Electronic Supplementary Material (ESI) for Chemical Communications.

Electronic supplementary information

Ag coated 3D-Cu foam as a lithiophilic current collector for enabling Li_2S-based anode-free batteries

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Experiments

preparations of current collectors.

2D-Cu foil (10 μm in thickness) and 3D-Cu foam (375 μm in thickness) were purchased from Guangdong Canrd New Energy Technology Co., Ltd. Before used, they were washed with nitric acid (10 wt%) to remove the oxide layer. Ag powder was purchased from Aladdin (>99.99%). The Ag@2D-Cu foil and Ag@3D-Cu foam were prepared by depositing 100 nm thickness Ag layer on 2D-Cu foil (single side) and 3D-Cu foam (both sides) using the thermal evaporation method with a deposition rate of 0.05 nm s^{-1}, respectively. Finally, these current collectors were cut into discs with a diameter of 18 mm.

Fabrication of CC/Li_2S cathode.

Cotton cloth (CC) was carbonized at 1000 ℃ for 3 h with a heating rate of 5 ℃ min^{-1} in an argon-filled tube furnace. Then it was cut into discs with a diameter of 12 mm. 350 mg Li_2S powder (Sigma Aldrich, 99.9%) and 150 mg acetylene black (Hefei Kejing material technology Co., Ltd., 99.9%) were mixed and then dispersed into 5 mL N-methyl-2-pyrrolidone (NMP) solvent. 150-600 μL slurry was dropped onto both sides of CC. Next, the electrode was dried at 60 ℃ for 12 h to remove the solvent. The obtained CC/Li_2S discs with a Li_2S mass loading of 3.8-14.6 mg cm^{-2} were used as cathodes. Graphene oxide and acetylene black modified polypropylene separator (GO/AB@PP) was prepared according to previous reports.[1]

Characterizations.

An X-ray diffractometer (XRD, SHIMADZU-7000) with a Cu Ka radiation was applied to identify the crystal structure. Raman signals were detected by a Raman spectrometer (LabRAM HR evolution) with excitation wavelength of 532 nm. A field-emission scanning electron microscope (SEM, SIRION-100) was used to investigate micromorphology. The element information was recorded by an energy dispersive spectrometer (EDS, Oxford) in SEM.

Electrochemical measurement.

Lithium metal was used as an anode and current collectors were used as cathodes to assemble 2025-type button batteries to study the deposition behavior of lithium on

the current collectors. Battery assembly was performed in an Ar-filled glove box, in which the concentration of O_2 and H_2O were both less than 0.5 ppm. 1 M LiTFSI and 1 wt% $LiNO_3$ in DOL/DME (v/v = 1: 1) solution was applied as the electrolyte, and polypropylene membrane (Celgard 2400) was used as a separator. For the full battery assembly, CC/Li_2S electrode was used as a cathode and current collector was used as an anode. Constant current charge-discharge tests were conducted on a Neware battery test station (5V/20mA). The batteries were initially charged to 3.8 V and then discharged/charged within the voltage range of 1.7-2.8 V for the cycle and rate performances test. Cyclic voltammetry (CV) measurement was performed at a scanning speed of 0.05 mV s^{-1} on an electrochemical workstation (CHI660D, Chenhua). Electrochemical impedance spectroscopy (EIS) measurement was conducted on the above electrochemical workstation in the frequency range between 0.1 Hz and 100 K Hz.

Reference

1. H. Cheng, H. Liu, H. Jin, N. Cai, C. Gao, S. Zhao and M. Wang, J. Mater. Chem. A, 2020, 8, 16429-16436.

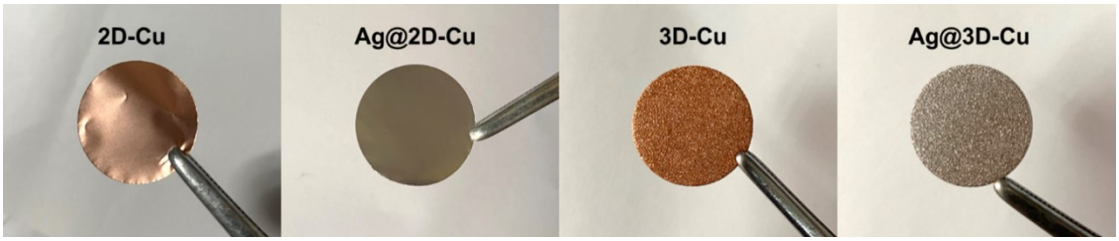

Fig. S1 Optical photos of 2D-Cu, Ag@2D-Cu, 3D-Cu and Ag@3D-Cu samples.

