Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Electronic Supplementary Information

Highly stretchable polymer conductors based on as-prepared PEDOT: PSA/n-PAA hydrogels

*Xiangyan Ma‡, Wanping Cai‡, Sai Zhang, Jiahong, Guo, Xing Peng, Zhoutong Qiu, Jie Ying, Jikui Wang**

School of Materials Science and Engineering, East China University of Science and Technology, 130 Meilong Road, Shanghai 200237, PR China

Corresponding Author:

*E-mail: Wang326@ecust.edu.cn

Author Contributions

‡Xiangyan Ma and Wanping Cai contributed equally to this work

Electronic supplementary information includes:

Movie Caption S1, S2

Figures S1 – S7

Supporting Movie Caption

Movie S1 Illustration of a stretchable LED array. Successful operation of LED arrays interconnected with PEDOT: PSA/n-PAA hydrogels between stretching and releasing.

Movie S2 Stretch and release contrast of the PEDOT: PSA/n-PAA hydrogel and the pure PAA hydrogel. By using PEDOT: PSA as a conductive component, the PEDOT: PSA/n-PAA hydrogels present well stretch performance comparing to the pure PAA hydrogel and good environmental stability.

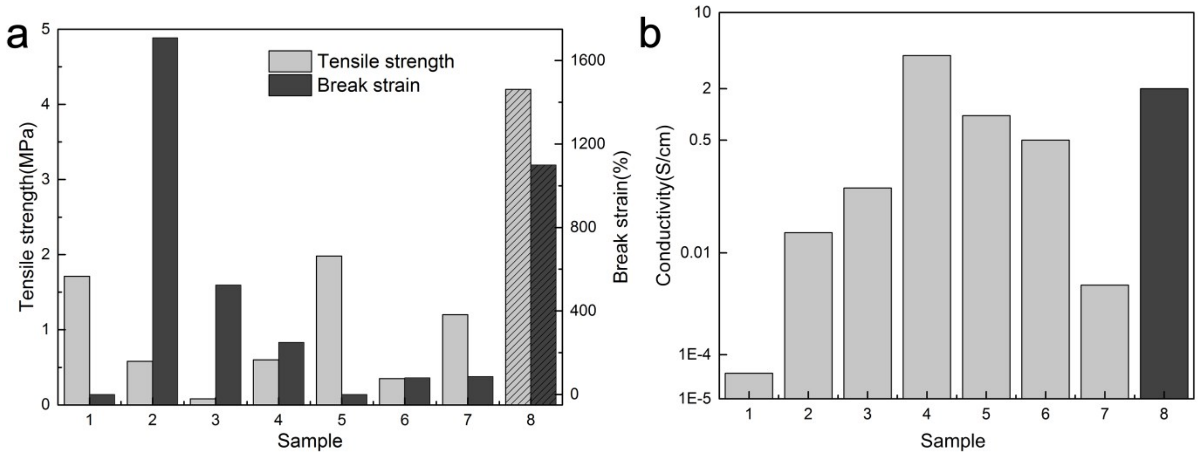

Fig.S1 Comparison of comprehensive properties of conducting hydrogels with other recently reported include 1 PAAm-rGO-Ag-PEDOT/PSS, 2 PNAGA-PAMPS/PEDOT:PSS, 3 PEDOT:PSS-PAAm, 4 PEDOT:PSS/PPEGMA-PAA, 5 PSS-PDMAAm-PEDOT, 6 PPy-RGO, 7 PEDOT:PSS/PAAm, 8 Our work.

Table S1 Comparison of comprehensive properties with other recently reported conducting hydrogels.

sample	Tensile strength/(MPa)	Break strain/%	Electrical conductivity/(S/cm)	Ref.
PAAm-rGO-Ag-PEDOT/PSS	1.71	0.75	3.91×10^{-5}	ref.36
PNAGA-PAMPS/PEDOT:PSS	0.22-0.58	817-1709	0.002-0.022	ref.28
PEDOT:PSS-PAAm	0.047-0.08	350-525	0.11	ref.11
PEDOT:PSS/PPEGMA-PAA	0.6	250	4.3	ref.9
PSS-PDMAAm-PEDOT	1.98	0.76	1	ref.34
PPy-RGO	0.35	80	0.5	ref.37
PEDOT:PSS/PAAm	0.3-1.2	74-86	2.6×10^{-3}	ref.38
PEDOT: PSA/n-PAA	0.5-4.2	400-1100	2	Our work

Fig.S1 and table S1 is the comparison of some properties including tensile strength, break strain and electrical conductivity with recently reported conducting gels, where can clearly demonstrate an improvement over similar state-of-the-art stretchable conducting polymer systems. Most conductive hydrogels are still mechanically weak and low conductivity in the recent reports. In this work, our group prepared the PEDOT: PSA/n-PAA hydrogels which was highly stretchable and mechanically strong (tensile strength 4.2 MPa) via the chemical crosslinking the carboxyl group. In addition, the conductivity is 2.00 S/cm which is largely higher than the conducting hydrogels and it can maintain the conductivity under repeatedly loadunload.

