

Supplementary Information

3D soft lithiophilic current collector of Ag-CuNW/MWCNT@melamine foam enables dendrite-free lithium metal anode

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Experimental section

Preparation of CuNW: CuNW were synthesized by the polyol reflux method: CuCl (2 mmol) and NH₄Cl (0.3 mmol) were added to ethylene glycol (30 mL) which was stirred for 30 min until the solution became light green. Oleylamine (6 mmol) was then added dropwise to the solution. The solution was transferred to a 100 mL round-bottomed flask, stirred and heated to 110 °C for 10 minutes, then the reaction was heated to 200 °C, refluxed and held for 20 minutes. When the reaction was complete, the product was rapidly cooled with cold water. Finally, the sample was washed and dispersed in cyclohexane.

Preparation of 3D soft lithiophilic current collector: The prepared CuNW and MWCNT were homogeneously mixed at a mass ratio of 3:1, and adjusted to a concentration of 10 mg mL⁻¹. Additionally, the MF (thickness of 1 mm, diameter of 12 mm) was completely immersed in the mixed slurry and removed after half a minute followed by repeated squeeze, and then vacuum drying at 60 °C for 12 h, to obtain the CuNW/MWCNT@MF composites. It was then immersed in a 1 mM AgNO₃ solution for 45 s followed by vacuum drying at 60 °C for 1 h.

Finally, the 3D lithiophilic current collector was obtained by heating at 245 °C for 1 h in a glycerol atmosphere.

Characterizations

X-ray diffraction (XRD) measurements were carried out on an X-ray diffractometer (Rigaku MiniFlex 600) using Cu K α radiation ($\lambda = 0.15405$ nm) in the 2θ range from 5° to 85°. The morphology and microstructure were characterized by SEM (Regulus-8100). N₂ adsorption-desorption isotherms and pore size distribution measurements were performed on a fully automated Specific Surface and Porosity (AUTOSORB IQ, USA).

Electrochemical measurements

CR2032-type coin cells were assembled in an Ar-filled glove box ($O_2 < 0.01$ ppm, $H_2O < 0.01$ ppm) using Ag-CuNW/MWCNT@MF or Cu foil as the working electrode and Li foil as the reference and counter electrode. The electrolyte used here was 1 M LiPF₆ solvated in 1:1:1 ethylene carbonate/dimethyl carbonate/diethyl carbonate (EC/DMC/DEC, MTI Inc.). A celgard 2500 membrane was used as the separator to evaluate the electrochemical performance. All cells were left to stand for 12 h after assembly to allow the electrolyte to fully contact the material. These cells were first electrochemically activated for 5 cycles at 50 μ A over a voltage range of 0 to 1 V. For the CE tests, a certain amount of Li was first plated on the current collector and then stripped away 1 V (vs Li⁺/Li) during each cycle. Symmetrical cells were fabricated by pre-deposited 60 mAh cm⁻² Li at 1 mA cm⁻² on the current collector then charged/discharged at various current densities and capacities in each cell. Electrochemical impedance spectra (EIS) measurements were performed on a PGSTAT 302N electrochemical workstation (Aptar, Switzerland) with a chosen amplitude of 5 mV and a test frequency range of 0.01~10⁵ Hz.

For full cell tests, LiFePO₄ (LFP) electrodes were used as cathodes coupled to Ag-CuNW/MWCNT@MF-Li (60 mAh cm⁻² deposited Li) and Cu foil-Li (6 mAh cm⁻² deposited Li) anode. The anode slurry was made by mixing LFP (active material): C (conductive carbon black): PVDF (binder) in N-methyl pyrrolidone (NMP) at a mass ratio of 8:1:1. Then, the homogeneously mixed slurry was coated on aluminum foil sheets using an automatic coating

and drying machine. After coating, the sample was dried in a vacuum oven at 80 °C for 12 h and then cut into round pole pieces with a diameter of 12 mm. The mass loading of the LFP cathode was approximately 2.5 mg cm⁻². The full cell was operated at a voltage range of 2.5~4.2 V. To standardize the tests, 50 μL of electrolyte was used for each button cell.

