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Electronic Supplementary Information

# Facile Fabrication of Covalently-bonded Carbon-Sulfur Composite from Lignin for High-Capacity Potassium Storage

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## Experimental Section

### Material Synthesis

All reagents in this experiment were applied directly without any further purification. The C-S-450 composite was synthesized via a one-step pyrolysis process: a mixture of lignin (dealkaline, Mucklin, AR) and cyclo-$S_8$ in a mass ratio of 1:4 was thoroughly ground in a mortar and then carbonized at 450 °C under an argon atmosphere in a tubular furnace. For comparison, the C-450 sample without sulfur was obtained by direct pyrolysis of pristine lignin under identical conditions. The C-450/$S_8$ composite was prepared through a melt-diffusion method by uniformly grinding C-450 with cyclo-$S_8$ at a mass ratio of 73:27, sealing the mixture in an evacuated glass tube, and heating at 155 °C for 12 hours.

### Material Characterization

Scanning electron microscopy (SEM) images were collected from a Hitachi FlexSEM1000 II. Transmission electron microscopy (TEM) images were collected from a JEOL JEM-2100F. Raman spectra were performed with a HORIBA LabRAM HR Evolution spectrometer with an excitation wavelength of 532 nm. Fourier transform infrared (FTIR) spectra were acquired on a Thermo 1701383S. Powder X-ray diffraction (PXRD) analysis was carried out on a miniflex600 diffractometer (Rigaku) with Cu Kα radiation, scanning at a rate of $10^{\circ}$ $min^{-1}$. Thermogravimetric analysis (TGA) was conducted using a Mettler-Toledo TGA2 in $N_2$ atmosphere, with a heating rate of 10 °C $min^{-1}$. For X-ray photoelectron spectroscopy (XPS), data were collected with a Thermo Scientific K-Alpha+. The C-S-450 electrodes for SEM tests were carefully extracted from the disassembled cells inside a glovebox filled with argon (The content of $O_2$ and $H_2O$ was below 0.1 ppm), followed by rinsing with the solvent diethyl carbonate (DEC) to remove residual salts.

### Electrochemical Measurements

The C-S-450 composite and the reference sample C-450 were used as active materials for battery tests. The electrode slurry was prepared by thoroughly mixing the active material, Super P, and a binder (consisting of sodium carboxymethyl cellulose and styrene-butadiene rubber in a 1:1 mass ratio) at a mass ratio of 7:2:1. After degassing, the slurry was coated onto a copper current collector and vacuum-dried at 60 °C for 12 hours. The mass loading of the active material was controlled within the range of 1.0~2.0 mg $cm^{-2}$. Metallic potassium (Sigma Ltd.) served as the

counter electrode in the K-ion batteries, with glass fiber (Whatman) used as the separator. The electrolyte for electrochemical testing consisted of 5 M potassium bis(fluorosulfonyl)imide (KFSI) dissolved in ethylene carbonate (EC) and enthyl methyl carbonate (EMC), (v/v, 1:1). All CR2032 coin cells were assembled in an argon-filled glove box with both $H_2O$ and $O_2$ contents maintained below 0.1 ppm. Galvanostatic charge/discharge tests were conducted at room temperature (~25 °C) with a Land battery testing system. Specific capacity values were calculated based on the total mass of C-S-450 or C-450 active materials, with tests performed with a voltage window of 0.01~3.0 V. The separator for Li-S batteries tests was polypropylene diaphragm (PP diaphragm), and the ester- and ether-based electrolyte were purchased with 1 M $LiPF_6$, EC: DEC (v/v, 1:1), 5% FEC and 1 M LiTFSI DME: DOL (v/v, 1:1), 2% $LiNO_3$. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) measurements were carried out on an electrochemical workstation (CHI 760E).

