

## Supporting Information

### **Ultrahigh conductivity and antifreezing zwitterionic sulfobetaine hydrogel electrolyte for low-temperature resistance flexible supercapacitors**

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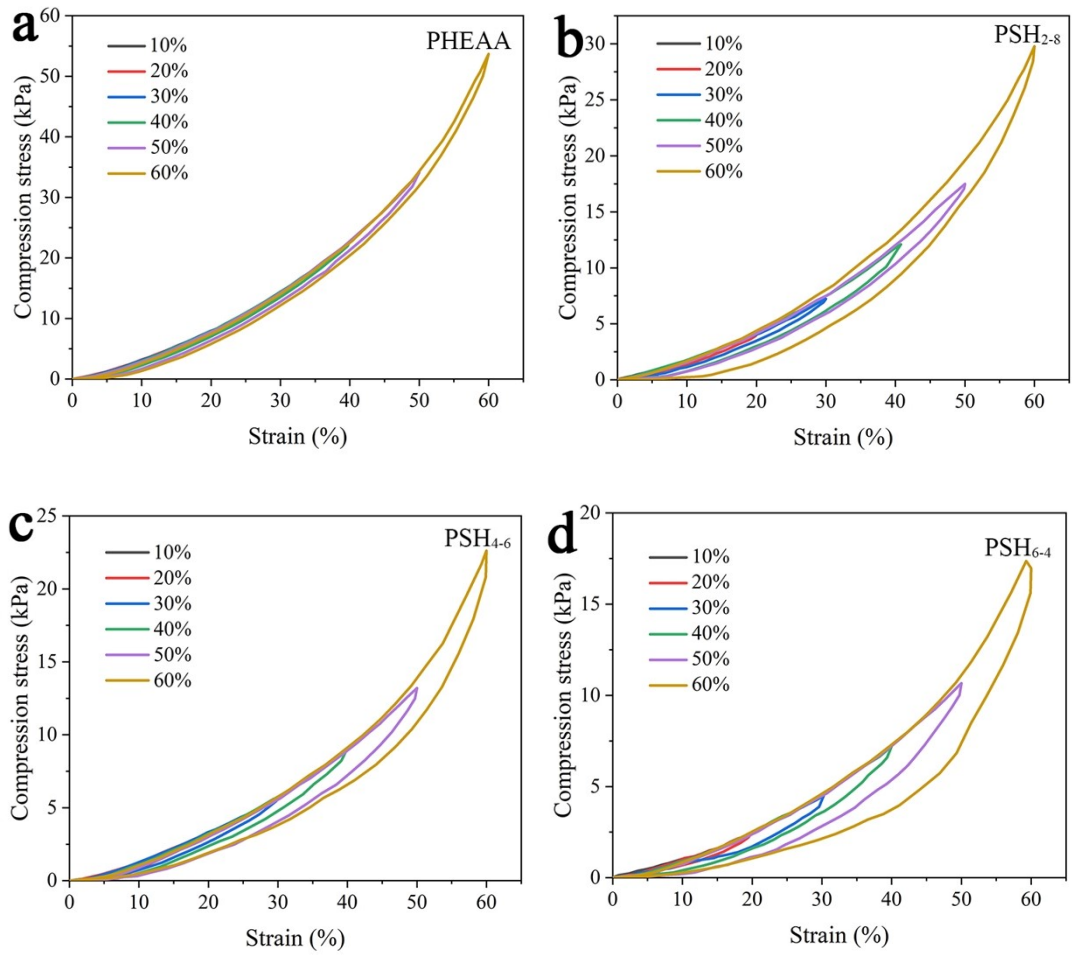


