Electronic Supplementary Information

Hydrogel electrolytes for zinc dendrite regulation: balancing conductivity and modulus

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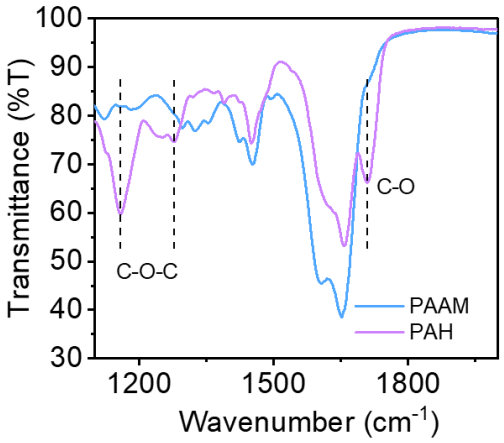

Fig. S1 FTIR spectra of PAAM and PAH hydrogels.

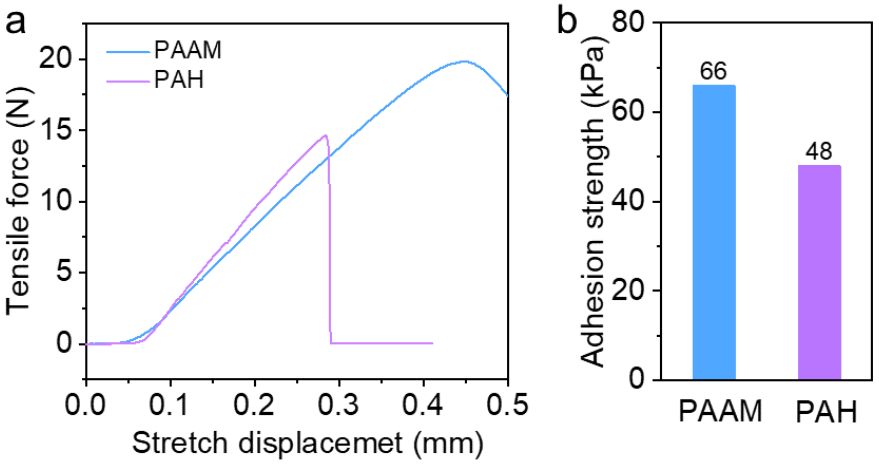

Fig. S2 Adhesion performance of hydrogel electrolytes. (a) The tension adhesion curves and (b) corresponding adhesion strength values.

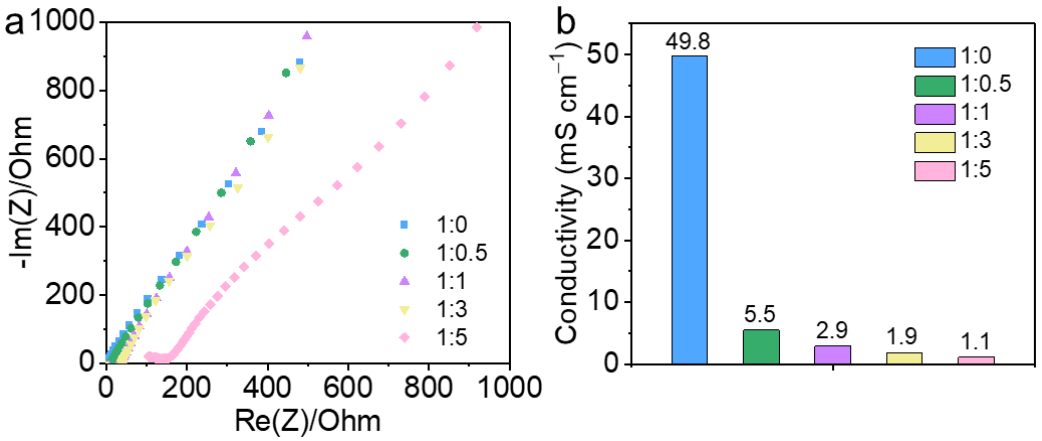

Fig. S3 Conductivity measurements. (a) Electrochemical impedance spectra of electrolytes with different monomer concentration ratios (AM : HEMA) and (b) conductivity results.

