Synergistic crosslinking effect of illite and hydroxyethyl cellulose on improving the properties of PVA based hydrogels

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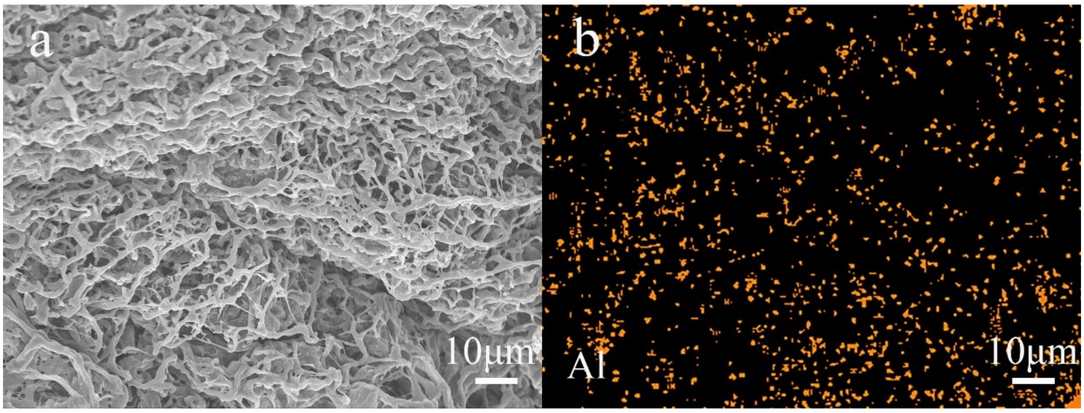