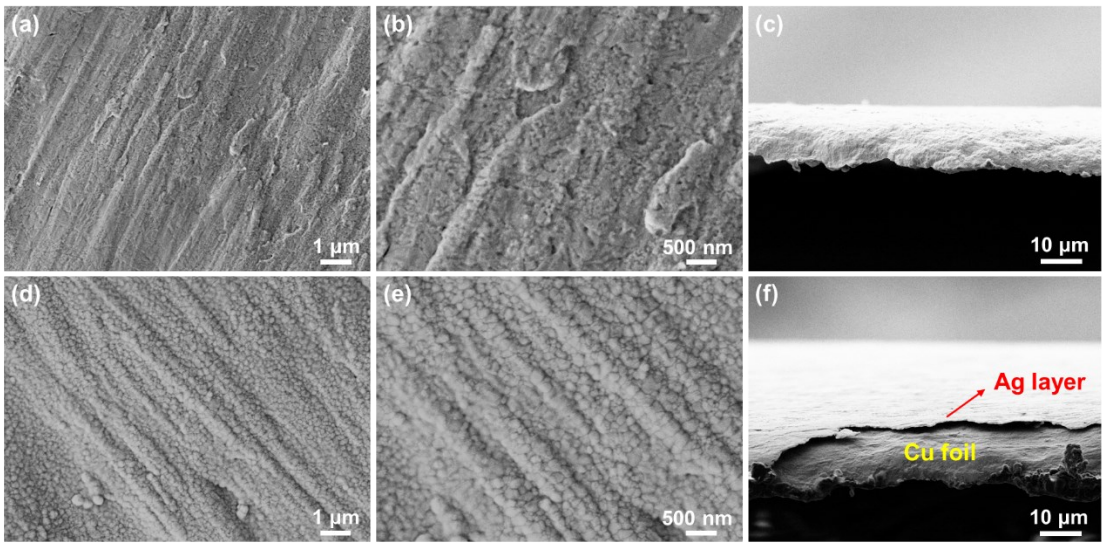

Fig. S2 SEM images of (a-c) 2D-Cu and (d-f) Ag@2D-Cu.

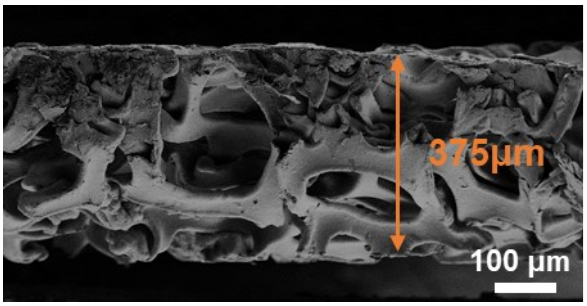

Fig. S3 Cross-sectional SEM image of Ag@3D-Cu sample.

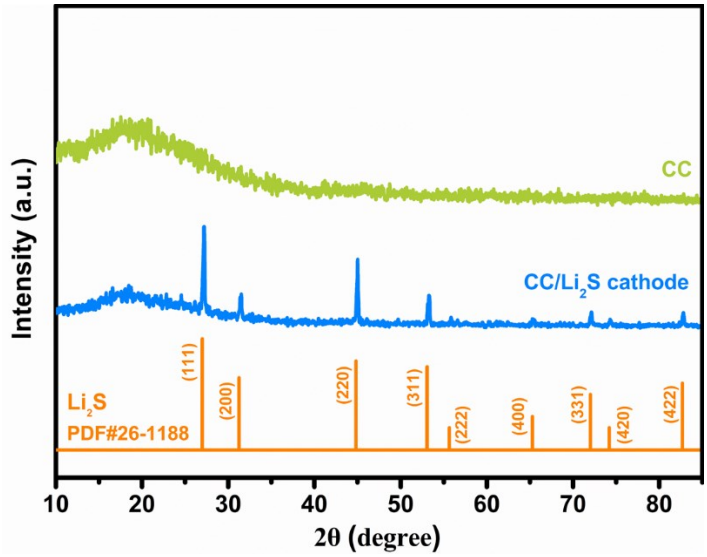

Fig. S4 XRD patterns of CC and CC/Li_2S samples.