**before reaction**

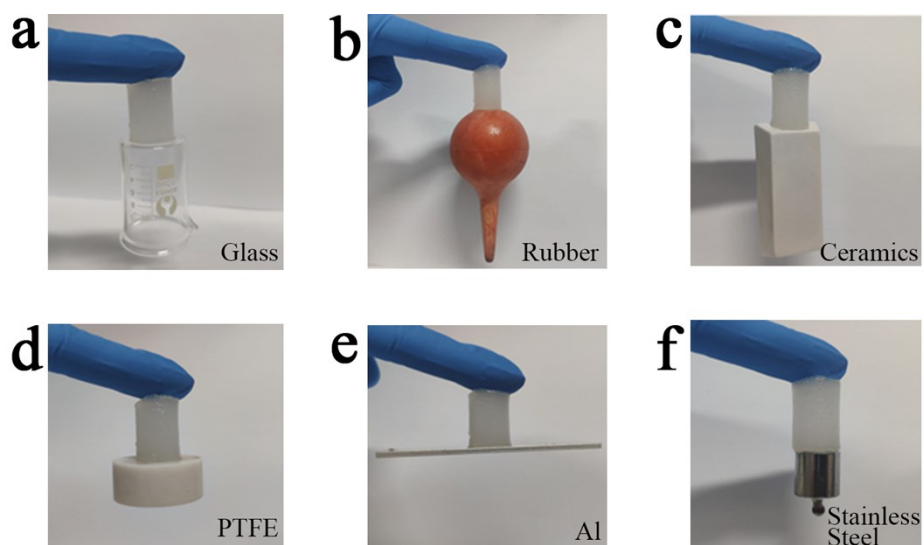


**after reaction**

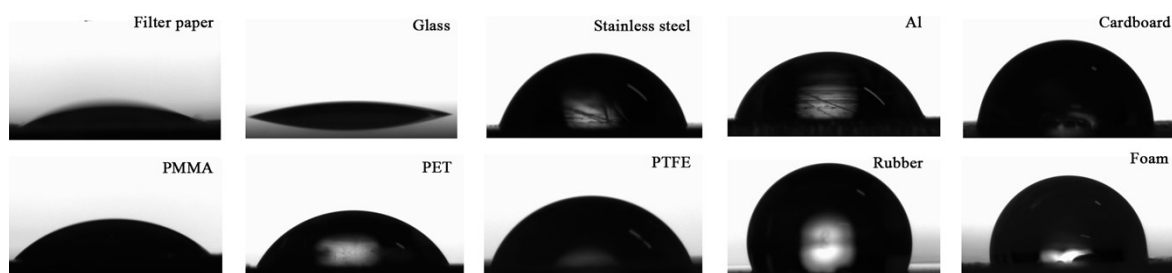
**Fig S1.** Images left to right showing the hydrogels before and after the reaction (PHEAA, PSH<sub>2-8</sub>, PSH<sub>4-6</sub>, PSH<sub>6-4</sub>, PSH<sub>8-2</sub> and PSBMA)



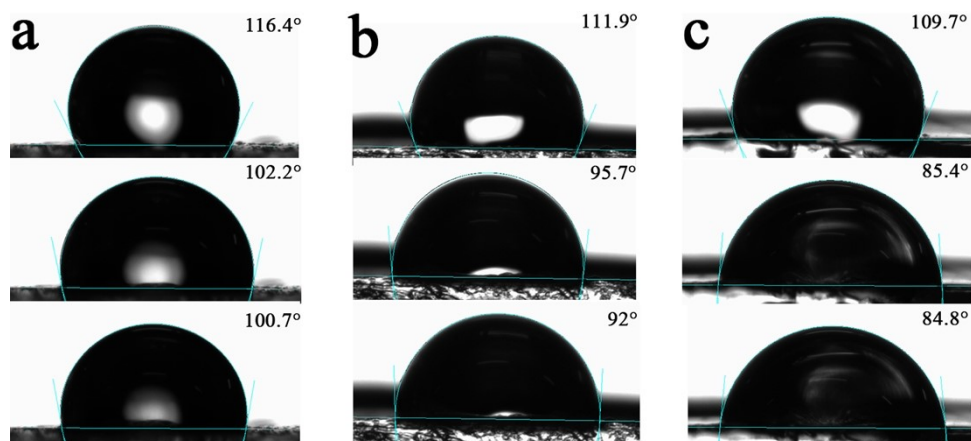
**Fig S2.** Compressive stress-strain curves of (a) PHEAA, (b) PSH<sub>2-8</sub>, (c) PSH<sub>4-6</sub>, (d) PSH<sub>6-4</sub>.



