

Supporting Information

VGCF 3D conducting host coating on glass fiber filter for lithium metal anodes

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Experimental Section

Materials

Lithium metal (China Energy Lithium Co., Ltd), glass fiber filter (GF/A Whatman), VGCF (vapor grown carbon fiber, Showa Denko), The electrolyte of 1.0 M LiPF₆ in EC (ethylene carbonate, > 99.9%)/ DEC (diethylene carbonate, > 99.9%) (v/v = 1 : 1, water content of < 20 ppm) was purchased from Zhangjiagang Guotai-Huarong New Chemical Materials Company, and 5 wt% VC (vinylene carbonate, 98%, Aladdin) was added before use.

Fabrication of the VGCF@GF electrode

The VGCF@GF electrode was fabricated by two steps. Firstly, the GF was placed in the magnetron sputtering instrument for 2 min to introduce a Au-coating layer. The VGCF and polyvinylidene fluoride (PVDF, weight ratio = 8 : 2) were mixed in N-Methyl pyrrolidone (NMP) by magnetic stirring for 2 h. Then the slurry was casted on the Au-coated GF with a doctor blade and dried in the vacuum oven at 80 °C overnight. The loading mass of VGCF was about 4.0 ~ 5.5 mg cm⁻².

Characterization

The morphology of the electrodes was characterized by a scanning electron microscope (SEM, S-4800, Hitachi). Prior to the analyses, the Li-deposited electrodes were disassembled from the electrochemical cells, rinsed three times with DMC (dimethyl carbonate), and dried in the Ar-filled glove box. The contact angle was

measured by a drop shape analysis system (Powereach JC2000C1, Shanghai Zhongchen Digital Technique Equipment Co. Ltd., China).

Electrochemical measurement

The 2016-type coin cells were assembled in an Ar-filled glove box with the content of both O₂ and H₂O below 1.0 ppm. The as-prepared VGCF@GF electrode was cut into discs with the diameter of 12 mm, and used as the working electrode. The metallic Li foil with the diameter of 12 mm and thickness of 1 mm was used as the counter electrode. The Celgard 2400 membrane was used as the separator and the solution of 1 M LiPF₆ in EC/DEC (v/v = 1 : 1) with 5 wt% VC as the electrolyte. For all the batteries, a fixed amount of Li (2.0, 2.5 or 3.0 mAh cm⁻²) was deposited onto the VGCF@GF electrode, and then stripped away up to 1.5 V at different areal current densities. The Electrochemical impedance spectroscopy (EIS) was tested by the Autolab Potentiostat Galvanostat 302N in the frequency range from 100 mHz to 100 kHz. All of the electrochemical tests were measured at 25 °C.

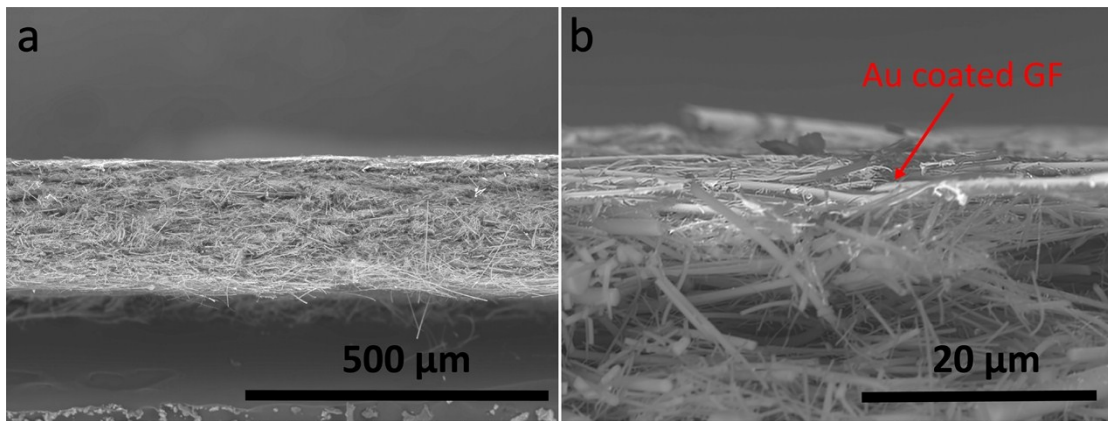


Figure S1. SEM images of the GF after magnetron sputtering.