Synthesis of the PSA copolymers:

1 g $Na_2S_2O_8$ (Sig-Aldrich) was dissolved in 400 ml deionized (DI) water, and then sealed preservation after complete dissolution. 5 g *p*-styrene sulfonate sodium (SSNa, Aladdin) and isopropanol (IPA) was added in DI water to completely dissolve with simultaneously stirring and heating at 80 ℃. Then 4 ml $Na_2S_2O_8$ aqueous solution prepared prior and AA (Aladdin) monomers were slowly added dropwise to the above solution for 1h. The copolymerization continued for 6h under the N_2 atmosphere. The products were precipitated by ethanol (Et), and then washed for 3 times with Et. After dried at 70 ℃ in oven, the copolymers P(SSNa-*co*-AA) were pulverized to powder. The powder was then redissolved completely in DI water to generate copolymer solution that was treated by ion exchange resin afterwards. Finally, the solution was dried at 70 ℃ in oven to produce PSA.

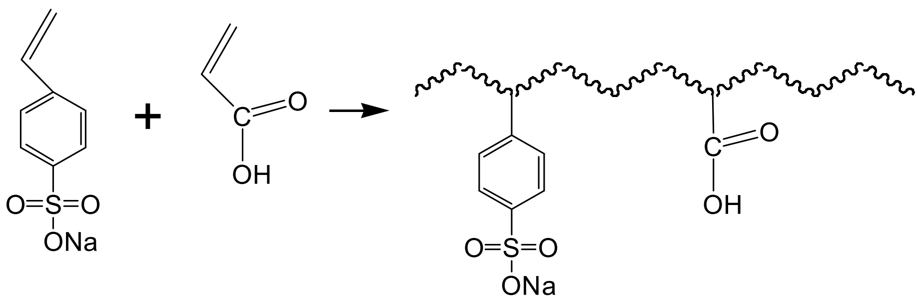

Fig. S2 Fabrication process of PSA copolymers

Microstructure demonstration of PEDOT: PSS/n-PAA hydrogels

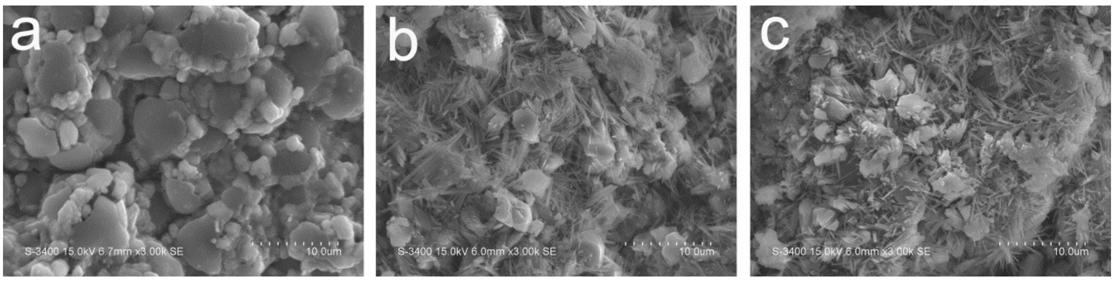

Fig.S3 SEM images of the PEDOT section of the PEDOT: PSS/n-PAA hydrogels include a) 5.84 wt% PEDOT, b) 7.04 wt% PEDOT, c) 8.16 wt% PEDOT, respectively.

The SEM images of PEDOT: PSS/n-PAA hydrogels are shown in fig.S2. The PAA chains could crosslink better at lower PEDOT: PSS weight percent. However, most PAA chains could not crosslink due to the stiffness of PEDOT: PSS with increasing the weight ratio of PEDOT: PSS. Hence, the introduction of carboxyl group of PSA can solve the problem.

