Supporting Information

for

Transparent, Mechanically Robust, Conductive, Self-healable, and Recyclable Ionogels for Flexible Strain Sensor and Electroluminescent Device

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1. Materials and Methods

1.1. Materials

2,2,2-Trifluoroethyl acrylate (TFEA, 98%) was purchased from Shanghai Qinba Chemical Co., Ltd., China. 1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI], 98%) was obtained from Monils Chemical (Shanghai) Co., Ltd., China. Polyvinylpyrrolidone (PVP) with the average molecular weights of 1250 kg/mol was obtained from Acros. Acrylamide (98.0%, AAm), 2-hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone (98.0%, Irgacure 2959), and ethanol (99%, EtOH) were obtained from Adamas-beta® (Co., Ltd., China). All chemicals were used as received without further purification.

1.2. Preparation of PVP based self-healing ionogel

The PVP based ionogels were prepared by the one-pot photo-initiated free radical polymerization process. To prepare the prepolymer solutions, certain amounts of PVP, AAm, TFSI, and photo-initiator Irgacure 2959 were dissolved in [EMIM][TFSI] to form a transparent solution. Different compositions were obtained by varying the mass ratio between PVP and the other components (see Table S1 for details). The molar percentage of Irgacure 2959 to TFEA and AAm was fixed at 1.0 mol%. The prepolymer solution was put into a vacuum defoaming apparatus for 5 min to remove bubbles, and then transferred into a closed glass mold coated by two polyethylene terephthalate (PET) substrates in the glove box under N_2, and crosslinked under UV light (365 nm) for 2 h.

1.3. Mechanical characterization

Tensile testing was performed on an electronic tensile machine (Instron 5965) with a 100 N load cell in an ambient condition (25 °C, 60% RH), and the stretching rate was set at 100 mm/min. The elastomer samples were cut into a dumbbell shape with a dimension of 20.0 × 2.0 × 1.7 mm^3. The cyclic tensile tests were recorded at a speed of 100 mm/min with a strain of 100%.

1.4. Electrical characterization

Ionogel samples with 8 mm diameter and 1.8 mm thickness were prepared using a PDMS mold, and subsequently incorporated into a coin cell (CR2032) using a stainless-steel disc of 8 mm diameter as the working electrode, and a nickel foam as a spacer. The electrical characterization of the coin cells was performed on an electrochemical workstation (CHI660e, CH Instruments, USA). The initial voltage was the measured open voltage; the frequency ranged from 1 Hz to1 MHz; and the AC amplitude was 0.01 V. The coin cells were allowed at the desired temperature for at least 30 min to reach the equilibrium before the measurements were taken. The toughness of ionogels in different proportions is determined by integrating the stress-strain curve of the corresponding ionogel, that is, the area enclosed between the stress-strain curve and the horizontal coordinate.

1.5. Evaluation of self-healing properties

The original elastomer was cut in half using a blade, and the severed sections were reconnected at room temperature for different times (1 h, 6 h, 12 h, and 24 h). The fractured

samples were allowed to self-heal over different periods of time and then subjected to tensile tests as described above. The mechanical properties of the repaired samples were tested using a universal tensile testing machine. The tensile strength and elongation of elastomers healed for different times were obtained from the tensile test. The self-healing efficiency (E) of the elastomer was calculated as follows:

$$E = \sigma_h/\sigma_0 \times 100\%$$

where σ_0 and σ_h correspond to the tensile strength for the original and healed samples, respectively.

1.6. Fabrication of the flexible strain sensor

The flexible strain sensor was assembled using a rectangle sample with a dimension of 40 × 10 × 1 mm^3. Two ends of the sample were attached with copper foil which connect to the electrochemical workstation, leaving 20 mm ionic conduct distance (L) in the middle, and the tensile strain were ranged from 0%-300%. The relative resistance change curves was obtained by the electrochemical workstation (CHI660e, CH Instruments, USA).

1.7. Other characterization

Fourier-transform infrared (FT-IR) spectra were recorded on a PerkinElmer Frontier. Each measurement included an average of about 32 scans from 4000 to 500 cm^{-1}. Thermogravimetric analysis (TGA) measurements were performed on a TGA Discovery 5500 with a temperature range from room temperature up to 600 °C at a scanning rate of 10 °C/min under a flowing N_2

environment. Optical test was performed on a UV-Vis spectrophotometer (UV-1800 Shimadzu). Ionogel samples with a thickness of 1.0 mm were tested with a wavelength range of 200 to 800 nm. The reference for measuring transparency was air.

