

Supporting Information

Pulsed Current Constructs 3DM Cu/ZnO Current Collector Composite Anode for Free-Dendritic Lithium Metal Batteries

Table S1. The supplier of raw materials.

Raw Materials	Supplier
Copper Mesh	Anping Hang Eagle Wire Mesh Products Co., Ltd
Lithium Foil	Tianjin Zhongneng Lithium Industry Co., Ltd
Znic Foil	Xinye Electronic Materials Factory
MnSO ₄ ·H ₂ O	Shanghai EPB Chemical Reagent Co., Ltd
ZnSO ₄ ·H ₂ O	Sinopharm Chemical Reagent Co., Ltd

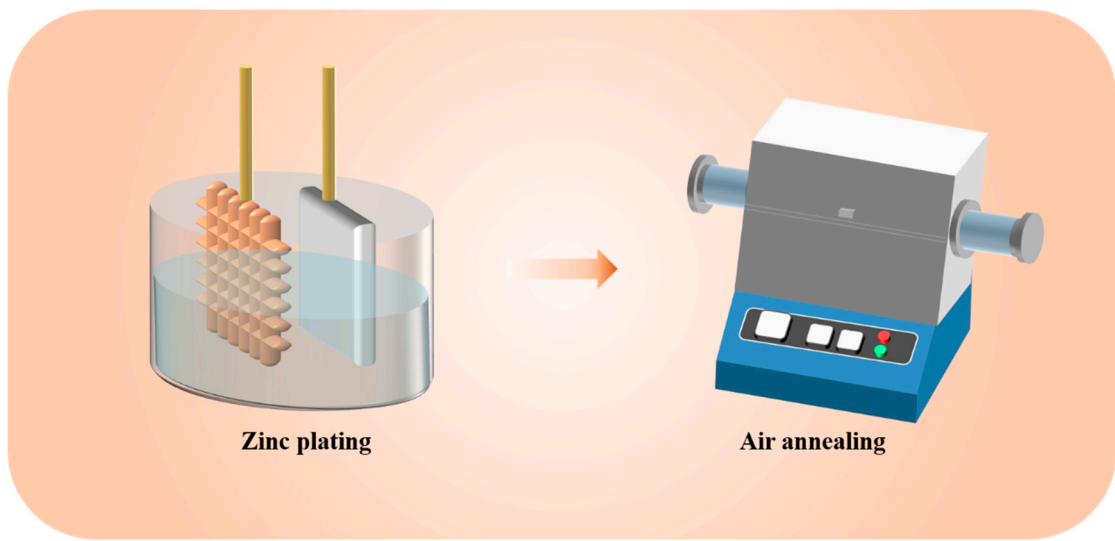


Figure S1. Schematic diagram of preparation of zinc oxide coating.