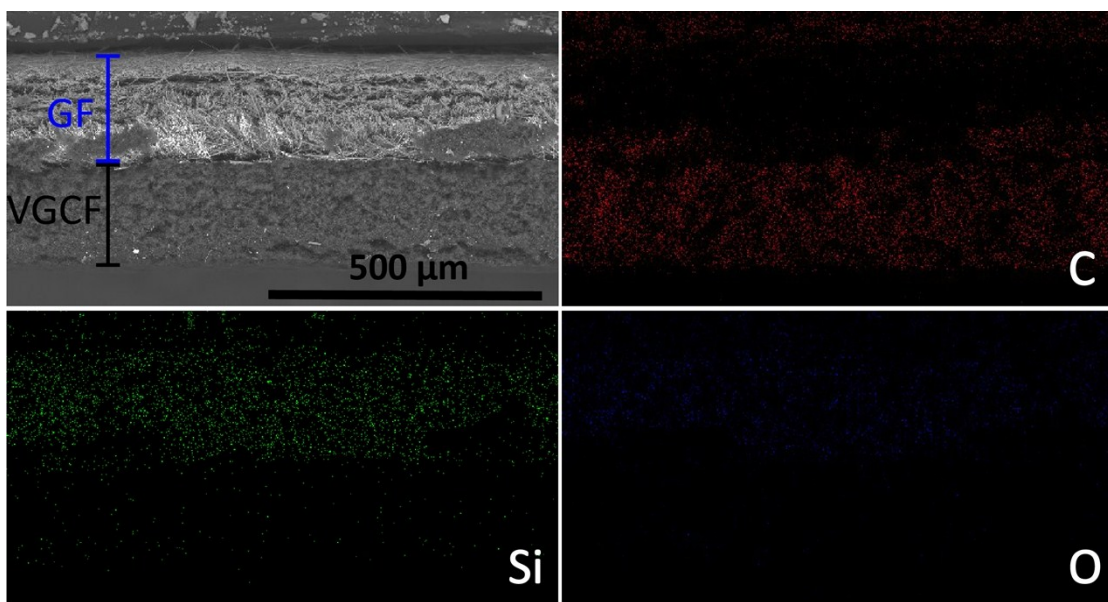


Figure S2. Side view SEM image and element mapping of the VGCF@GF electrode.

