Supporting Information

Bottom-top Channeling Li Nucleation and Growth by Gradient Lithiophilic 3D Conductive Host for Highly Stable Li Metal Anodes

Xiaolin Yan, Qingfei Zhang, Wanjie Xu, Qingshui Xie, Pengfei Liu, Qiulin Chen, Hongfei Zheng, Laisen Wang, Zi-Zhong Zhu, and Dong-Liang Peng**

X. Yan, Q. Zhang, W. Xu, Dr. Q. Xie, P. Liu, Q. Chen, H. Zheng, Dr. L. Wang, Prof. D.-L. Peng

State Key Lab of Physical Chemistry of Solid Surface, Collaborative Innovation Center of Chemistry for Energy Materials, College of Materials, Xiamen University, Xiamen 36105, China

Dr. Z-Z Zhu

Collaborative Innovation Center for Optoelectronic Semiconductors and Efficient Devices, Department of Physics, Xiamen University, Xiamen 36105, China

E-mail: xieqsh@xmu.edu.cn (Q. Xie), dlpeng@xmu.edu.cn (D. Peng)

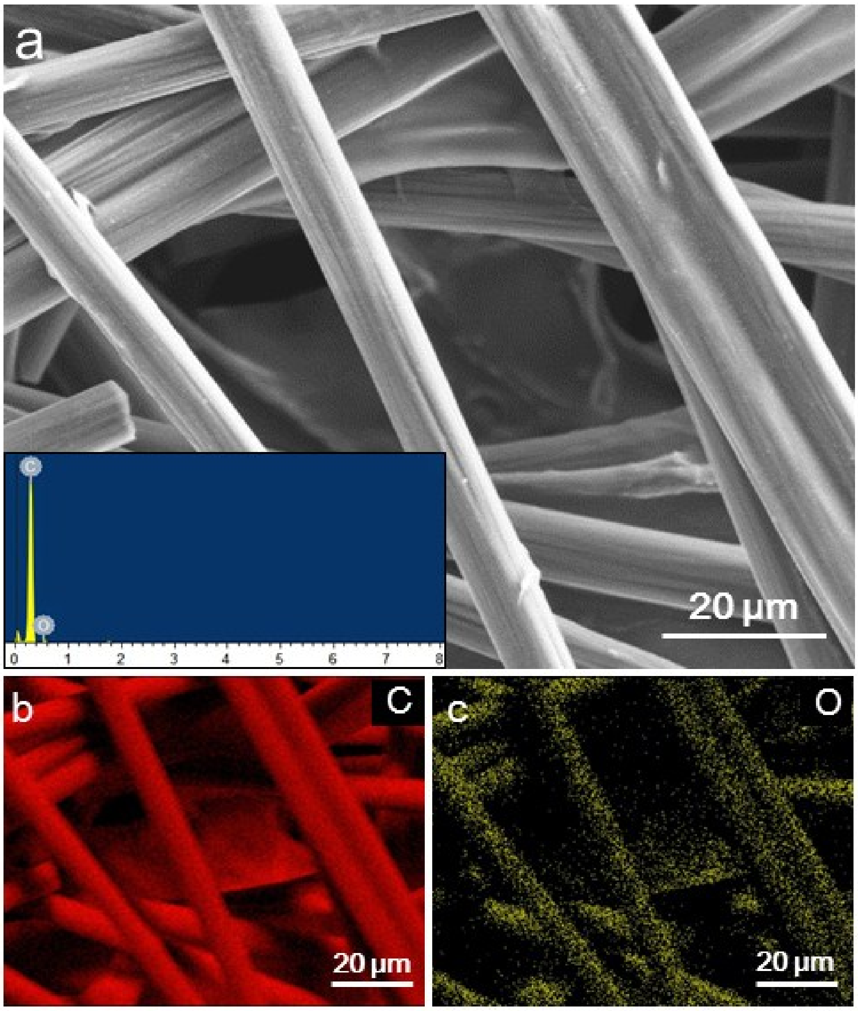


Figure S1. EDS and element mappings of 3D CP matrix.