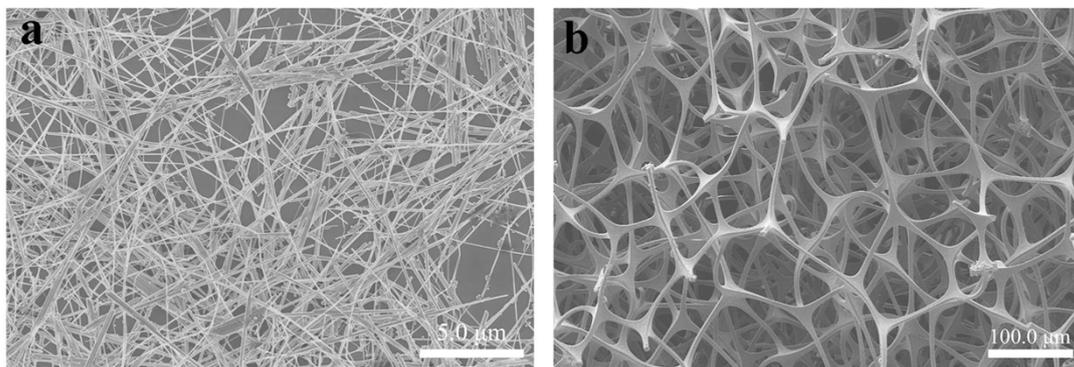


Fig. S1. SEM of (a) CuNWs and (b) bare MF.

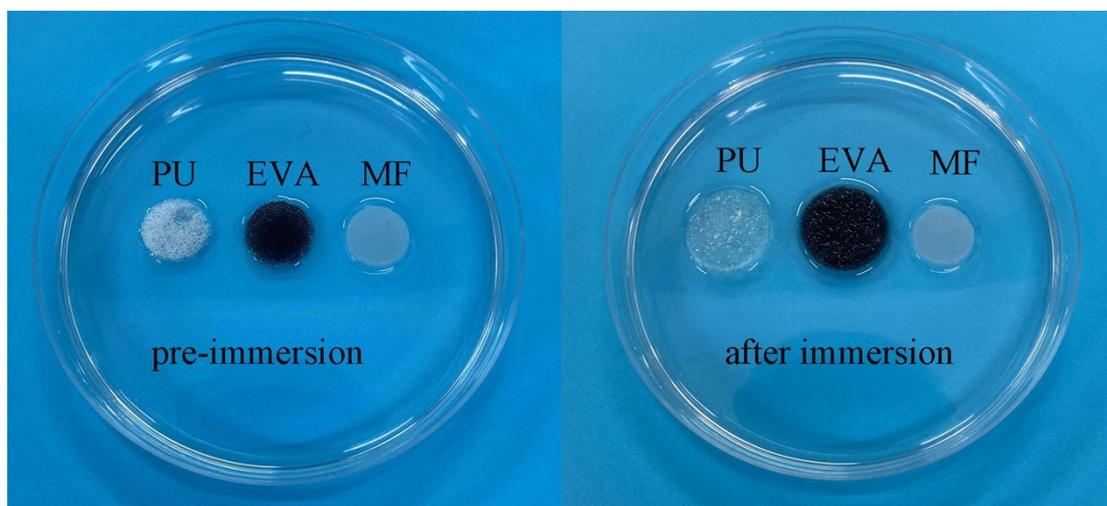


Fig. S2. Immersion diagram of different foams in electrolyte solution.

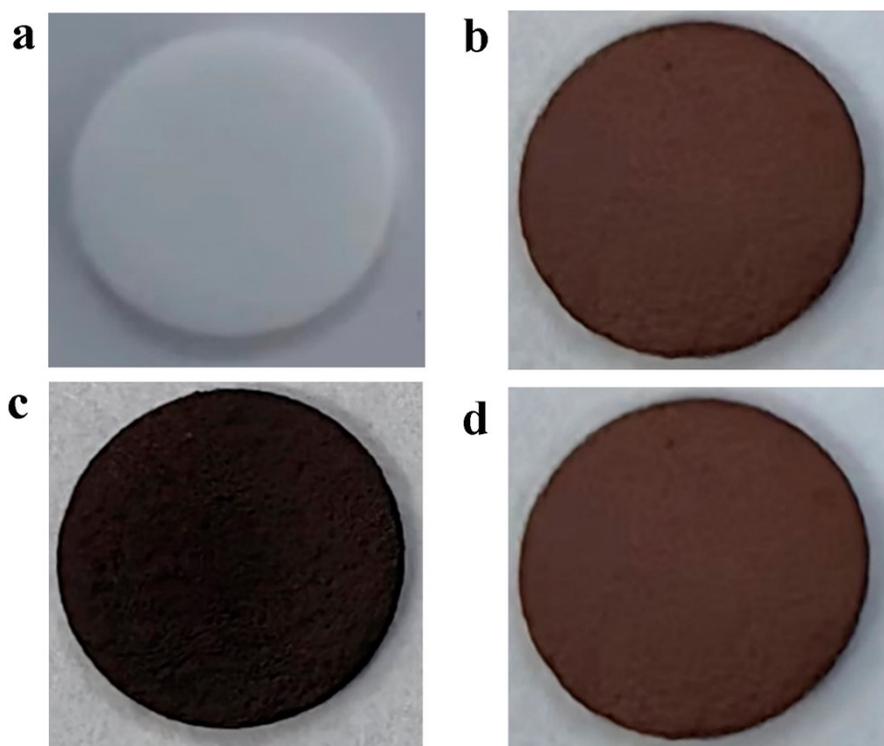


Fig. S3. (a) Bare MF, (b) MF loaded with CuNW/MWCNT composites, (c) immersed in AgNO_3 solution, and (d) reduced by glycerol atmosphere.