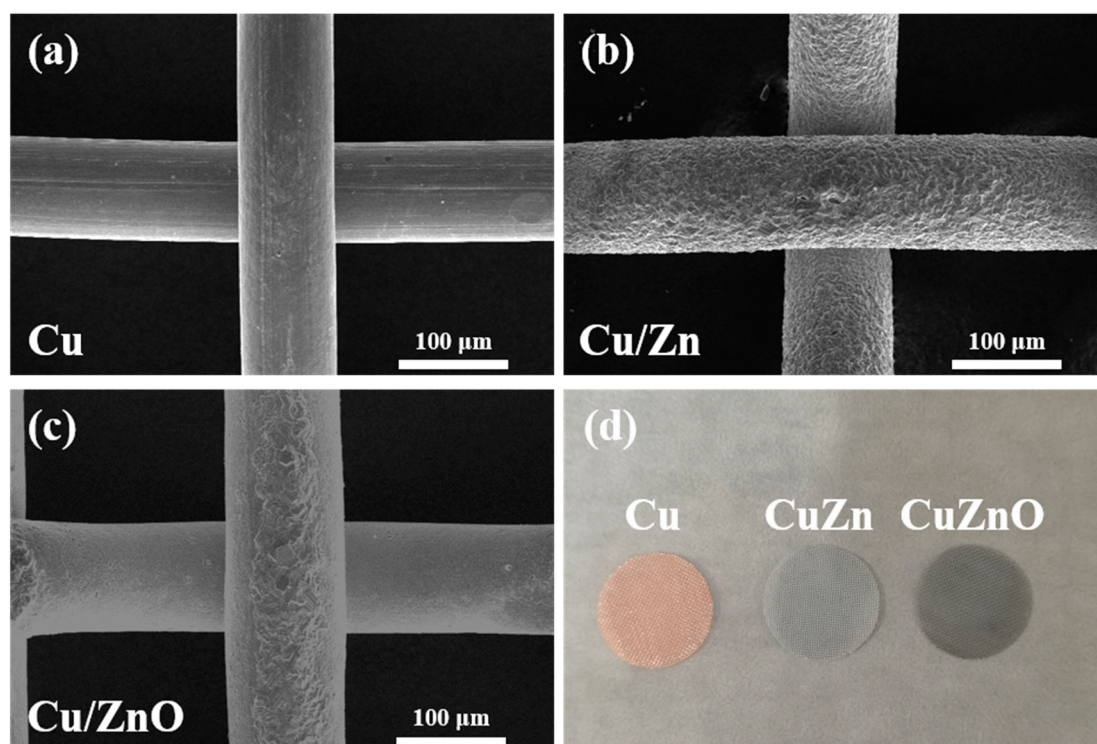


Figure S2. SEM image of (a) 3DM Cu, (b) 3DM Cu/Zn, (c) 3DM Cu/ZnO and (d) Sample comparison.

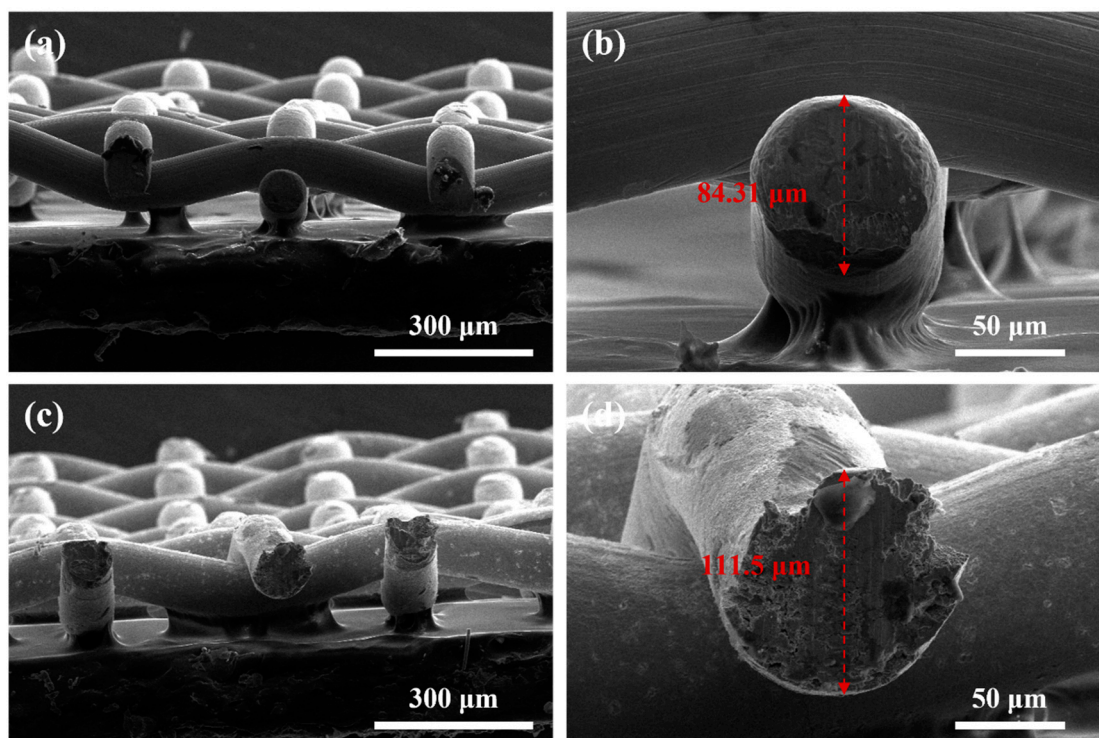


Figure S3. The cross-sectional SEM images of (a,b) 3DM Cu and (c,d) Cu/ZnO.