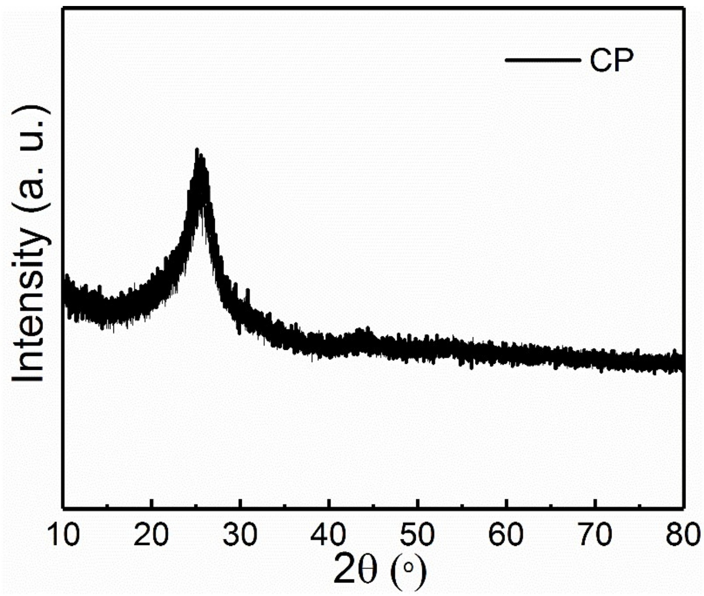


Figure S2. XRD pattern of 3D CP matrix.

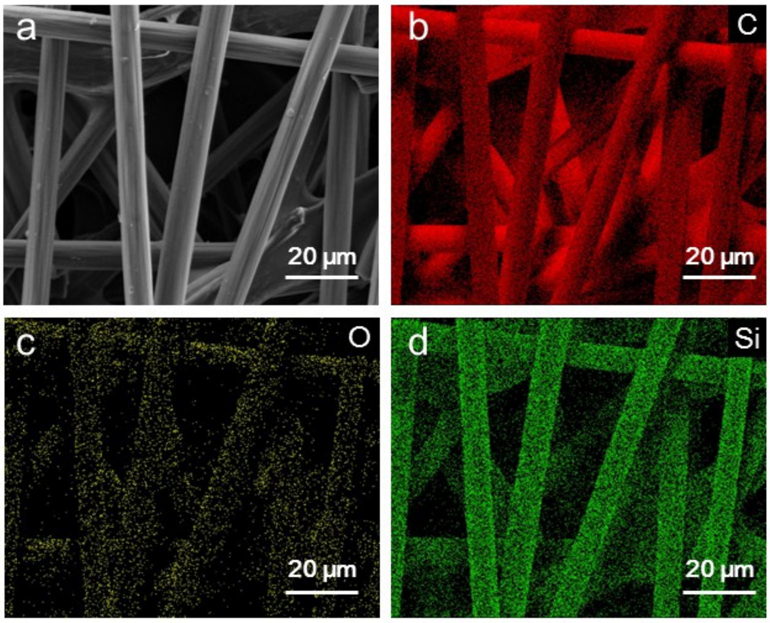
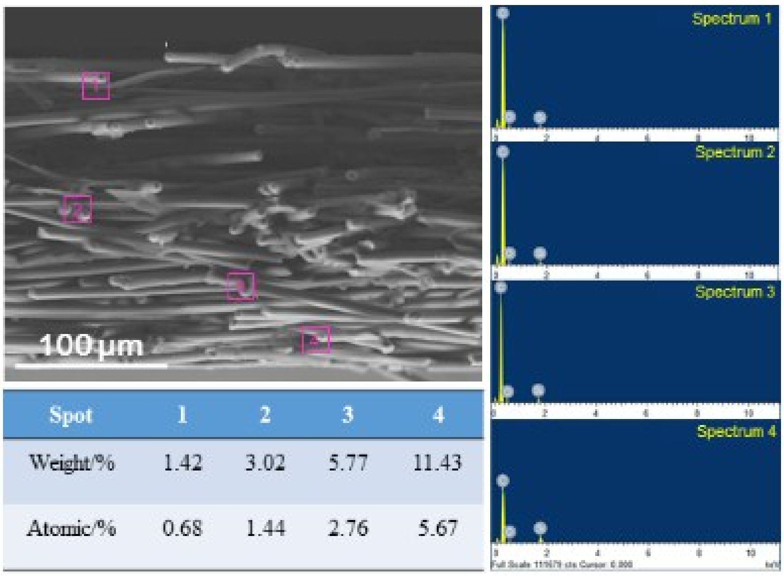


Figure S3. SEM image and corresponding Element mappings of GSCP electrode.



Spot	1	2	3	4
Weight/%	1.42	3.02	5.77	11.43
Atomic/%	0.68	1.44	2.76	5.67

Figure S4. Cross-section SEM image and corresponding energy-dispersive X-ray spectroscopy data of GSCP electrode at different areas.