Mechanical properties of PEDOT: PSS/n-PAA hydrogels

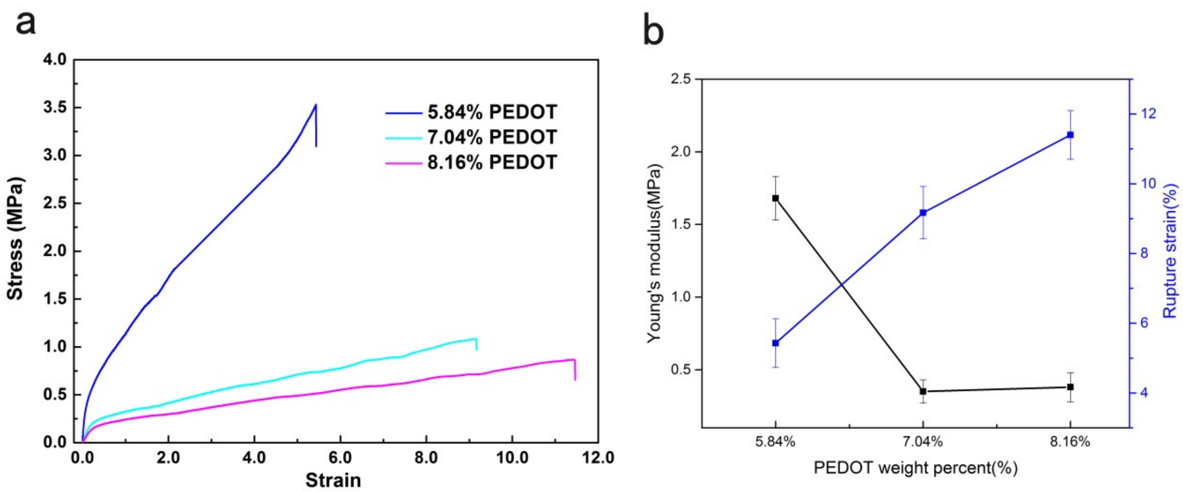

Fig.S4 Stretch-insensitive behavior of resistance for PEDOT: PSS/n-PAA hydrogels. a) Stress-strain curves for the PEDOT: PSS/n-PAA hydrogels for different weight ratios of PEDOT: PSS. b) The Young’s modulus and rupture strain of the hydrogels for various weight ratios of PEDOT: PSS.

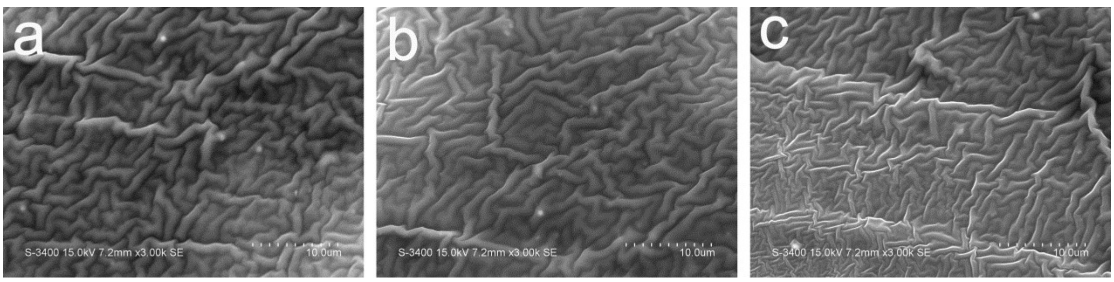

Fig.S5 SEM images of PEDOT: PSA/n-PAA hydrogels after tensile cycles. (a) 1th. (b) 500th. (c)

1000th.

Demonstration of PEDOT: PSA/n-PAA hydrogels for interconnects

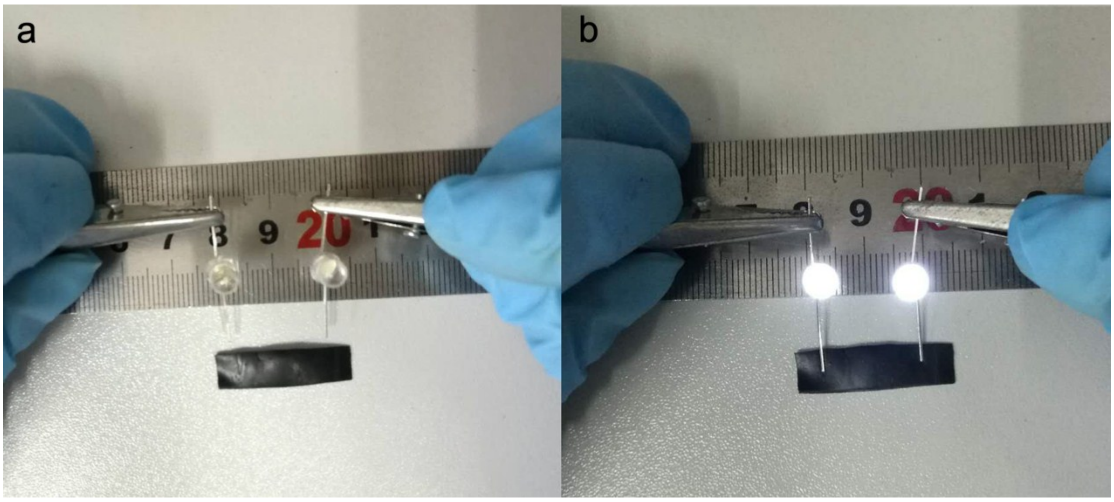

Fig.S6 Optical images of the PEDOT: PSA/n-PAA hydrogels

Comparison of PEDOT: PSA (S)/n-PAA hydrogels for environmental stability

Fig. S7 Photographs of the PEDOT: PSS/n-PAA hydrogel and PEDOT: PSA/n-PAA hydrogel

before and after being dried in air for 1 d.