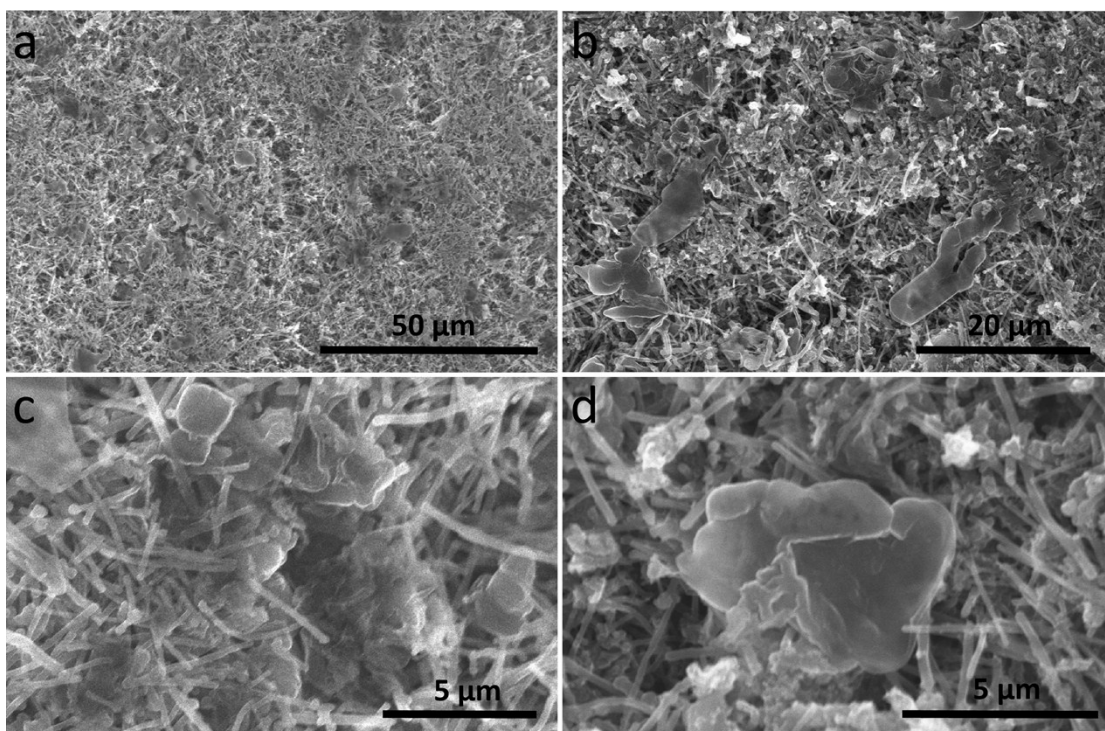


Figure S3. Bottom view SEM images of the VGCF@GF electrode for 2.5 mAh cm^{-2} of Li deposition at the current density of 0.5 mA cm^{-2} after 50 cycles.

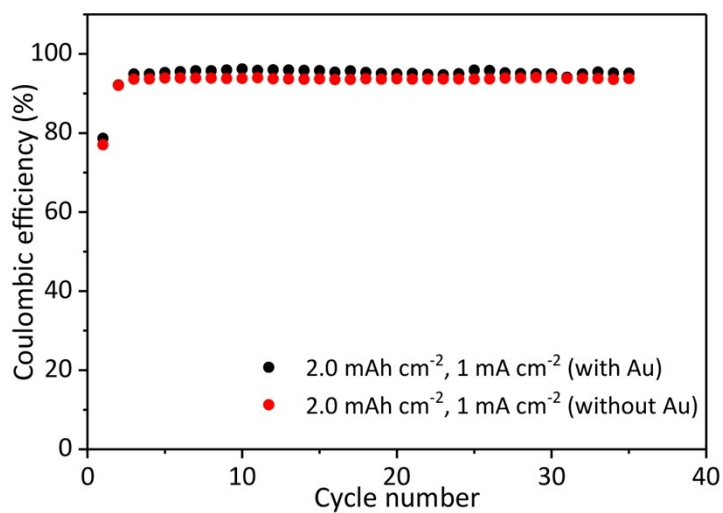


Figure S4. Comparison of the coulombic efficiency (CE) of VGCF@GF electrode with/without Au coating at the current density of 1.0 mA cm^{-2} for 2.0 mAh cm^{-2} .