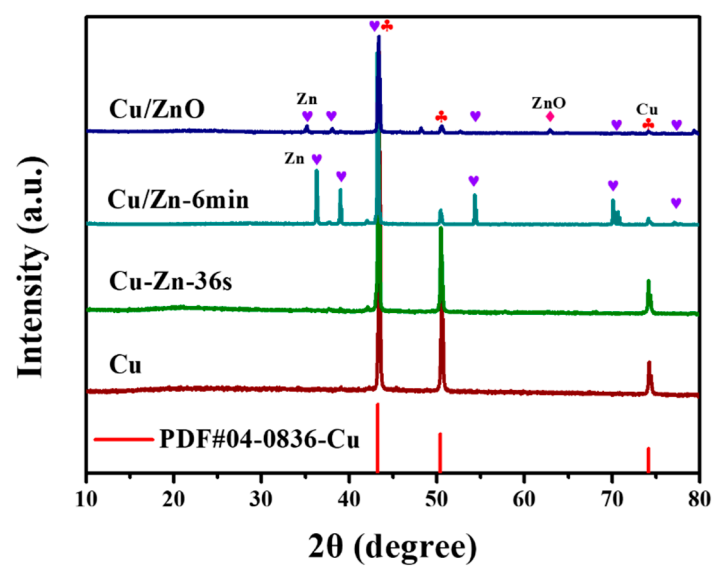


Figure S4. XRD pattern of 3DM Cu, 3DM Cu/Zn and 3DM Cu/ZnO.