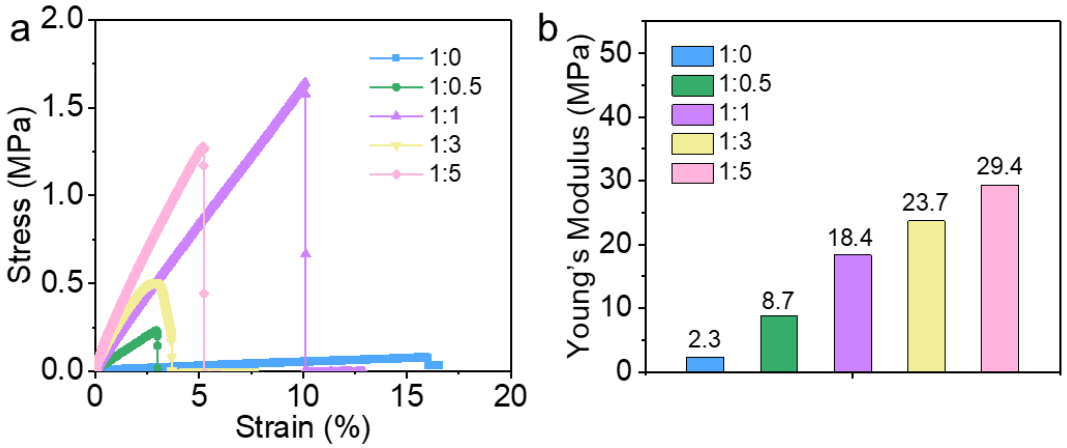

Fig. S4 Modulus measurements. (a) Stress-strain curves of the electrolytes and (b) Young's modulus.

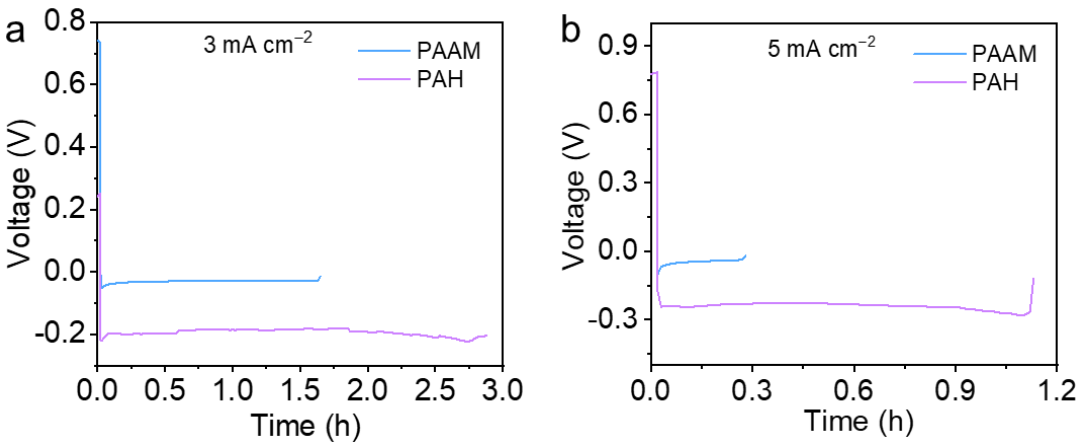

Fig. S5 Voltage evolution of zinc half-cells upon plating current at (a) 3 mA cm^{-2} and (b) 5 mA cm^{-2}.

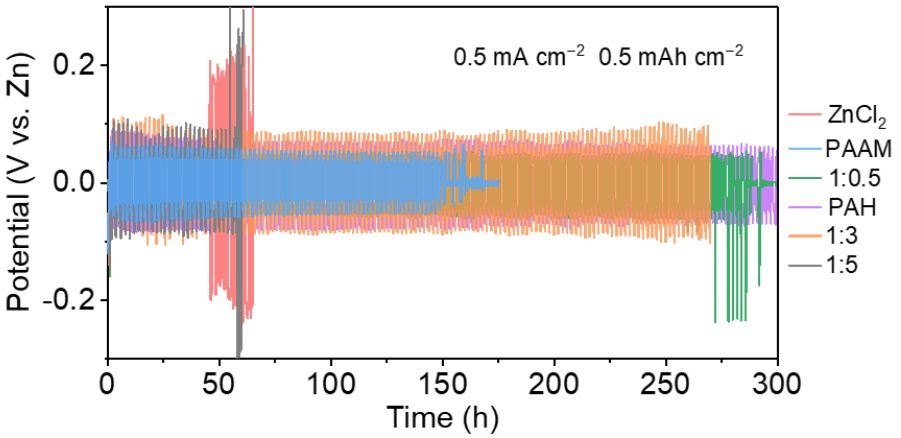

Fig. S6 Long-term cycling of symmetric Zn cells using $ZnCl_2$, PAAM, and PAH hydrogel electrolytes with varying *c*(AM):*c*(HEMA) ratios (1:0.5, 1:1, 1:3, and 1:5).

