In-situ self-assembled 3D zincophilic heterogeneous metal layer on zinc metal surface for dendrite-free aqueous zinc-ion batteries

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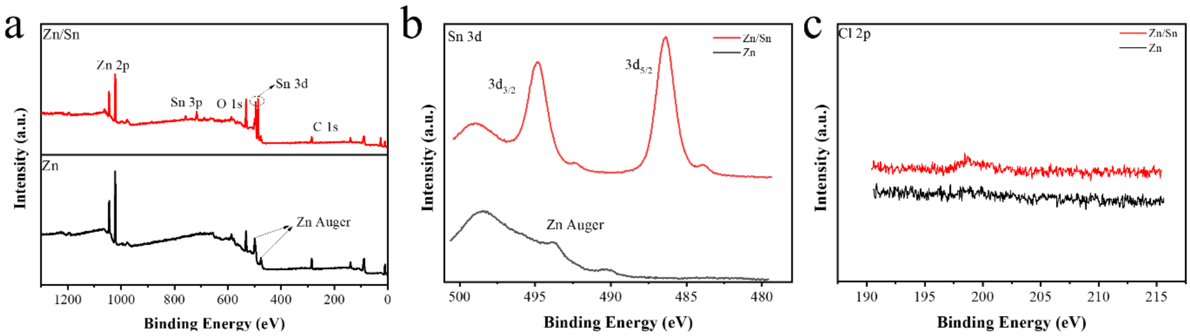


Figure S1. Full XPS spectra (a) and high-resolution XPS spectra of Sn 3d (b) and Cl 2p (c) for the Zn and Zn/Sn anodes.

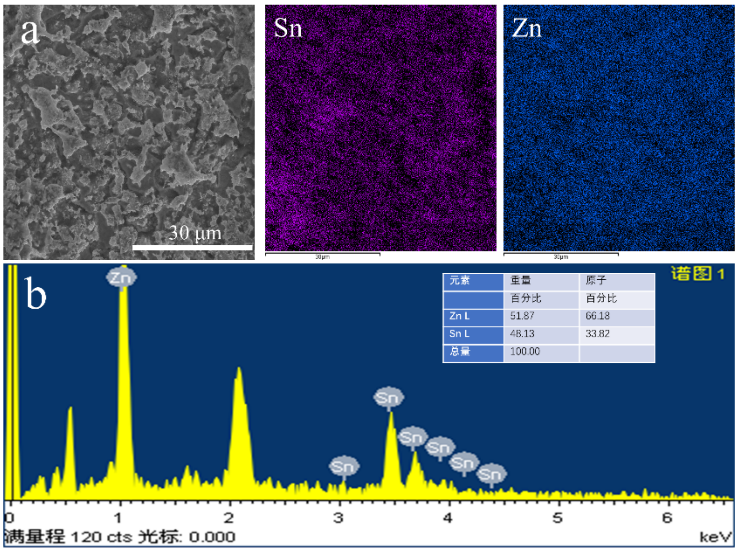


Figure S2. (a) SEM images of Zn/Sn and the corresponding EDS mapping of the Zn/Sn anode. EDS content analysis of Zn and Sn elements (b).

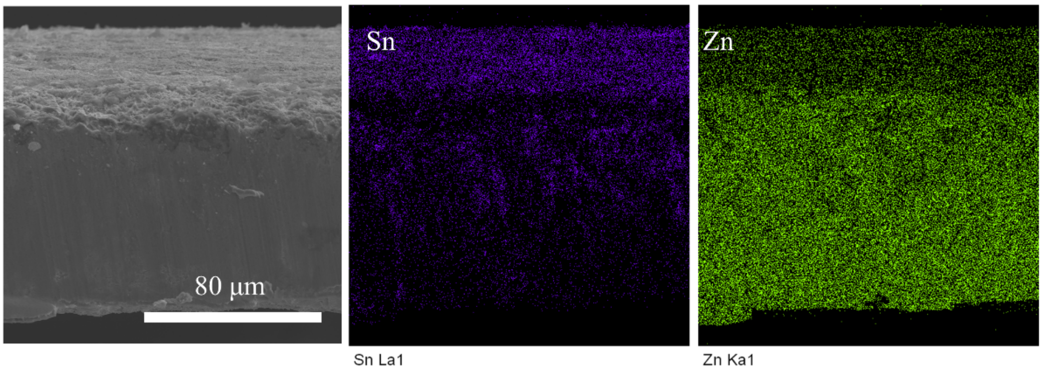


Figure S3. Cross-section SEM image and the corresponding EDS mapping of Zn/Sn anode.

