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## Supporting Information

## Facile Fabrication of a Highly-Conductive Hydrogel through Filling with Polyiodide

Ji-Hua Zhu, Hongtao Yu\*, and Hong Meng

S1: the polyiodide solution was fabricated as follows: 0.1 mL HI (57%) was added to 5 mL  $H_2O$ , then keep under daylight until the color of the solution remain unchanged. Finally, a brown solution was obtained.



HI aqueous solution

Polyiodide solution

Figure S1. Digital photograph of the HI (0.15 M) and polyiodide aqueous solutions.



**Figure S2.** Digital photographs of the PAM hydrogel (7.57 g) and its dehydrated (0.89 g) form can be switched under several turns of dehydrate and hydrate. Even if the hydrogel is dehydrated, it can be rehydrated quickly.



Figure S3. (a) FT-IR spectra and (b) TGA diagrams of PAM and PAMPI, respectively.



Figure S4. EIS plots of (a) PAMPI (Inset is the local enlargement at the frequency region 1000~0.01 Hz and the corresponding equivalent circuit), and (b) PAM/KI hydrogel.



**Figure S5.** The relationship between the conductivity and the exposure time of PAMPI in the air.



Figure S6. Stable cyclic voltammogram curves of the PAM hydrogel wires filled with or without electrolytes in a two-electrode configuration from -3 V $\sim$  3 V (scan rate: 100 mV/s).



**Figure S7.** Digital photograph of the hydrogel wire device used to transmit current to illuminated diode bulbs.

Movie S1. The PAMPI hydrogel wire was used to transmit current to light the diode bulb.

**Movie S2.** The PAM/K<sub>3</sub>Fe(CN)<sub>6</sub> hydrogel wire was used to transmit current to light the diode bulb.

Movie S3. The PAM/KI hydrogel wire was used to transmit current to light the diode bulb.

Gels	Conductive factors	Max conductivity (S cm <sup>-1</sup> )	Ref.
PAAM-PVP	LiCl	0.02	[1]
PACMO/PC	LiTFSI	0.00079	[2]
PNIPAM-clay- PDAEA	Clay	0.002	[3]
PVA/TTSBI	Fe <sup>3+</sup>	0.00216	[4]
PNIPAM	MXene	0.01092	[5]
PVA-PDA	GO	0.0027	[6]
PAM-co-PAA	CNTs	0.082	[7]
PDA-PPy-PAM	PPY	0.12	[8]
P(AAm-co-HEMA)	PANI	0.0824	[9]
Gelator	Polyiodide	0.011	[10]
PAM	Polyiodide	0.15	This work

Table S1. Comparison of the Specific Capacitance of Micro-sized Carbon Fiber-Based Electrodes

## **Reference:**

[1] Zhang H, Niu W, Zhang S. Extremely stretchable, sticky and conductive double-network ionic hydrogel for ultra-stretchable and compressible supercapacitors. Chemical Engineering Journal, 2020, 387: 124105.

[2] Gao Y, Shi L, Lu S, et al. Highly stretchable organogel ionic conductors with extremetemperature tolerance. Chemistry of Materials, 2019, 31(9): 3257-3264.

[3] Di X, Hang C, Xu Y, et al. Bioinspired tough, conductive hydrogels with thermally reversible adhesiveness based on nanoclay confined NIPAM polymerization and a dopamine modified polypeptide. Materials Chemistry Frontiers, 2020, 4(1): 189-196.

[4] Zhang X, Cai J, Liu W, et al. Synthesis of strong and highly stretchable, electrically conductive hydrogel with multiple stimuli responsive shape memory behavior. Polymer, 2020, 188: 122147.

[5] Zhang Y, Chen K X, Li Y, et al. High-strength, self-healable, temperature-sensitive, MXene-containing composite hydrogel as a smart compression sensor. ACS applied materials & interfaces, 2019, 11(50): 47350-47357.

[6] Wang M, Chen Y, Khan R, et al. A fast self-healing and conductive nanocomposite hydrogel as soft strain sensor. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 2019, 567: 139-149.

[7] Han L, Liu K, Wang M, et al. Mussel-inspired adhesive and conductive hydrogel with long-lasting moisture and extreme temperature tolerance. Advanced Functional Materials, 2018, 28(3): 1704195.

[8] Han L, Yan L, Wang M, et al. Transparent, adhesive, and conductive hydrogel for soft bioelectronics based on light-transmitting polydopamine-doped polypyrrole nanofibrils. Chemistry of Materials, 2018, 30(16): 5561-5572.

[9] Wang Z, Chen J, Cong Y, et al. Ultrastretchable strain sensors and arrays with high sensitivity and linearity based on super tough conductive hydrogels. Chemistry of Materials, 2018, 30(21): 8062-8069.

[10] Kubo W, Murakoshi K, Kitamura T, et al. Quasi-solid-state dye-sensitized  $TiO_2$  solar cells: effective charge transport in mesoporous space filled with gel electrolytes containing iodide and iodine. The Journal of Physical Chemistry B, 2001, 105(51): 12809-12815.