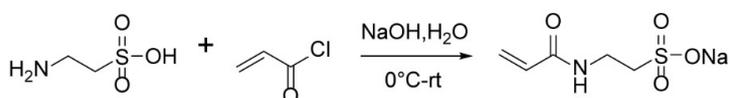


## Supporting Information

### Experimental section

**Materials:** Acrylamide (AM), 2-ketoglutaric acid, and 2-amino-ethanesulfonic acid were purchased from Damas-beta reagent. methacrylamido propyl trimethyl ammonium chloride (MPTAC, in 50% aqueous solution) was purchased from Sinopharm Chemical Reagent Co. Ltd., China. 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide (MTT) was obtained from Aldrich Chemical Co. (Shanghai, China). Hoechst 33258 was supplied by Beyotime Biotechnology Co. Ltd. (Shanghai, China).

### Synthesis of 2-acrylamido-ethanesulfonic sodium



2-amino-ethanesulfonic acid (3.0 g, 25 mmol) and NaOH (5.76 g, 150 mmol) were dissolved in 30 mL deionized water, and then acryloyl chloride (6.0 mL, 75 mmol) was dropped at a rate of 0.1 mL/min to maintain the reaction temperature no more than 0 °C. After the solution was stirred for additional 2 hours at room temperature, the solvent was removed out by spinning evaporator, and the residue was recrystallized from ethanol to give the product. Yield: 4.19 g (87%); <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>): δ (ppm) = 2.59 (dd, 2H, -CH<sub>2</sub>-), 3.39 (m, 2H, -CH<sub>2</sub>-), 5.52 (dd, 1H, =CH<sub>2</sub>), 6.01 (dd, 1H, =CH<sub>2</sub>), 6.13 (dd, 1H, =CH-), 8.01 (s, 1H, -NH-). <sup>13</sup>C NMR (150 MHz, DMSO-*d*<sub>6</sub>): δ (ppm) = 36.03 (2C, -CH<sub>2</sub>-), 50.92 (2C, -CH<sub>2</sub>-), 125.38 (2C, =CH<sub>2</sub>), 132.37 (1C, =CH-), 164.85 (1C, C=O). ESI-MS: Calcd for 2-acrylamido-ethanesulfonic sodium (C<sub>5</sub>H<sub>7</sub>NNaO<sub>4</sub>S): 199.9999 [M]<sup>-</sup>, found: 200.0015 [M]<sup>-</sup>.

### Nuclear magnetic resonance (NMR) spectroscopy

The spectra were recorded at 25 °C on AVANCE III 500 MHz (Bruker Biospin, Switzerland) at working frequency of 500 MHz for the <sup>1</sup>H nuclei. All chemical shifts were reported in ppm relative to the signals corresponding to the residual non-deuterated solvents (DMSO-*d*<sub>6</sub>: δ = 2.50 ppm).

The spectra were recorded at 25 °C on AVANCE III 500 MHz (Bruker Biospin, Switzerland) at working frequency of 150 MHz for the <sup>13</sup>C nuclei. All chemical shifts were reported in ppm relative to the signals corresponding to the residual non-deuterated solvents (DMSO-*d*<sub>6</sub>: δ = 39.52 ppm).

### High-resolution mass spectrometry (HRMS)

maXis Impact + 1290 infinity (Bruker Daltonics Inc., America) was used to record the spectra in negative ion mode. The performance and resolution were verified using Tunemix (Agilent Technologies, USA) with resolution > 40,000 (FWHM) @ 1222 m/z. Mass calibration was done using Tunemix (internal calibration) between m/z 50 and 3,000 in the same acquisition mode with accuracy of 10 ppm. One acquisition consisted of infusing the sample at 50 μM in CH<sub>3</sub>OH and H<sub>2</sub>O (v : v = 1 : 1) in 2 min, followed by 1-min infusion of Tunemix. The capillary voltage was set to

4500 V and the ion energy at 5 eV. The system was controlled by Compass Hystar (Bruker Daltonics Inc., America). Before injection of the sample, a blank solution of CH<sub>3</sub>OH and H<sub>2</sub>O (v : v = 1 : 1) was injected as reference.