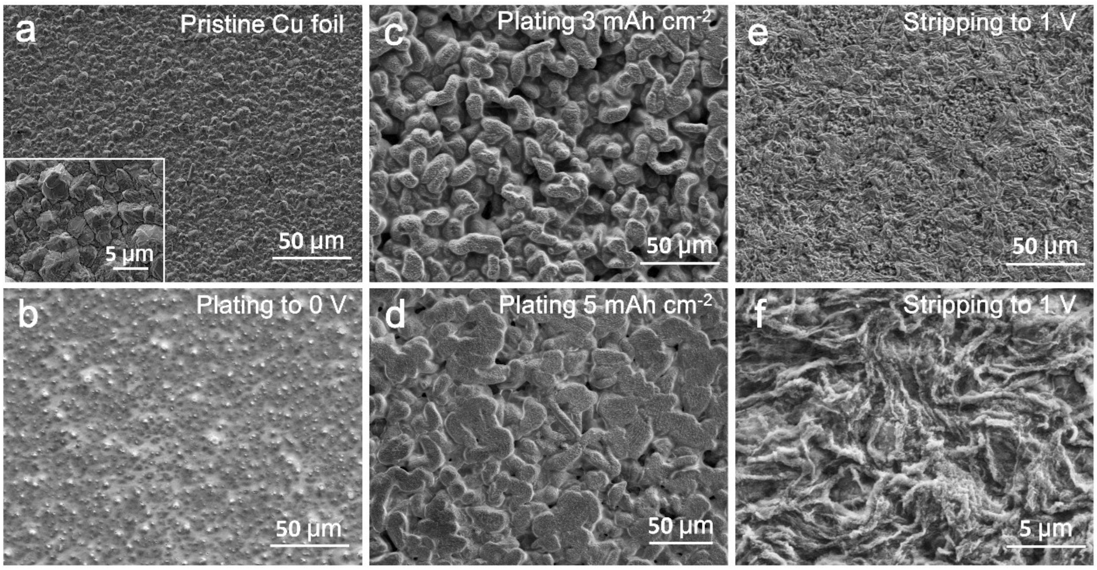


Figure S5. SEM images of (a) pristine Cu foil, top surface morphology of Cu foil (b) plating to 0 V, (c) plating 3 mAh cm^{-2} of Li metal, (d) plating 5 mAh of Li and then (e, f) stripped to 1 V.