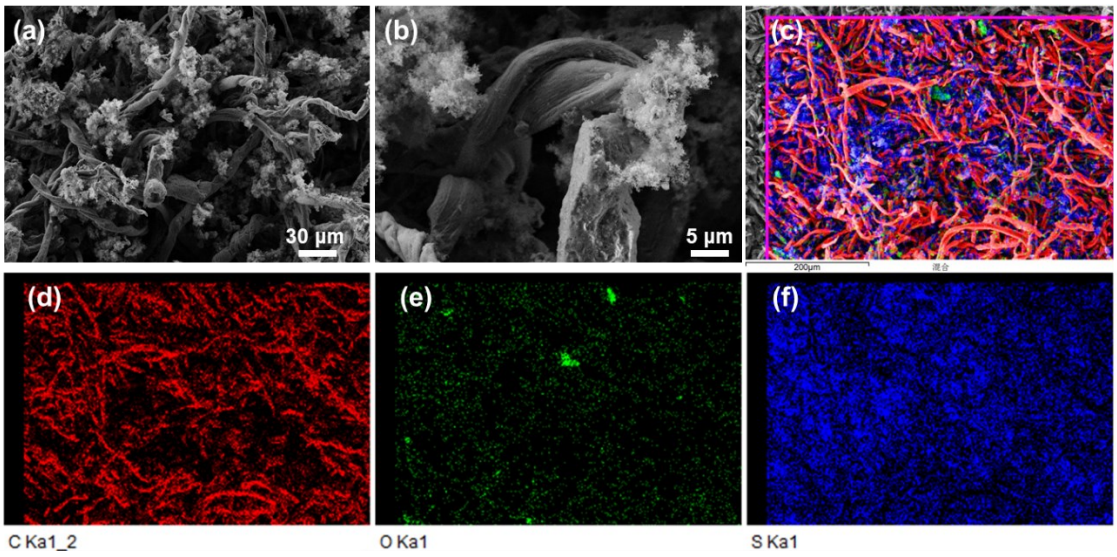

Fig. S5 (a, b) SEM images and (c-f) EDS elemental mapping images of CC/Li_2S cathode.

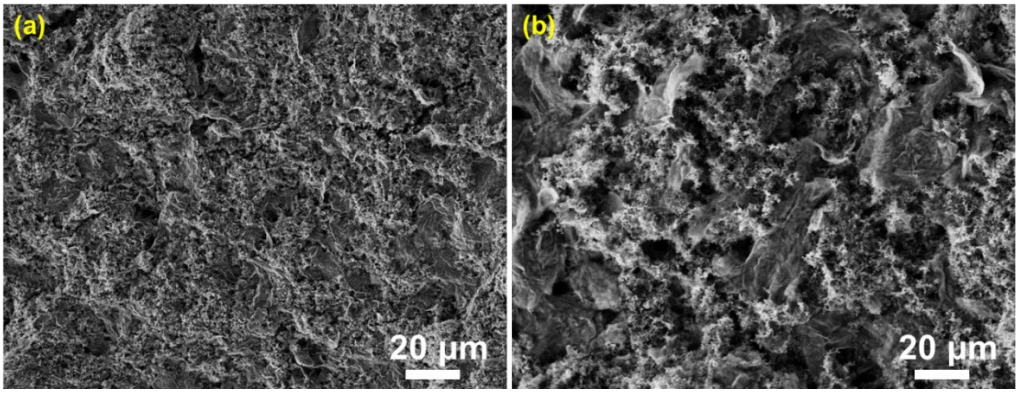

Fig. S6 (a, b) Top-view SEM images of GO/AB@PP separator.