#### **Synthesis of poly(AASA-co-AM)**

2-acrylamido-ethanesulfonic sodium (1.8 g), acrylamide (1.44 g), and 2-ketoglutaric acid (40 mg) were dissolved in deionized water (8 mL) in a 20-mL beaker under vigorous stirring in dark condition by a magnetic bar. After 1 h, the stirring was stopped, and the beaker was moved to a vacuum oven for removal of formed gas bubbles in solution at 0.01 MPa. The solution in the beaker was then exposed to ultraviolet (UV) light (intensity: 200 mW cm<sup>-2</sup>) at 365 nm for 2 h. The polymerized product was taken out and purified three times by precipitation in methanol. The solid product was collected and dried into white powder with a yield of 95.6%. Before application, the white powder (1.0 g) was fully dissolved in 10 mL deionized water (heat is optional) and the gas bubbles in the solution were removed by vacuum oven at 0.01 MPa. Finally, the solution was filled into a glass template (size: 65.0 mm × 15.0 mm × 1.5 mm) and dried under 40 °C into a transparent film with the thickness of 1.0 μm.

#### **Synthesis of poly(MPTAC-co-AM)**

Methacrylamido propyl trimethyl ammonium chloride (2.0 g), acrylamide (1.3 g), and 2-ketoglutaric acid (45 mg) were dissolved in deionized water (8 mL) in a 20-mL beaker under vigorous stirring in dark condition by a magnetic bar. After 1 h, the stirring was stopped, and the beaker was moved to a vacuum oven for removal of formed gas bubbles in solution at 0.01 MPa. The solution was exposed to ultraviolet (UV) light (intensity: 200 mW cm<sup>-2</sup>) at 365 nm for 5 h. The product was taken out and reprecipitated in methanol for three times. The solid product was collected and dried into white powder with a yield of 98.2%. The powder (1.0 g) was then dissolved in 7.5 mL deionized water, and the gas bubbles in the solution were removed by vacuum oven at 0.01 MPa. The prepared solution was used for in-situ formation of hydrogel poly(AASA-co-AM)/poly(MPTAC-co-AM).

#### **IR spectroscopy**

All specimens were tested at 25 °C with Nicolet iS50 (Thermo, America) FT-IR spectrometer by pressed-disk technique. The dried compound powder was mixed with KBr (weight ratio to KBr = 1: 50) and pressed into a disk prior to IR scanning. The measurement was repeated three times for each sample and the air background was subtracted.

#### **Diffusion coefficient**

Diffusion ordered spectroscopy (Dosy) using stimulated echo (STE) pulse sequence was performed on a Bruker Avance NEO 600.41 MHz spectrometer (Bruker Biospin, Switzerland), equipped with a Z-gradient probe. The temperature was controlled at 25°C. The diffusion coefficients were determined using the relationship between the echo signal intensities and the field-gradient parameters:  $I(g) = I(0) \exp[\gamma^2 \delta^2 g^2 (\Delta - \delta/3) D]$ , where  $I(g)$  and  $I(0)$  are the echo signal intensities with and without the field gradient pulse, respectively;  $\gamma$  is the gyromagnetic ratio of H;  $\delta$  is the duration of the gradient pulses;  $g$  is the field gradient strength;  $\Delta$  is the diffusion time; and  $D$  represents the diffusion coefficient. Here,  $\delta$  and  $\Delta$  were fixed to 1.5 ms and 0.4 s, respectively. Standard decay

curves were recorded with 16 increments and the gradient strength linearly increased from 2% to 95% of the probe's maximum value. The maximum gradient strength was 5.35 G/mm.

### **X-Ray Diffraction (XRD)**

Dried hydrogel of poly(AASA-*co*-AM)/poly(MPTAC-*co*-AM) was cut to rectangle strips (2.0 cm × 2.0 cm), which were kept between two glass slides prior to the diffraction tests on an X-Ray diffraction instrument (Mode: SmartlabSE). The final spectra were obtained by deducting the background of glass slide.