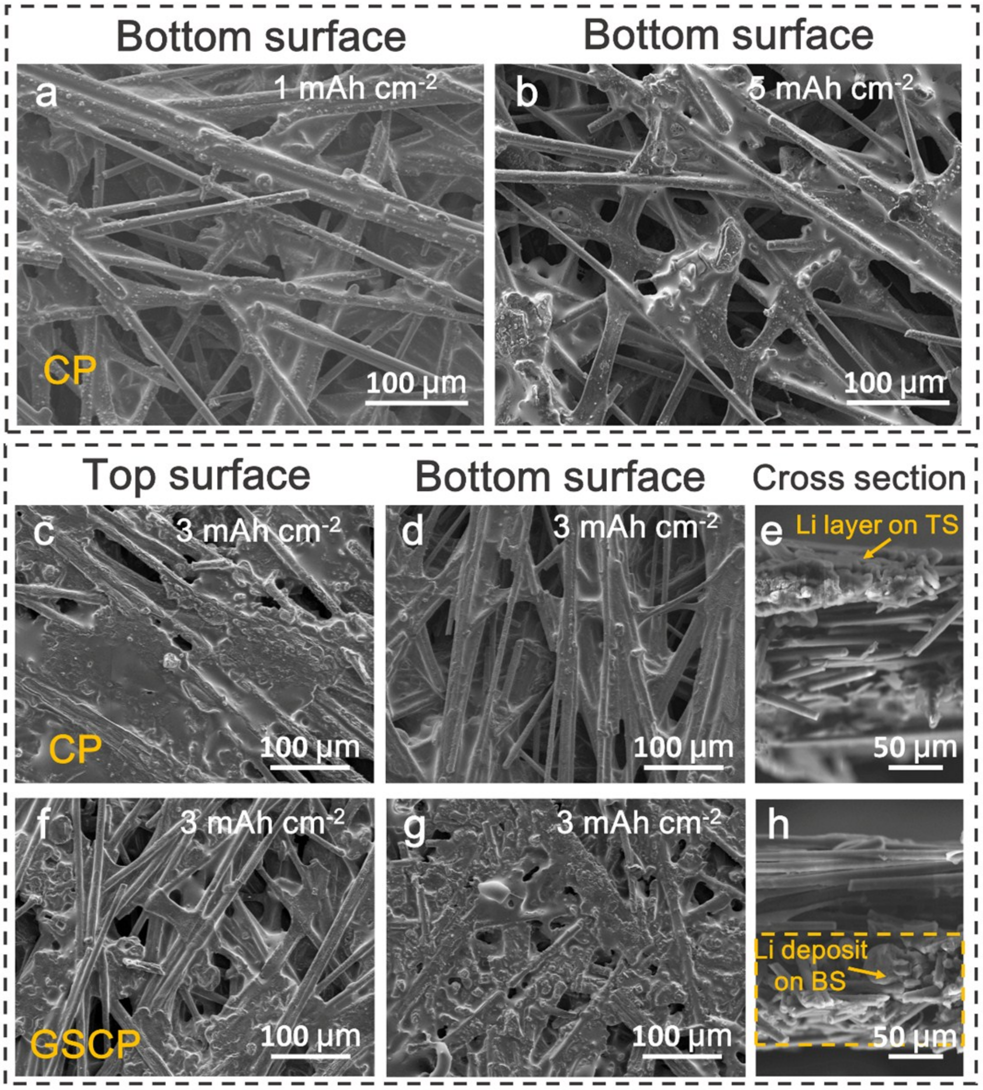


Figure S6. SEM images of bottom surface for CP electrode after depositing (a) 1 mAh cm^{-2}Li metal, (b) 5 mAh cm^{-2} Li metal. SEM images of top surface, bottom surface and cross-section for (c-e) CP electrode and (f-h) GSCP electrode after depositing 3 mAh cm^{-2} Li metal.

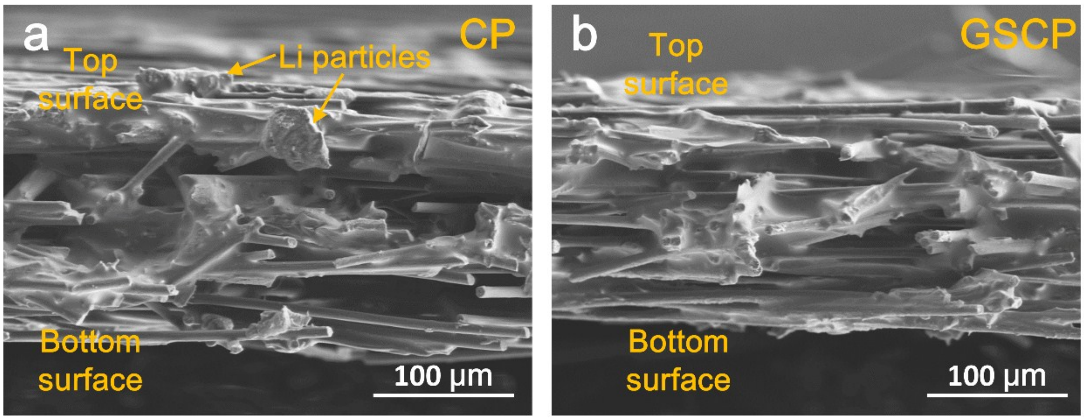


Figure S7. Cross-section SEM images of (a) CP and (b) GSCP after depositing 5 mAh cm^{-2} of Li and then stripped to 1 V at 1 mA cm^{-2}.

