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## **Electronic Supplementary Information (ESI) For**

# Conductive Hydrogel-Based Flexible Strain Sensor with Superior Chemical Stability and Stretchability for Mechanical Sensing in Corrosive Solvents

Hong Zhang<sup>a</sup>, Mingqiang Yue<sup>a</sup>, Tingting Wang<sup>a, b, c</sup>, Jinqing Wang<sup>b, c\*</sup>, Xianzhang Wu

<sup>b, c</sup>, and Shengrong Yang<sup>b, c\*</sup>

<sup>a</sup> College of Chemical Engineering, Northwest Minzu University, Key Laboratory for

Utility of Environmental-Friendly Composite Materials and Biomass in University of

Gansu Province, Lanzhou 730030, China.

<sup>b</sup> State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, China.

<sup>c</sup> Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China.

\* Corresponding authors,

jqwang@licp.cas.cn (J. Q. Wang);sryang@licp.cas.cn (S. R. Yang)

Fax: 0086-931-4968019

Tel: 0086-931-4968076

### **Preparation of GO**

First, 4.2 g potassium persulfate ( $K_2S_2O_8$ ), 4.2 g  $P_2O_5$ , 5 g graphite powder were added into 20 mL H<sub>2</sub>SO<sub>4</sub> in batches, and the mixed solution was stirred at 80 °C for 5h. After cooling to room temperature, 0.25 L of ultrapure water was slowly added to the mixed solution and allowed to stand overnight. The precipitate was washed several times and suction filtered to remove excess acid and dried at 60 °C. For oxidization, the pretreated graphite powder (5 g) was slowly added to concentrated H<sub>2</sub>SO<sub>4</sub> (250 mL) in an icewater bath state and thoroughly stirred. 30 g of KMnO<sub>4</sub> was slowly added under stirring; then the temperature was raised to 35 °C and stirred for 2 h. Then, 400 mL of ultrapure water was slowly added dropwise for dilution and stirred for 2 h. The mixture was further diluted with ultrapure water (1.2 L), and H<sub>2</sub>O<sub>2</sub> (30 mL, 30%) was added dropwise until the solution turned bright yellow; stirring was continued, and the solution was allowed to stand overnight to precipitate. The precipitate was washed neutral with hydrochloric acid and washed several times with deionized water until it could not be centrifuged. The precipitate was washed with hydrochloric acid to neutrality and washed several times with deionized water until it could not be centrifuged; and the resulting product was dialyzed against ultrapure water for two weeks. Finally, the GO suspension was obtained by the treatment of ultrasonication.

NIPA M (g)	PEDOT:PSS (PH1000) (13 mg/mL) (mL) (V <sub>P</sub> /V <sub>H</sub> %)	GO (5 mg/mL) (mL)	H <sub>2</sub> O (mL)	BIS (mg)	KPS (g)	TEMED (μL)
8.475	0.00 (0.0 %)	20	30	1.155	0.135	100
8.475	0.05 (1.7 %)	20	23	1.155	0.135	100
8.475	0.15 (5.1 %)	20	23	1.155	0.135	100
8.475	0.50 (8.5 %)	20	25	1.155	0.135	100
8.475	0.70 (11.9 %)	20	23	1.155	0.135	100

Table S1 Composition formulations of hydrogels

Note:  $V_P$ : volume of PEDOT:PSS solution;  $V_H$ : volume of prepared hydrogel.  $V_P/V_H$  is the volume ratio of the PEDOT:PSS to the prepared hydrogel.

### Characterizations

The conductivity of the hydrogel was tested using a Keithley 2450 digital source meter (2450, Tektronix, USA). A field emission scanning electron microscope (SEM, JSM-6701F, Japan Electronics Co., Ltd.) is used to observe the internal network structure of the hydrogel, and a thin layer of gold is sprayed on the surface to obtain a clearer picture. Fourier transform infrared spectroscopy (FTIR, IFS 66 v/s, Germany), Raman spectroscopy (Raman, LabRAM HR Evolution, France), X-ray powder diffraction (XRD, EMPYREAN, Netherlands) characterizations were performed for the structure and composition analysis of hydrogels.

### **Conductivity Test**

A Keithley 2450 digital source meter was used to test the resistance of the hydrogel cylinders with a height of 20 mm and a diameter of 6 mm. The water content of all hydrogels used for the test was 78%. The conductivity of hydrogel was calculated using the following formula:

$$\sigma = \frac{L}{S} \cdot \frac{1}{R}$$

where  $\sigma$ , L, S, and R represent the conductivity, length, cross-sectional area, and resistance of the hydrogel, respectively. All tests were repeated three times at room temperature, and G(8.5%)PP-h was selected as the test sample. The measured conductivity was 0.084 S/m.