### **Zeta-potential measurement**

The zeta potentials of zwitterionic polymers were measured on a NanoBrook 90 Plus PALS. poly(AASA-*co*-AM) and poly(MPTAC-*co*-AM) were dispersed in water at a concentration of 0.10 g/mL, respectively. The pH value of the solution was adjusted by 1 mol/L HCl solution or 1 mol/L NaOH solution.

### **SEM**

Microscopic network structure of hydrogel of poly(AASA-*co*-AM)/poly(MPTAC-*co*-AM) was recorded on an electron microscope (S-4800, SYST TA PRO 1156) with the primary electron energy of 5 kV. We observed the hydrogel by focusing on the surface. After treatment in pH 7.0 deionized water and pH 10.0 NaOH solution, the hydrogel samples were immersed in liquid nitrogen and then lyophilized in a freeze-dryer (Model: FD-1A-50, Shanghai Bilon Instrument Manufacturing Co., Ltd). The samples were attached to a silicon wafer with adhesive carbon tape and coated with a thin layer of gold (~5 nm) prior to the SEM observation. The micrographs were recorded at room temperature and a pressure of  $8.8 \times 10^{-7}$  Pa.

### **Tensile testing**

All specimens were tested at 25 °C. A sample of rectangular shape (25.0 mm × 10.0 mm × 1.5 mm) was clamped by the grips of movable and stationary fixtures in a screw-driven device using universal testing machine equipment (Model: HY-0580, purchased from Shanghai hengyi precise instrument limited company) that pulled the sample until breaking while recording the applied load and elongation. The testing speed was 20.0 mm/min. The load cell (50 N) and extensometer were calibrated before use. The tests complied with rules specified by the international standard norms.

### **Gauge Factor testing**

All specimens were tested at 25 °C. The changes of the strain of the hydrogel were recorded by universal testing machine equipment (Model: HY-0580). The changes of the impedance of the hydrogel were recorded by Keysight 34401A Digit Multimeter. The rate of change in impedance (%) was obtained by equation:  $\Delta R / R_0 (\%) = (R - R_0) \times 100\% / R_0$

where  $R$  is the real-time impedance under different strains, and  $R_0$  is the initial impedance without strain. The strain sensitivity GF was calculated by formula:  $GF = (\Delta R / R_0) / \varepsilon$

where  $\varepsilon$  is the strain generated by the hydrogel.

### **Self-healing testing**

All specimens were tested at 25 °C. A hydrogel strip with the size of 25.0 mm × 10.0 mm × 1.5 mm

was cut to half and dyed with fluorescent green and rhodamine B, respectively. Then, two pieces of the hydrogel were put together. After the self-healing process was kept for 1, 2, 4 and 8 hours, respectively, the self-healing efficiency was evaluated based on their stress-strain tensile results, which was compared with the intact hydrogel.

### **Interfacial adhesion**

The interfacial adhesion between hydrogel and pigskin was measured with a standard 90° peeling test on a tensile testing machine, equipped with a 90° peeling fixture. The pigskin was washed with water and wiped with paper tissues before use. A 50 N load cell was used for all the tests. The peeling speed was constant at 20 mm min<sup>-1</sup>. One side of the hydrogel was covered with a rigid tape backing to prevent the elongation along the peeling direction, the other side was tightly attached to the pigskin surface. Thus, the measured interfacial adhesion was equal to the steady-state peeling force per width of the bilayer. The hydrogel and pigskin were kept in sealed environment to reduce the water loss. The plateau force was then divided by the width of the hydrogel strip to get the interfacial adhesion value. Meanwhile, adhesion tests of different substrates were done using the same technique. The substrates include wood, paper, glass, nitrile glove, frog organs, leaf, rubber and ceramics. The substrates were washed and wiped with paper tissues before use. The adhesion on wood was tested using a hydrogel strip (60.0 mm× 10.0 mm) with its half part being adhered to a wood stick. The strip was pulled to lift a weight of 1 kg.