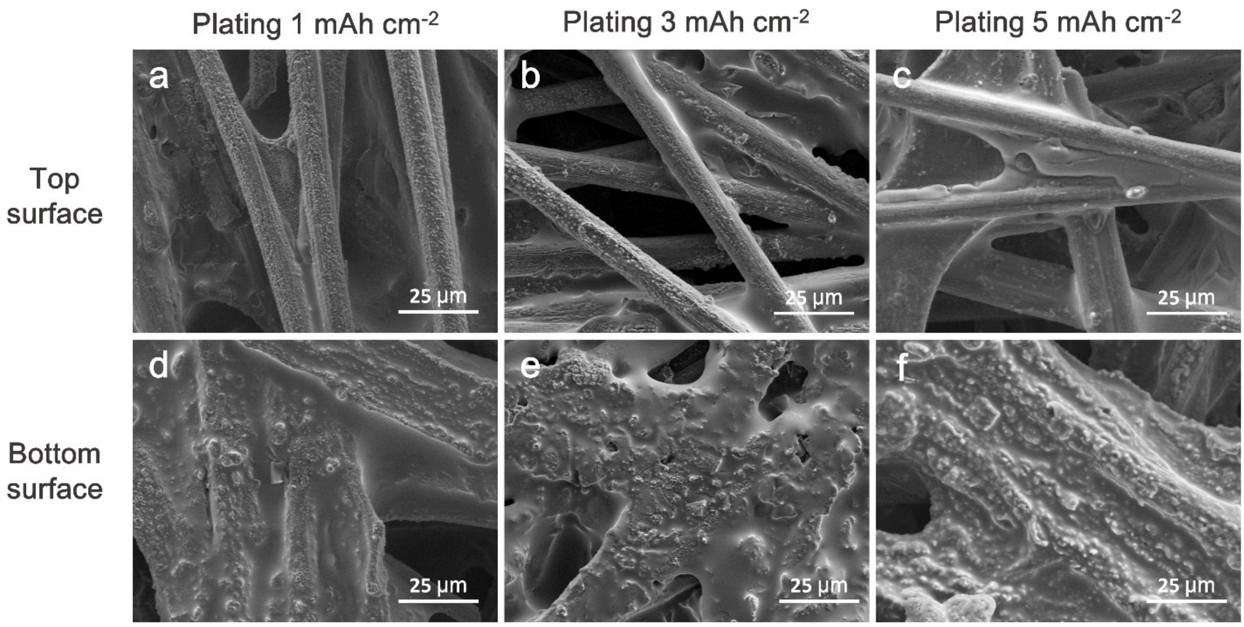


Figure S8. SEM images of top and bottom surfaces for GSCP electrode after depositing (a and d) 1 mAh cm^{-2}, (b and e) 3 mAh cm^{-2} and (c-f) 5 mAh cm^{-2} of Li metal.

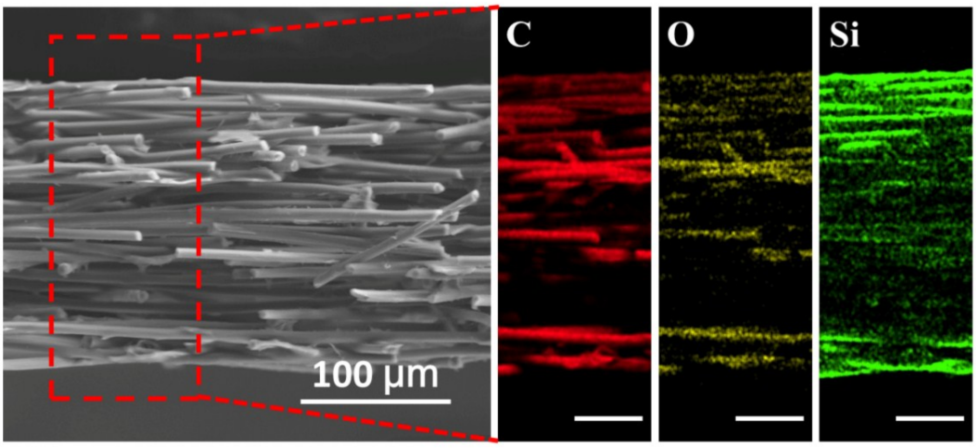


Figure S9. SEM image and corresponding element mappings of SCP electrode (The scale bar in element mapping is 50 µm).

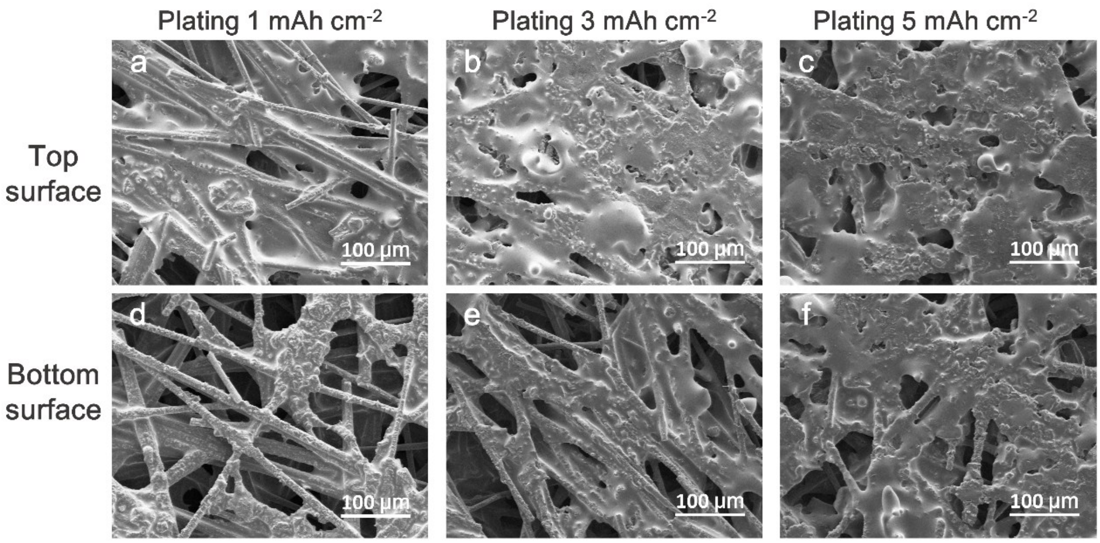


Figure S10. SEM images of top and bottom surfaces of SCP after plating (a and d) 1 mAh cm^{-2}, (b and e) 3 mAh cm^{-2}, and (c and f) 5 mAh cm^{-2} of Li.