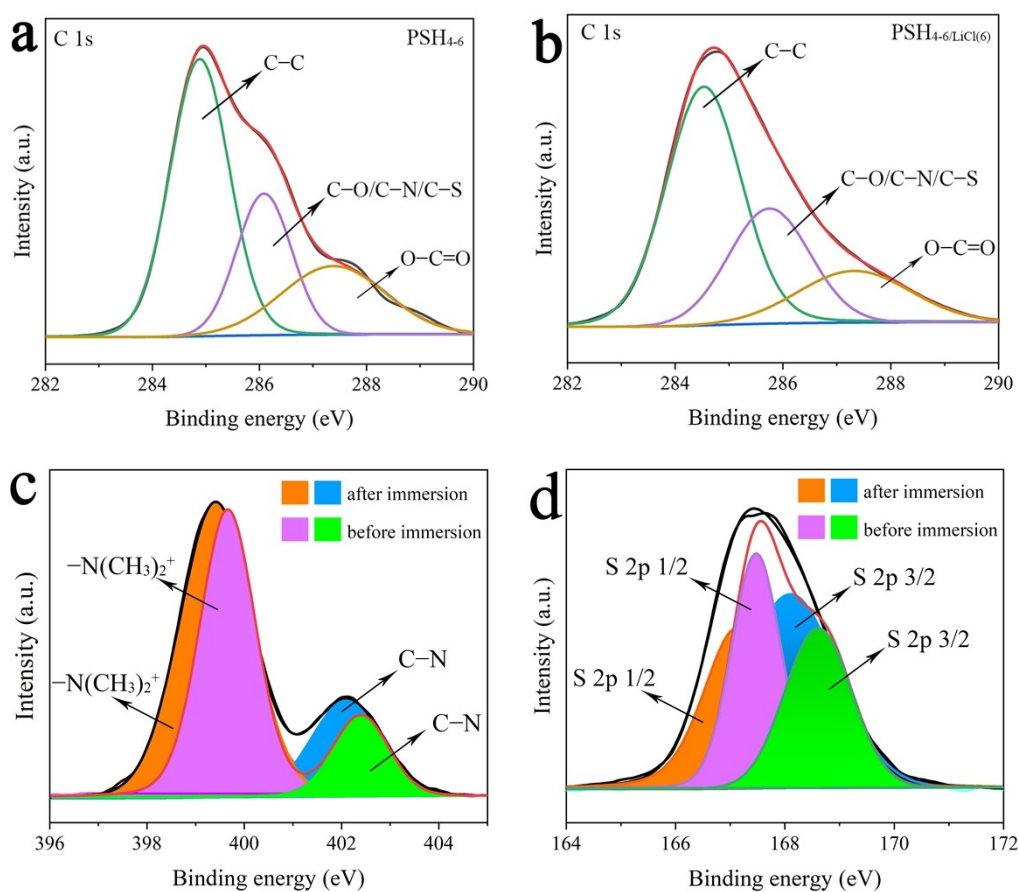
**Fig S3.** Hydrogels adhered to various materials (a) glass, (b) rubber, (c) ceramics, (d) PTFE, (e) Al, (f) stainless steel.