Fig. S1. Al element EDS mapping of PH_3I_6 hydrogel

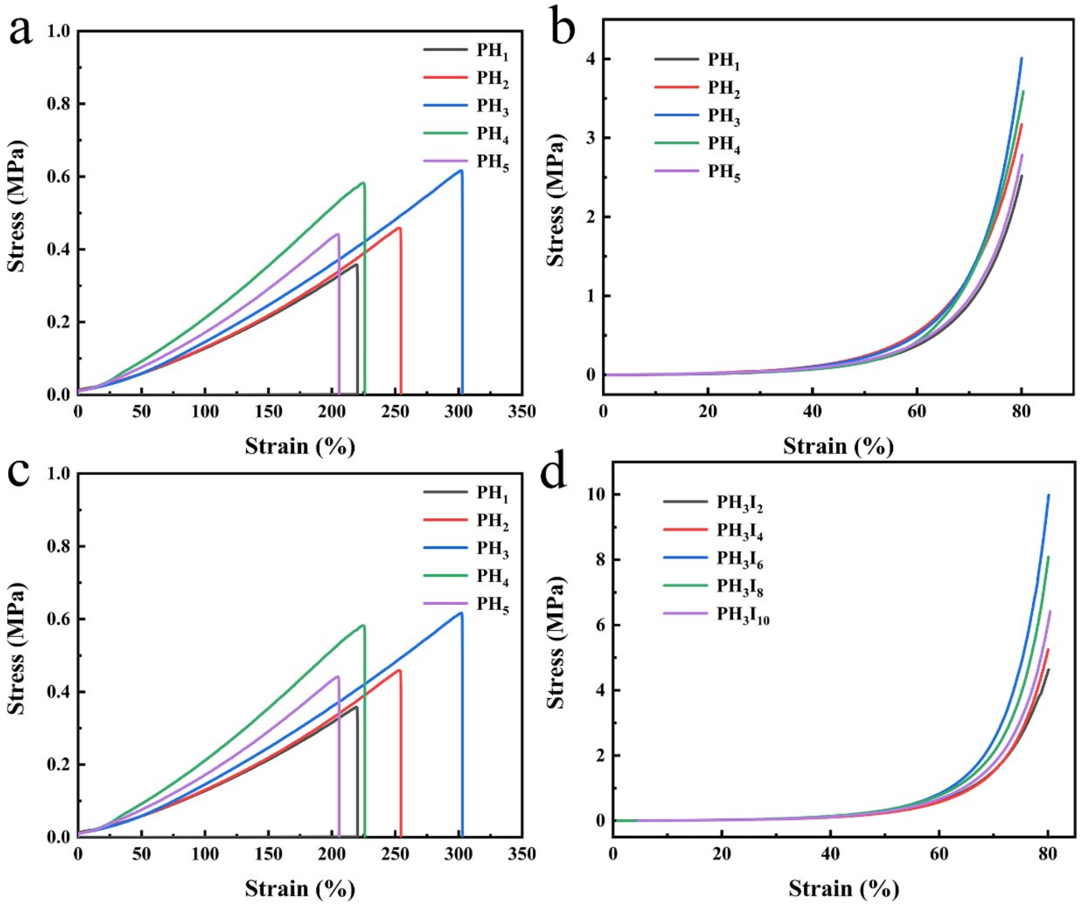

Fig. S2. (a) Tensile stress-strain curves of PH*x* hydrogels, (b) compressive stress-strain curves of PH*x* hydrogels, (c) tensile stress-strain curves of PH_3Ix, and (d) compressive curves of PH_3Ix.

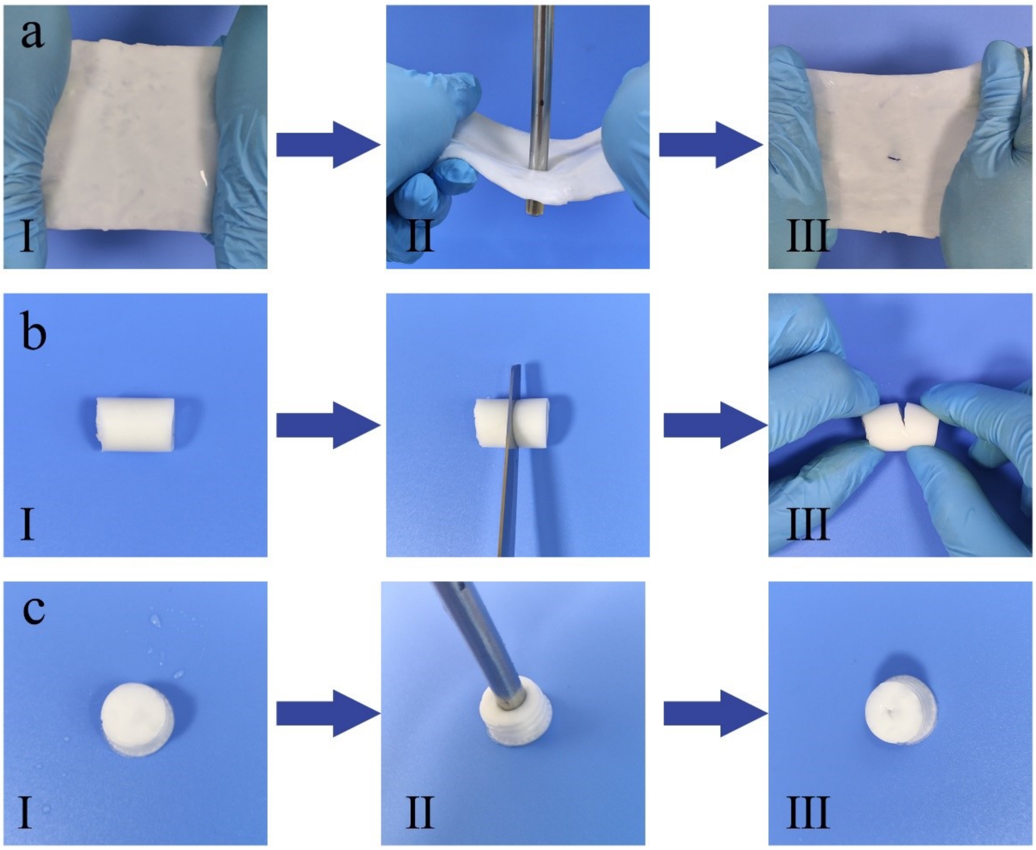

Fig. S3. Destructive experiment of PVA hydrogel.