### **Cytotoxicity tests**

NCTC clone 929 mouse fibroblasts cell was purchased from the Cell Bank of Type Culture Collection of the Chinese Academy of Sciences (Shanghai, China). The cytotoxicity of hydrogel was evaluated against NCTC clone 929 mouse fibroblasts cell by MTT assay according to the ISO 10993-5 international standard. All samples were pretreated according to ISO 10993-12 international standard. Typically, a freeze-dried hydrogel piece was milled and sterilized by UV lamp for 4 h and immersed in culture medium for 24 h. The medium containing hydrogel extracts was filtered and diluted into different concentrations: 0.12, 0.25, 0.50 mg/mL. L929 cells were seeded into a 96-well culture plate with a density of 1×10<sup>4</sup> cell per well, and incubated in the liquid extraction for 24, 48 and 72 h, respectively. After the liquid extraction was removed, 20 μL MTT solution (5 mg mL<sup>-1</sup>) and 180 μL MEM medium were added to each well. The plate was taken into an incubator with 5 % CO<sub>2</sub>, and kept at 37 °C for 4 h. After removing the MTT solution and MEM medium, 100 μL DMSO was added into each well to dissolve the formazan. The plate was put into an incubator with 5 % CO<sub>2</sub> and kept at 37 °C for 6 h. For the control group, the cells were seeded into MEM medium without liquid extraction. Meanwhile, the MEM medium without cells were used as a blank group. All experiments were proceeded for six groups. The optical density (OD) of the formazan was quantified by using an enzyme standard instrument at 570 nm, where the OD reflected the level of cytotoxicity.

The cell growth was recorded by a fluorescent inverted microscope (TH4-200 Olympus). L929 cells were seeded into a 6-well culture plate with a density of 2 × 10<sup>5</sup> cell per well and cultured with liquid extraction of each hydrogel sample for 24 h. After the liquid extraction was removed, 2 mL 4% paraformaldehyde fix solution was added to each well for 10 min. Then the fix solution was absorbed, and 6 mL PBS solution was used to wash the cells for three times. 1 mL nuclear stain

(Hoechst 33258, 10  $\mu\text{g}/\text{mL}$ ) was added to the well, and kept for 8 min. After the nuclear stain was removed, 6 mL PBS solution was used to wash the cells for three times. Finally, the 6-well culture plate was transferred to the fluorescence microscope for the observation and photographing.

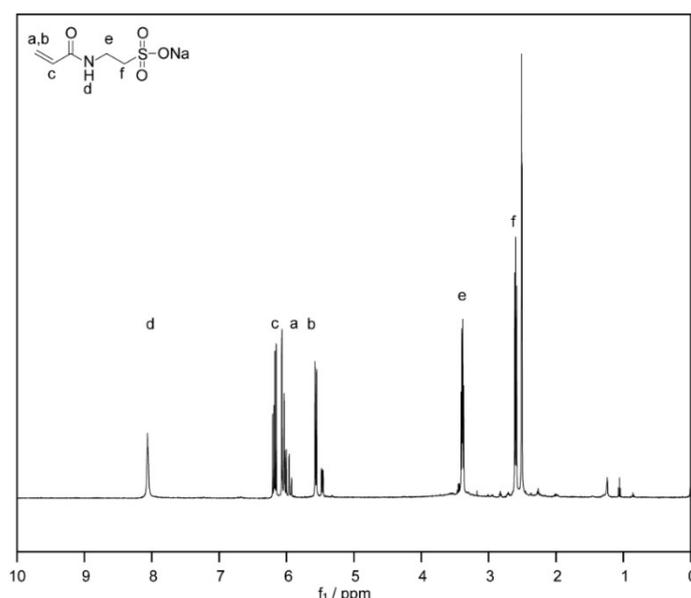
### Pithing for frog surgery

Pithing would destroy the frog brain, which is located between two eyes of the frog. A probe was inserted quickly into the cranial vault which was moved into the cranial vault from side to side to destroy the brain and spinal cord. At this time, the frog was relaxed completely, indicating that it has been executed successfully. Pithing method was relatively painless to the frog. With this operation, the frog would be insensitive to pain. After applying liquid poly(MPTAC-co-AM) at the surface of the frog heart, the thin solid film of poly(AASA-co-AM) was covered to form hydrogel sensor in situ. Two electrodes were implanted in the hydrogel, and the heartbeat was monitored by the hydrogel sensor, in which the electric signals were collected by a Keysight 34401A Digit Multimeter

