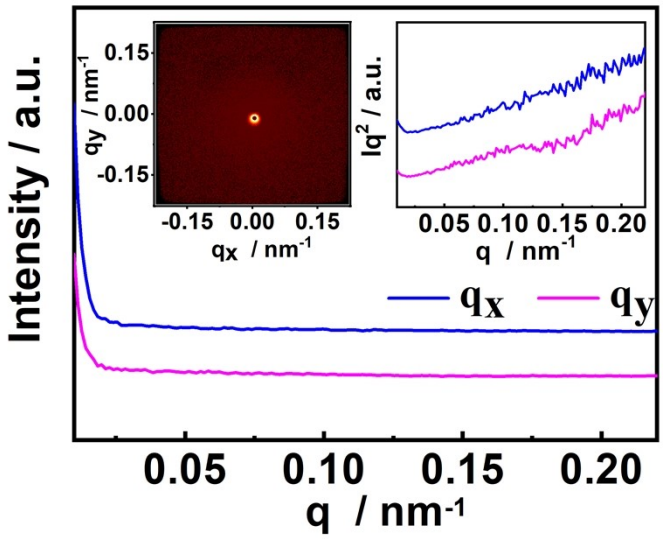

Fig. S6. SAXS curves of the PEDOT: PSS/PAA-Fe^{3+} conductive gel after EG treatment.

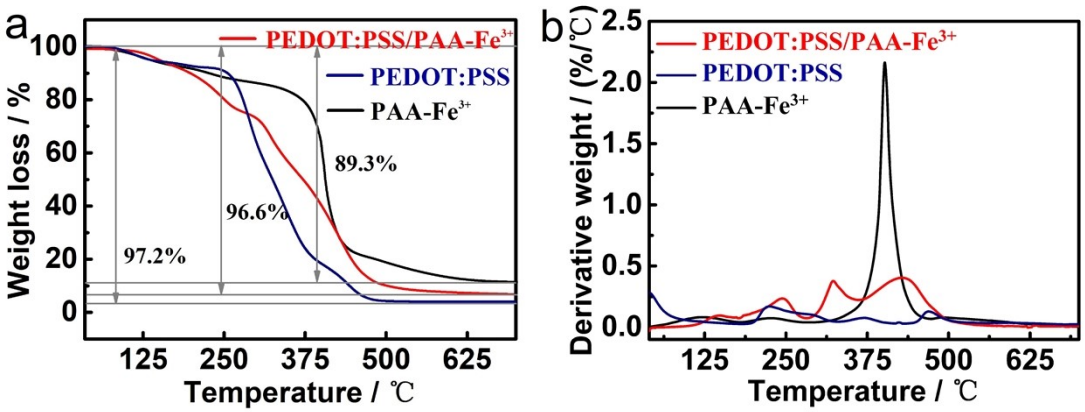

Fig. S7. TG (a) and DTA (b) curves of pure PAA-Fe^{3+}, PEDOT: PSS and PEDOT: PSS/PAA-Fe^{3+} conductive gels.