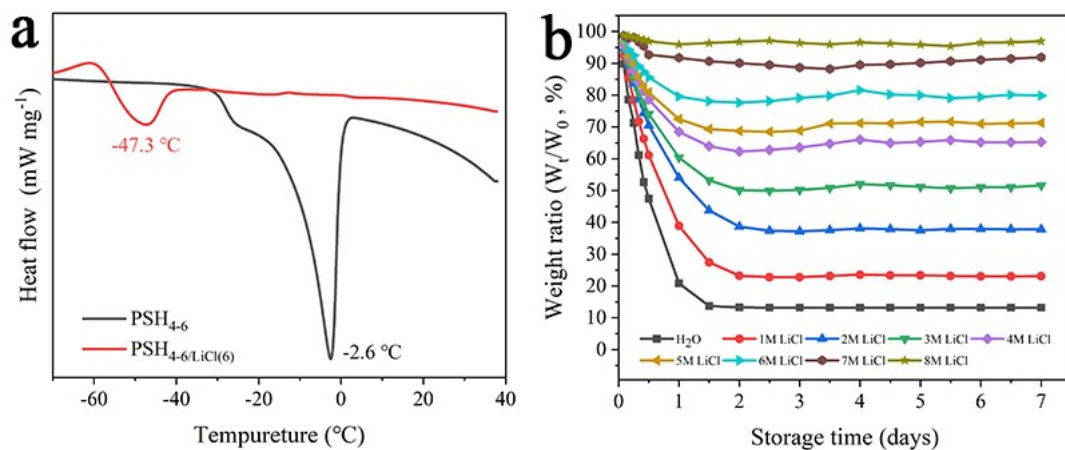
**Fig S4.** Contact angle of water droplet on different substrates



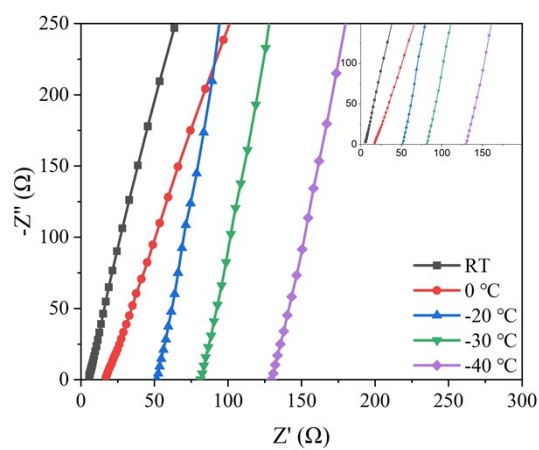
**Fig S5.** Change of contact angles of water at 0 second, 10 seconds and 15 seconds for different hydrogel, (a) PSH<sub>2-8</sub>, (b) PSH<sub>4-6</sub>, (c) PSH<sub>6-4</sub>.