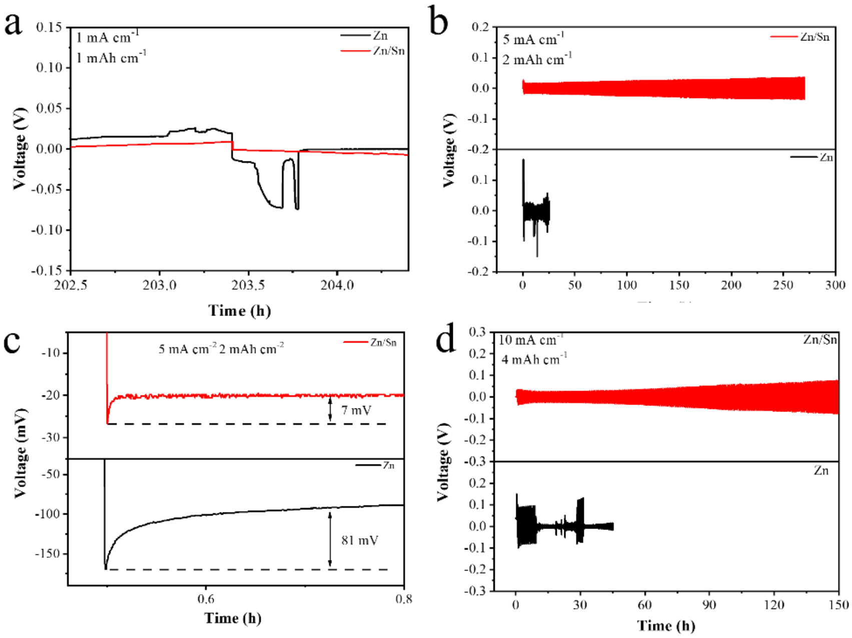


Figure S4. Detailed voltage profiles of above symmetric cells at specific cycling times of 101th at a current density of 1 mA cm^{-2} for 1 h. Zinc stripping/plating in Zn and Zn/Sn symmetric cells at 5 mA cm^{-2} for 0.4 h (b). (c) The nucleation overpotential of the Zn and Zn/Sn symmetric cells at 5 mA cm^{-2} and 2 mA h cm^{-2}. Stripping/plating performance of Zn/Sn and Zn symmetric cells with 4 mAh cm^{-2} at 10 mA cm^{-2} (d).

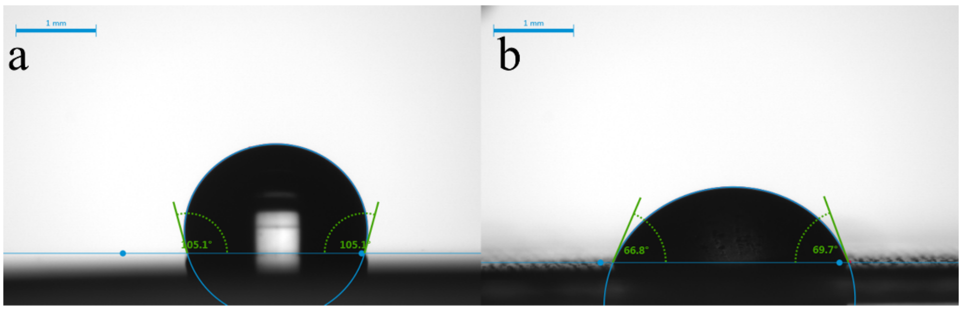


Figure S5. Contact angle measurements of Zn and Zn/Sn anode.

Table S1 The performance comparison of the similar anode materials reported in literatures.