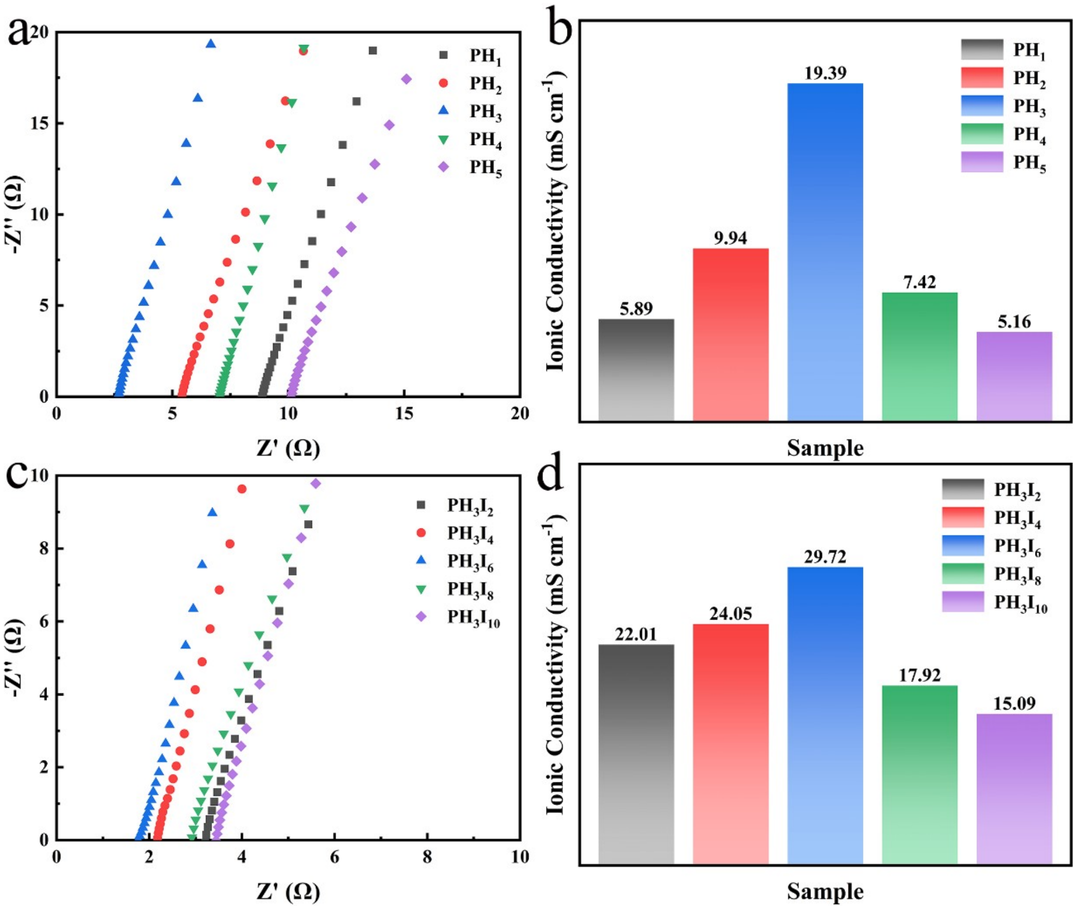

Fig. S4. (a) EIS curves of PH_x hydrogel electrolytes, (b) ionic conductivity of PH_x hydrogel electrolytes, (c) EIS curves of PH_3I_y hydrogel electrolytes, and (d) ionic conductivity of PH_3I_y hydrogel electrolytes.

