Supporting Information

Self-Assembled 2-Hydroxyphosphonoacetic Acid Protective Layer Enables Dendrite-Free Zn Anodes

Yingying Song,‡[a] Fengcan Ma,‡[b] Qinghua Xiao,[b] Sidan He,[b] Hongqiang Wang,*[,a] Qinghong Wang*[,b]

[a]School of Materials Science and Engineering, Shandong University of Technology, Zibo, Shandong, 255000, P. R. China

[b]School of Chemistry and Materials Science, Jiangsu Normal University, Xuzhou 221116, P. R. China

‡ These authors have contributed equally to this work.

* Corresponding authors.

E-mail: hwang@sdut.edu.cn (H. Wang);

wangqh@jsnu.edu.cn (Q. Wang)

1. Experimental section

1.1 Preparation of Zn-HPAA anode

Briefly, 0.6 mmol L^{-1} HPAA-Dimethyl sulfoxide (HPAA-DMSO) self-assembled solution was prepared by dissolving HPAA in DMSO and stirring for 2 h at room temperature. High-purity argon gas was then introduced into the prepared solution for 20 min to remove oxygen. After that, Zn anodes were immersed into the HPAA-DMSO solution for different assembly time (30 min, 1 h, and 2 h). Throughout the assembly process, argon gas was used to maintain an inert environment. Finally, the obtained Zn-HPAA anodes were dried by blowing Ar gas over the entire surface to remove the residual DMSO.

1.2 Characterization

The morphologies and structures of bare Zn and Zn-HPPA electrodes were conducted by field-emission scanning electron microscopy (SEM, Hitachi, SU-8010), powder X-ray diffractometer (XRD, Bruker D8 Advanced), Fourier transform infrared spectrometer (FT-IR, Bruker VERTEX 80 v), and atomic force microscope (AFM, BRUKER Dimension Icon). The elemental compositions for the surface of Zn-HPAA anode were carried out on X-ray photoelectron spectroscope (XPS, Thermo Scientific, K-Alpha). The contact angles on the surface of bare Zn and Zn-HPPA were measured on the Ramé Hart MODEL 260 instrument. The thicknesses of self-assembled protective layers of Zn-HPAA were measured by an ellipsometer (J.A. Woollam, M-2000V).

1.3 Electrochemical measurements

Symmetric cells (CR2032 coin) were assembled with bare Zn or Zn-HPAA electrodes, glass fiber as separator, and 2 M $ZnSO_4$ as electrolyte. The coulombic efficiency (CE) test of Zn striping/plating was conducted on a Cu foil. Galvanostatic charge-discharge measurements of the symmetric cells were recorded on a battery testing system (LAND, CT2001A). Tafel curves, linear sweep voltammetry (LSV), cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS) measurements were recorded on CHI 760e electrochemical station. The electrochemical performance of Zn//V_2O_5 and Zn-HPAA//V_2O_5 full cells were also investigated, using bare Zn and Zn-HPPA as anodes, commercial V_2O_5 as cathode material, and 2 M $ZnSO_4$ as electrolyte.

Table S1. Different protective layers on the Zn anodes and their electrochemical performance.

Zinc Anode	Current density (mA cm^{-2})	Areal density (mAh cm^{-2})	Time (h)	Reference
ATP@Zn	5.0	1	2800	[1]
DTPMP-Zn	5	0.5	1300	[2]
ZnTAPP-NTCDA-POP@Zn	0.5	0.5	1200	[3]
SCM@Zn	2	1	500	[4]
C-840/Zn	10	10	400	[5]
HqTpCOF@Zn	1	1	700	[6]
PYBZ@Zn	1	1	1900	[7]
Zn@FCTF-5	1	1	1600	[8]
PVDF-SBA15@Zn	3	0.6	1650	[9]
MOF-PVDF coated Zn	3	0.5	500	[10]
SPANI-Zn	0.5	0.5	1500	[11]
Zn-HPAA	**1**	**1**	**2500**	**This work**
	30	**1**	**650**	