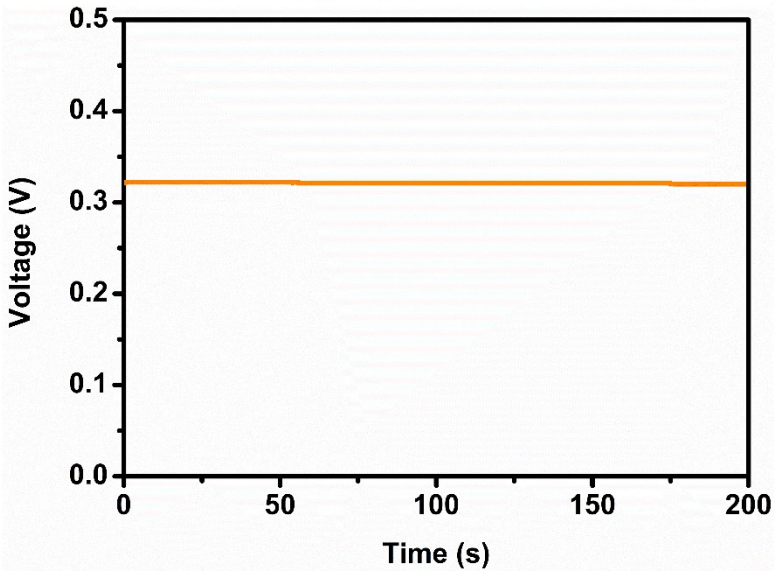

Fig. S7 Voltage-time profile of the assembled Ag@3D-Cu||Li_2S battery.

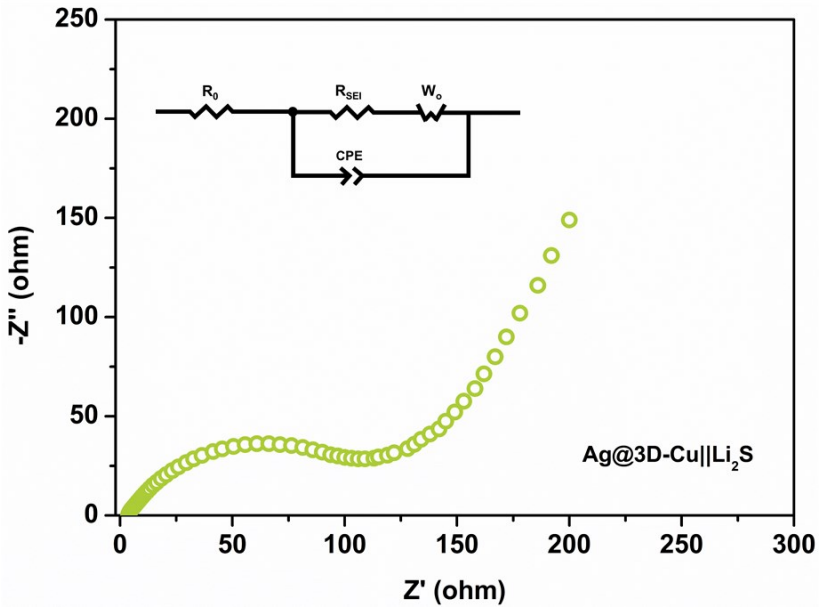

Fig. S8 EIS profile of the assembled Ag@3D-Cu||Li_2S battery, the inset is the equivalent circuit model.

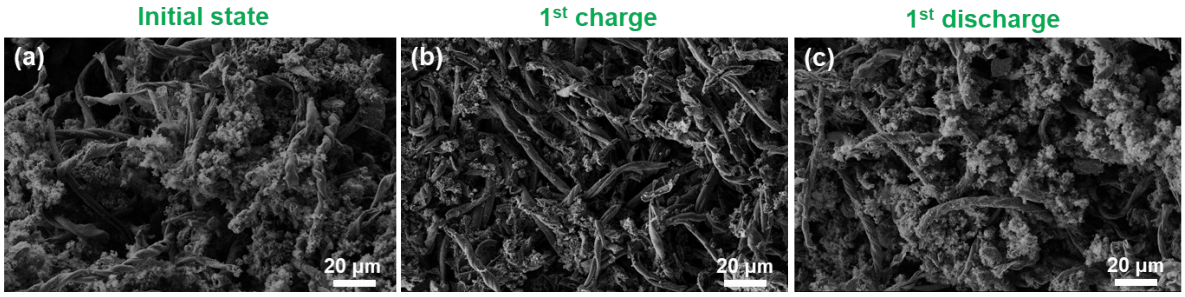

Fig. S9 SEM images of CC/Li_2S cathode (a) at initial state, (b) after 1st charging and (c) after 1st discharging.