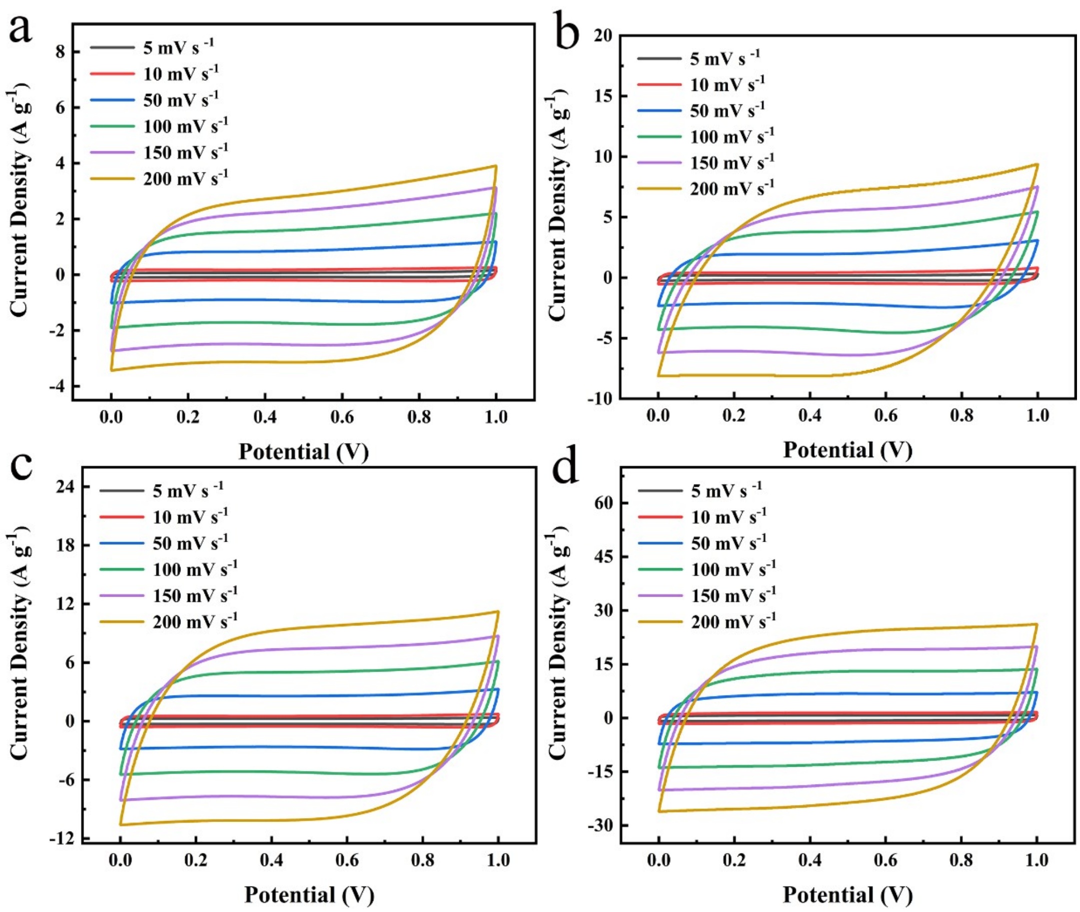

Fig. S5. CV curves of (a) PVA, (b) PI_6, (c) PH_3, and (d) PH_3I_6 assembled supercapacitors at scan rates from 5 to 200 mV s^{-1}.