Anode materials	Voltage hysteresis	Lifespan	Ref.
rGO coated zinc foil	≈20 mV (1 mA cm^{-2})	300 h (1 mA cm^{-2}, 1 mAh cm^{-2})	[1]

Carbon fiber framework	30 mV (1 mA cm−2)	350 h (1 mA cm^{-2}, 1 mAh cm^{-2})	[2]
Ultrathin TiO_2-coated zinc anode	57 mV (1 mA cm^{-2})	150 h (1 mA cm^{-2}, 1 mAh cm^{-2})	[3]
CNT scaffold-stabilized Zn anodes	36 mV (0.1 mA cm^{-2})	1800 h (0.1 mA cm^{-2}, 0.5 mAh cm^{-2})	[4]
Nanoporous $CaCO_3$-coated zinc anode	80 mV (0.25 mA cm^{-2})	836 h (0.25 mA cm^{-2}, 0.05 mAh cm^{-2})	[5]
Kaolin coated zinc foil	≈70 mV (4.4 mA cm^{-2})	800 h (4.4 mA cm^{-2}, 1.1 mAh cm^{-2})	[6]
Polyamide layer/zinc foil	100 mV (0.5 mA cm^{-2})	8000 h (0.5 mA cm^{-2}, 0.25 mAh cm^{-2})	[7]
PAM/Zinc plated copper mesh	25 mV (0.2 mA cm^{-2})	350 h (0.2 mA cm^{-2}, 1 mAh cm^{-2})	[8]
$Ti_3C_2T_x$ MXene@Zn Paper	83 mV (1 mA cm^{-2})	84 h (1 mA cm^{-2}, 1 mAh cm^{-2})	[9]
Al_2O_3-coated zinc anode	36.5 mV (1 mA cm^{-2})	500 h (1 mA cm^{-2}, 1 mAh cm^{-2})	[10]
3D flexible carbon nanotubes	27 mV (2 mA cm^{-2})	200 h (1 mA cm^{-2}, 2 mAh cm^{-2})	[11]
Eutectic $Zn_{88}Al_{12}$ alloys	≈20 mV (0.5 mA cm^{-2})	2000 h (0.5 mA cm^{-2}, 0.5 mAh cm^{-2})	[12]
3D Zn/Sn anode	30 mV (1 mA cm^{-2})	900 h (1 mA cm^{-2}, 1 mAh cm^{-2})	**Our work**

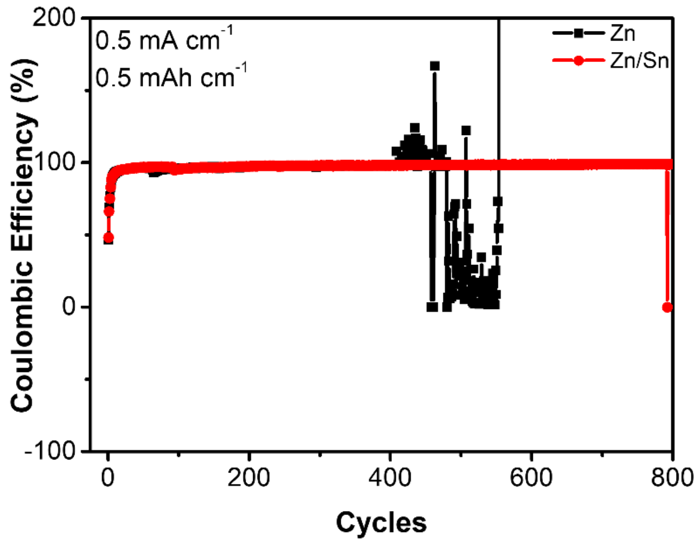


Figure S6. CE of Zn plating/stripping in the Zn-Cu and Zn/Sn-Cu half-cells at 0.5 mA cm^{-2}.