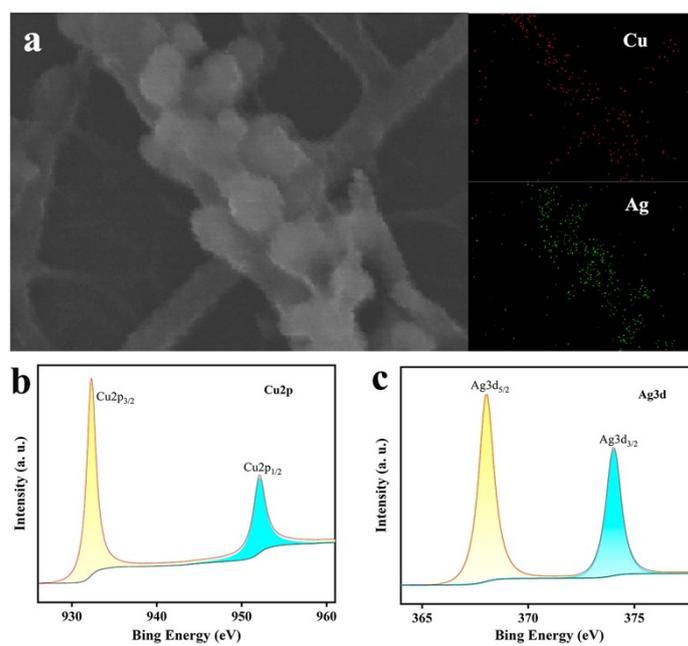


Fig. S4. (a) EDS elemental mappings of Cu and Ag. XPS images of Ag-CuNW/MWCNT@MF (b)Cu2p, (c)Ag3d.



Fig. S5. Resistance test of Ag-CuNW/MWCNT@MF.



Fig. S6. Ag-CuNW/MWCNT@MF electrode after deposition of 90 mAh cm^{-2} Li.

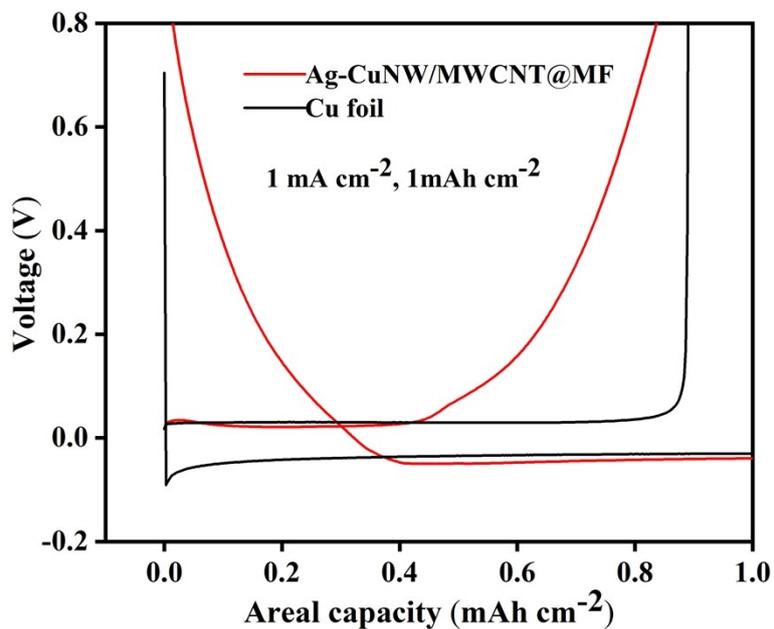


Fig. S7. The first charge/discharge curves of the half-cells of Li||Ag-CuNW/MWCNT@MF and Li||Cu foil.