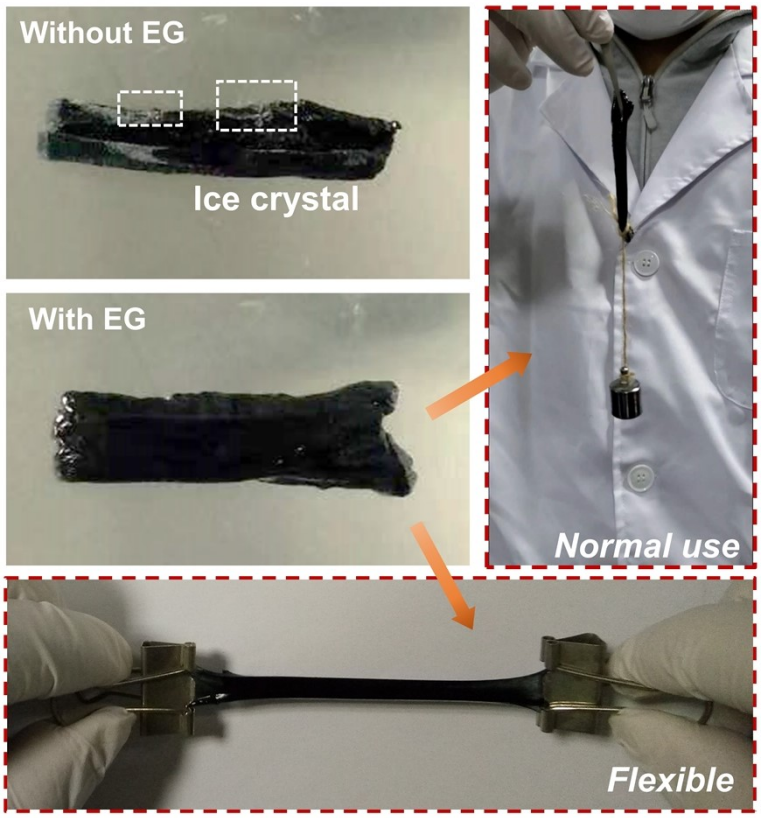

Fig. S8. Optical images showing anti-freezing properties of the gel after EG treatment.

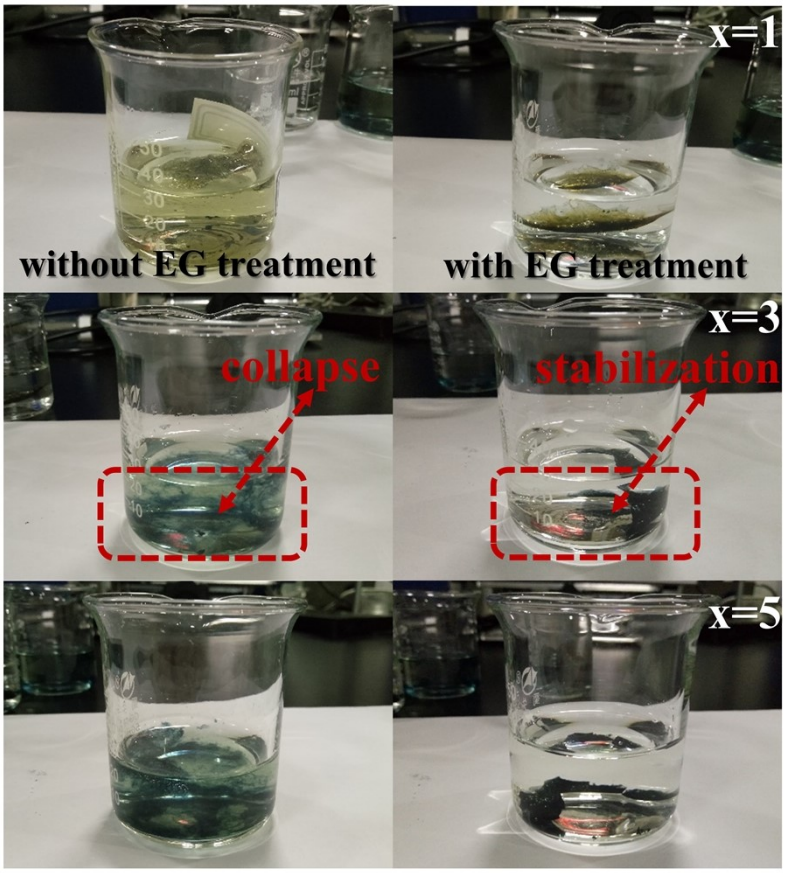

Fig. S9. Optical images showing water-resistance properties of the gel after EG treatment.

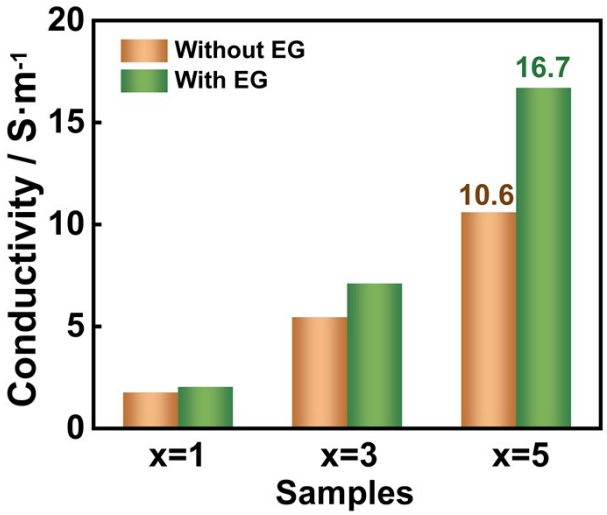

Fig. S10. The electrical conductivity of PEDOT: PSS/PAA-Fe^{3+} gel before and after EG treatment.