Figure S1-S6.

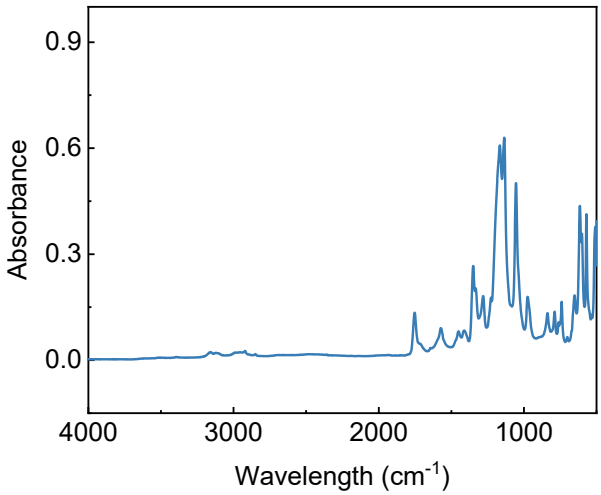

Figure S1. FT-IR spectra of the PVP-based ionogel from wavelength 500 to 4000 cm^{-1}.

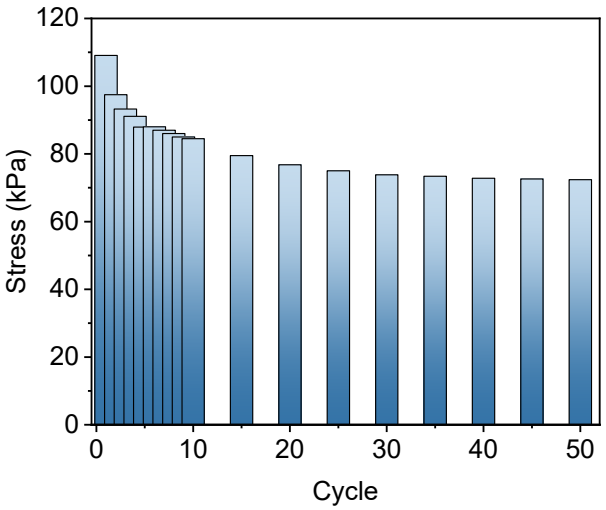

Figure S2. Variation of stress with number of cycles in 50 tensile-unloading cycle tests at 200% strain.

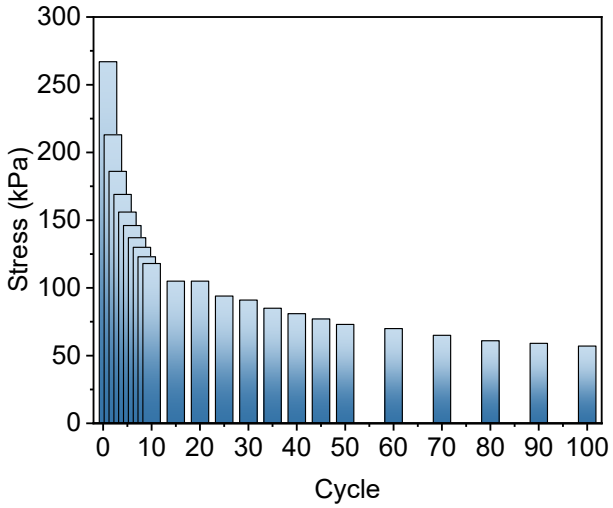

Figure S3. Variation of stress with number of cycles in 100 tensile-unloading cycle tests at 500% strain.