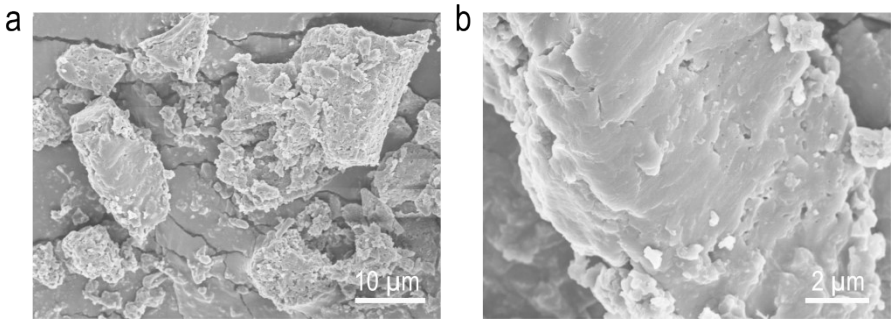


**Fig. S1.** (a-b) SEM images of pristine lignin.

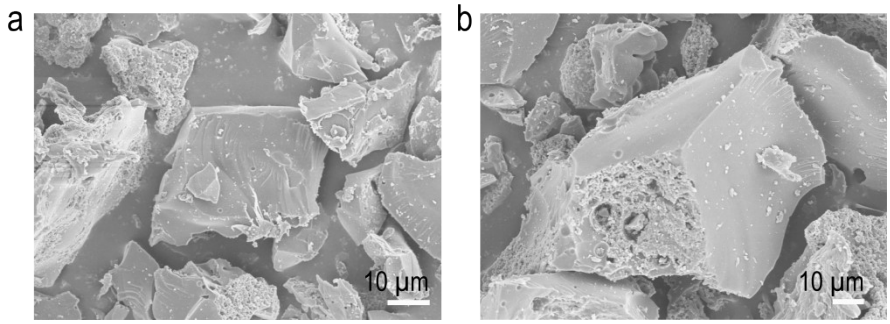


**Fig. S2.** (a-b) SEM images of C-S-450.

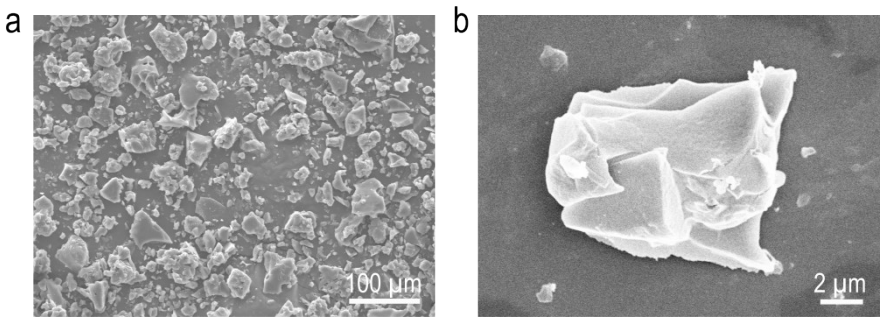


**Fig. S3.** (a-b) SEM images of C-450.

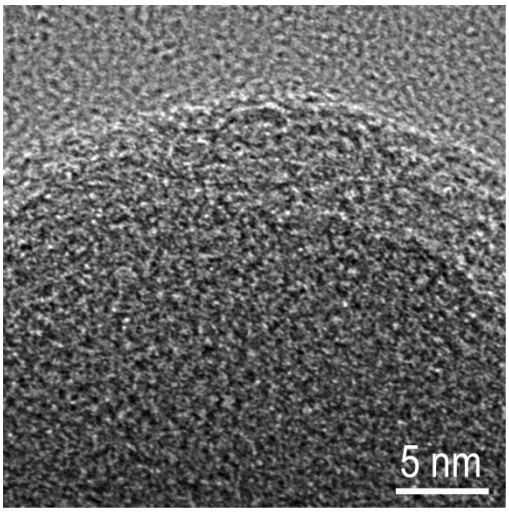


**Fig. S4.** HRTEM image of C-S-450.

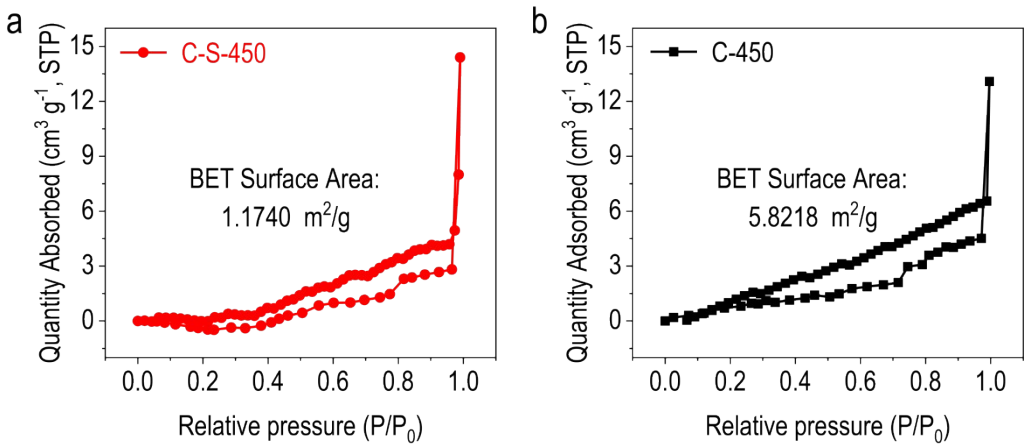


**Fig. S5.** $N_2$ adsorption and desorption isoterms of C-S-450 and C-450.

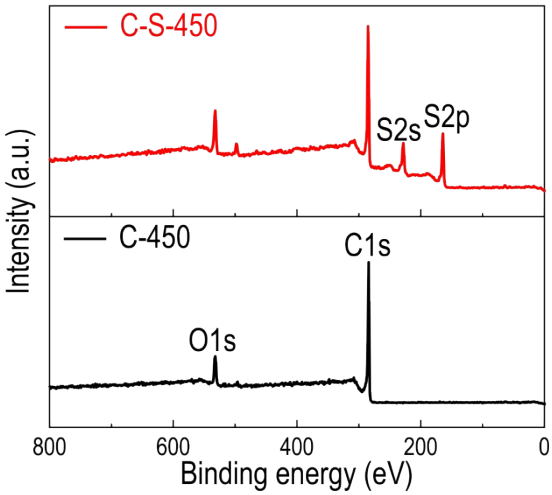