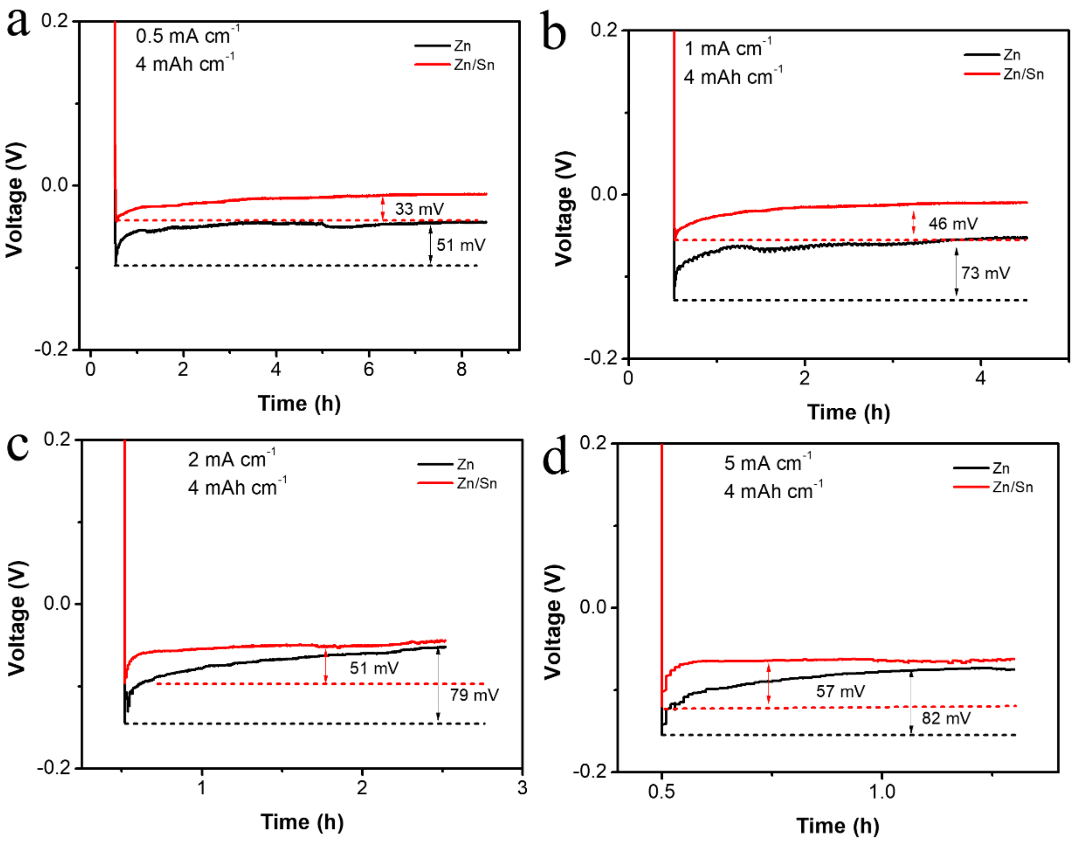
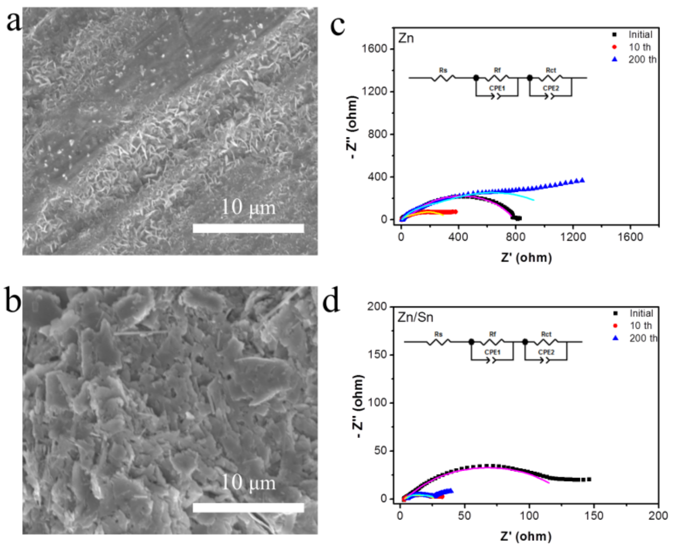


Figure S7. The nucleation overpotential on Cu matrix for Zn and Zn/Sn electrode at a different density of 0.5 mA cm^{-2}(a), 1 mA cm^{-2}(b), 2.0 mA cm^{-2}(c), and 5.0 mA cm^{-2}(d).



