Supporting Information

Experimental section

1. Materials:

Acrylic acid (AA), *N,N'*-Methylenebisacrylamide (MBA), 1,6-dibromohexane were purchased from Adamas-beta reagent. Sodium hydroxide (NaOH), hydrochloric acid (HCl) solution (36 wt.%), triethylamine, ethanol(99%), acetone(99%) were purchased from Sinopharm Chemical Reagent Co., Ltd. Ammonium persulfate (APS) was purchased from General-Reagent.

2. Preparation of hexamethylenebis(triethylammonium bromide) (BQA):

Triethylamine (0.25 mol) was mixed with 1,6-dibromohexane (0.1 mol) in ethanol (150 mL), and refluxed for 48 h at 80 °C. After removal of solvent by evaporation, white solid was formed. The white solid was washed thoroughly with acetone, and dried under vacuum to give solid product with the yield of 90%.

3. Preparation of PA/BQA hydrogel:

Monomer, AA (6.9 mmoL, 0.5 g), BQA(0.67 mmoL, 0.3 g) were dissolved in 2 mL deionized water under vigorous stirring by a magnet bar at room temperature. We then added crosslinker, MBA (0.0325 mmoL, 5 mg), and initiator, APS (0.11 mmoL, 25 mg) to the above solution and continued to stir mixture to give transparent solution. The as-prepared precursor solution was degassed by vacuum pump, and injected into a hermetically-sealed glass mold. The sample was then polymerized under ultraviolet light (365nm) for 1 h to give rise to the hydrogel of PA/BQA after removal of the mold.

4. 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 1H nuclei. All chemical shifts were reported in ppm relative to the signals corresponding to the residual non-deuterated solvents (D_2O: $\delta = 4.79$ ppm). The spectra were recorded at 25 °C on AVANCE III 500 MHz (Bruker Biospin, Switzerland) at working frequency of 150 MHz for the 13C nuclei. All chemical shifts were reported in ppm relative to the signals corresponding to the residual non-deuterated solvents.

5. Infrared spectroscopy:

All specimens were tested at 25 °C with an attenuated total reflectance Fourier transform infrared spectrometer (ATR-FTIR) (Thermo Scientific Nicolet IS10). All samples were dried at 50 °C for 12 hours in a vacuum oven before measurement at the scanning range of 4000 to 800 cm^{-1}. The variation of asymmetric stretching vibration band of C=O was recorded for revealing the phase transition of hydrogel with water swelling.

6. UV-visible spectroscopy:

The absorption spectra were conducted on a UV-vis spectrophotometer (UV-2400, SDPTOP) to test the specimens transmittance in a range of 280 – 700 nm at 25 °C. All specimens were prepared with a size of 1 cm × 1cm × 1 mm.

7. Raman spectroscopy:

Raman spectra were recorded on a Renishaw in Via Reflex spectrometer with 532 nm laser (50% power: 25 mW; exposure time: 4 s) as light source. All Raman data were collected at a spectral resolution of 1 cm^{-1}, and 5 scans were accumulated.

8. Zeta-potential measurement:

The zeta potentials of zwitterionic polymers were measured on a NanoBrook 90 Plus PALS. BQA and PA 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.

9. Water swelling ratio (WSR):

The hydrogel sample was dried at 60 °C for 24 h, which was then weighed and immersed in deionized water until saturation with water sorption at room temperature. The WSR was calculated with the equation of (Ws - Wd) / Wd, where Ws and Wd are the weight of the swollen and dried samples, respectively.

10. Differential Scanning Calorimetry (DSC):

DSC was performed on Q2000 DSC at a heating rate of 10 °C/min using DSC/TG pan of Al_2O_3. The temperature range was from 25 to 90 °C, and dry N_2 was used as carrier gas at a flow rate of 50 mL/min.

Figures

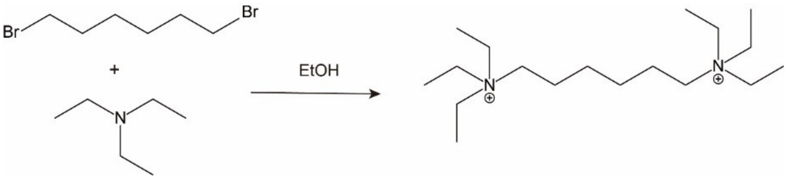

Fig. S1 Synthetic procedure of bis-quaternary ammonium cation.