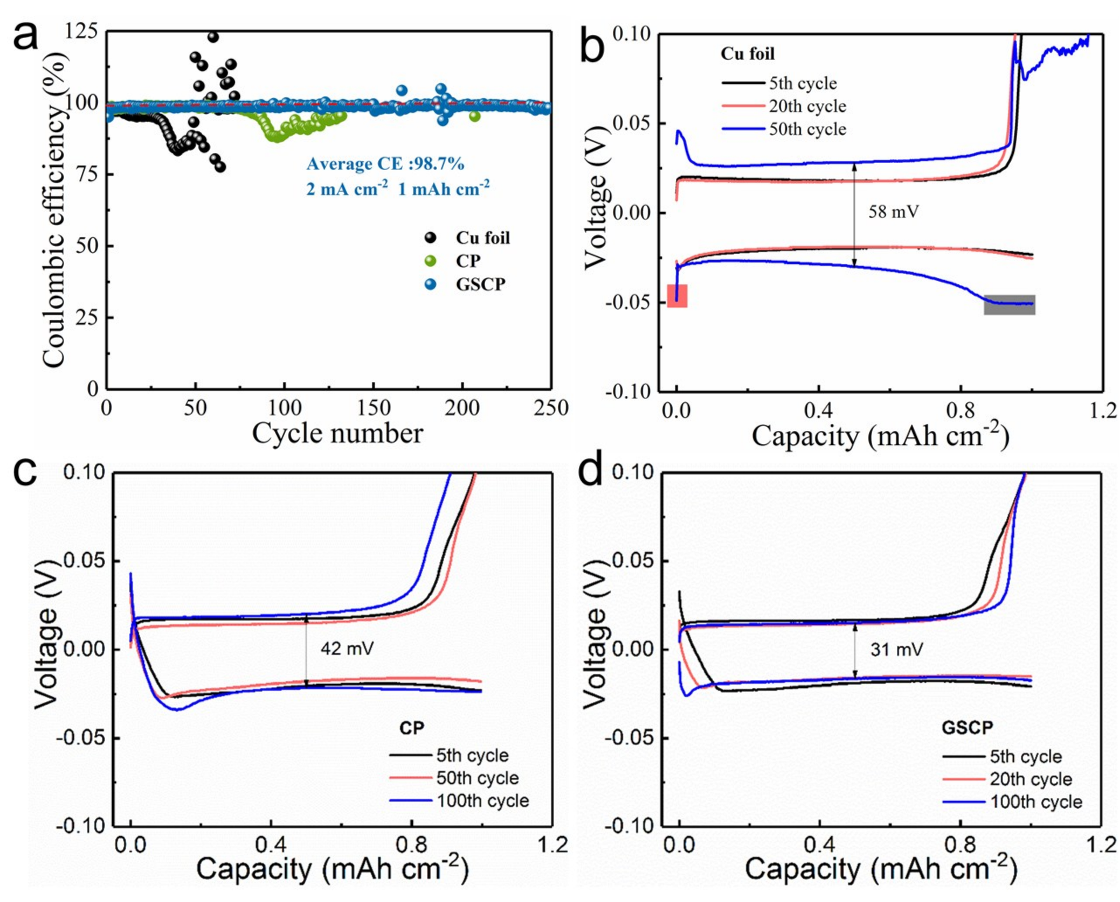


Figure S11. (a) CE performances of Cu foil, CP and GSCP electrodes at 2 mA cm^{-2} for 1 mAh cm^{-2} and corresponding voltage profiles of (b) Cu foil, (c) CP and (d) GSCP electrodes.

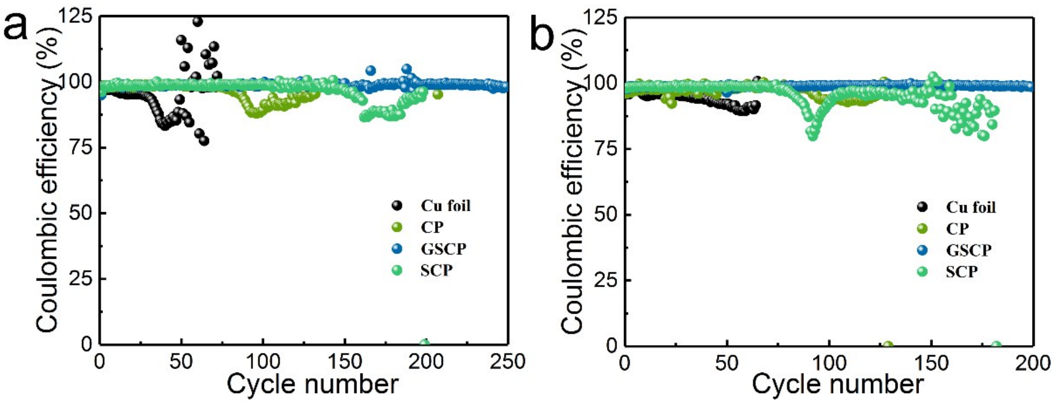


Figure S12. (a) CE performances of Cu foil, CP, GSCP and SCP electrodes at (a) 2 mA cm^{-2} for 1 mAh cm^{-2} and (b) 2 mA cm^{-2} for 2 mAh cm^{-2}.