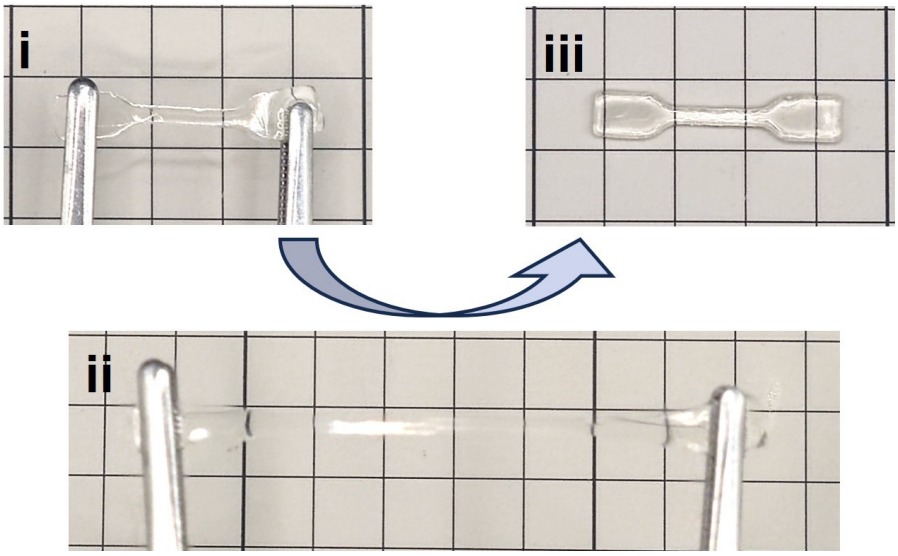

Figure S4. The dumbbell-shaped Ionogel-7.5 sample was able to fully recover its original length within 3 minutes after it had been extended 5 times its original length.

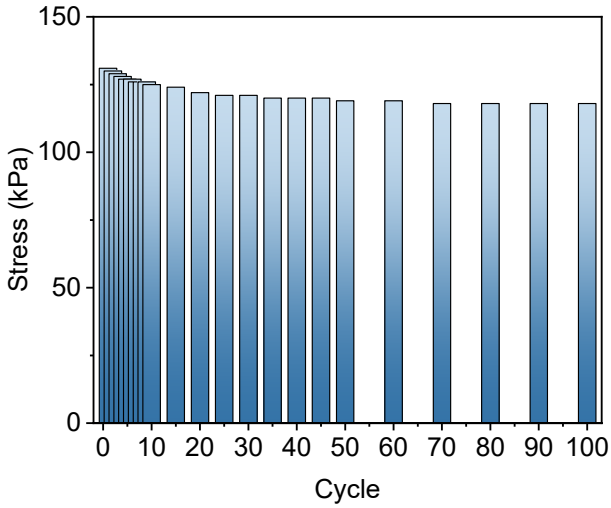

Figure S5. Variation of stress with number of cycles in 100 compression-unloading cycle tests at 50% strain.

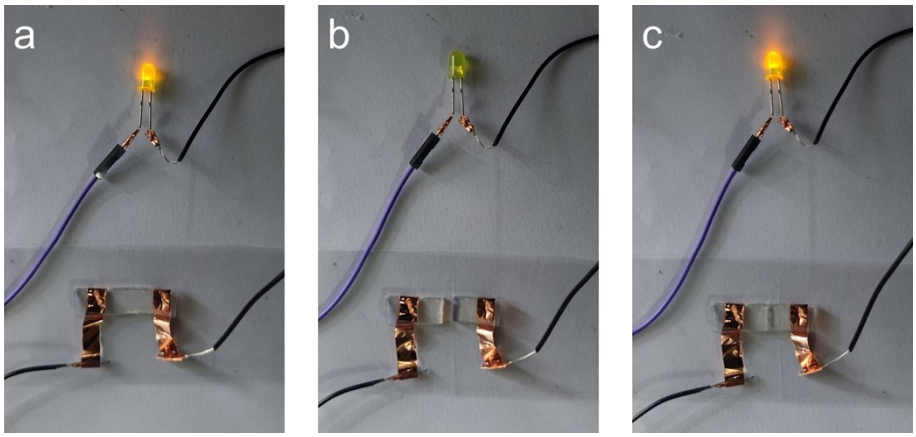

Figure S6. The electrical self-healing properties of the ionogel are demonstrated by cutting and subsequently healing an ionogel to illuminate a light-emitting diode (LED).