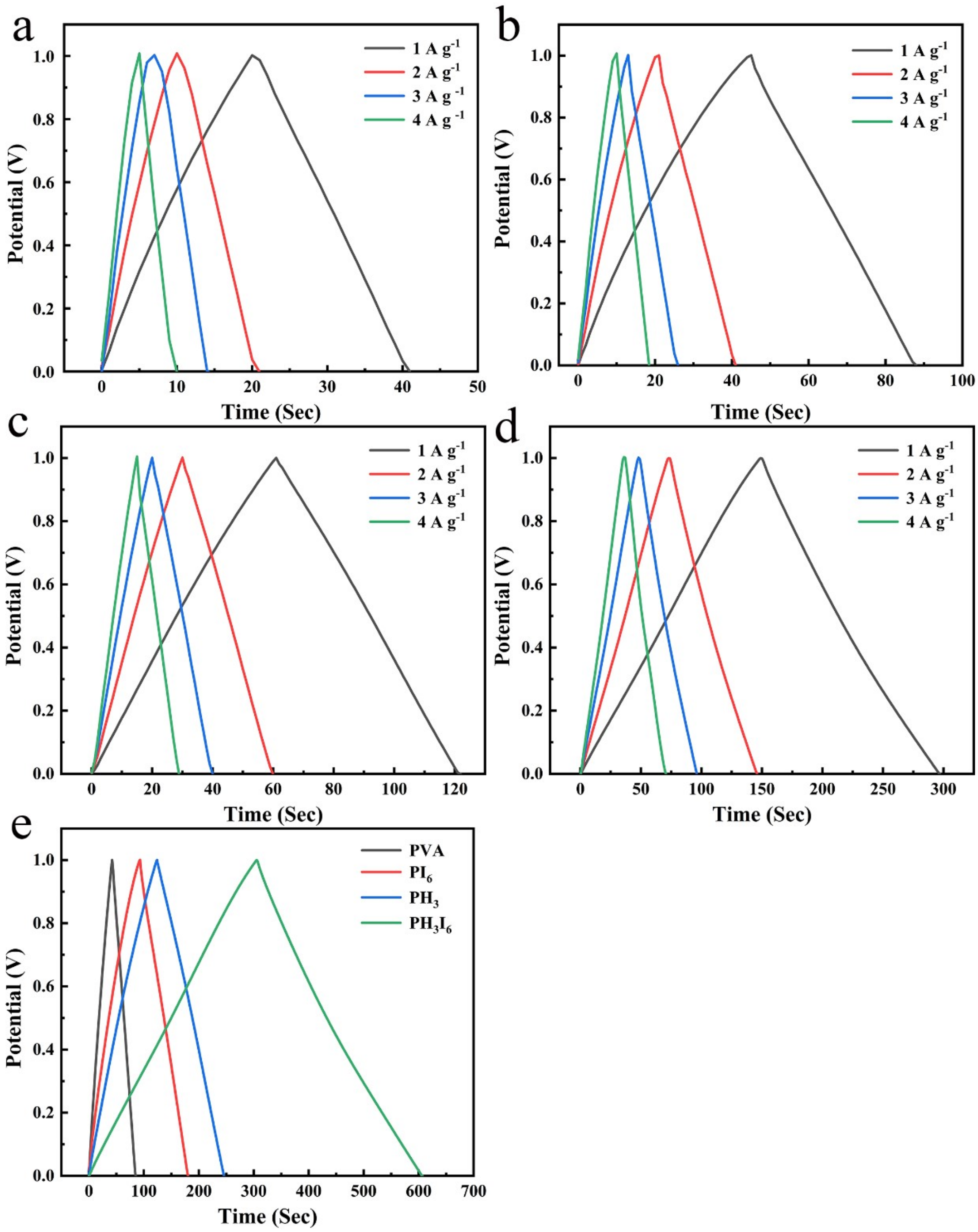

Fig. S6. GCD curves of (a) PVA, (b) PI_6, (c) PH_3, and (d) PH_3I_6 hydrogel-based supercapacitors at current densities from 1 to 4 A g^{-1}, and (e) GCD curves of PH_3I_6 hydrogel-based supercapacitor at 0.5 A g^{-1}.

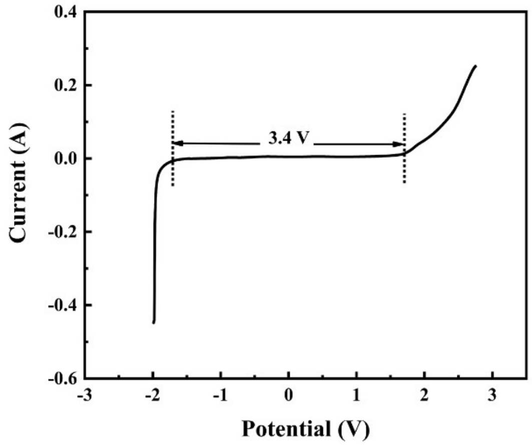

Fig. S7. Voltage stability window of PH_3I_6 hydrogel electrolyte at 5 mV s^{-1}.

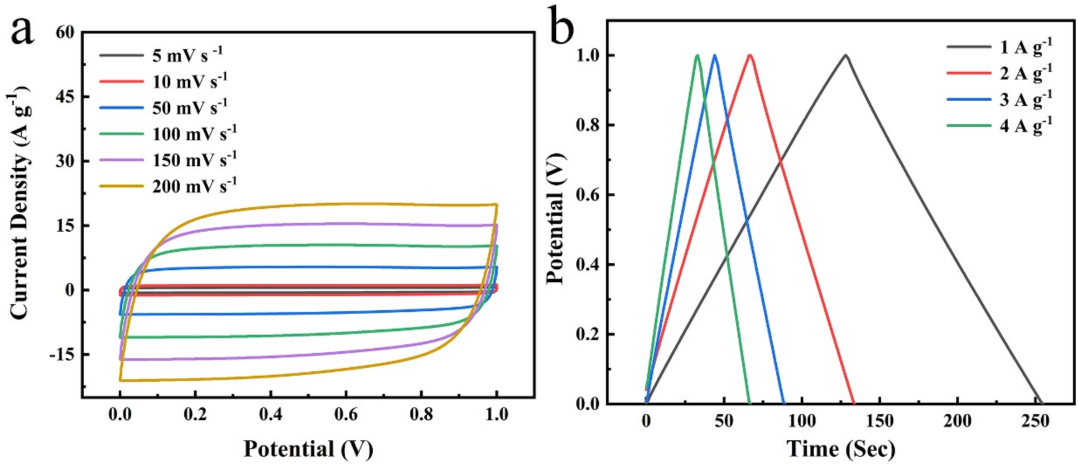

Fig. S8. (a) CV curves and (b) GCD curves of supercapacitor with PH_3I_6 hydrogel electrolyte immersed in 6 M KOH at different scan rates.