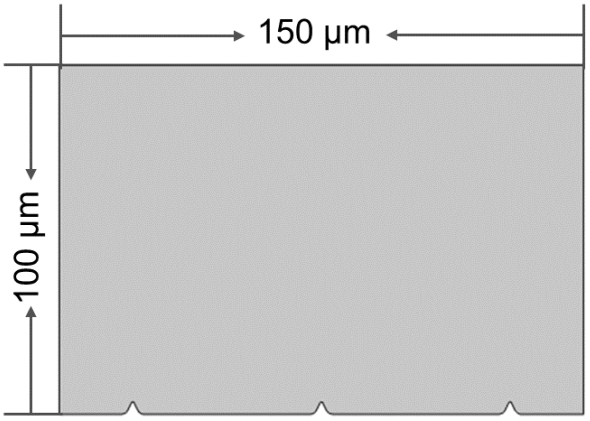

Fig. S7 Schematic of the geometry for simulations.

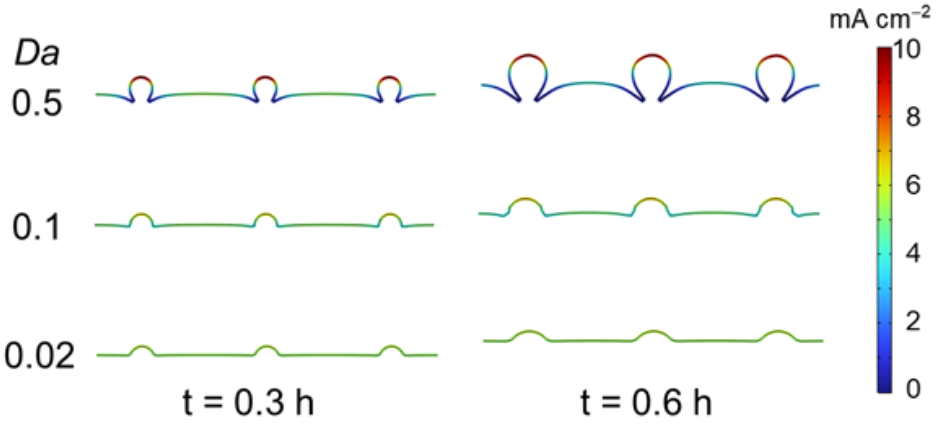

Fig. S8 The current density distribution at the electrode surface during zinc deposition.

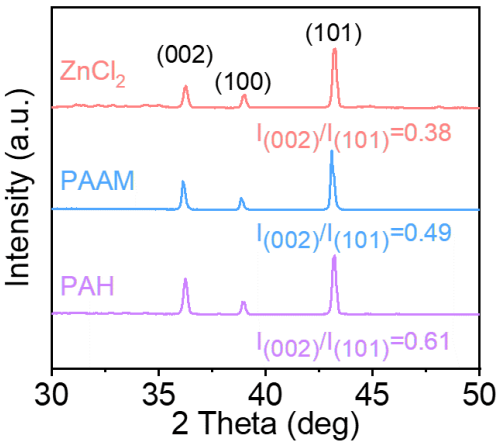

Fig. S9 XRD patterns of zinc anodes after 2 hours of deposition at current density of 5 mA cm^{-2} with $ZnCl_2$, PAAM and PAH electrolytes.

Table S1. Modulus summary of hydrogel electrolytes.

Hydrogels	Young's Modulus (MPa)	References
PAH	10-30	This work
Aniso-CMC	18	PNAS., **2024**, 121, e2322944121.
PVA-416	8	Adv. Mater., **2023**, 35, 2211673.
Zn alginate	5.2	Adv. Mater., **2024**, 36, 2313772.
α-HP/PVA	4.9	Energy Environ. Sci., **2024**, 17, 7919-7931.
DCZ-gel	2.1	Nano-Micro Lett., **2024**, 16, 106.
MMT-PAM	0.3	Energy Storage Mater., **2023**, 55, 236-243.
NASHE-0.5	0.2	Adv. Energy Mater., **2024**, 14, 2400063.
P-MC	0.03	Energy Environ. Sci., **2024**, 17, 8570-8581.
Gelatin	0.01	Small, **2023**, 19, 2207610.

Table S2. Parameters used in the simulation.

Symbol	Value	Name
W_cell	150 [μm]	Computational cell width
H_prot	4 [μm]	Height of protrusion
W_prot	4 [μm]	Width of protrusion
H_sep	100 [μm]	Height of separator
T	293.15 [K]	Temperature
T_cycle	3 [min]	Cycle time
i_app	56.52 [A m^{-2}]	Applied current density (forward cycle only)
σ_$ZnCl_2$	5 [S m^{-1}]	conductivity of $ZnCl_2$
σ_PAAM	4.98 [S m^{-1}]	conductivity of PAAM
σ_PAH	0.285 [S m^{-1}]	conductivity of PAH
ρ_Zn	7.14 [g cm^{-3}]	Density of zinc metal
M_Zn	65.38 [g mol^{-1}]	Molecular weight of zinc metal
α_c	0.5	Cathode transfer coefficient
α_a	0.5	Anode transfer coefficient
n	2	Electrons transferred