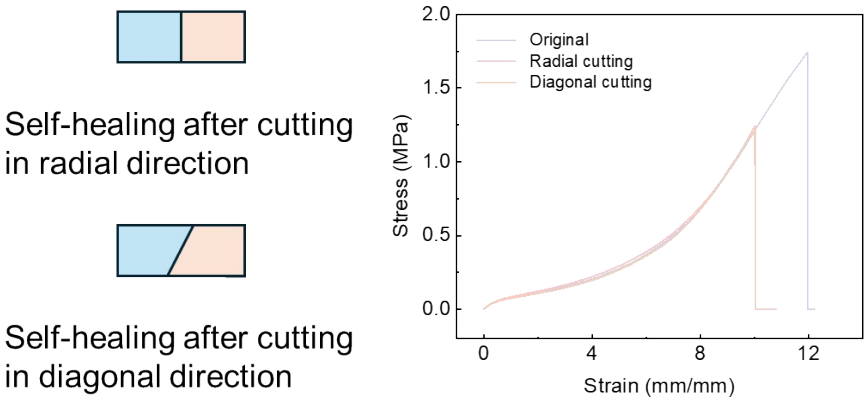

Figure S7. Stress–strain curves of the Ionogel-7.5 healed at room temperature with different cutting directions.

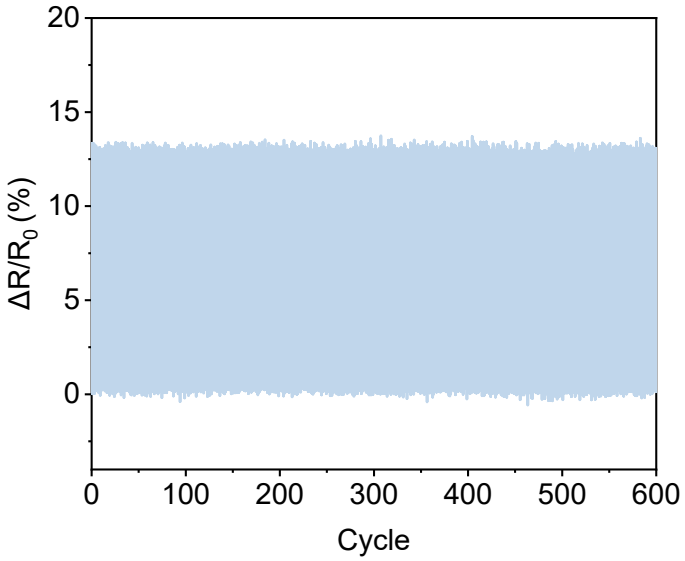

Figure S8. Relative resistance response under repeated loading-unloading processes with a strain of 20% for 600 cycles.

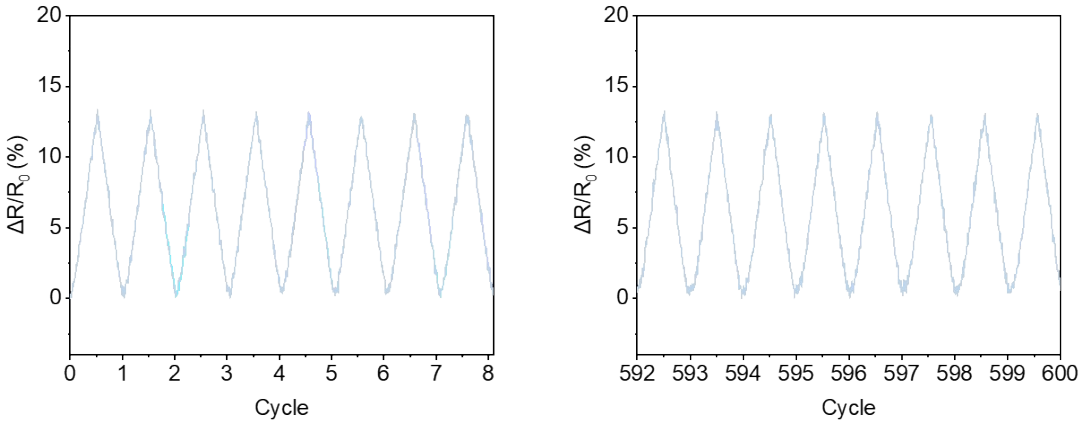

Figure S9. Zoomed-in comparison of response curves for the initial (left) and final (right) 8 consecutive cycles.