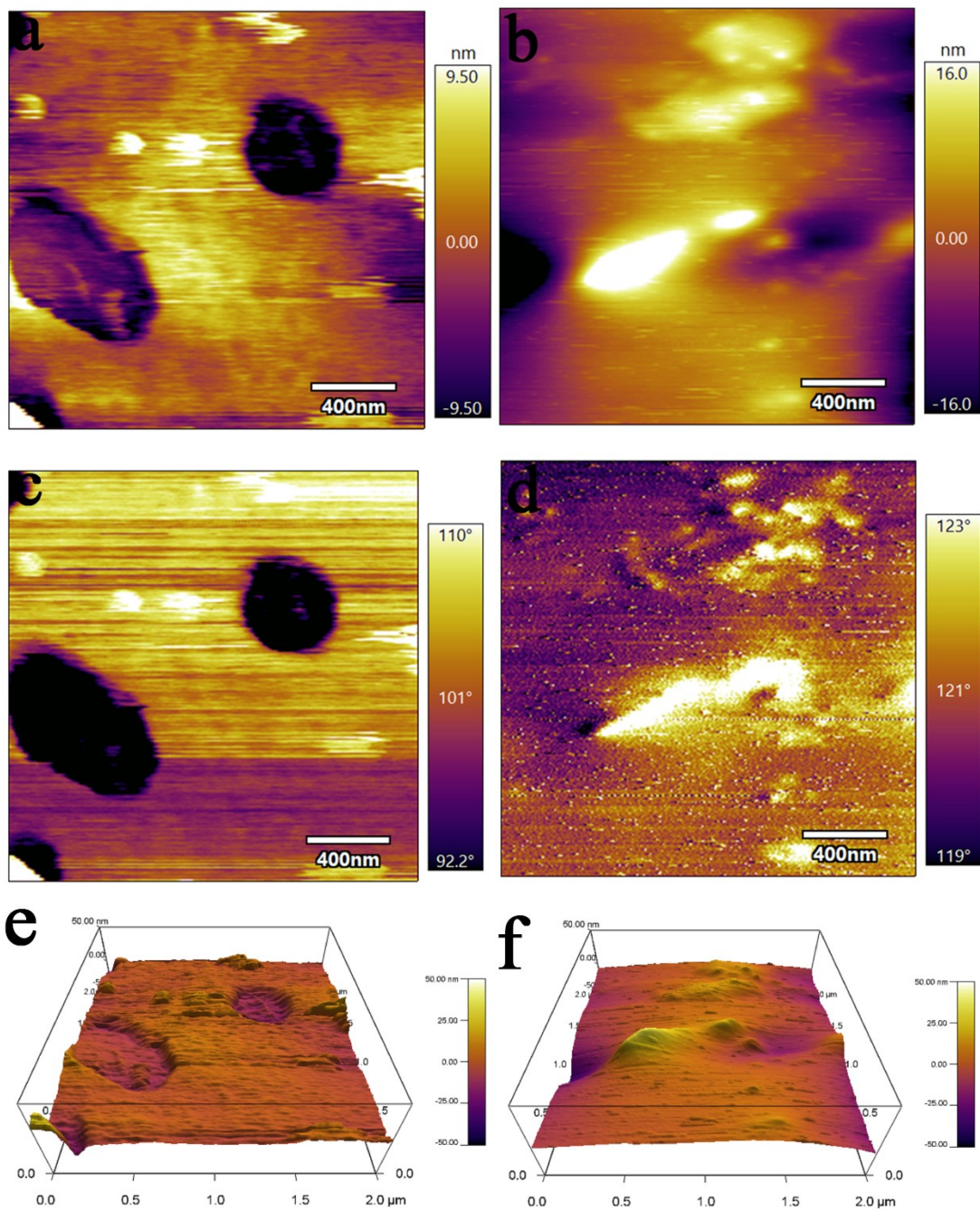
**Fig S6.** C 1s spectra of (a) PSH<sub>4-6</sub> and (b) PSH<sub>4-6</sub>/LiCl(6). (c) N 1s and (d) S 2p spectra of PSH<sub>4-6</sub> and PSH<sub>4-6</sub>/LiCl(6) for comparison.



**Fig S7.** (a) DSC results of PSH<sub>4-6</sub> hydrogel without and with 6 M LiCl. (b) Water loss rate of PSH<sub>4-6</sub> hydrogels after immersing in different concentrations of LiCl solution at room temperature.

