

## **Supporting Information**

### **Natural Polysaccharide Strengthened Hydrogel Electrolyte and Biopolymer Derived Carbon for Durable Aqueous Zinc Ion Storage**

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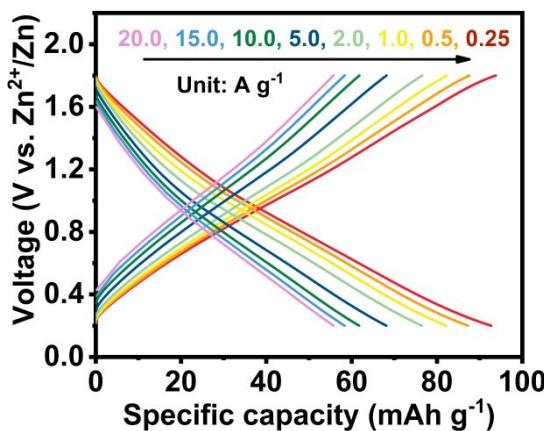
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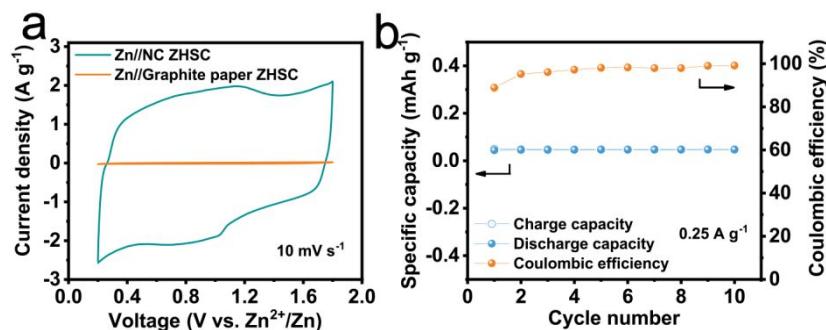
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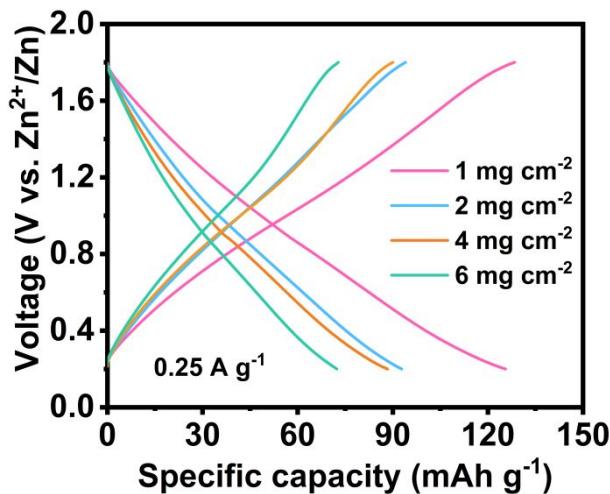
<sup>¶</sup>These authors contribute equally to this work.



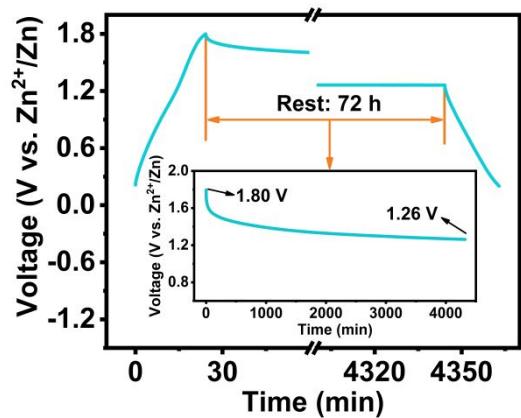
**Figure S1.** GCD curves of the aqueous ZHSC.



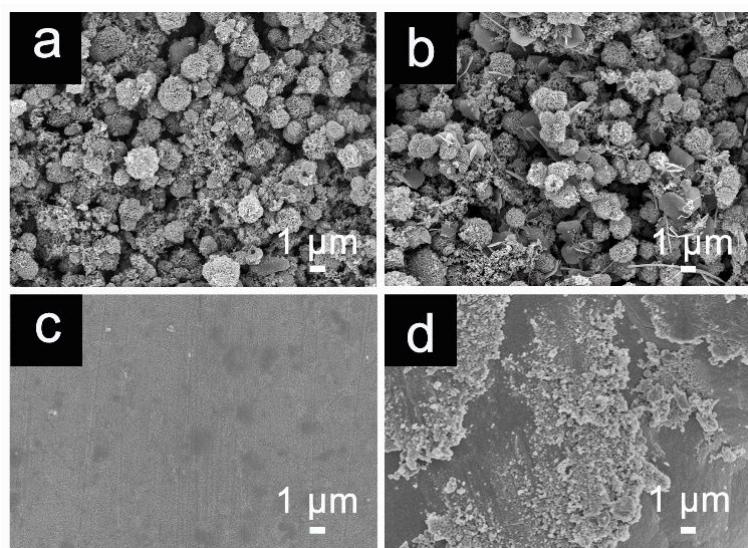
**Figure S2.** (a) CV curves of the aqueous Zn//NC and Zn//graphite paper ZHSCs. (b) Specific capacity and Coulombic efficiency of Zn//graphite paper ZHSC.



