Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2023

Supporting Information

A low-hysteresis, self-adhesive and conductive PAA / PEDOT: PSS

hydrogel enabled body-conformable electronics

Qiang Gao1*, Chao Li1, Mingxu Wang1, Jiadeng Zhu2, Chunxia Gao1

¹School of Chemistry and Chemical Engineering, Yangzhou University, Yangzhou,

225002, China

²Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831,

USA

Corresponding author: *Dr. Qiang Gao (gaoqiang@yzu.edu.cn)*



Fig. S1 The multilevel structure of PEDOT: PSS.



Fig. S2 SEM images and pore size of PAA and PPH with different x values after freezedrying. (a) pure PAA, (b) x=1: 1, (c) x=1: 3, (d) x=1: 5, (e) stretch the hydrogel (x=1:3) after 300%, (f) stretch the hydrogel (x=1:3) after fracture (elongation is about 600%).

It can be seen from the SEM images that the pore size of the hydrogel presents a trend of decreasing with the increase of PEDOT: PSS content. This may be due to PEDOT: PSS increasing the degree of crosslinking of hydrogels through providing hydrogen bonds. When the hydrogel is strained, the internal network tends to order along the stress direction. However, when the stress is high enough, the internal network of the hydrogel breaks.



Fig. S3 ATR-FTIR spectra of PPHs.



Fig. S4 Rheological test of hydrogels. (a) Forming temperature test of PAA hydrogels and (b) polymerization tracking of PAA hydrogels at the molding temperature of 65°C. (c) Forming temperature test of PPH and (d) polymerization tracking of PPH at the molding temperature of 77°C.

The rheological behavior of the forming hydrogel is monitored as a function of temperature or time, following the shear storage modulus G' and the loss modulus G". The storage modulus G' characterizes the elastic and the loss modulus G" the viscous part of the viscoelastic behavior. The values of G' represent the stored energy, while G" stands for the deformation energy lost by internal friction during shearing. Until the gelation point G" is larger than G'. This crossing point of G' and G" represents the transition from liquid-like behavior to solid-like behavior and is taken as the indication of the gelation temperature or time of the hydrogel. Due to the hydrogen bond between PEDOT: PSS and PAA, the addition of PEDOT: PSS increases both G', G" and the viscosity of the hydrogel precursors. The test gelation time of the hydrogel containing PEDOT: PSS does not appear to intersect because the machine has an error of 90 seconds, within which time it has already gelated.



Fig. S5 Dynamic mechanical analysis of (a) PAA hydrogels and (b) PPH (x=1:3).

Higher storage modulus and loss modulus commonly means better elasticity and stronger viscidity. Obviously, when more PEDOT: PSS was added to the mixed system, both the storage modulus and loss modulus of the sample increase. This is because the addition of PEDOT: PSS forms more physical crosslinking points, which increases the overall crosslinking degree of the hydrogel system and thus increases the elasticity, that is, the storage modulus increases. Moreover, the increase of intermolecular force leads to the rise of loss modulus. Also, when the temperature increases, dehydration at high temperatures improves the rigidity and modulus of the sample, indicating an increasing modulus.



Fig. S6 Energy loss coefficient of different hydrogel samples



Fig. S7 The model is used to test the adhesion strength.



Fig. S8 The swelling degree of hydrogels in deionized water over time.



Fig. S9 The optical photos of volume change of pure PAA hydrogels and PPH (x=1: 3, x=1: 5) in DI water for 0-5 h.



Fig. S10 The conductivity of different samples

Tab. S1 Comparison of the main parameters of PPH-based strain sensors in this workand previous mainstream studies.

Component	Mechanical property		Strain Sensor		Ref.
	Tensile strain	Tensile	Detection	Gauge	
	(%)	stress (MPa)	strain	Factor	
			range (%)	(GF)	
MXene/PAA/ACC	450%	300 kPa	0.3-30%	1.51	[1]
			30-450%	10.79	
PAA/PAM/MXene/T	560.82±19.56%	251±50 kPa	0-75%	2.401	[2]
А			75-150%	4.769	
			150-250%	10.536	
CNS/PAA/Fe ³⁺	1800%	104 kPa	0-600%	4.37	[3]
TOCNF/PANI/PAA	982%	74.98kPa	300%	8	[4]
PAA/rGO	600%	400kPa	100%	0.31	[5]
			500%	1.32	
TOCNF-GN/PAA	850%	32kPa	350%	5.8	[6]
PAA/PANI	1160%	0.3MPa	0-800%	0.6	[7]
			800-	1.05	
			1130%		
PAA-PDA-Fe ³⁺	780.75%	22.78 kPa	0-250%	1.565	[8]
			250-600%	4.292	
PAANa/PP/PVA	380%	1.97MPa	0-200%	0.57	[9]
rGO/CMCNa/PAA	1200%	66.9kPa	0-300%	1	[10]
PAA/PEDOT:PSS	600%	21.48 kPa	0-250%	1.52	This
			250-400%	4.33	work
			400-600%	7.73	