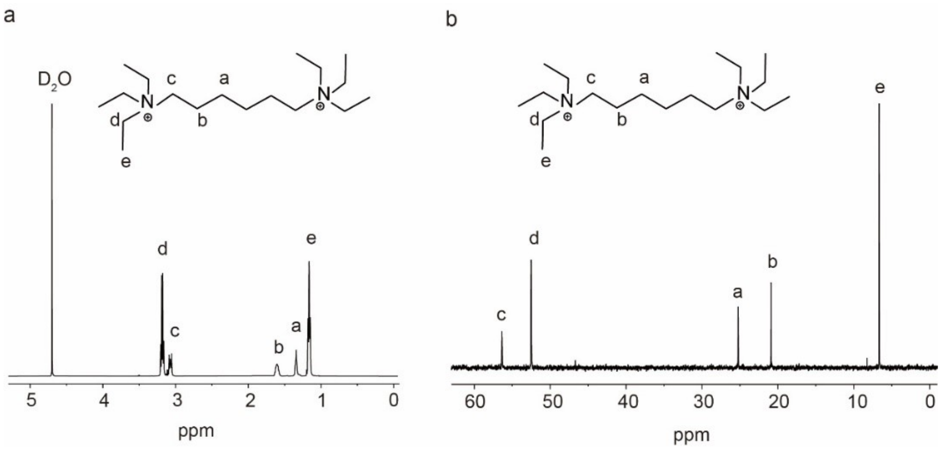

Fig. S2 (a) ^{1}H NMR spectrum of bis-quaternary ammonium cation with D_2O as solvent. (b) ^{13}C NMR spectrum of bis-quaternary ammonium cation with D_2O as solvent.

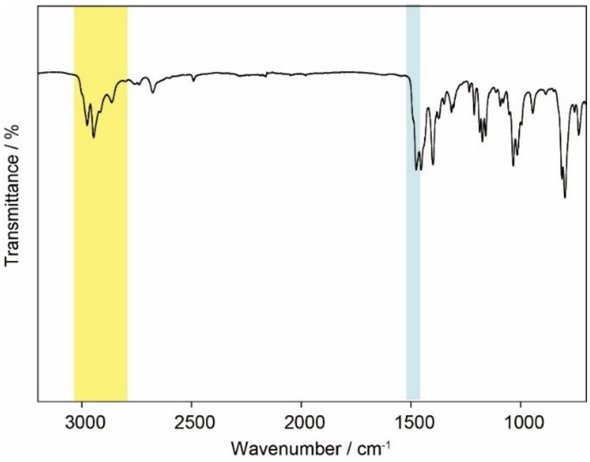

Fig. S3 ATR-FTIR spectrum of bis-quaternary ammonium cation. The absorption peaks at 2946 cm^{-1} and 2864 cm^{-1} belong to the stretching vibration of the CH_3 and CH_2, respectively (yellow area). The characteristic peak at 1475 cm^{-1} was attributed to the quaternary ammonium groups (blue area).

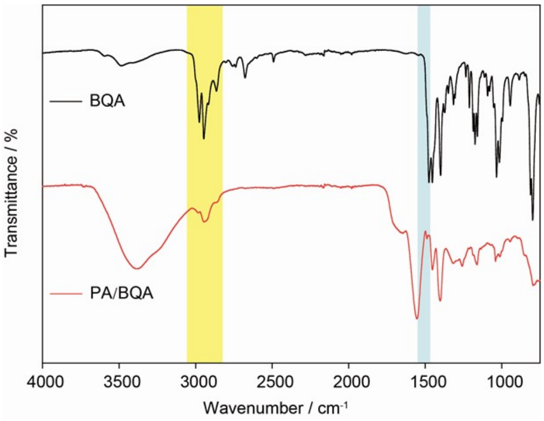

Fig. S4 ATR-FTIR spectra of neat BQA and hydrogel of PA/BQA

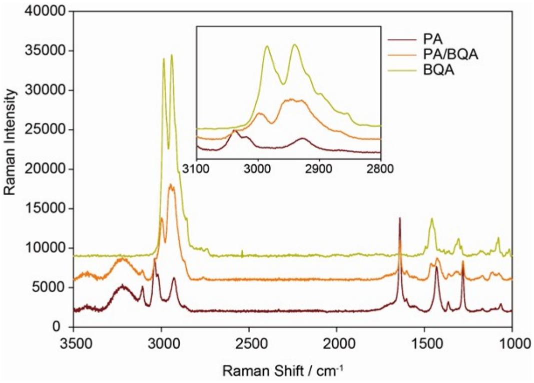

Fig. S5 Raman spectra of BQA, PA, and hydrogel of PA/BQA.

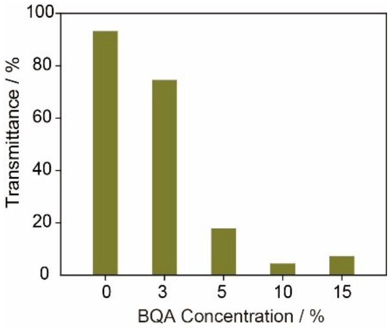

Fig. S6 Transmittance of PA/BQA with different content of BQA. The samples were tested at 550 nm.