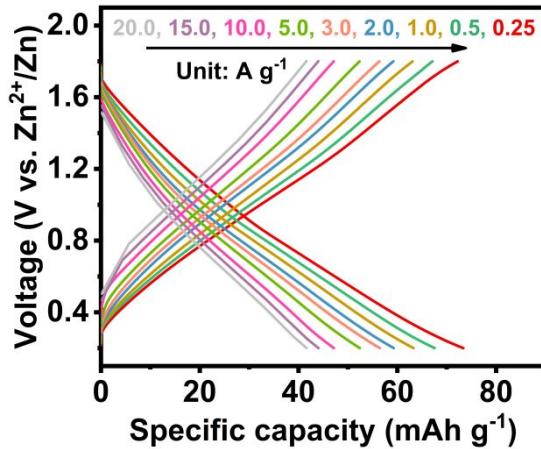
**Figure S3.** GCD curves of the ZHSCs under various NC mass loadings.



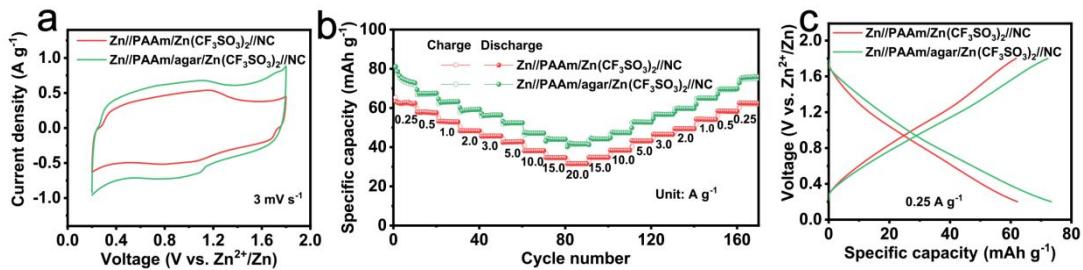
**Figure S4.** Self-discharge curve of the aqueous ZHSC.



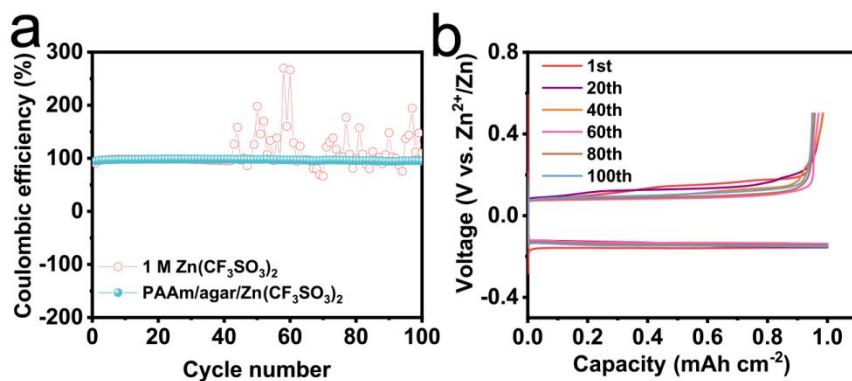
**Figure S5.** SEM images of NC cathode (a) before cycling and (b) after cycling, Zn anode (c) before cycling and (d) after cycling.



**Figure S6.** GCD curves of the quasi-solid-state ZHSC.



**Figure S7.** (a) CV curves of the quasi-solid-state Zn//PAAm/Zn( $\text{CF}_3\text{SO}_3$ )<sub>2</sub>//NC and Zn//PAAm/agar/Zn( $\text{CF}_3\text{SO}_3$ )<sub>2</sub>//NC ZHSCs. (b) Rate performance of the quasi-solid-state ZHSCs. (c) GCD curves of the quasi-solid-state ZHSCs in different hydrogel electrolytes.



**Figure S8.** (a) Coulombic efficiencies of Zn//Cu cells in aqueous and hydrogel electrolytes. (b) Voltage curves of Zn//Cu cell in hydrogel electrolyte.