e

	Rs	Rf	Rct
Zn-0	3.62	45.21	741.30
Zn-10	4.22	57.73	274.70
Zn-200	5.26	197.50	925.80

f

	Rs	Rf	Rct
Zn/Sn-0	3.05	1.35	126.90
Zn/Sn-10	3.39	6.89	16.69
Zn/Sn-200	4.67	9.51	14.64

Figure S8. SEM images of Zn (a) and Zn/Sn (b) deposition at 1 mA cm^{-1} for 50 cycles. Electrochemical impedance spectroscopy measurements of Zn (c) and Zn/Sn (d) symmetric cells after different numbers of cycles and the corresponding fitting data (e-f).

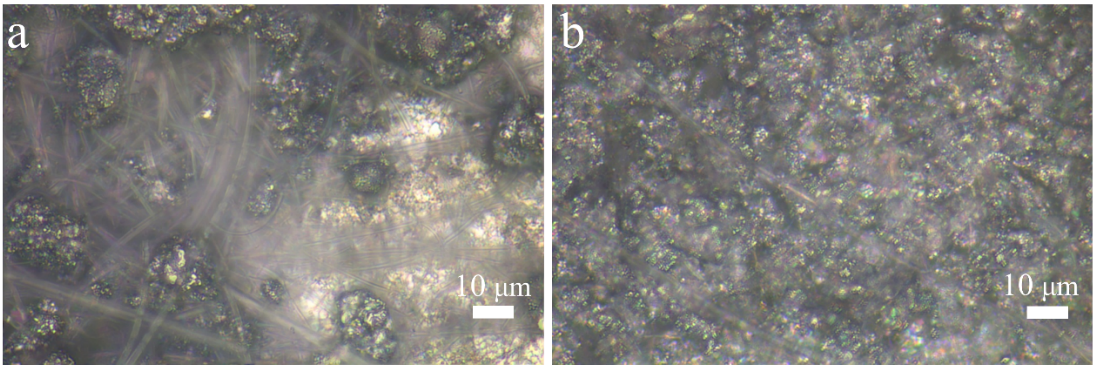


Figure S9. The optical microscopy images of Zn^{2+} ions deposition morphology on bare Zn(a) and (b) Zn/Sn symmetrical cells at a current density of 1 mA cm^{-2}.

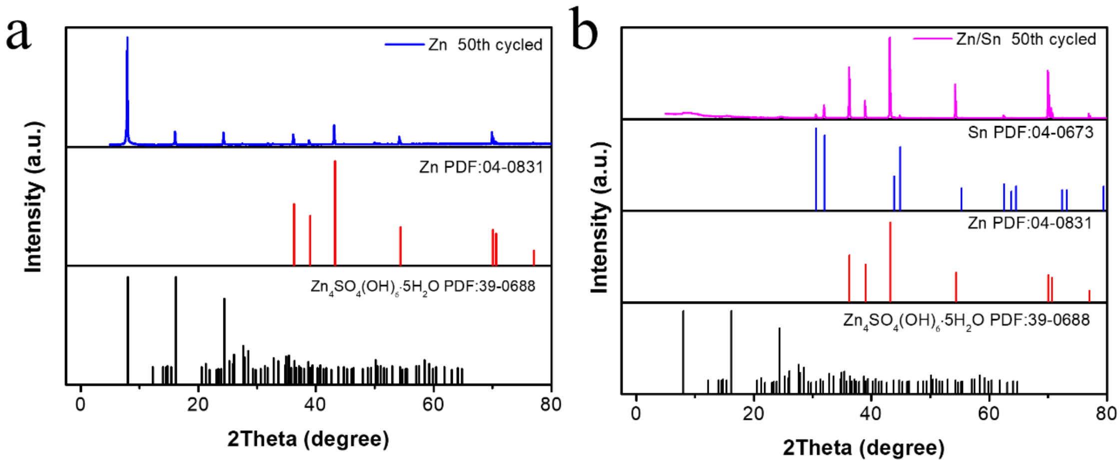


Figure S10. XRD patterns of cycled Zn and Zn/Sn electrodes in 2 M $ZnSO_4$.