Reference

- X. Li, L. He, Y. Li, M. Chao, M. Li, P. Wan, L. Zhang, Healable, Degradable, and Conductive MXene Nanocomposite Hydrogel for Multifunctional Epidermal Sensors, ACS Nano. 15 (2021) 7765–7773.
- [2] M. Qin, W. Yuan, X. Zhang, Y. Cheng, M. Xu, Y. Wei, W. Chen, D. Huang, Preparation of PAA/PAM/MXene/TA hydrogel with antioxidant, healable ability as strain sensor, Colloids and Surfaces B: Biointerfaces. 214 (2022) 112482.
- [3] F. Lu, Y. Wang, C. Wang, S. Kuga, Y. Huang, M. Wu, Two-Dimensional Nanocellulose-Enhanced High-Strength, Self-Adhesive, and Strain-Sensitive Poly(acrylic acid) Hydrogels Fabricated by a Radical-Induced Strategy for a Skin Sensor, ACS Sustainable Chem. Eng. 8 (2020) 3427–3436.
- [4] Y. Jiao, Y. Lu, K. Lu, Y. Yue, X. Xu, H. Xiao, J. Li, J. Han, Highly stretchable and self-healing cellulose nanofiber-mediated conductive hydrogel towards strain sensing application, Journal of Colloid and Interface Science. 597 (2021) 171– 181.
- [5] X. Jing, H.-Y. Mi, X.-F. Peng, L.-S. Turng, Biocompatible, self-healing, highly stretchable polyacrylic acid/reduced graphene oxide nanocomposite hydrogel sensors via mussel-inspired chemistry, Carbon. 136 (2018) 63–72.
- [6] C. Zheng, K. Lu, Y. Lu, S. Zhu, Y. Yue, X. Xu, C. Mei, H. Xiao, Q. Wu, J. Han, A stretchable, self-healing conductive hydrogels based on nanocellulose supported graphene towards wearable monitoring of human motion, Carbohydrate Polymers. 250 (2020) 116905.

- [7] R.M. Pallares, X. Su, S.H. Lim, N.T.K. Thanh, Fine-tuning of gold nanorod dimensions and plasmonic properties using the Hofmeister effects, J. Mater. Chem. C. 4 (2016) 53–61.
- [8] H. Zhou, S. Li, H. Liu, B. Zheng, X. Jin, A. Ma, W. Chen, High-Performance Flexible Sensors of Self-Healing, Reversibly Adhesive, and Stretchable Hydrogels for Monitoring Large and Subtle Strains, Macromol. Mater. Eng. 305 (2020) 1900621.
- [9] J.-Y. Gong, F.-C. Sun, Y.-C. Pan, A.-M. Fei, S.-F. Leicheng, F.-P. Du, Y.-F. Zhang, Stretchable and tough PAANa/PEDOT: PSS/PVA conductive hydrogels for flexible strain sensors, Materials Today Communications. 33 (2022) 104324.
- [10] L. Wu, Y. Hu, P. Tang, H. Wang, Y. Bin, High stretchable, pH-sensitive and selfadhesive rGO/CMCNa/PAA composite conductive hydrogel with good strainsensing performance, Composites Communications. 24 (2021) 100669.

Supplementary Videos

Video S1 shows in the circuit with PPH as the conductor, the brightness of the LED gradually decreased with increasing strain from 0 to 300%. (MP4)

Video S2 shows PPH-based sensor can control the movement of the manipulator by the movement of the human hand. (MP4)

Video S3 shows that PPH-based sensors can replace commercial electrodes for stable transmission of ECG signals. (MP4)

Video S4 shows that PPH-based TENG can convert machines into electricity and light LED bulbs. (MP4)