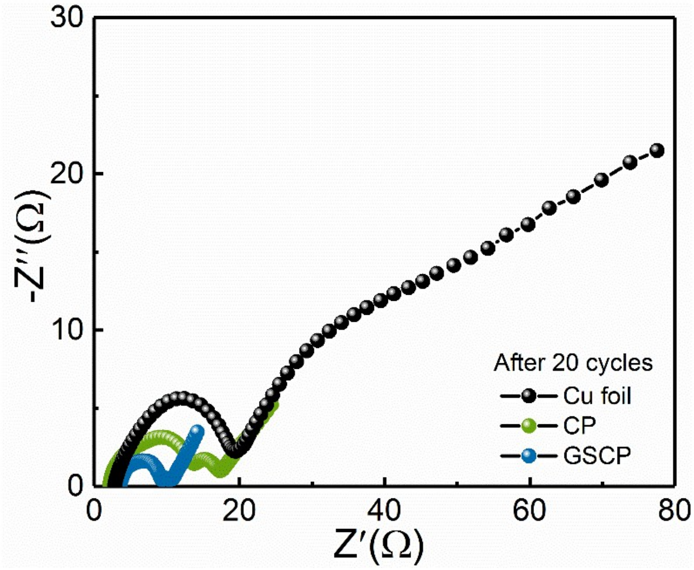


Figure S13. Electrochemical impedance spectra of Cu foil, CP and GSCP electrodes before cycling.

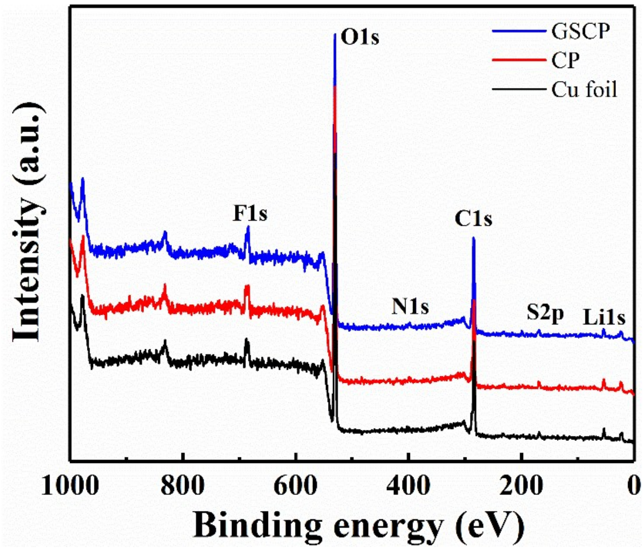


Figure S14. XPS full spectra of Cu foil, CP and GSCP electrodes after 50 cycles.

In full XPS spectra, C, O, F, S, Li and N elements are detected. These elements may come from electrolyte decomposition. The LiTFSI electrolyte can be reduced to $Li_2S_xO_y$, C_2F_xLi, Li_3N and LiF. The $LiNO_3$ can be reduced to Li_3N and LiN_xO_y. The DOL and DME can react to form $ROCO_2Li$, RCH2OLi, Li_2CO_3 et al[1-3].

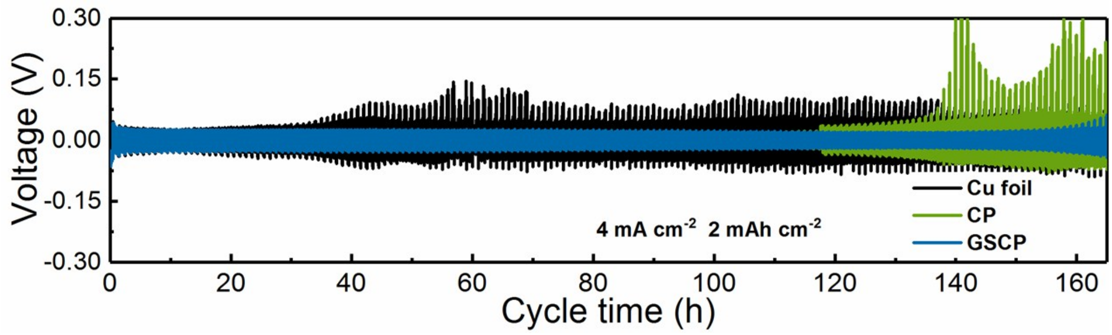


Figure S15. Cycling performance of Li|Cu@Li, Li|CP@Li and Li|GSCP@Li symmetrical cells at 4 mA cm^{-2} for 2 mAh cm^{-2}.