**Table S1.** Comparisons of the mechanical properties with recently reported hydrogel electrolytes.

Electrolyte	Tensile strength (kPa)	Compressive strength (kPa)	Ref.
PolySH/LiCl	5	42, 75%	1
PVA/NFC/ZnSO <sub>4</sub>	23.1	26.2, 90%	2
$\kappa$ -CG/PAAm/Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	34.6	75.3, 75.0%	3
PAM-co-PAA/k-CG/ZnSO <sub>4</sub>	42.5	~110, 60%	4
PAM with 7.5 M ZnCl <sub>2</sub>	~40	-	5
SPMA-Zn3	58	70, 50%	6
P(GMA <sub>3</sub> -AAm <sub>7</sub> )-borate	17.7	-	7
PAAm/agar/Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	78.9	118.0, 60.0%	This work

**Table S2.** Comparisons of the ionic conductivity with recently reported hydrogel electrolytes.

Electrolyte	ionic conductivity (S m <sup>-1</sup> /mS cm <sup>-1</sup> )	Ref.
PHE	6.33 mS cm <sup>-1</sup> (25 °C)	8
P(AM-co-DMAEMA)-AMP/ gelatin	~1.35 S m <sup>-1</sup> (25 °C) 0.43 S m <sup>-1</sup> (-40 °C)	9
PVA/glycerol/CaCl <sub>2</sub>	1 S m <sup>-1</sup>	10
P(AMPS <sub>0.3</sub> -co-AAM <sub>0.4</sub> )	0.82 S m <sup>-1</sup> (-20 °C) 1.12 S m <sup>-1</sup> (100 °C)	11
HPAAN/PDA	0.32 S m <sup>-1</sup>	12
AFHE	5.88 mS cm <sup>-1</sup> (20 °C)	13
cross-linked lignin hydrogel	10.35 mS cm <sup>-1</sup> (25 °C)	14
PAAm/agar/Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	1.55 S m <sup>-1</sup>	This work

**Table S3.** Comparisons of the obtained capacity with previously reported aqueous ZHSCs.

Materials	Electrolyte	Voltage range (V)	Specific capacity/ Capacitance	Ref.
MnO <sub>2</sub> nanorods	ZnSO <sub>4</sub>	0-2	54.1 mA h g <sup>-1</sup> (0.1 A g <sup>-1</sup> )	15
BC-CNa	ZnSO <sub>4</sub>	0.2-1.8	51.4 mA h g <sup>-1</sup> (0.2 A g <sup>-1</sup> )	16
EG/PANI	ZnSO <sub>4</sub>	0.1-1.5	172 F g <sup>-1</sup> (0.13 A g <sup>-1</sup> )	17
AC	Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	0-1.8	170 F g <sup>-1</sup> (0.1 A g <sup>-1</sup> )	18
MXene-rGO2	ZnSO <sub>4</sub>	0.2-1.6	128.6 F g <sup>-1</sup> (0.4 A g <sup>-1</sup> )	19
PC	ZnSO <sub>4</sub>	0.3-1.9	75.2 mA h g <sup>-1</sup> (0.22 A g <sup>-1</sup> )	20
NMXC	ZnSO <sub>4</sub>	0.2-1.8	83.9 mA h g <sup>-1</sup> (0.1 A g <sup>-1</sup> )	4
NC	Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	0.2-1.8	92.8 mAh g <sup>-1</sup> (0.25 A g <sup>-1</sup> )	This work

**Table S4.** Comparisons of the cycling stability with the ZHSCs in different electrolytes.

Materials	Electrolyte	Cycling stability	Ref.
ACNS	$\kappa$ -CG/PAAm/Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	102.7% after 10000 cycles	3
NMXC	PAM-coPAA/k-CG/ZnSO <sub>4</sub>	96.4% after 10000 cycles	4
AC	SPMA-Zn hydrogel electrolyte	95.3% after 5000 cycles	6
AC	SA-Zn double network hydrogel electrolyte	95.4% after 2000 cycles	21
HPC-600	gelatin/Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	102.1% after 10000 cycles	22
WC-6ZnN-12U@CC	PAM-ZnSO <sub>4</sub>	81.4% after 2000 cycles	23
VOPO <sub>4</sub> ·2H <sub>2</sub> O	UV-crosslinked ZIP with 0.65 M Zn(OTf) <sub>2</sub> salt	63% after 50 cycles	24
AC	Redox SA-Zn-Br double network hydrogel electrolyte	87.7% after 5000 cycles	25
OPCNF-20	gelatin/ZnSO <sub>4</sub>	92.1% after 1000 cycles	26

NC	PAAm/agar/Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub> hydrogel electrolyte	112.4% after 10000 cycles	This work
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