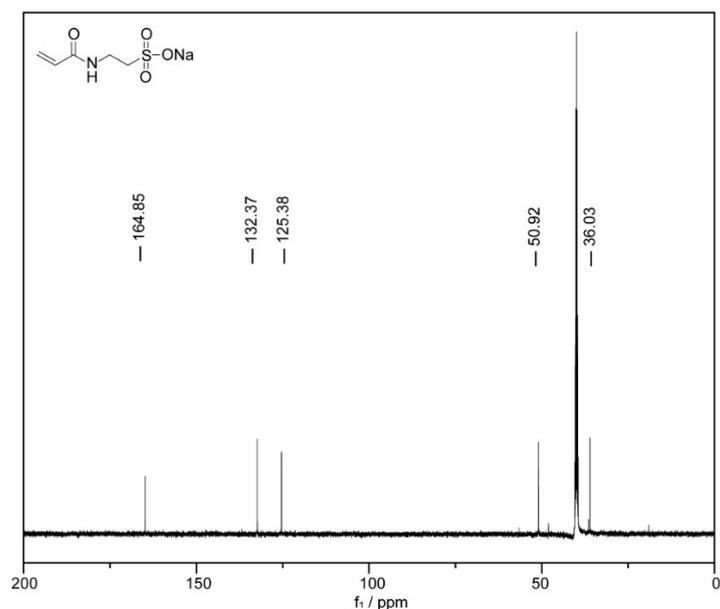
### Cyclic tensile testing

All specimens were tested at 25  $^{\circ}\text{C}$ . A rectangular hydrogel strip (25.0 mm  $\times$  10.0 mm  $\times$  1.5 mm) was clamped by the grips of movable and stationary fixtures in a screw-driven device using universal testing machine equipment (Model: HY-0580, purchased from Shanghai hengyi precise instrument S3 limited company). The strip was loaded to generate 10%, 20% and 50% strain and then unloaded for complete recovery of the strain. The number of cycles was set to 300 for each strip sample. The strip was stretched at a rate of 10 mm/min<sup>-1</sup> and the retrace rate remained the same. Two electrodes were implanted inside the hydrogel strip. The changes of the impedance were recorded by a Keysight 34401A digit multimeter.

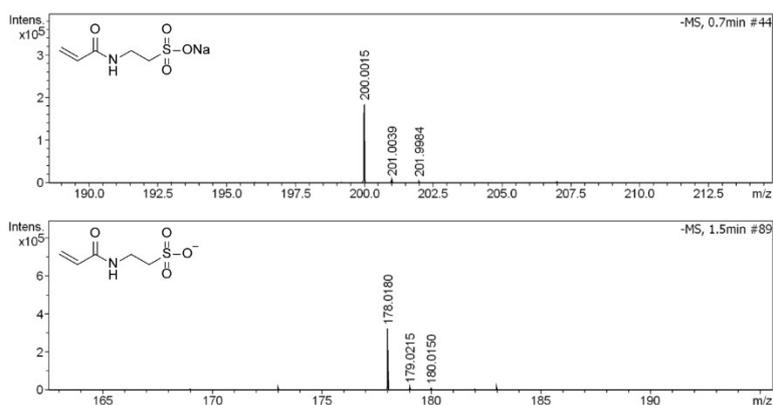
### Supplementing Figures



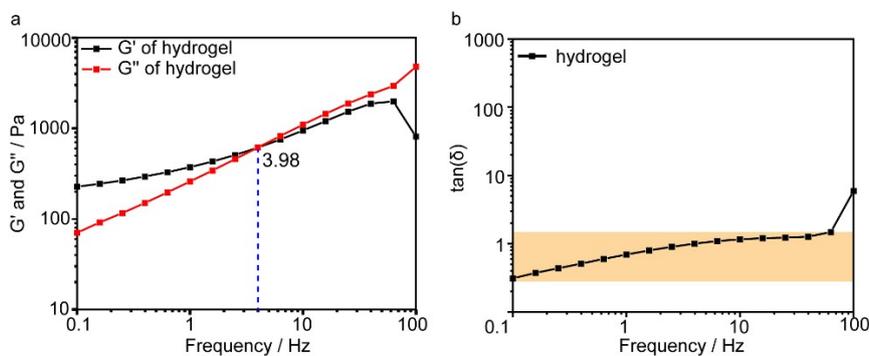
**Figure S1.** <sup>1</sup>H NMR spectrum of 2-acrylamido-ethanesulfonic sodium. DMSO-*d*<sub>6</sub> was used as deuterated solvent.