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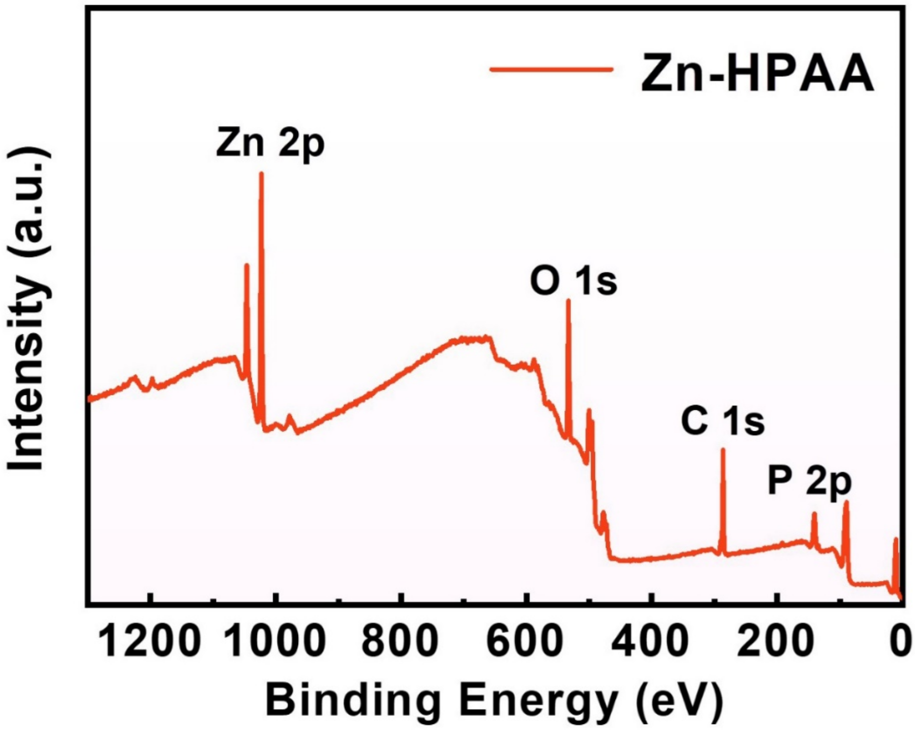

Figure S1. XPS survey spectrum of Zn-HPAA.

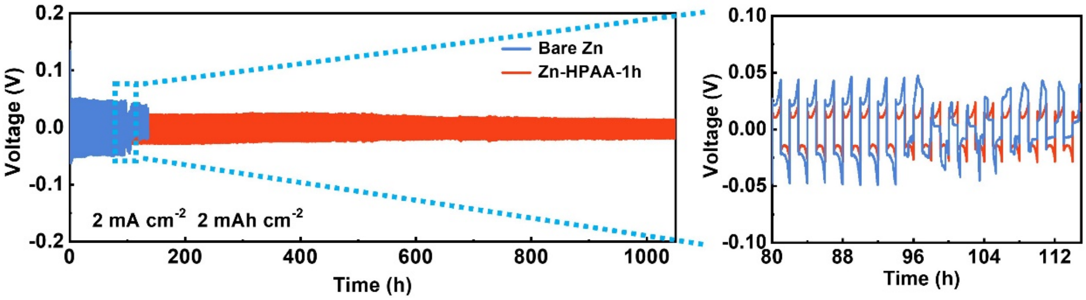

Figure S2. Long-term cycling performance of bare Zn and Zn-HPAA-1h symmetric cells at 2.0 mA cm^{-2} and 2.0 mAh cm^{-2}.

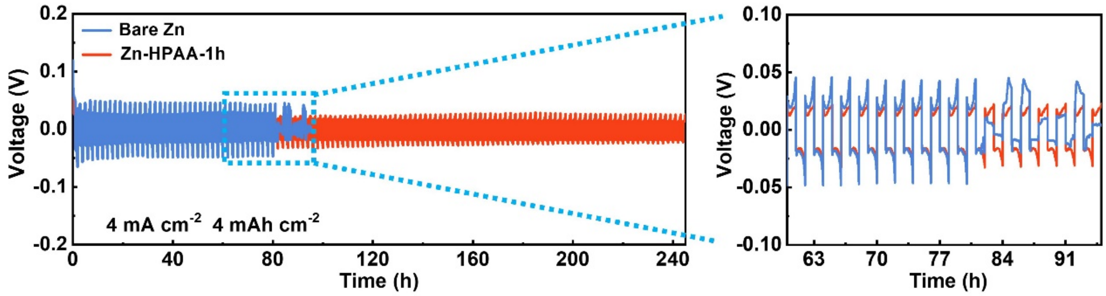

Figure S3. Long-term cycling performance of bare Zn and Zn-HPAA-1h symmetric cells at 4.0 mA cm^{-2} and 4.0 mAh cm^{-2}.

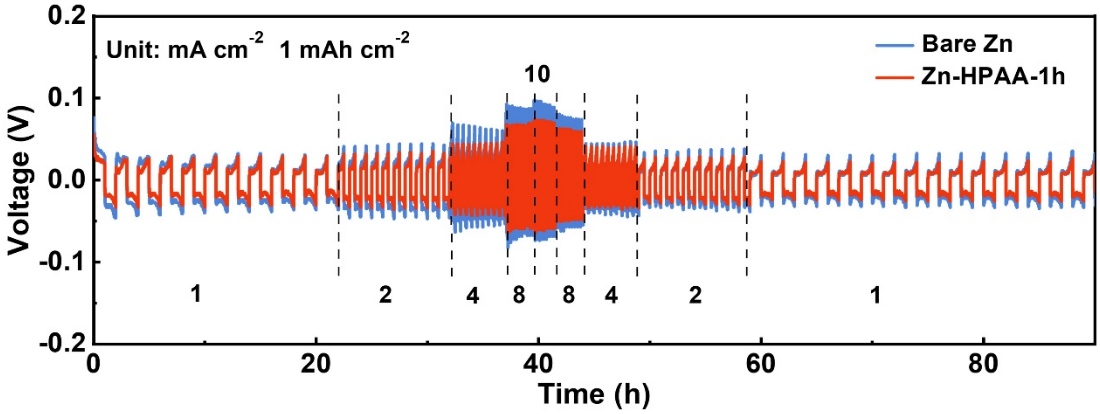

Figure S4. Rate capacity bare Zn and Zn-HPAA-1h symmetric cells at current densities from 1 to 10 mA cm^{-2} with capacity of 1 mAh cm^{-2}.