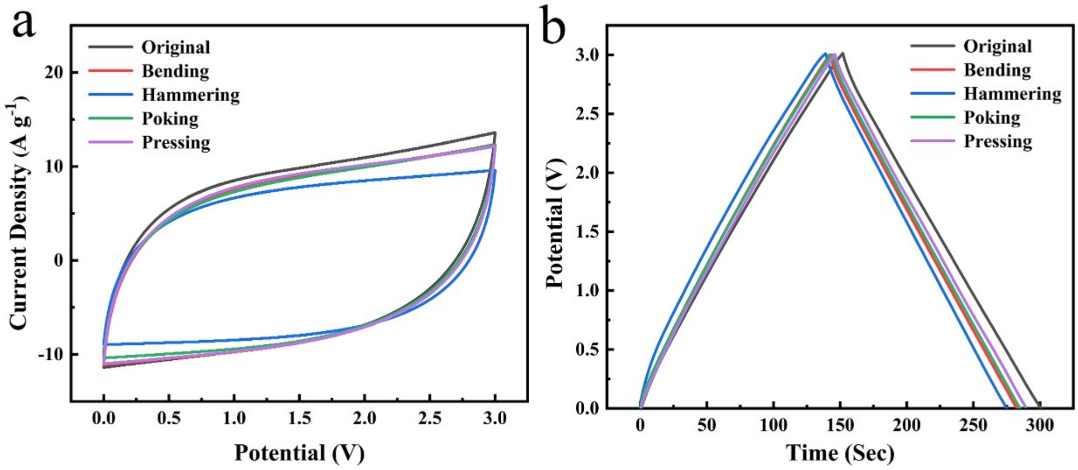

Fig. S9. (a) CV curves and (b) GCD curves of PH_3I_6 hydrogel-based supercapacitor under different conditions.

Table S1 The mechanical properties and ionic conductivity of PVA/HEC/illite hydrogel compared with previous hydrogels

Hydrogel	Stain (%)	Stress (Mpa)	Conductivity (mS cm^{-1})	Ref
PVA/CNF	200	0.84	15	[S1]
FT/PAM/PVA	410	0.4	14.63	[S2]
PVA/PAM/CS	452	0.41	32.16	[S3]
PUL/PVA	330	0.136	30.1	[S4]
PVA/gelatin/LiCl	1100	1.7	21.14	[S5]
PVA/N-CS	457	0.36	33	[S6]
PEI/PVA	1223	0.034	21.49	[S7]
PVA/$Zn(OTf)_2$	760	0.16	7.95	[S8]
PVA/HEC/illite	**500**	**1.41**	**29.72**	**This work**

Table S2 Rs and Rct of supercapacitors assembled with different hydrogel electrolytes

Hydrogel electrolyte	Rs (Ω)	Rct (Ω)
PVA	5.34	8.86
PI_6	3.68	4.63
PH_3	0.92	1.56
PH_3I_6	0.61	0.51

Table S3 Electrochemical performance of supercapacitor based on the PH_3I_6 hydrogel electrolyte compared with previous studies about flexible supercapacitor

Hydrogel electrolyte	E ($Wh\ kg^{-1}$)	P ($W\ kg^{-1}$)	Cycling stability	Ref
PVA/Agar/GO-EMIMBF4	4.2	498.7	80% after 5000 cycles	[S9]
PVA/MXene	4.6	300	99.4% after 5500 cycles	[S10]
PVA/TiO_2/CNFs	5.3	773.1	84.3% after 5000 cycles	[S11]
PVA	10.8	16	96.5% after 20000 cycles	[S12]
PVA/Phytic acid	13.7	150	78% after 5000 cycles	[S13]
PVA/DCBQ	18.8	450	73% after 10000 cycles	[S14]
PVA/HRG	6.1	1000	93% after 5000 cycles	[S15]
PVA/$LiClO_4$	2.3	150	94% after 5000 cycles	[S16]
PVA/HEC/illite	**10.14**	**250**	**96.52% after 10000 cycles**	**This work**

Table S4 Specific capacitance (C), energy density (E), and power density (P) of supercapacitors under different current densities

Current Density ($A\ g^{-1}$)	C ($F\ g^{-1}$)	E ($Wh\ kg^{-1}$)	P ($W\ kg^{-1}$)
0.5	300	10.42	125
1	292	10.14	250
2	284	9.86	500
3	282	9.79	750
4	272	9.44	1000

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