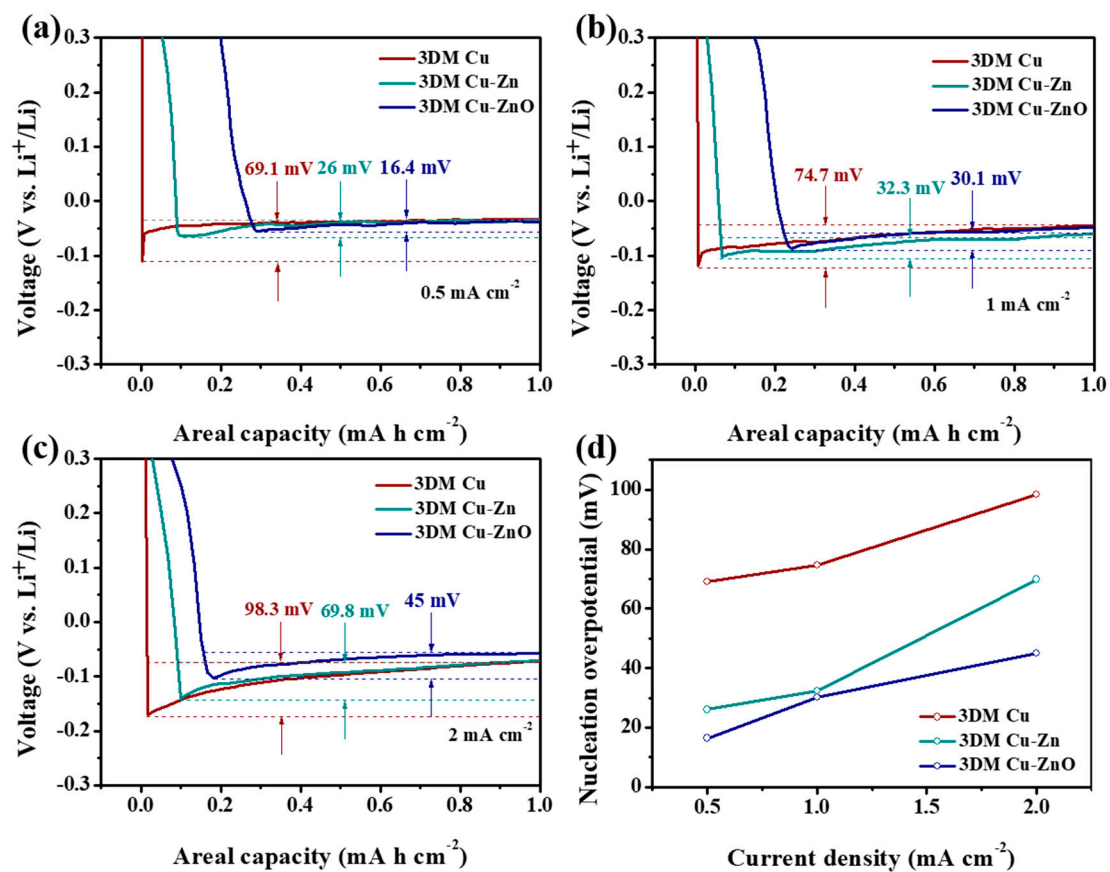


Figure S5. The nucleation of 3DM Cu, 3DM Cu/Zn and 3DM Cu/ZnO at current density of 0.5, 1 and 2 mA cm^{-2} .

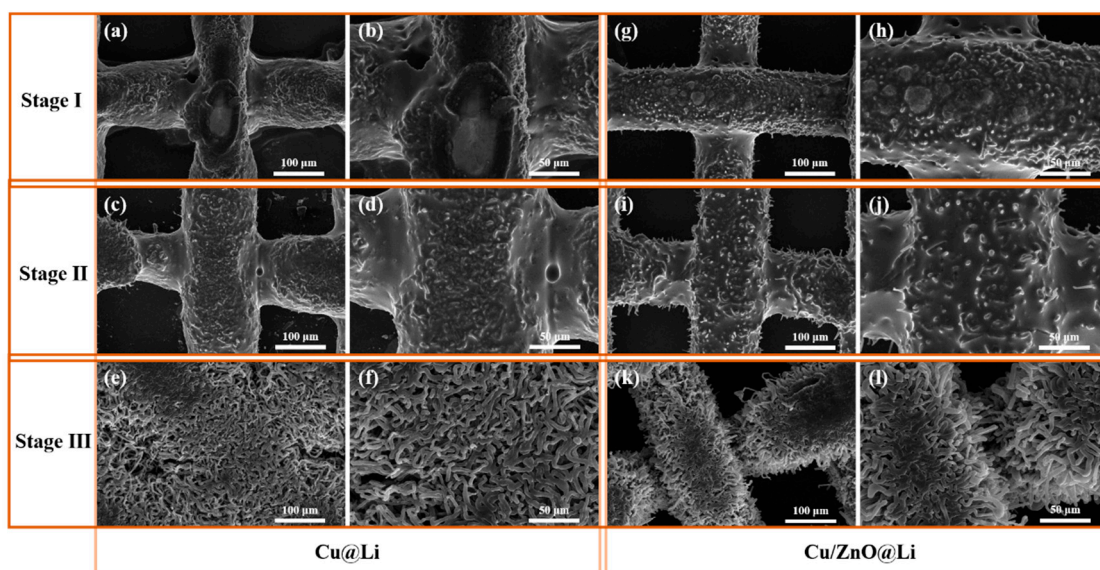


Figure S6. SEM images of (a-f) bare Cu and (g-l) Cu/ZnO mesh current collectors along Li plating of 1 (stage I), 3 (stage II) and 6 mAh cm⁻² (stage III).