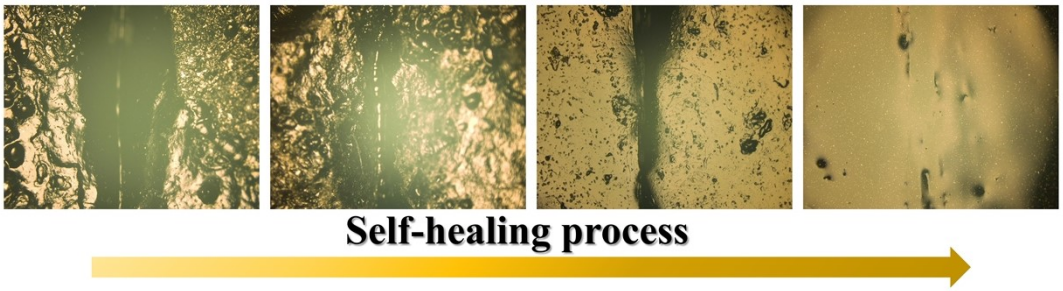

Fig. S11. Digital images of the incision in the self-healing process.

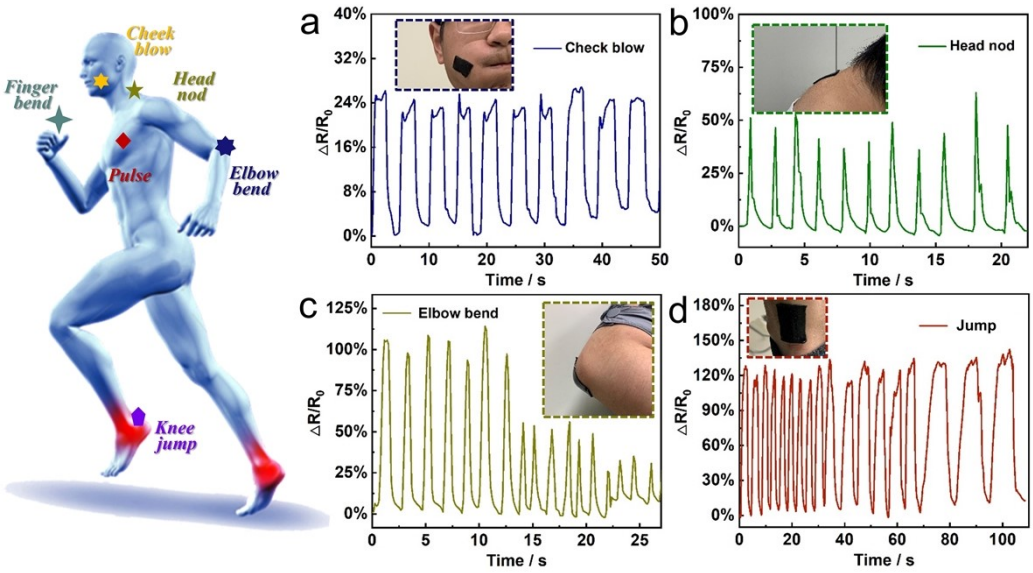

Fig. S12. Real-time resistance changes of conductive gels used as wearable strain sensors for monitoring various human activities including check blowing (a), head nodding (b), elbow bending (c), and jumping (d).

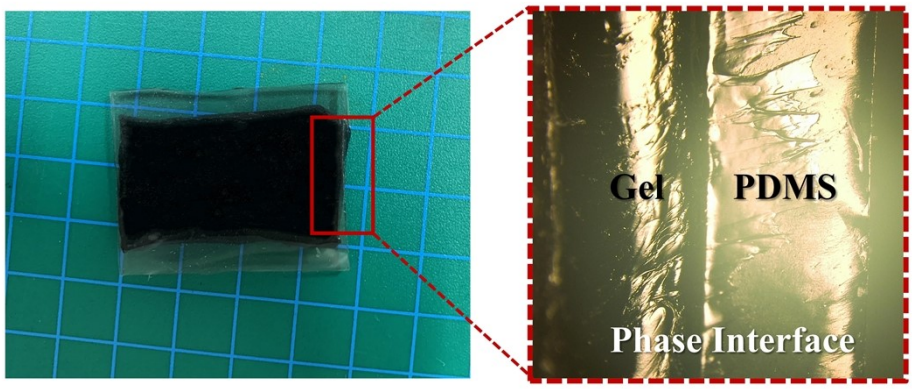

Fig. S13. A SEM image of interfaces between the gel and PDMS.