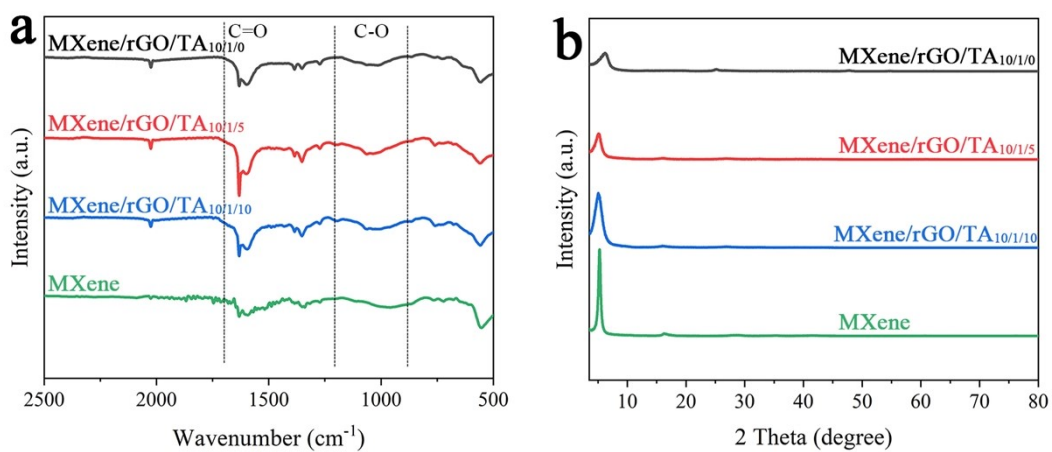


**Fig S8.** The EIS curves of PHEAA/LiCl(6) hydrogels at different temperatures.

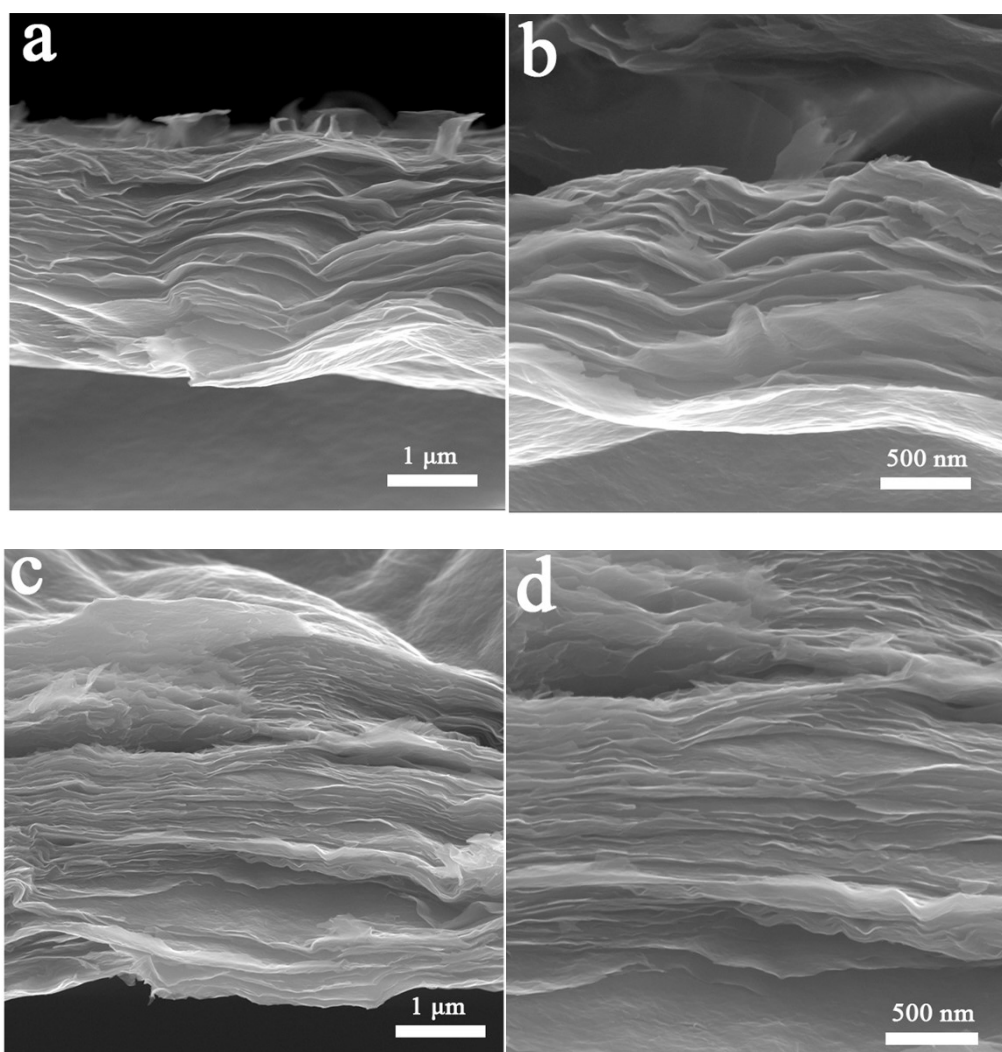


**Fig S9.** In-situ AFM graph of PSH<sub>4-6</sub> hydrogel without/with 6 M LiCl at the same location. The height retrace (a), phase retrace (c) and 3D images (e) of PSH<sub>4-6</sub> hydrogel.