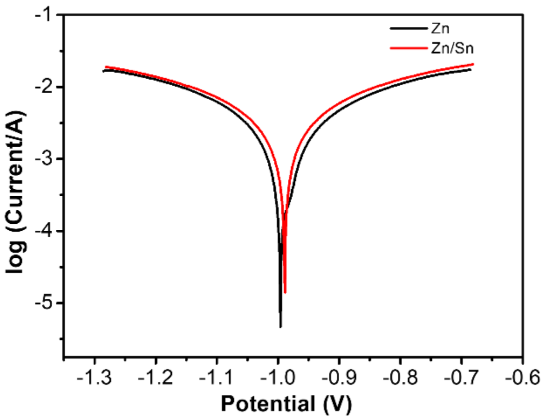


Figure S11. Tafel curves of bare Zn and Zn/Sn in 2 M $ZnSO_4$ electrolyte.

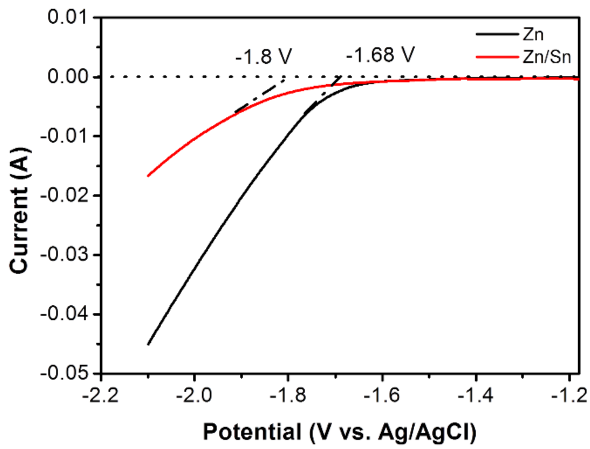


Figure S12. LSV curves of Zn and Zn/Sn electrode in 1 M aqueous Na_2SO_4 electrolyte at a scan rate of 5 mV s^{-1}.

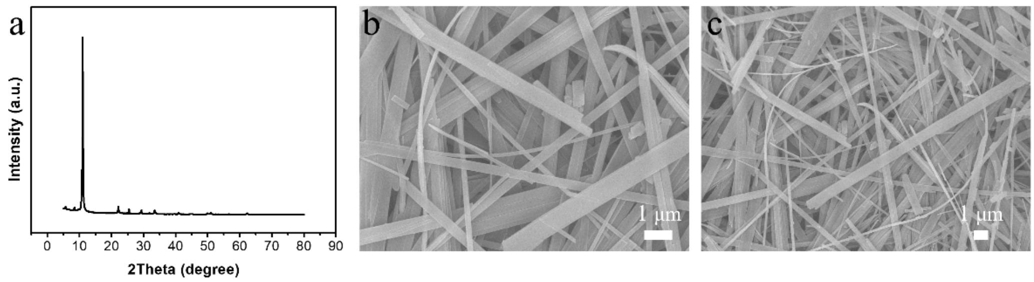


Figure S13. XRD pattern (a) and SEM images (b-c) of $CaV_6O_{16}\cdot 3H_2O$ obtained in a similar way as reported[13].

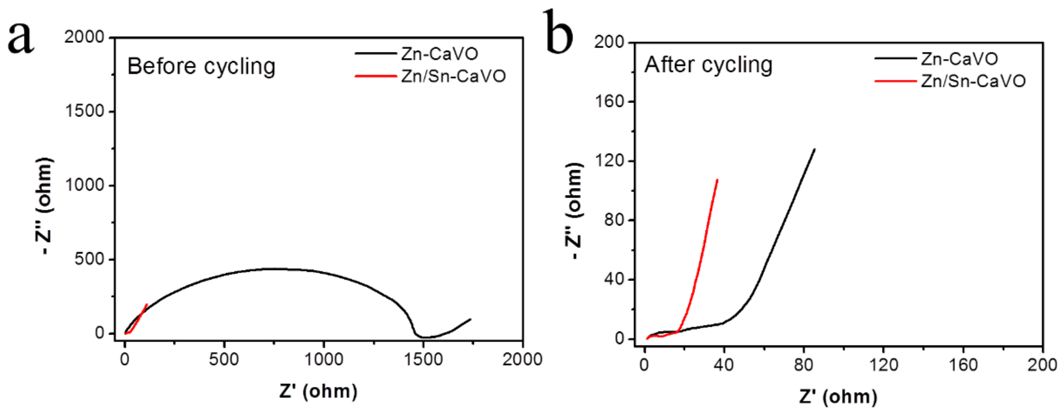


Figure S14. EIS spectra of Zn-CaVO and Zn/Sn-CaVO cells before the test and after 5th cycles.