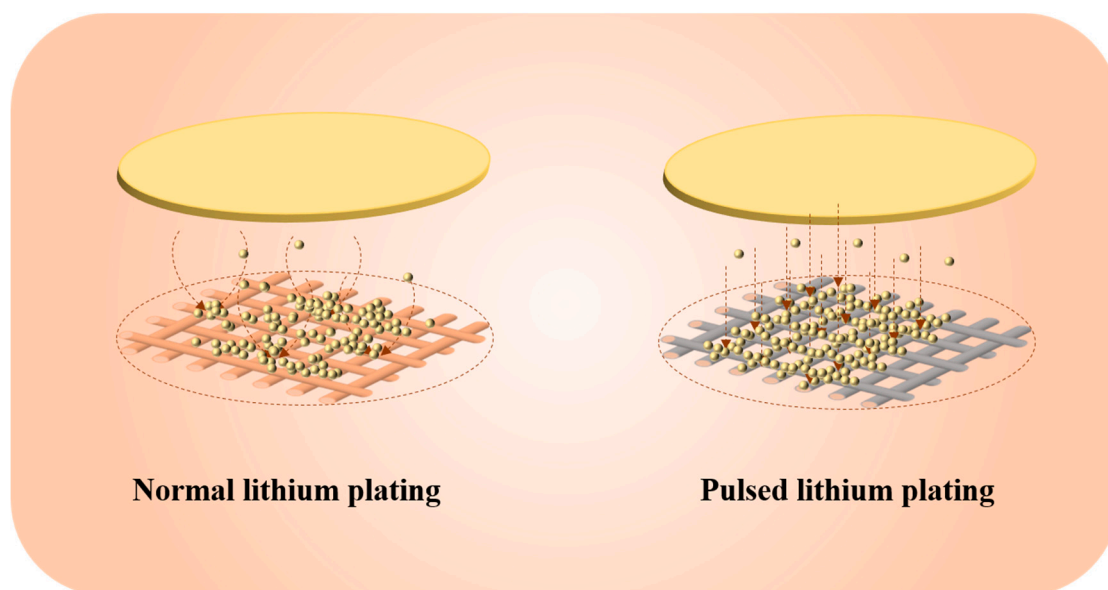


Figure S7. Schematic diagram of lithium electroplating deposition.

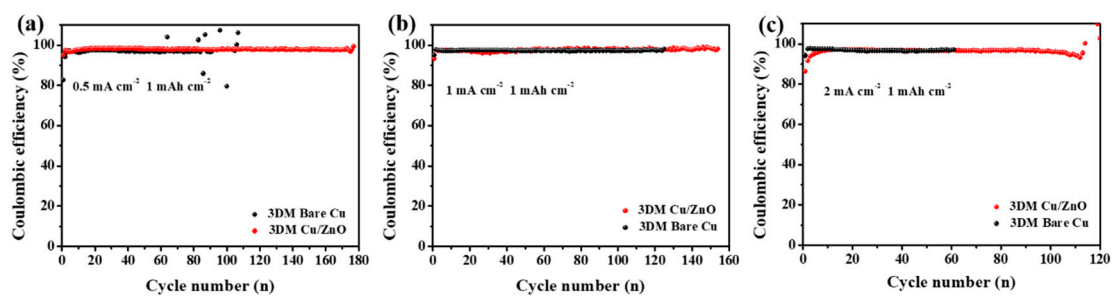


Figure S8. The CE of the 3DM Cu@Li and Cu/ZnO@Li-P composite anode during the Li plating/stripping at different current densities with the capacity of 1 mAh cm⁻².

