Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

Electronic Supplementary Information

Flexible and wearable strain sensors based on tough and self-adhesive ion conducting hydrogels

Zhenwu Wang,[ab] Jing Chen,[b] Liufang Wang,[b] Guorong Gao,[b] Yang Zhou,[b] Rong Wang,[b] Ting Xu,[b] Jingbo Yin,*[a] and Jun Fu *[b]

a School of Materials Science and Engineering, Shanghai University, 99 Shangda Road, Shanghai 200444, People's Republic of China. Email: jbyin@oa.shu.edu.cn
b Cixi Institute of Biomedical Engineering & Polymers and Composites Division, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Zhongguan West Road 1219, Zhenhai District, Ningbo, 315201, People's Republic of China. Email: fujun@nimte.ac.cn

Experimental Section

Materials

Poly(vinyl alcohol) (99% hydrolyzed, degree of polymerization 1750, analytical grade) was purchased from Shanghai Chemical Reagent Co. Ltd. The [2-(methacryloyloxy) ethyl] dimethyl-(3-sulfopropyl) (SBMA, 97%) and 2-hydroxy-2-methylpropio- phenone (1173, 97%) were purchased from Aladdin Reagent Co., Ltd., China. All aqueous solutions were prepared with deionized (DI) water.

Preparation of ion conductive hydrogels

SBMA was dissolved into 5 mL deionized water containing 0.1 mol% 1173 under sonication to form solution A. Meanwhile, 1 g PVA was added to 5 ml of deionized water under stirring at 90 °C for 2 h as solution B. Then, solutions A and B were mixed by vigorous stirring to form a homogeneous mixture. After removing bubbles, the

SBMA in mixture was initiated by UV irradiation (365 nm in wavelength, 1 J m^{-2}) for 5 min to produce PSBMA. Finally, the solution was frozen at –20 °C for 3 h and thawed at room temperature for 1 h to form PVA/PSBMA hydrogel.

Characterization of hydrogels

The hydrogel samples were tested by using an Instron 5567 Instrument (Instron Inc., MA), which were coated with silicon oil to prevent water loss during testing. Three samples were tested for each hydrogel. For tensile tests, PVA/PSBMA hydrogels with thickness of 1 mm were cut into dumbbell-shaped bars 2 mm wide and 10 mm long and uniaxially tested at 100 mm/min. Cylinder-shaped samples (5-8 mm height and 20 mm diameter) were compression tested at 10%/min. The lap shear test was performed to measure the adhesive strength of PVA/PSBMA hydrogels on various substrates with the contacting area of 14 mm × 8 mm at 100 mm/min until full separation. The hydrogels were quenched in liquid nitrogen, fractured, freeze-dried and gold sputtered on the fracture surface for imaging by scanning electron microscopy (SEM, Hitachi S-4800, Japan).

Electrical test on strain sensors

The real-time electrical signals of the hydrogels at different strains were recorded by using a CHI660E Electrochemical workstation. The relative change of the resistance was calculated by Ohm's law ($R = U/I$) on the basis of the application of a constant voltage to the strain sensors inducing changes in the electrical current under different strains.

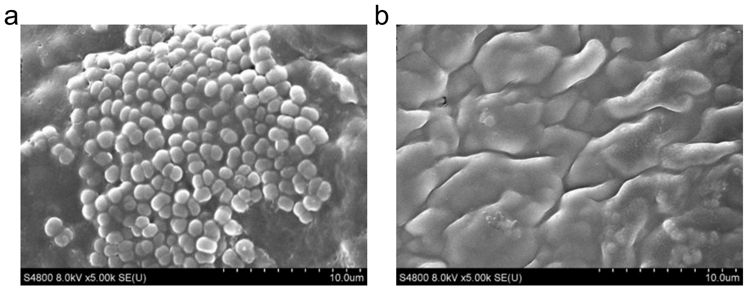

Figure S1. SEM image of (a) PVA/PSBMA mixture and (b) PVA/PSBMA hydrogels.