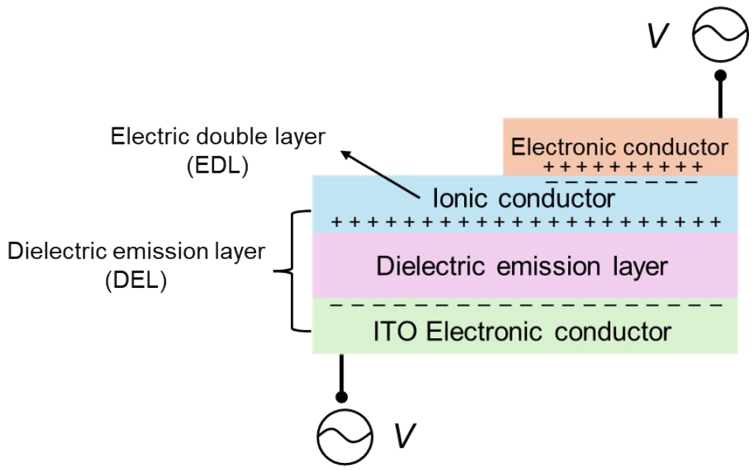

Figure S10. Illustration of the working principle of the flexible electroluminescent device. The electronic conductor and ionic conductor constitute the electric double layer (EDL) capacitor, whereas the dielectric emission layer (DEL) and the ionic conductor form the DEL capacitor.

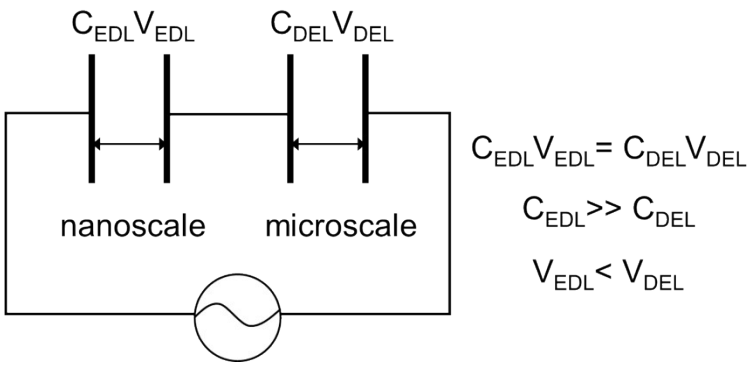

Figure S11. The equivalent circuit of the flexible electroluminescent device illustrates the connection of three capacitors in a series configuration.

Table S1-S2.

Table S1. Nomenclature of the ionogels with different polymer mass fractions and monomer molar ratios.

Samples	PVP mass fraction (%)	Monomer molar ratio (TFEA/AAm)	IL mass fraction (%)
Ionogel-2.5	2.5	2:1	60
Ionogel-5	5	2:1	60
Ionogel-7.5	7.5	2:1	60
Ionogel-10	10	2:1	60
Ionogel-15	15	2:1	60

Table S2. Summary of the mechanical properties of the ionogels with different compositions at the deformation rate of 100 mm min^{-1} under ambient conditions.

Composition	Maximum tensile strength (MPa)	Strain at break (%)	Young's modulus (kPa)	Toughness (MJ/m^3)
Ionogel-2.5	0.92	1485	72	5.14
Ionogel-5	1.10	1298	78	5.16
Ionogel-7.5	1.74	1195	108	6.80
Ionogel-10	1.98	993	128	5.63
Ionogel-15	2.03	889	144	5.06

Table S3. A rough comparison of the overall performance between this work and previously reported recyclable ionogel materials.

Ref.	Mechanical strength	Transparency	Ionic conductivity	Self-healing efficiency	Strength after self-healing
This work	1.74 MPa	92.28%	1.02 mS cm^{-1}	70%	1.32 MPa
Angew. Chem. Int. Ed. **2022**, *61*, e202212512	8 MPa	Transparent	0.1 mS cm^{-1}	1.6%	0.13 MPa
ACS Sustainable Chem. Eng. **2023**, *11*, 15031–15042	49.7 kPa	90%	1.0 mS cm^{-1}	NA	NA
Adv. Mater. **2022**, *34*, 2203049	0.25 MPa	90%	1.06 mS cm^{-1}	100%	0.25 MPa
Adv. Mater. **2021**, *33*, 2105306	0.55 MPa	92%	1.27 mS cm^{-1}	100%	0.55 MPa
ACS Appl. Mater. Interfaces **2023**, *15*, 28664–28674	0.5 MPa	91.9%	1.0 mS cm^{-1}	96.4%	0.48 MPa
ACS Appl. Mater. Interfaces **2021**, *13*, 20653–20661	0.2 MPa	87%	0.3 mS cm^{-1}	90%	0.18 MPa

Movie S1. Demonstration of the electrical sensing properties of the ionogel by lighting up a small LED bulb.