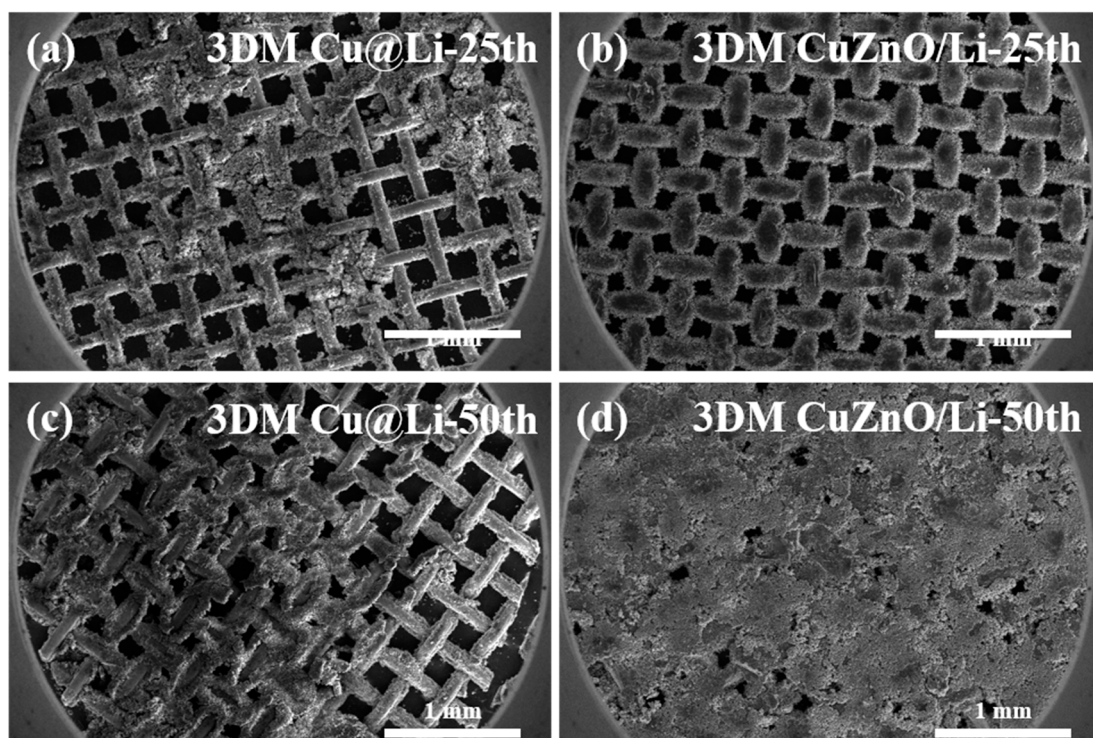


Figure S9. SEM image of after 25th and 50th cycles lithium stripping which the surface of (a, c) 3DM Cu@Li anode and (b, d) 3DM Cu/ZnO@Li-P composite anode.

Table S2. Comparison of the cycling performance of Li metal anodes with decorating ZnO.

Materials	Current density (mA cm^{-2})/Areal capacity (mAh cm^{-2})	Cycling stability	Electrolyte
MCuF@Li ^[1]	1/2	600 h	1 M LiTFSI in DOL/DME (v/v=1) with 1 wt% LiNO ₃
Li@LZMNF ^[2]	1/1	1200 h	1 M LiTFSI in DOL/DME (v/v=1) with 1 wt% LiNO ₃
Li@MXene/ZnO ^[3]	1/1	250 h	1 M LiTFSI in DME with 1% FEC
LiSZ ^[4]	1/1	900 h	1 M LiTFSI in DOL/DME (v/v=1) with 2 wt% LiNO ₃
Cu/ZnO@Li-P (This work)	1/1	1500 h	1 M LiTFSI in DOL/DME (v/v=1) with 0.2 M LiNO ₃

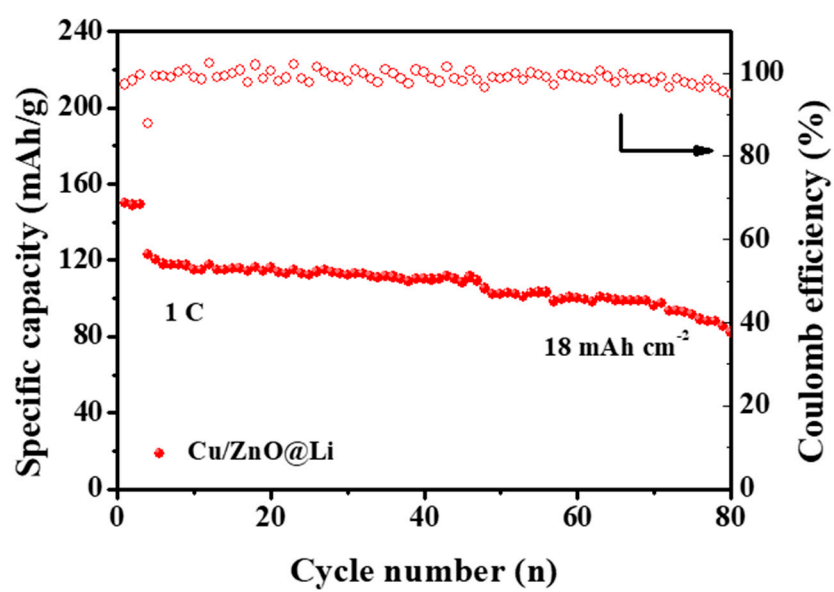


Figure S10. Cycling performance of full-cells with 3DM Cu/ZnO@Li-P composite anode at 1 C with three activation cycles at 0.1 C in the beginning.

