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Supporting Information

A highly stable elastic electrode via direct covalent crosslinking for strain sensor

Linping Wang,^a Ben-Lin Hu,^{*a}, Fengyuan Zhang,^a Yinxia Zhang,^{ab} Junming Li,^{ab} Tianhua Xu ^{ac} and Run-Wei Li ^{*a}

^a CAS Key Laboratory of Magnetic Materials and Devices, and Zhejiang Province Key Laboratory of Magnetic Materials and Application Technology, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China

^b Nano Science and Technology Institute, University of Science and Technology of China, Suzhou 215123, China

^c College of Materials Science and Opto-Electronic Technology, University of Chinese Academy of Sciences, Beijing 100049, China

*Corresponding author. E-mail address: <u>hubenlin@nimte.ac.cn</u>; <u>runweili@nimte.ac.cn</u>;



Figure S1. a) Wide-scan XPS spectra and b) high-resolution S 2p XPS spectra of crosslinked PEDOT:PSS and pristine PEDOT:PSS



Figure S2. ATR-FTIR spectra of PSS, epoxydodecane, and their product.



Figure S3. a) Strain-stress curves and b) tensile strain at break of bulk crosslinked PEDOT:PSS films using PEGDE crosslinkers with different molecular weights and heated at different temperatures. Sample "PEGDE500-120 °C" represents crosslinked PEDOT:PSS with PEGDE500 reacted at 120 °C.



Figure S4. Strain-stress curves of bulk crosslinked PEDOT:PSS thick films heated at various temperatures from 60 °C to 180 °C. Tests of each sample were conducted 3 times. Sample "180 °C-1" represents the crosslinked PEDOT:PSS heated at 180 °C for the first strain-stress test. The ratio of PEGDE 6k was 50 wt% for all samples.



Figure S5. Strain-stress curves of bulk crosslinked PEDOT:PSS thick films mixed with different weight ratio of PEGDE. Tests of each sample were conducted 3 times. Sample "30%-2", for example, represents the crosslinked PEDOT:PSS with PEGDE ratio of 30 wt% for the second strain-stress test. All the films were crosslinked at 120 °C.



Figure S6. Optical microscope images of crosslinked PEDOT:PSS with the PEGDE ratio at a) 50 wt% and b) 70 wt%.



Figure S7. a) AFM height images and b) phase images of crosslinked PEDOT:PSS.



Figure S8. a) AFM height images and b) phase images of pristine PEDOT:PSS.

Mixed materials	Conductivity (S m ⁻¹)	Break	Modulus (MPa)	R/R0 at 50% strain	R/R0 at 100% strain	Ref.	
	60800	133%	55	/	1	Sci. Adv. 2017, 3, e1602076	
Ionic liquid+DMSO	42000	112%	28	/	1		
	28800	176%	47	/	1		
Triton X-100	7800	57%	/	1.6	/	Adv. Mater. 2016, 28, 4455	
Zonyl	/	/	/	2	18	Chem. Mater. 2012, 24, 373	
Sorbitol	67100	67%	1.9	2	12	ACS Appl. Mater. Interfaces 2019, 11, 26185	
WPU+Sorbitol	~40000	~45%	~60	1.15 at 30% strain		Nat. Commun 2020, 11, 4683	
PVA+DMSO	<5000	3.50%	/	~10	~150		
PAA+DMSO	12500	45%	/	~5	25	J. Mater. Chem. C, 2020, 8, 6013	
PMAA+DMSO	<5000	3%	/	>200	>200		
PEO1000K	5260	38%	/	1	/	ACS Appl. Mater. Interfaces 2015, 7, 18415	
PVA89k+DMSO	14200	55%	/	1	/		
PR-PEGMA	11000	150%	~800	1.5	2.5	Science 2022, 375, 1411	
PEGM+PEGDM	55000	13%	<10	1.1	30	Adv. Funct. Mater. 2020, 2001251	
PEGDM+FS30	~300000	1	/	1	/	Science 2021, 373, 88	
PEGDE	13100	50%	251	2.2	9.5	This work	

Table S1. Mechanical and electrical properties of crosslinked PEDOT:PSS in this work

 compared to representative stretchable PEDOT:PSS conductors reported in literature.



Figure S9. Relative changes in resistance of bulk PEDOT:PSS film under cyclic strains of 30% versus cycle time.



Figure S10. Relative changes in resistance of crosslinked PEDOT:PSS thin films under cyclic strains of 50% versus cycle times.

Table S2. Key parameters of C/C₀ linear fitting curve

Linear fitting curve	at 10% strain	at 20% strain	at 30% strain	at 40% strain	at 50% strain
Intercept	0.991	0.989	0.995	0.996	1.001
Slope	0.717	0.720	0.740	0.749	0.723
R square	0.993	0.992	0.999	0.997	0.999



Figure S11. Relative change in capacitance of sensors under cyclic stretching of 30% and 50% versus cycle times.