Figure S1. TEM image of GO nanosheet.



**Figure S2.** GO/PEDOT:PSS/PNIPAM hydrogel without chemical crosslinker of BIS is easily damaged in 0.4 wt% HF solution at the temperature of 25 °C.



Figure S3. The content influence of PEDOT:PSS on the conductivity of hydrogels.



**Figure S4.** Compressive stress-strain curves and column line charts of the GPP-h with different contents of PEDOT:PSS.



Figure S5. GPP-h can easily adhere to a variety of surfaces including glass, fiber, carton, plastic, steel, and PTFE.



**Figure S6.** Time evolution of the healable process for the GPP-h determined by the real-time current measurement.



**Figure S7.** The resistance change rate of the hydrogel after being immersed in ethanol, alkali solution with pH of 14 and DMSO solution for 2 days.

Materials	Elongation at break (%)	Sensing Stability (cycles)	Sensing Ranges (%)	Conductivity (S m <sup>-1</sup> )	Adhesion	Ref.
GO/PEDOT:PSS/PNIPAM	2512	4000 (30%-S)	≥ 200	0.084	YES	This work
PNIPAAm/PANI	290	350 ( N/G)	≤ 120	0.068	YES	[1]
TA@CNC	800	≈250 (20%-S)	≤ 100	N/G	YES	[2]
MXene/PVA/PAAm	$\approx 1600$	N/G	200-350	N/G	NO	[3]
Poly(NIPAM-co-β-CD)/CNT/PPY	515	N/G	N/G	34.93	NO	[4]
MXene/HAPAM/PNIPAM	1400	N/G	N/G	1.092	NO	[5]
CNT/Laponite/PNIPAM	1062	N/G	N/G	0.200	YES	[6]
GO/PNIPAM	2800	N/G	N/G	NO	NO	[7]
PAAm/PEDOT:PSS	525	1000 ( 50%-S)	50	1.000	NO	[8]
GO/PDA/PAM	3500%	N/G	N/G	0.001	YES	[9]
PVA/PSBMA	400%	N/G	300	0.01	YES	[10]
PANI/P(AAm-co-HEMA)	530%	N/G	N/G	8.14	NO	[11]

Table S2 Comparison of the conductive hydrogels-based artificial electronic skins

Note: 'S' indicates 'strain' in the Table. 'N/G' indicates 'not given' in the references.

#### Reference

- [1] Z. Wang, H. Zhou, W. Chen, Q. Li, B. Yan, X. Jin, A. Ma, H. Liu, W. Zhao, ACS Appl. Mater. Interfaces, 2018, 10, 14045.
- [2] C. Cui, C. Shao, L. Meng, J. Yang, ACS Appl. Mater. Interfaces, 2019, 11, 39228.
- [3] H. Liao, X. Guo, P. Wan, G. Yu, Adv. Funct. Mater., 2019, 29, 1904507.
- [4] Z. Deng, Y. Guo, X. Zhao, P.X. Ma, B. Guo, Chem. Mater., 2018, 30, 1729.
- [5] Y. Zhang, K. Chen, Y. Li, J. Lan, B. Yan, L. Shi, R. Ran, ACS Appl. Mater. Interfaces, 2019, 11, 47350.
- [6] Z. Deng, T. Hu, Q. Lei, J. He, P.X. Ma, B. Guo, ACS Appl. Mater. Interfaces, 2019, 11, 6796.
- [7] K. Shi, Z. Liu, Y.Y. Wei, W. Wang, X.J. Ju, R. Xie, L.Y. Chu, ACS Appl. Mater. Interfaces, 2015, 7, 27289.
- [8] Y.Y. Lee, H.Y. Kang, S.H. Gwon, G.M. Choi, S.M. Lim, J.Y. Sun, Y.C. Joo, Adv. Mater., 2016, 28, 1636.
- [9] L. Han, X. Lu, M. Wang, D. Gan, W. Deng, K. Wang, L. Fang, K.Liu, C. W. Chan, Y. Tang, L.T. Weng, and H. Yuan, Small, 2017, 13, 1601916.

[10] Z. Wang, J. Chen, L. Wang, G. Gao, Y. Zhou, R. Wang, T. Xu, J. Yin, J. Fu, J. Mater. Chem. B, 2019, 7, 24-29.

[11] Z. Wang, J. Chen, Y. Cong, H. Zhang, T. Xu, L. Nie, Jun Fu, Chem. Mater. 2018, **30**, 8062–8069.