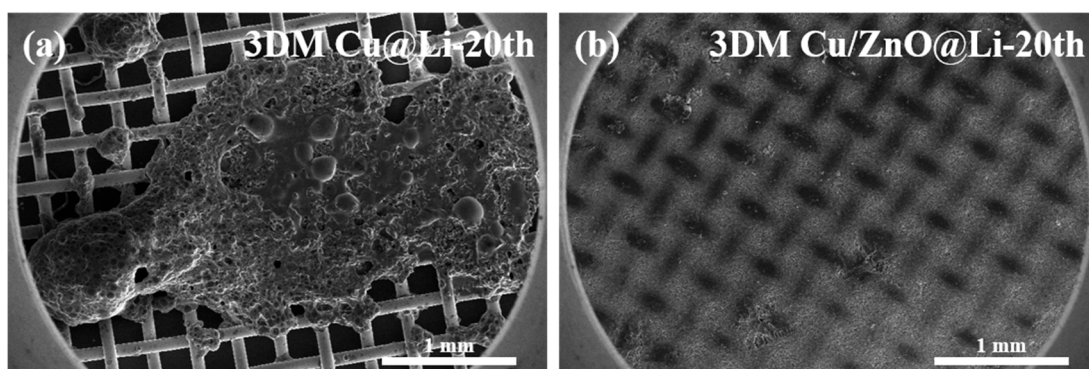


Figure S11. SEM images of (a) 3DM Cu@Li and (b) 3DM Cu/ZnO@Li-P composite anodes after 20 cycles lithium stripping in a full-cell.

Table S3. Electrochemical parameters of the equivalent circuit for cells constructed from the prepared electrodes.

Sample	R_1 (Ω)	R_2 (Ω)	CPE (F)	W (Ω S ^{-0.5})
3DM Cu@Li before cycling	6.122	25.49	1.14×10^{-5}	20.38
3DM Cu@Li after cycling	5.627	110.3	1.07×10^{-5}	77.25

Sample	R_1 (Ω)	R_2 (Ω)	R_3 (Ω)	CPE ₁ (F)	CPE ₂ (F)	W (Ω S ^{-0.5})
3DM Cu/ZnO@Li before cycling	5.794	56.55	10.19	5.45×10^{-6}	3.64×10^{-4}	30.81
3DM Cu/ZnO@Li after cycling	4.165	4.004	10.57	9.36×10^{-4}	1.44×10^{-2}	1819