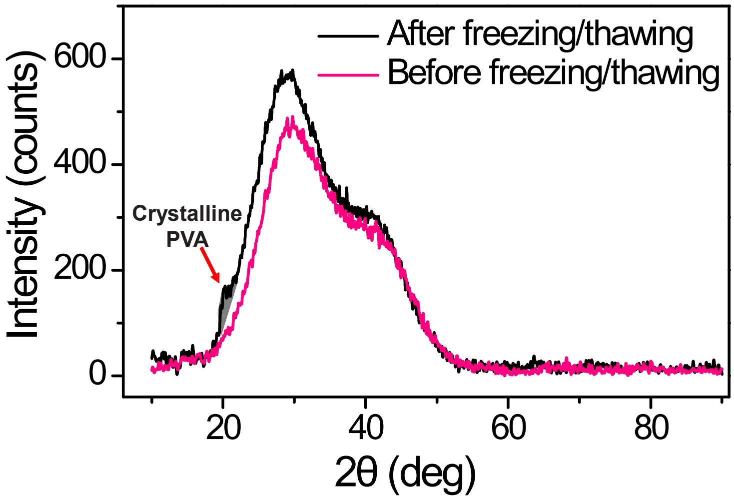

Figure S2. X-ray powder diffraction profiles of as-prepared hydrogels with 15wt% PSBMA before and after freezing/thawing.

The X-ray diffraction profiles of as-prepared hydrogel samples are shown in Figure S1 after subtraction of a straight baseline. After comparing two curves, a weak peak in the 2θ range 18-21° (gray part) was found, demonstrating the presence of crystalline PVA aggregates, which is in accordance with the previous article[1].

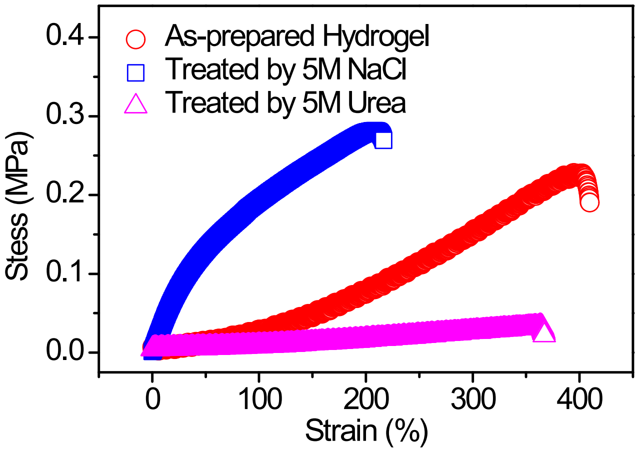

Figure S3. Representative tensile stress-strain curves of the hydrogels with different conditions.

To eliminate the effect of the electronic interaction and hydrogen bonds, we respectively soaked the hydrogels (with 15wt% PSBMA) in 5M NaCl solution (for the electronic interactions) and 5M Urea solution (for hydrogen bonds) for 3h. Then, the representative tensile stress-strain curves are shown in Figure S3. As we can see, the hydrogels treated by 5M NaCl solution present shorter fracture strain (210%) than the as-prepared hydrogels (400%). Besides, the hydrogels treated by 5M Urea solution exhibit much lower ultimate tensile strength (0.034 MPa) than the as-prepared samples (0.228 MPa). These results may explain the effects of the electronic interaction and hydrogen bonds

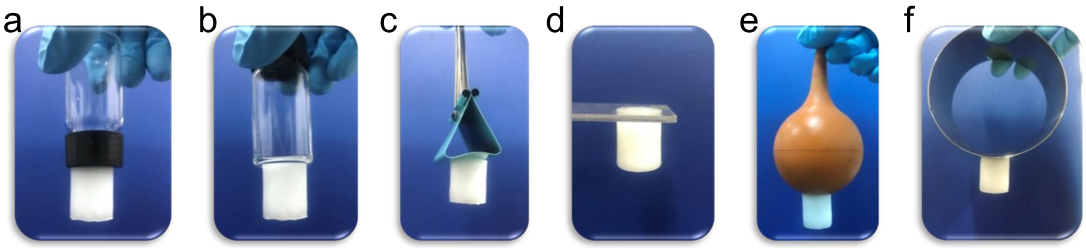

Figure S4. Hydrogel weighing 10 g can adhere to (a) polypropylene, (b) glass, (c) varnish, (d) polyethylene, (e) rubbers and (f) steel freely.

