Supporting Information

Lithiophilic liquid metal layer induced lithium plating/stripping in 3D

Cu matrix to mitigate lithium dendrites and volume expansion

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Volume of the porous structure (Vp): The theoretical pores-utilization rate calculation is as follows: take the electrodes with 1.4 cm diameter and 0.01cm height as an example. First, the theoretical volume of Cu foam (V₁) is calculated by the multiplication of bottom area and height. The actual volume (V₂) of pure Cu is obtained by dividing the average weight of the Cu foam (M₂) with the Cu density (ρ_{Cu}). The volume of the porous structure (Vp) is calculated by subtracting V1 and V2:

 $Vp=V_1-V_2=S\cdot h-M_2/\rho_{Cu}$

$$1.5386 \text{ cm}^2 \times 0.01 \text{ cm} - 0.1 \text{ g} \div 8.96 \text{ g} \text{ cm}^{-3} \approx 4.2 \cdot 10^{-3} \text{ (cm}^3)$$

The area specific capacity of theoretical lithium storage (C_{LiI}): According to the above calculation results, Vp≈4.2·10⁻³ cm³, therefore, the specific areal capacity of theoretical lithium storage (C_{Li1}) with porous structures can be calculated according to the density, area and specific capacity:

 $m_{Li2} = V p \cdot \rho_{Li}$

$4.2 \cdot 10^{-3} \text{ cm}^3 \times 0.5324 \text{ g cm}^{-3} \approx 2.23 \cdot 10^{-3} \text{ (g)}$

 $C_{Li1}=m_{Li2}\cdot 3860 \text{ mAh } g^{-1}/S$

$$2.23 \cdot 10^{-3} \text{ g} \times 3860 \text{ mAh g}^{-1} \div 1.5386 \text{ cm}^2 \approx 5.6 \text{ (mAh cm}^{-2)}$$

Table S1. Equivalent-circuit fitting and detailed analysis on the obtained EIS spectra of different Li capacity MGC anode.

Equivalent circuit	R1 Rsei Rct W1 CPE1 CPE2	
capacity	R _{SEI} (ohm)	Rct (ohm)
0 mAh cm ⁻² (Initial)	6.54	47.01
0.5 mAh cm ⁻²	33.92	25.98
2 mAh cm^{-2}	15.24	11.38
6 mAh cm ⁻²	14.32	7.06
12 mAh cm ⁻²	13.82	6.83
24 mAh cm ⁻²	17.13	7.56
0 mAh cm ⁻² (Stripping)	16.22	27.58



Fig. S1. Scheme of preparation process of LM composite.



Fig. S2. Digital photos showing the diffusion of Au in LM and spontaneous alloying.



Fig. S3. XRD patterns of AuGa₂ formation process.



Fig. S4. Nyquist plots of fresh cells with different current collectors.



Fig. S5. Photos of Cu foam and MGC electrodes before and after Li plating with different capacities.



Fig. S6. Li plating behaviors on MGC and Cu foam electrodes with a capacity of 1 mAh cm^{-2} under 1 mA cm^{-2} .



Fig. S7. SEM image of Cu foam after Li plating with a capacity of 1 mAh cm⁻² under 1mA cm⁻².



Fig. S8. SEM image of Cu foam after Li plating with a capacity of 5 mAh cm⁻² under 1mA cm⁻².



Fig. S9. SEM image of Cu foam after Li stripping with a capacity of 5 mAh cm⁻² under 1mA cm⁻².



Fig. S10. SEM image of MGC after Li plating with a capacity of 1 mAh cm⁻² under 1mA cm⁻².



Fig. S11. SEM image of MGC after Li plating with a capacity of 5 mAh cm⁻² under 1mA cm⁻².



Fig. S12. SEM image of MGC after Li stripping with a capacity of 5 mAh cm⁻² under 1mA cm⁻².



Fig. S13. Cross-sectional SEM images with deposition of different amount of Li on Cu foam and MGC.



Fig. S14. Cross-section SEM image of MGC after Li plating with a capacity of 5 mAh cm⁻² under 1mA cm⁻².



Fig. S15. Cross-section SEM image of Cu foam after Li plating with a capacity of 5 mAh cm⁻² under 1mA cm⁻².



Fig. S16. Porous utilization of MGC under different lithium deposition loads.



Fig. S17. (a) Cross-sectional SEM images of MGC with 3mAh cm⁻² Li plating and (b-f) EDS elemental distribution mapping of MGC after 50 cycles at 1 mAh cm⁻².



Fig. S18. Cross-section SEM image of MGC after Li plating with a capacity of 3 mAh cm⁻² under 1mA cm⁻².



Fig. S19. Cross-section SEM image of Cu foam after Li plating with a capacity of 3 mAh cm⁻² under 1mA cm⁻².



Fig. S20. Cyclic voltammograms of MGC anode at a scan rate of 0.1 mV $\rm s^{-1}$.



Fig. S21. The cycling performance of MGC ||LFP full cell



Fig. S22. Electrochemical performances of different full cells assembled with MGC, Cu Foam/Au and Cu Foam/LM anodes. (a) Long-term cycling performance and selected cycle voltage profiles of (b) Cu Foam/LM, (c) Cu Foam/Au, (d) MGC.



Fig. S23. (a) Long-term cycling performance of MGC||NCA full cell at 1 C and (b) corresponding charge/discharge curves.



Fig. S24. Disassembled cycled pouch cells.



top



bottom

Fig. S25. Photos of cycled MGC anode from MGC||LFP pouch cell.



Fig. S26. Flexibility and power display of MGC||LFP pouch cell.