**Fig. S6.** XPS survey spectra of C-S-450 and C-450.

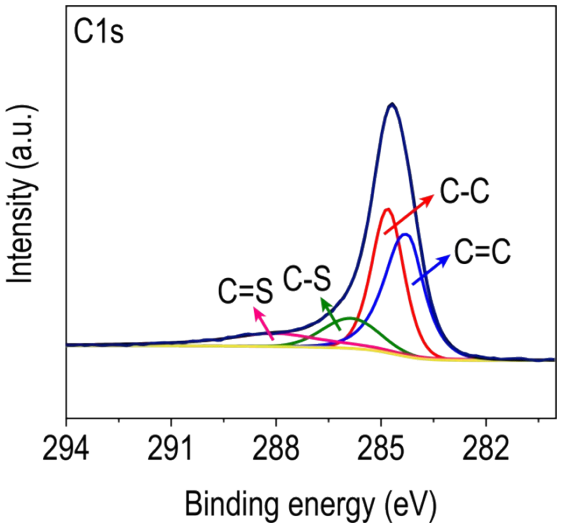


**Fig. S7.** High-resolution XPS spectra of C1s for C-S-450.

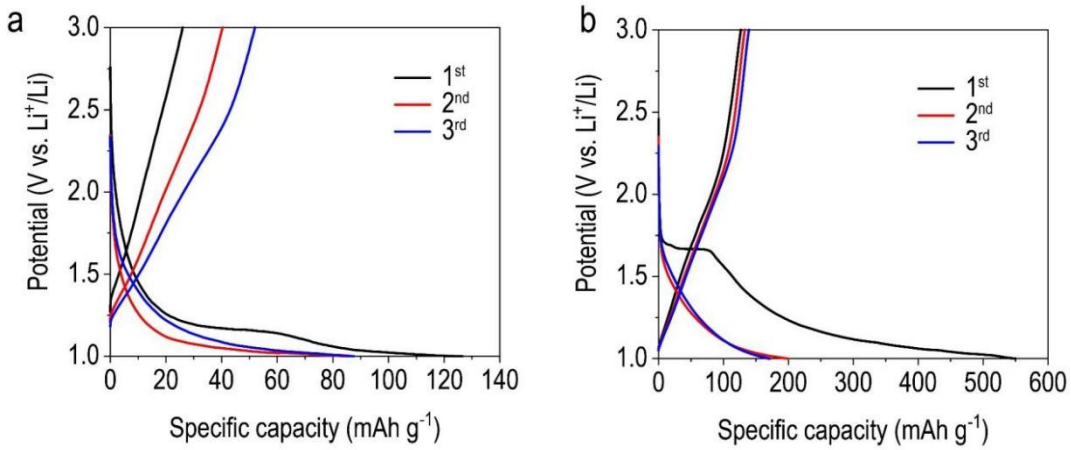


**Fig. S8.** (a) The first third charge-discharge curves of C-S-450 at 0.06 A $g^{-1}$ using ester-based electrolyte. (b) The first third charge-discharge curves of C-S-450 at 0.06 A $g^{-1}$ using ether-based electrolyte.