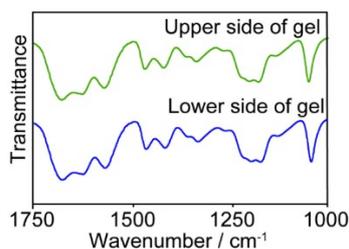
**Figure S2.**  $^{13}\text{C}$  NMR spectrum of 2-acrylamido-ethanesulfonic sodium.  $\text{DMSO-}d_6$  was used as deuterated solvent.



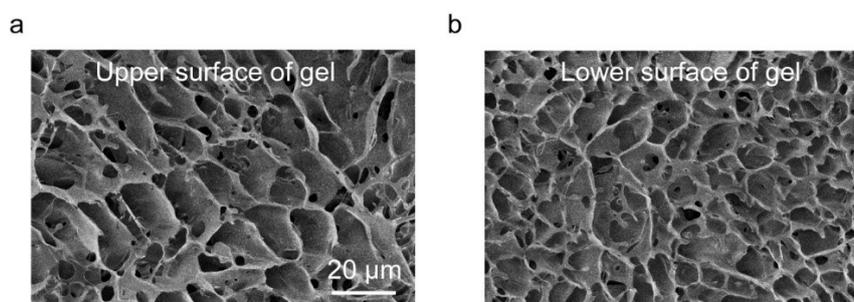
**Figure S3.** ESI-MS Spectra of 2-acrylamido-ethanesulfonic sodium. Calcd for 2-acrylamido-ethanesulfonic sodium ( $\text{C}_5\text{H}_7\text{NNaO}_4\text{S}$ ): 199.9999 [M] $^-$ , found: 200.0015 [M] $^-$ , Calcd for  $\text{C}_5\text{H}_8\text{NO}_4\text{S}$ : 178.0180 [M] $^-$ , found: 178.0180 [M] $^-$ .



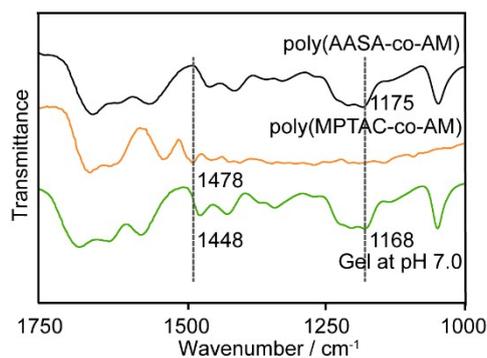
**Figure S4.** Rheological properties. (a) Dynamic viscoelastic behavior of hydrogel of poly(AASA-*co*-AM)/poly(MPTAC-*co*-AM). (b)  $\text{Tan}(\delta)$ -to-frequency curve of the hydrogel.



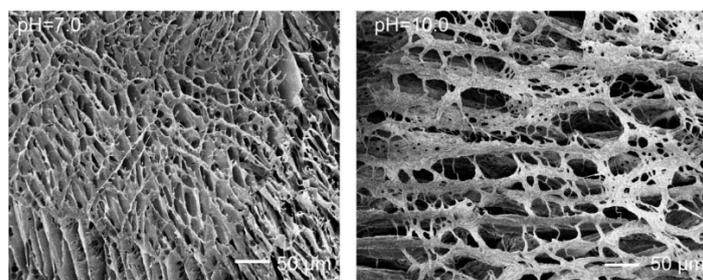
**Figure S5.** FT-IR spectra of the upper and lower surfaces of poly(AASA-co-AM)/poly(MPTAC-co-AM) hydrogel. We have tested the upper and lower surface of our hydrogel with FT-IR spectrometer. The results showed that there were no significant differences between the upper and lower surface, which indicated poly(MPTAC-co-AM) has completely permeated into the anionic film of poly(AASA-co-AM) to form a hydrogel layer.