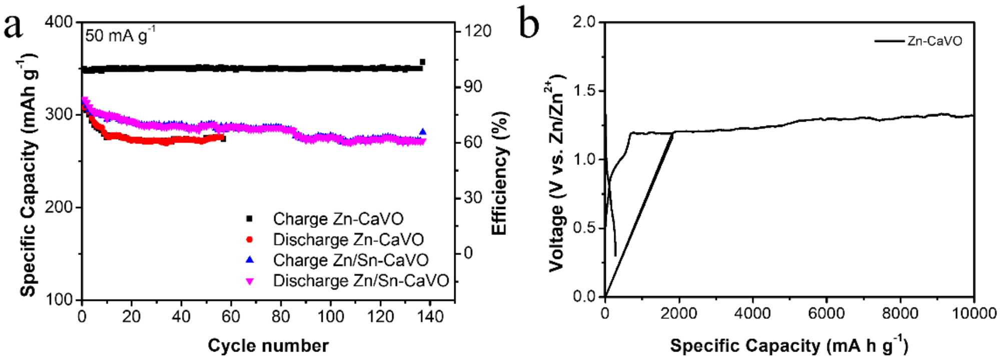


Figure S15. (a) Cycling performance of Zn-CaVO and Zn/Sn-CaVO cells at a current density of 50 mA g^{-1}. (b) The charge and discharge curve of the last lap of Zn-CaVO cell.

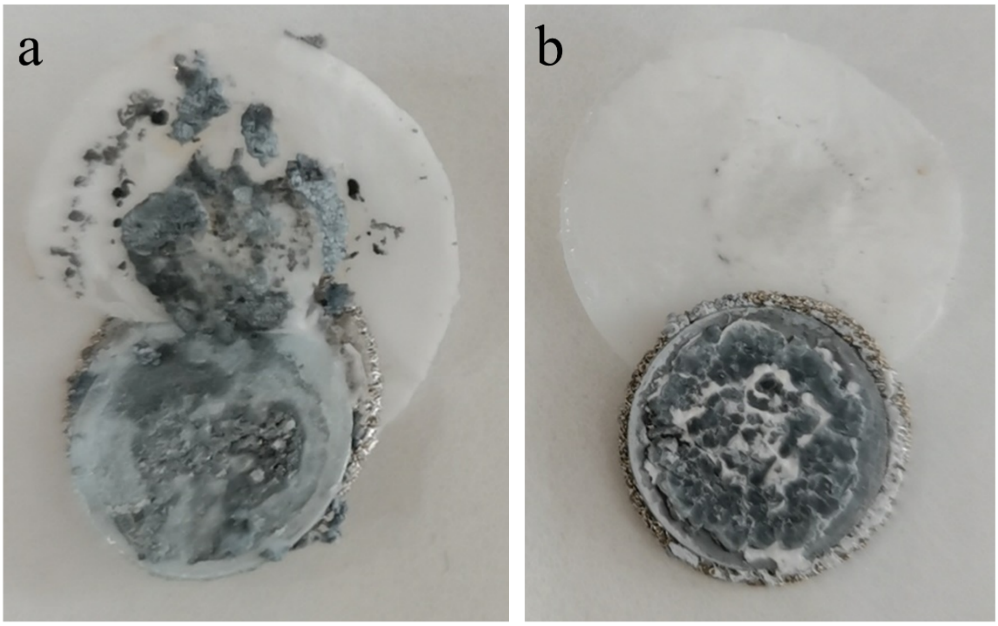


Figure S16. Optical image of bare Zn (a) and Zn/Sn (b) electrodes electrode after 800 cycles in full battery.

References

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