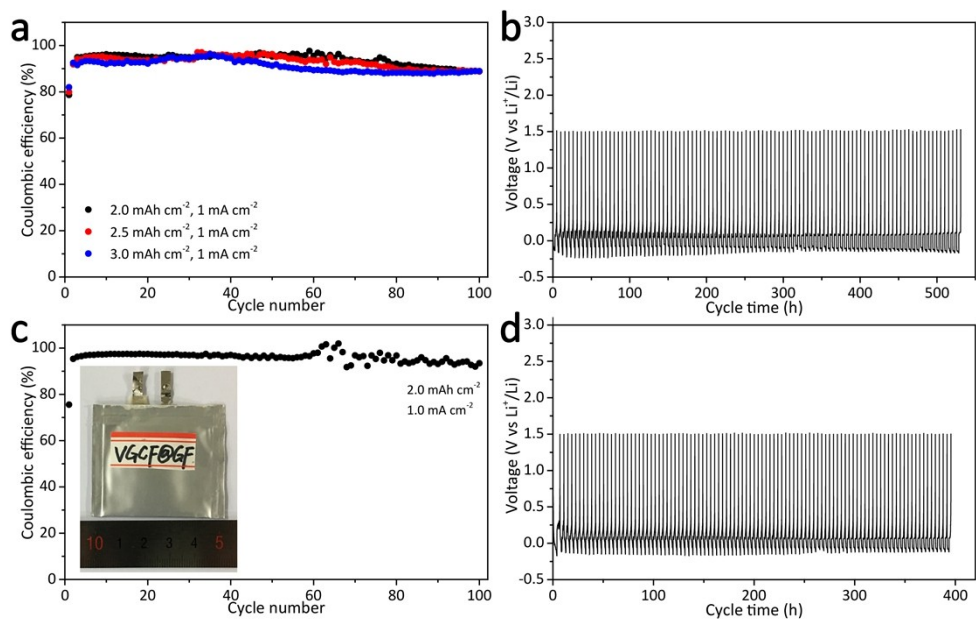


Figure S5. (a) CE of Li deposition/stripping on the VGCF@GF electrode at the current density of 1.0 mA cm⁻² for different capacities. (b) Voltage stability of the VGCF@GF electrode at 1.0 mA cm⁻² for 2.5 mAh cm⁻². (c) CE and (d) voltage stability of the VGCF@GF electrode assembled in the pouch cell at 1.0 mA cm⁻² for 2.0 mAh cm⁻².

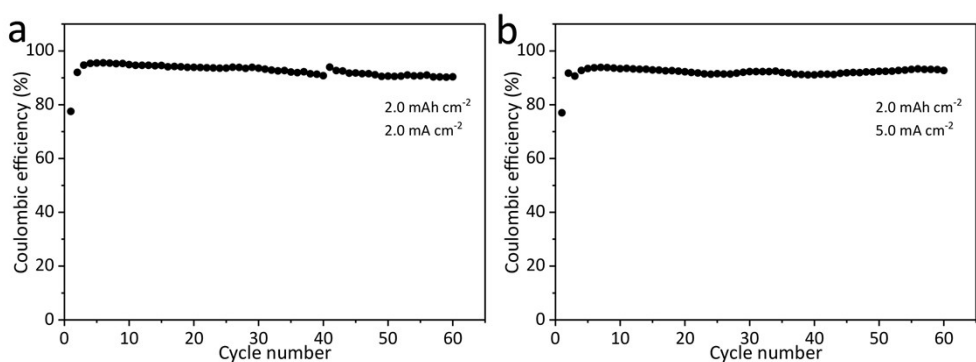


Figure S6. CE of Li deposition/stripping on the VGCF@GF electrode for 2.0 mAh cm⁻² at the current density of (a) 2.0 mA cm⁻² and (b) 5.0 mA cm⁻².

Table S1. The comparison of electrochemical performance of the VGCF@GF electrode in this work with some reported works employing the concept of 3D-conductive host or protective layer.

References	Electrolyte	Capacity (mAh cm ⁻²)	Cycle life/Current density	Coulombic efficiency after cycling
Lithium-coated polymeric matrix ¹	Carbonate	1	~194 h/1 mA cm ⁻²	Not mentioned
LiF artificial SEI ²	Carbonate	1	160 h/1 mA cm ⁻²	Not mentioned
CNT 3D matrix ³	Carbonate	1	Not mentioned	~60% (100 cycles)
PDMS thin film modified ⁴	Carbonate	1	800 h/0.5 mA cm ⁻²	93% (100 cycles)
SiO ₂ @PMMA coating layer ⁵	Carbonate	2	Not mentioned	87% (50 cycles)
3D Cu current collector ⁶	Ether	0.5	600 h/0.2 mA cm ⁻²	Not mentioned
GF modified Cu foil ⁷	Ether	0.5	Not mentioned	93% (90 cycles)
This work	Carbonate	2.5	965 h/ 0.5 mA cm ⁻²	91.1 (100 cycles)

References

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