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Supporting Information

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A Lightweight and Binder-free Electrode Enabled by Lignin Fibers@Carbon-Nanotubes and Graphene for Ultrastable Lithium-Sulfur Batteries

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Calculation of the electrolyte uptake amount.

Electrolyte uptake amount of the lignin fibers was calculated by the following equation.

Uptake amount (%) = $[(m_2 - m_1)/m_1] \times 100\%$

where m_1 correspond to the weight of lignin fibers, m_2 is related to the weights of the lignin fibers after the electrolyte absorption.

Preparation of lignin fibers.

The bleached wood pulp powder which contains lignin fiber and trace cellulose was obtained from shandong Yousuo Chemical Technology Co., Ltd. 50 g wood pulp powder was dissolved in 1.0 L ultrapure water, and added 100 mL NH₃.H₂O (27%). The dilute ammonia solution was heated at 80 °C for 3 h to get rid of cellulose and increase the specific surface area of lignin fibers. The obtained lignin fibers were filtered and washed with ethanol and water for several times (until PH=7-8).



Fig. S1 Digital images of (a) PVDF@CNT-GN, (b) LF@CNT-GN substrate.



Fig. S2 SEM images of LF@CNT-GN substrate after Li_2S_6 injection (a) Low magnification, (b) high magnification, and EDX elemental mapping for S, C and F from the area shown in the frame.



Fig. S3 (a) Raman spectrum and (b) XRD spectrum of the CNTs.



Fig. S4 Digital images of (a) PVDF@CNT-Al coated by an automatic coating machine, and (b) corresponding cathode electrode (inset: LF@CNT-GN electrode).



Fig. S5 Rate performance of (a) PVDF@CNT-Al@GN and (b) PVDF@SP-GN at different rates from 0.1 to 3 C within a potential window of 1.7–2.8 V vs. Li⁺/Li⁰ (inset: digital images of electrodes). (c) Cycling performance of PVDF@CNT-Al@GN electrodes at 1.0 mg cm⁻² sulfur loading at 1.0 C. (d) Cyclic voltammetry (CV) curves of the LF@CNT-GN and PVDF@SP-Al electrodes in the 3rd cycle at a scan rate of 0.1 mV s⁻¹.



Fig. S6 Electrochemical impedance spectra of the Li–S batteries using LF@CNT-GN and blank electrodes (a) before and (b) after 50 cycles at 0.1 C.



Fig. S7 SEM images of the PVDF@SP-Al at (a) high magnification and (b) low magnification, and the LF@CNT-GN at (c) high magnification and (d) low magnification.



Fig. S8 Typical SEM images of different separators after 200 cycles: (a) PVDF@SP-Al electrodes with 1.0 mg cm⁻² sulfur loading (inset: blank separator), (b) LF@CNT-GN electrodes with 1.0 mg cm⁻² sulfur loading.



Fig. S9 (a) The SEM images of LF@CNT-GN electrode with 4.3 mg cm⁻² sulfur loading. (b) The charge-discharge curves

of the electrodes at 0.1 C within a potential window of 1.7-2.8 V vs. Li⁺/Li⁰, with the sulfur loading of 4.3 and 9.2 mg cm⁻². SEM images of the LF@CNT-GN electrode with 4.3 mg cm⁻² sulfur loading after 200 cycles at (c) high magnification and (d) low magnification.

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