The corresponding graph of PSH<sub>4-6</sub>/LiCl(6) ((b) (d) (f)).

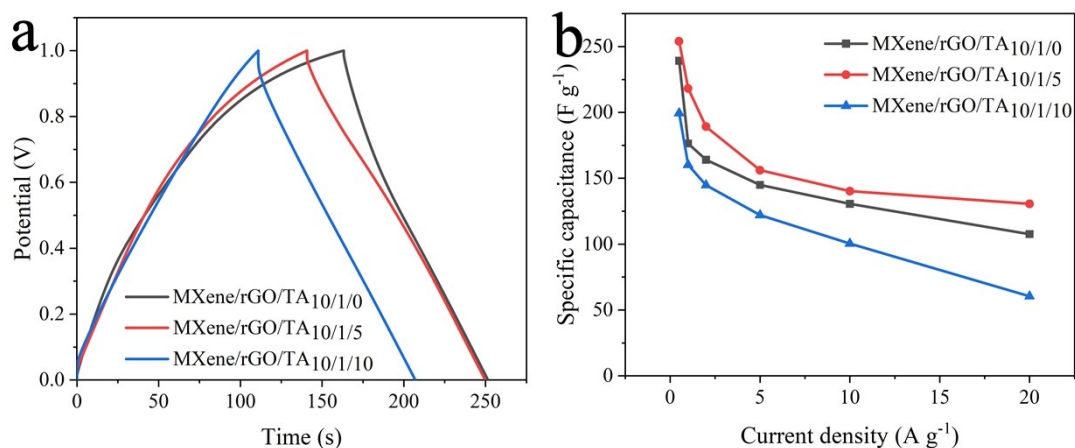


**Fig S10.** (a) FTIR spectra and (b) XRD spectra of  $\text{Ti}_3\text{C}_2\text{T}_x/\text{rGO}/\text{TA}$  composite films.

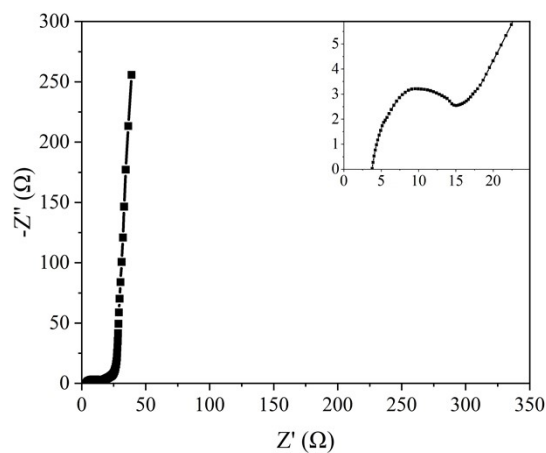


**Fig S11.** SEM images of  $\text{Ti}_3\text{C}_2\text{T}_x/\text{rGO}/\text{TA}_{10/1/0}$  (a and b),  $\text{Ti}_3\text{C}_2\text{T}_x/\text{rGO}/\text{TA}_{10/1/5}$  (c and d).





**Fig S12.** Electrochemical properties of supercapacitor constructed with as-prepared film. (a) GCD curves at a current density of 1 A g<sup>-1</sup>. (b) Specific capacitance versus different current densities (1-20 A g<sup>-1</sup>)



**Fig S13.** The EIS curves of flexible supercapacitors at room temperature.

**Table S1.** Different molar ratio hydrogels S 2p XPS data

Sample	S 2p 1/2	S 2p 3/2	Area ratio
PSH <sub>2-8</sub>	167.64 (eV)	168.79 (eV)	1.054
PSH <sub>4-6</sub>	167.47 (eV)	168.61 (eV)	1.135
PSH <sub>6-4</sub>	167.33 (eV)	168.47 (eV)	1.326
SBMA	167.18 (eV)	168.38 (eV)	1.553
PSH <sub>4-6</sub> /LiCl(6)	167.15 (eV)	168.20 (eV)	0.756