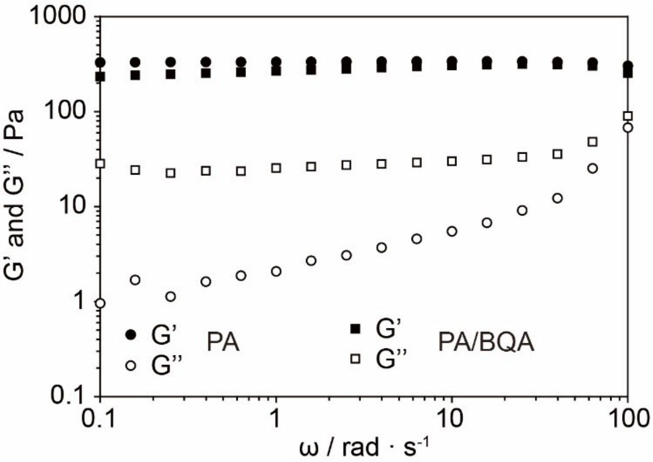

Fig. S7 Storage moduli (G′) and loss moduli (G″) of PA and PA/BQA hydrogels.

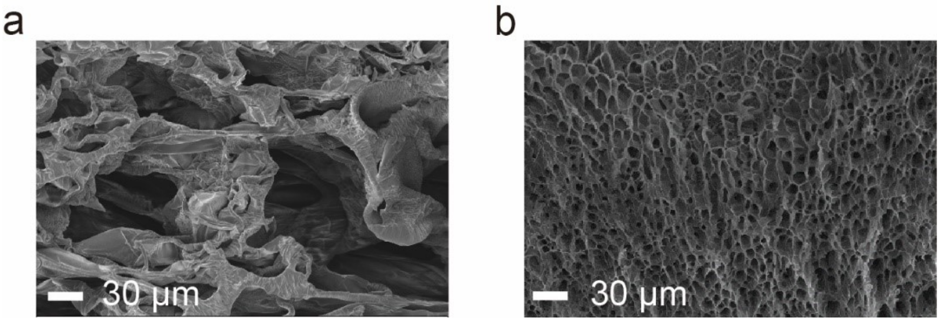

Fig. S8 SEM images of PA (a) and PA/BQA (b) hydrogels.

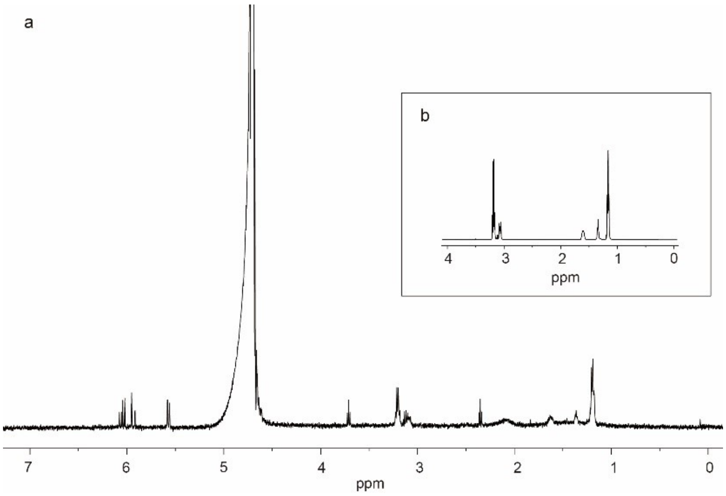

Fig. S9 (a) ^{1}H NMR spectrum of the leaching solution of PA/BQA. (b) ^{1}H NMR spectrum of BQA for the comparison. We immersed the hydrogel of PA/BQA in D_2O for 24 h, and then we measured the solution of D_2O by NMR to see if BQA was released into the D_2O solution.

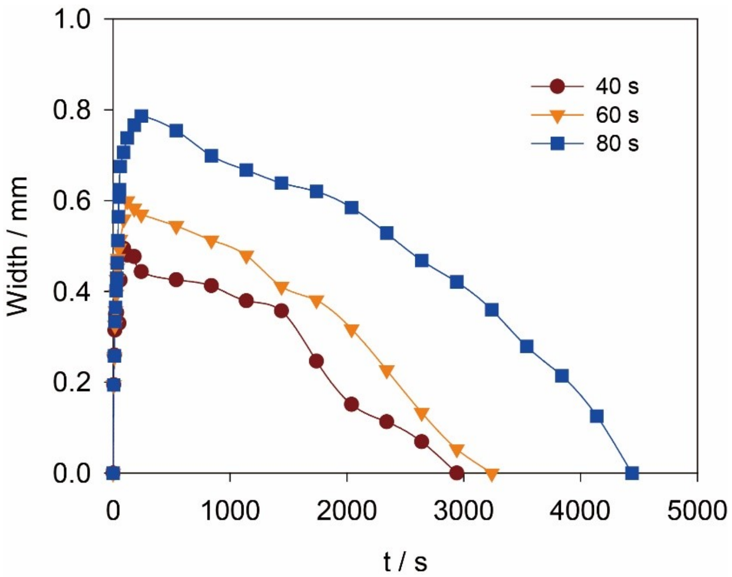

Fig. S10 Curves showing the gradual shrinking the phase transition region as a function of time after electrochemical treatments for different time.

Movie Legends

Movie S1. Electrochemistry-induced surface patterning of a triangle shape on the hydrogel of PA/BQA. The triangular pattern gradually appeared with the increase of power time. The video was displayed at its two times speed.

Movie S2. The change of phase transition degree and phase transition region with the prolongation of energization time. The video was displayed at its ten times speed.