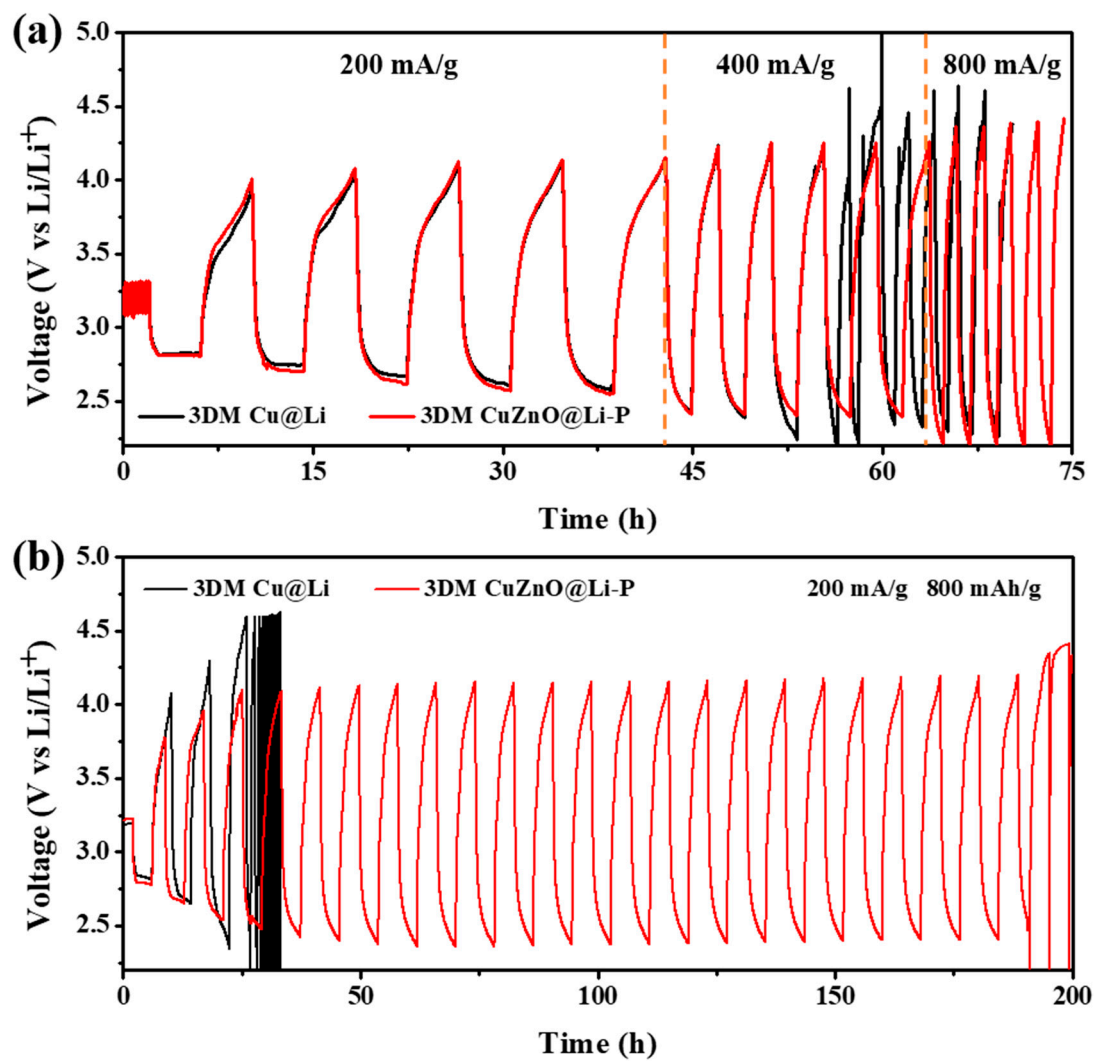


Figure S12. (a) Rate performance and (b) cycle performance of lithium-oxygen batteries constructed with 3DM Cu/ZnO@Li-P composite anode.

Reference

- [1] Zhou, Y.; Zhao, K.; Han, Y.; Sun, Z.H.; Zhang, H.T.; Xu, L.Q.; Ma, Y.F.; Chen, Y.S. A Nitrogen-Doped-Carbon/ZnO Modified Cu Foam Current Collector for High-Performance Li Metal Batteries. *J. Mater. Chem. A*. **2019**, 7, 5712.
- [2] Sun, C.Z.; Li, Y.P.; Jin, J.; Yang, J.H.; Wen, Z.Y. ZnO Nanoarray-Modified Nickel Foam as A Lithiophilic Skeleton to Regulate Lithium Deposition for Lithium-Metal Batteries. *J. Mater. Chem. A*. **2019**, 7, 7752.
- [3] Shen, Y.X.; Pu, Z.Y.; Zhang, Y.R.; Chen, Y.; Zhang, H.; Wang, N.T.; Qiu, H.L.; Li, Y.M. MXene/ZnO Flexible Freestanding Film as A Dendrite-Free Support in Lithium Metal Batteries. *J. Mater. Chem. A*. **2022**, 10, 17199.
- [4] Ni, C.K.; Mao, J.T.; Cheng, Z.L.; Pan, P.; Jiang, L.Y.; Wang, Z.X.; Zhang, M.M.; Zhang, Y.R.; Xing, Y.S.; Zeng, Y.; Chen, Q.; Hu, Y. Si/ZnO Framework: 3D Lithiophilic Structure for Dendrite-Free Lithium Metal Batteries. *J. Alloy. Compd.* **2021**, 25, 876.