**Table S2.** Summary of ionic conductivity of different conductive hydrogel at room temperature.

Components	Conducting ions	Conductivity (S m <sup>-1</sup> )	Refs
SBMA-HEA	LiCl	14.6	1
SBMA-PAM	NaCl	3.674	2
SBMA-CNF	ZnSO <sub>4</sub>	2.46	3
SBMA-PVA-PAM	ZnCl <sub>2</sub>	1.57	4
PVA-Thioctic acid	AlCl <sub>3</sub>	~0.23	5
TBOT-BA	LiTFSI	0.134	6
PAMPS-PAAm	LiCl	2.29	7
PVA-CNF	NaCl	3.2	8
PAMPS-MC	KOH	10.5	9
PAO/PEI	KOH	22.35	10
PVA/glycerol	CH <sub>3</sub> COONa	8.127	11
SBMA-HEAA	LiCl	25.8	Our Work

**Table S3.** Summary of antifreezing hydrogel electrolytes and supercapacitors

Electrode material	Electrolyte components	Conductivity (S m <sup>-1</sup> )	Capacitance	Refs
Activated carbon	SBMA-HEA/LiCl	1.26 at -40°C	134 mF cm <sup>-2</sup> at -30°C	12
Activated carbon	SBMA-AA/ZnCl <sub>2</sub>	1.56 at -60°C	~50 F g <sup>-1</sup> (0.5 A g <sup>-1</sup> ) at -60°C	13
Activated carbon	SBMA-AM/EG	0.151 at -50°C	62 F g <sup>-1</sup> (62.5 mA g <sup>-1</sup> ) at -50°C	14
Activated carbon	SBMA-AM-AMPS	3.4 at room temperature	12.5 F g <sup>-1</sup> (0.5 A g <sup>-1</sup> ) at -10°C	15
Carbon nanotubes	PVA/P(SBMA-AM)/ CaCl <sub>2</sub>	0.28 at -40°C	-	16
-	SBMA-AM-PVA/LiCl	7.95 at -45.3°C	-	17
Polypyrrole	MMT-AM/EMIMBF <sub>4</sub>	0.518 at -30°C	~35 mF cm <sup>-2</sup> at -30°C	18
Polyaniline	PVA/H <sub>2</sub> SO <sub>4</sub> /Glycerol	1.71 at -40°C	268 mF cm <sup>-2</sup> at -20°C	19
Graphene	VC-PVA PEDOT:PSS/EG	4.0 at -30°C	212.6 F g <sup>-1</sup> (0.1 A g <sup>-1</sup> ) at -20°C	20
Activated carbon	HPC/PVA/ Glycerol/LiClO <sub>4</sub>	0.57 at -40°C	143.6 F g <sup>-1</sup> (2 A g <sup>-1</sup> ) at -40°C	21
Graphene	PVA/EG/Zn(Tf) <sub>2</sub>	0.353 at -40°C	202.8 F g <sup>-1</sup> (0.2 A g <sup>-1</sup> ) at -20°C	22
Activated carbon	Carrageenan-PVA/EG	3.18 at -40°C	113.6 F g <sup>-1</sup> (3 A g <sup>-1</sup> ) at -40°C	23
Activated carbon	Carrageenan-AM/LiCl/KCl	1.9 at -40°C	73.4 F g <sup>-1</sup> (1 A g <sup>-1</sup> ) at -40°C	24
Activated carbon	PAMPS-AM/EG	0.1 at -30°C	55.5 F g <sup>-1</sup> (1 A g <sup>-1</sup> ) at -20°C	25
Activated carbon	Silk backbone/ EG/ChCl/ZnCl <sub>2</sub>	0.363 at -20°C	242.9 F g <sup>-1</sup> (0.2 A g <sup>-1</sup> ) at -18°C	26
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /rGO/TA	SBMA-HEAA/LiCl	2.21 at -40°C	133 F g <sup>-1</sup> (0.5 A g <sup>-1</sup> ) at -40°C	Our Work

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