Table S1. Summary of Coulombic efficiency with different 3D Li metal hybrid anodes.

Electrode	Current density	Capacity density	Coulombic efficiency (CE)	Ref.
3D-AGBN	1 1	6 12	average CE of ~97.3% for 50 cycles average CE of ~97.6% for 20 cycles	[4]
Ni@Li_2O-NW	1	2	a CE of 92.5% at 180 cycles	[5]
Ni_xN decorated Ni foams	1 2	1 1	a CE above 97.0% at 300 cycles a CE of ~98.6% after 150 cycles	[6]
Single atom Ni-nitrogen-doped graphene	0.5 2 4	1 1 1	average CE of 98.5% for 250 cycles a CE at ~97.7% after 150 cycles a CE of ~97.0% after 100 cycles	[7]
Cu porous current collector	0.5 1	1 1	a CE of 94.6% for 200 cycles a CE of 97% for 120 cycle	[8]
Stacked graphene coated Cu scaffold	0.5 5	1 1	a CE of ~97.1% for 200 cycles a CE of ~91.6% for 125 cycles	[9]
g-C3N4@Ni foam	0.5 1	1 2	a CE of 98% after 300 cycles a CE of 97% after 140 cycles	[10]
Ag@HKUST-1	0.5 1 1	1 1 5	a CE > 97% after 300 cycles a CE of 97.5% at 150 cycles a CE of 98.5% at 50 cycles	[11]
AgNP@SiO_2	0.5 1 2	1 1 1	a CE of 95% at 150 cycles a CE of 93% at 150 cycles a CE of 91% at 150 cycles	[12]
N-doped graphene/Ni foam	0.5 1 2	1 1 1	average CE of 99% for 200 cycles average CE of 98% for 200 cycles a CE of 98.3% for 200 cycles	[13]
GSCP	**1** **2** **5** **2** **3**	**1** **1** **1** **2** **3**	**average CE of 99.0% for 400 cycles** **average CE of 98.7% for 250 cycles** **average CE of 97.0% for 150 cycles** **average CE of 98.9% for 200 cycles** **average CE of 96.1% for 100 cycles**	**This work**

The units of current density and capacity density are mA cm^{-2} and mAh cm^{-2}, respectively.

Table S2. Summary of cycling performances of symmetrical cells with 3D Li metal anodes.

Electrode	Current density/ Capacity density	Cycle time	Cycle number	Ref.
CuBr modified Cu foam@Li	1/1	850	425	[14]
Ni_xN decorated Ni foams@Li	1/1 2/1 5/1	400 300 160	200 300 400	[6]
TiC-C core/shell nanowire skeleton@Li	0.5/1 1/1 3/1	800 400 133.3	200 200 200	[15]
Coiled Li	1/1 2/1	1000 275	500 275	[16]
Graphitized carbon fibers@Li	1/1 2/1	1000 300	500 300	[17]
SnO_2-CF@Li	1/1 3/1	700 120	350 180	[18]
PI-ZnO@Li	1/1 3/1 5/1	200 67 40	100 100 100	[19]
Porous Cu@Li	1/1 2/1 3/1	800 535 145	400 535 217	[20]
GSCP@Li	**1/1** **2/1** 4/2	**1350** **700** **160**	**675** **700** **160**	**This work**

The units of current density and capacity density are mA cm^{-2} and mAh cm^{-2}, respectively.

Table S3. Summary of cycling performances of LTO-based full cells with Li metal anodes.

Electrode	**Current density**	**C rate**	**Cycle number**	**Specific capacity**	**Ref.**
Cu-mesh@Li	overdose	4	500	87.4	[21]
Carbon cloth@Li	2.03	1	700	115	[22]
Coiled Li	overdose 1.68	1 5	2500 2000	141 129	[16]
PMF@Li	overdose	2	1000	~95	[23]
Lithiated Nafion-LiCl@Li	overdose	2	800	~100	[24]
Cu foam-ZnO@Li	~2.1	10	1000	~90	[25]
Al_2O_3 framework@Li	~4.8	4	1200	70	[26]
g-C_3N_4@Ni foam@Li	overdose	2	1000	~90	[13]
GSCP@Li	**~2**	**10**	**5000**	**97.0**	**This work**

The units of capacity density and Specific capacity are mAh cm^{-2} and mAh g^{-1}, respectively.

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