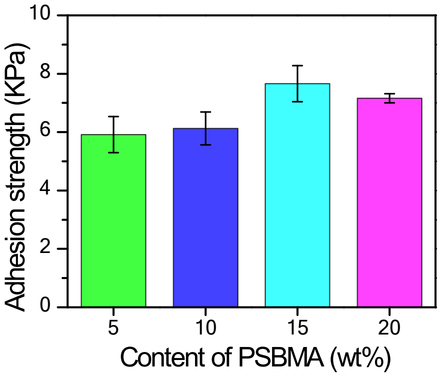

Figure S5. Adhesion strength of hydrogels to rubber with different content of PSBMA.

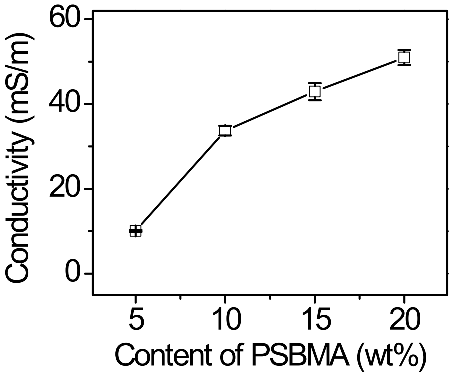

Figure S6. Conductivity of the hydrogels with various contents of PSBMA.

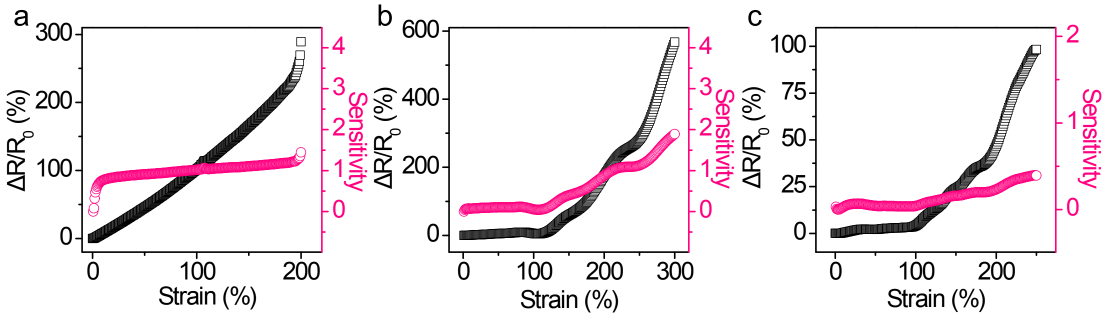

Figure S7. The dependence of resistance change ratio and sensitivity of hydrogels on tensile strain, (a) with 5wt% PSBMA, (b) with 15wt% PSBMA and (c) with 20wt% PSBMA.

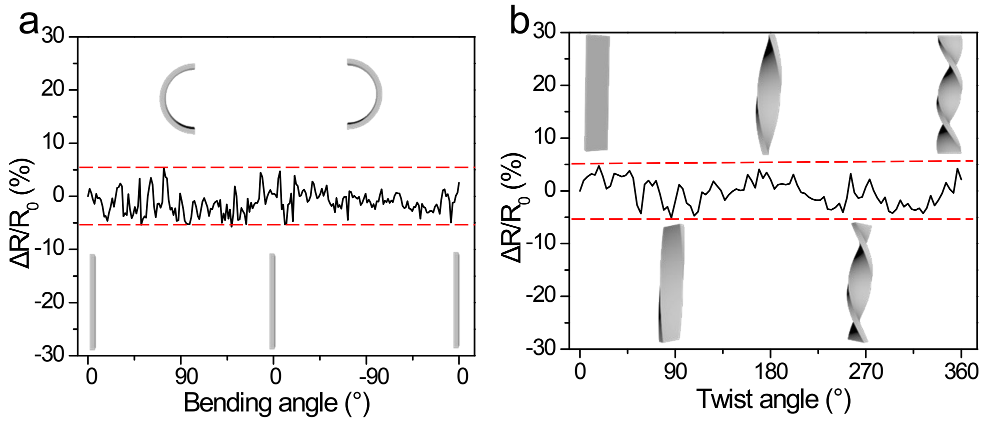

Figure S8. The dependence of resistance change ratio and sensitivity of hydrogels on different conditions. (a) resistance change ratio on bending angle, (b) resistance change ratio on twist angle.

Reference

1. R. Ricciardi, F. Auriemma, C. De Rosa and F. Lauprêtre, *Macromolecules*, 2004, **37**, 1921-1927.