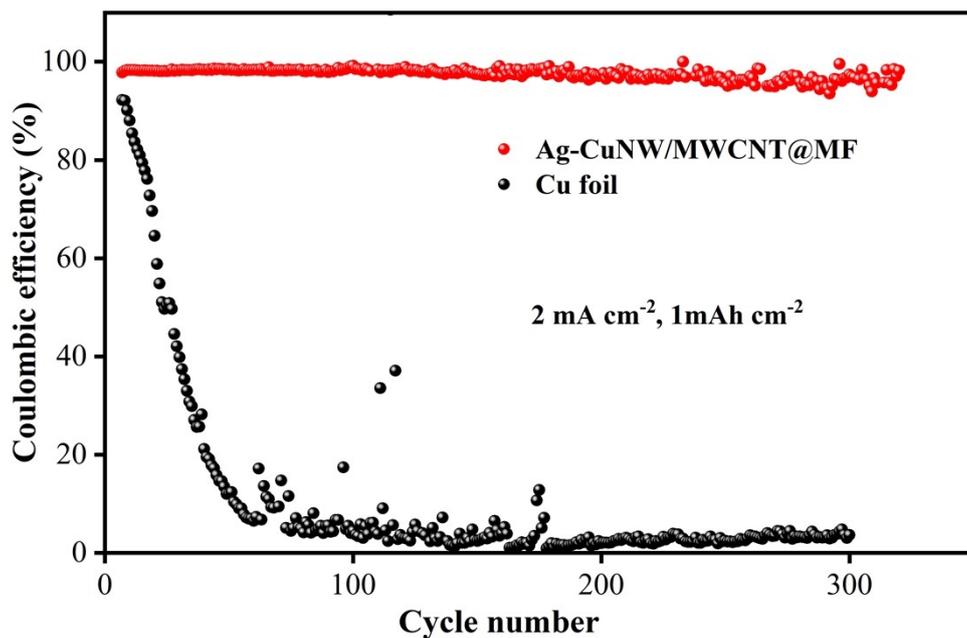


Fig. S8. Comparative CE plots of Ag-CuNW/MWCNT@MF and Cu foils at 2 mA cm^{-2} and 1 mAh cm^{-2} .

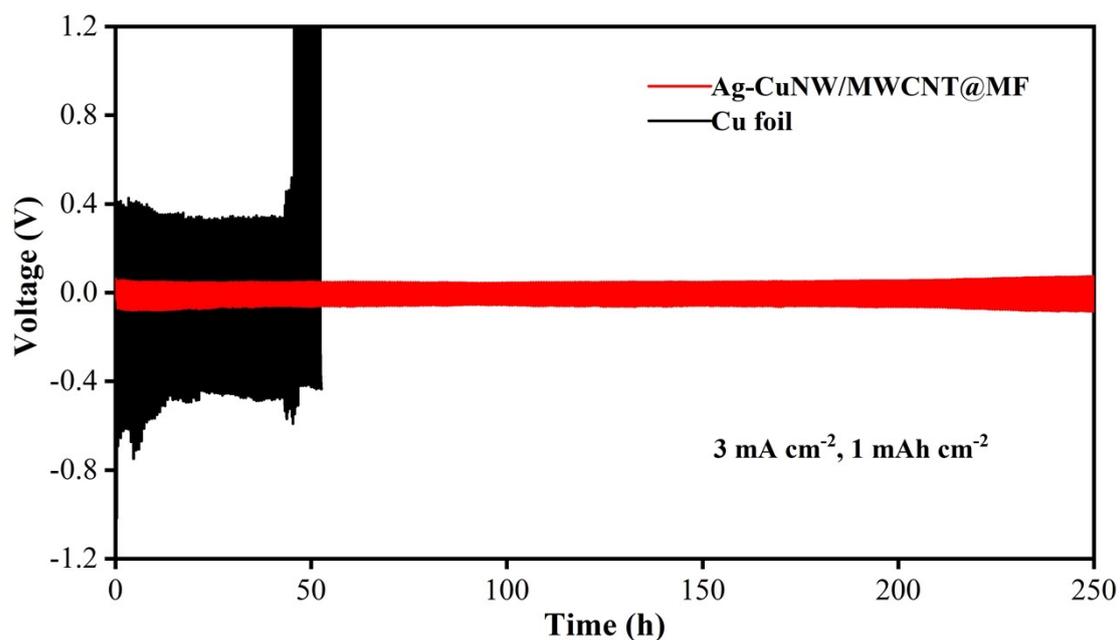


Fig. S9. Voltage-time curves of Ag-CuNW/MWCNT@MF-Li and Cu foil-Li symmetric cells at 3 mA cm^{-2} and 1 mAh cm^{-2} .

Table S1. Comparison of CE at 1mA cm^{-2} and 1mAh cm^{-2} for different current collectors.

Current collector	Number of cycles	Average CE(%)	Ref.
Ag-CuNW/MWCNT@MF	500	98.5	This work
Cu@Cu₃P mesh	500	97.74	46
ZMNF	300	98	47
h-CoCoPBAs@SWCNT	200	96	48
Ni₃S₂@Ni foam	400	98.3	49
NCNF	150	96	50
NCA	100	96.2	51
ZIF-8@RGO	350	98.48	52
GO-Zn/Cu	200	98	53