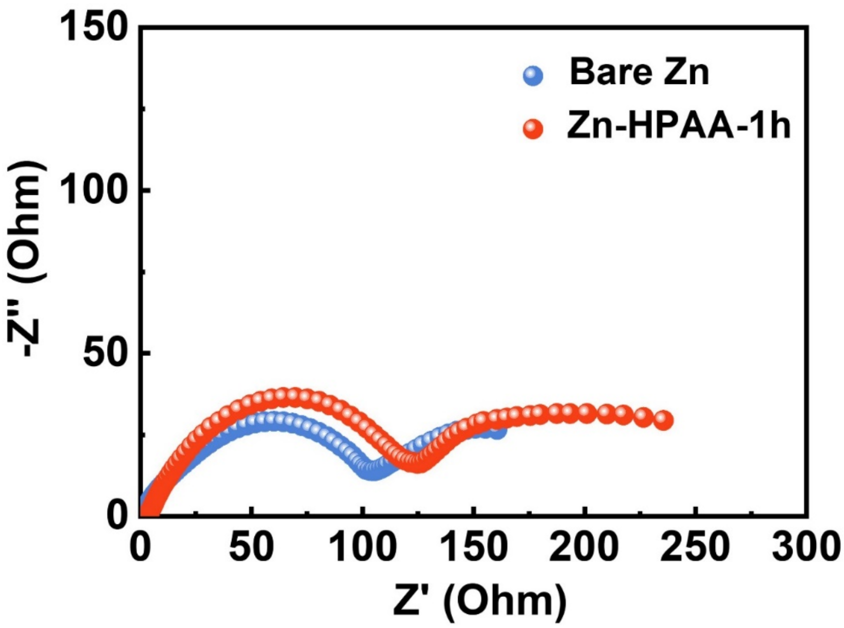

Figure S5. EIS of bare Zn and Zn-HPAA-1h symmetric cells.

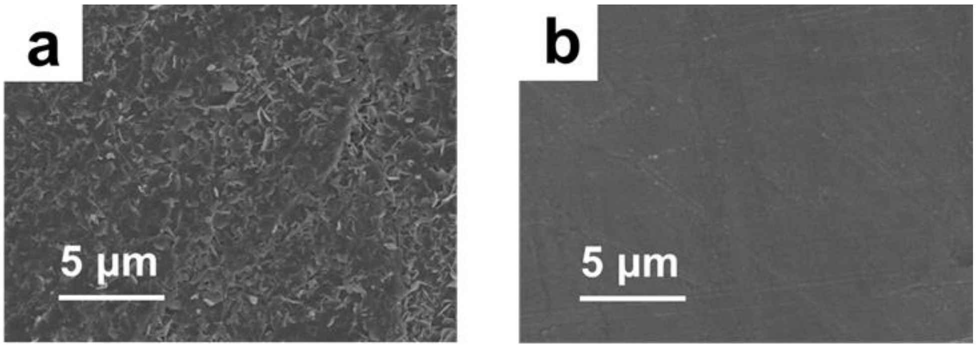

Figure S6. SEM images of (a) bare Zn and (b) Zn-HPAA-1h electrodes after soaking in 2.0 M $ZnSO_4$ for 6 h.

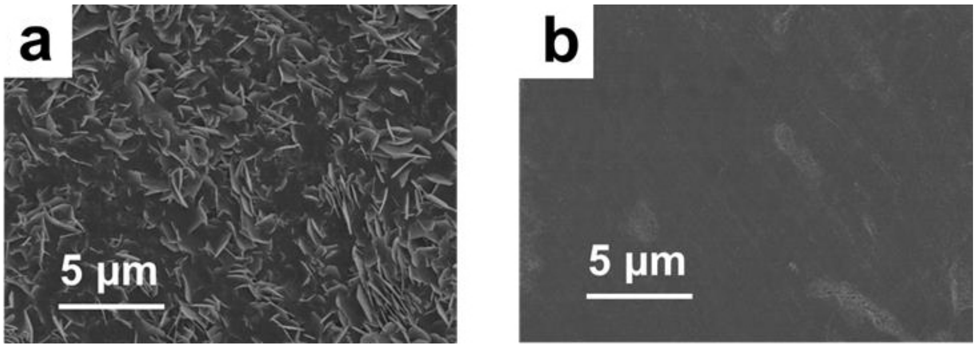

Figure S7. SEM images of (a) bare Zn and (b) Zn-HPAA-1h electrodes after soaking in 2.0 M $ZnSO_4$ for 12 h.