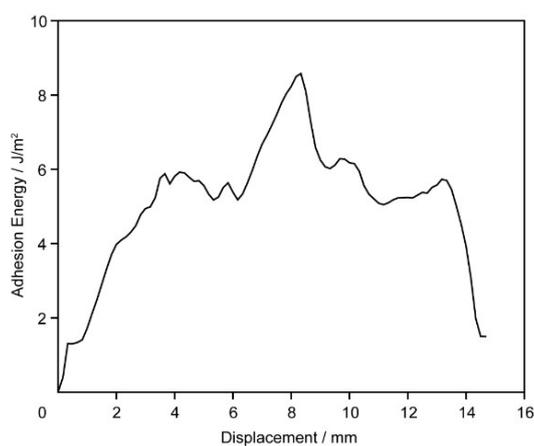
**Figure S6.** SEM images of the upper and lower surfaces of poly(AASA-co-AM)/poly(MPTAC-co-AM) hydrogel. The upper and lower surfaces did not show apparent difference in porous structures.



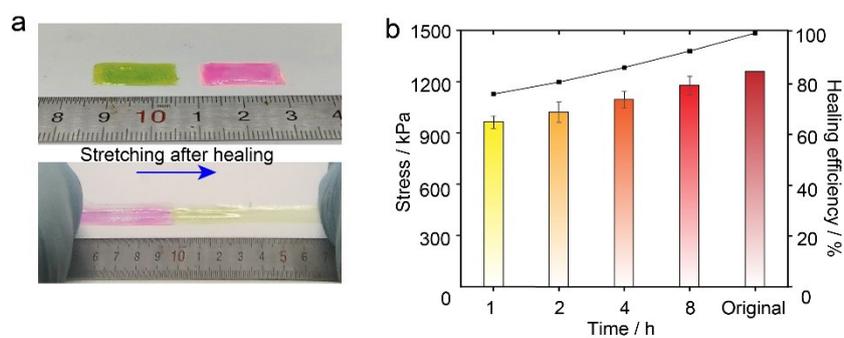
**Figure S7.** FT-IR spectra of poly(MPTAC-co-AM), poly(AASA-co-AM) and the hydrogel of poly(AASA-co-AM)/poly(MPTAC-co-AM) in pH 7.0, respectively.



**Figure S8.** SEM images of the hydrogel of poly(AASA-*co*-AM)/poly(MPTAC-*co*-AM) in pH 7.0 and 10.0.



**Figure S9.** Adhesion energy-displacement profile of the hydrogel of poly(AASA-*co*-AM)/poly(MPTAC-*co*-AM) on pigskin after being treated with NaOH solution (pH>9.0).



**Figure S10.** Self-healing testing. (a) Self-healing testing. Two pieces of hydrogel, dyed with fluorescent green and Rhodamine B, were connected for the self-healing tests. (f) Self-healing efficiency of the hydrogel.

## Supplementing Movies

**Movie S1.** Demonstration on the gelation process of two linear polymers of poly(AASA-*co*-AM) and poly(MPTAC-*co*-AM). The two linear ionic polymers were soluble in water. Once their aqueous solutions were mixed together, they would rapidly form hydrogel.

**Movie S2.** Self-adaptive adhesion of hydrogel sensor at the lower jaw of a frog. A piece of 10  $\mu\text{m}$ -thick hydrogel film sensor of poly(AASA-*co*-AM)/poly(MPTAC-*co*-AM) was tightly adhered at the lower jaw of a frog, which did not move or separate from the lower jaw, even when it was impacted by strong water flow. Such the self-adaptive adhesion allowed the hydrogel sensor to detect the dynamic motions of the lower jaw of frog and transduced them into electric signals.

**Movie S3.** Self-adaptive adhesion of hydrogel sensor at the surface of heart of a frog. A drop of aqueous solution containing 10 mg poly(MPTAC-*co*-AM) was applied on the heart, and a piece of 10 mg film of poly(AASA-*co*-AM) was covered over the coating of poly(MPTAC-*co*-AM). After waiting for 10 min, the hydrogel could be formed in situ to give a soft sensor.