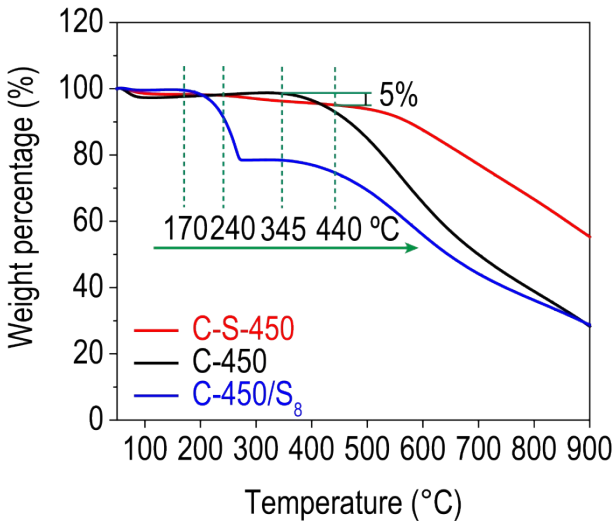


**Fig. S9.** TGA curves of C-S-450, C-450, and C-450/$S_8$.

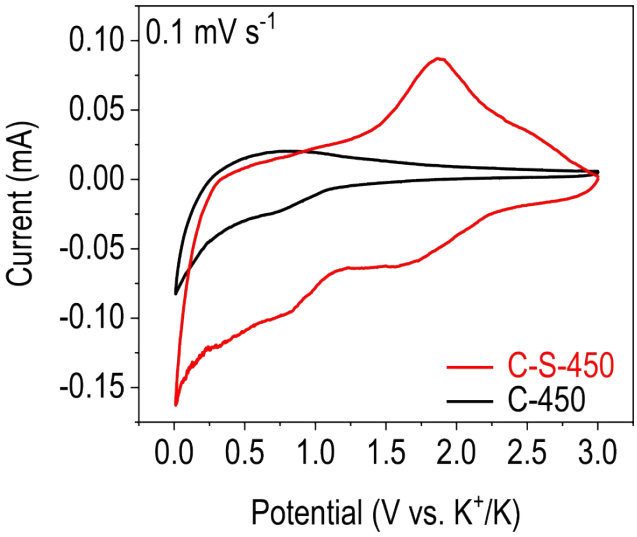


**Fig. S10.** CV curves of C-S-450 and C-450 at 0.1 mV $s^{-1}$ for the second cycle.

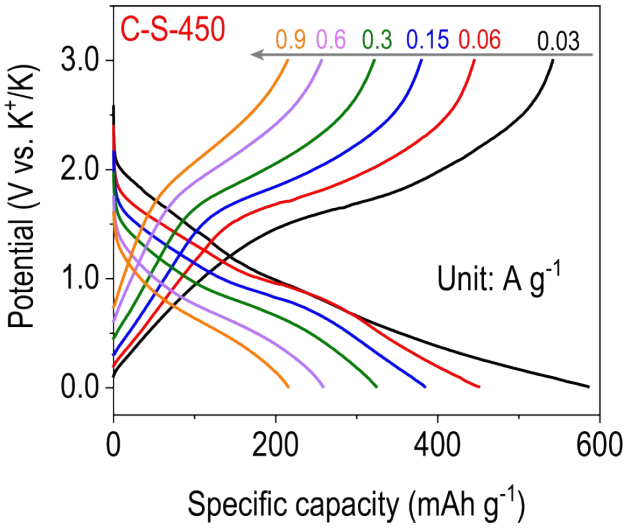


**Fig. S11.** The charge-discharge profiles of C-S-450 at different current densities.

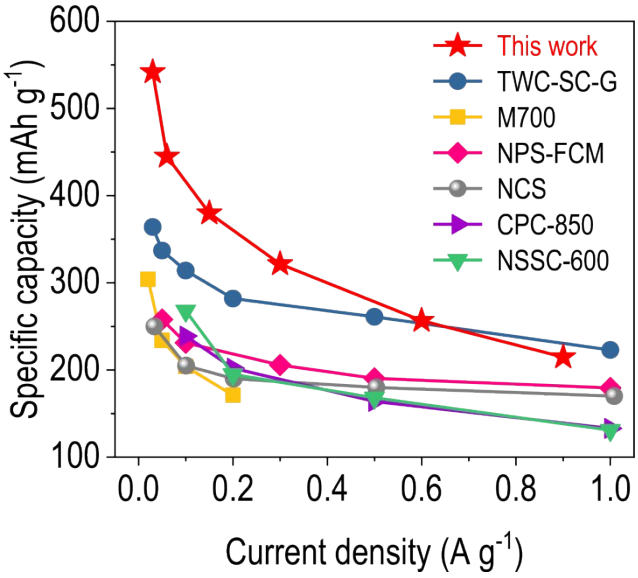


**Fig. S12.** Comparison of rate performance of C-S-450 with the reported biomass-derived carbon anodes for KIBs.[1-6]

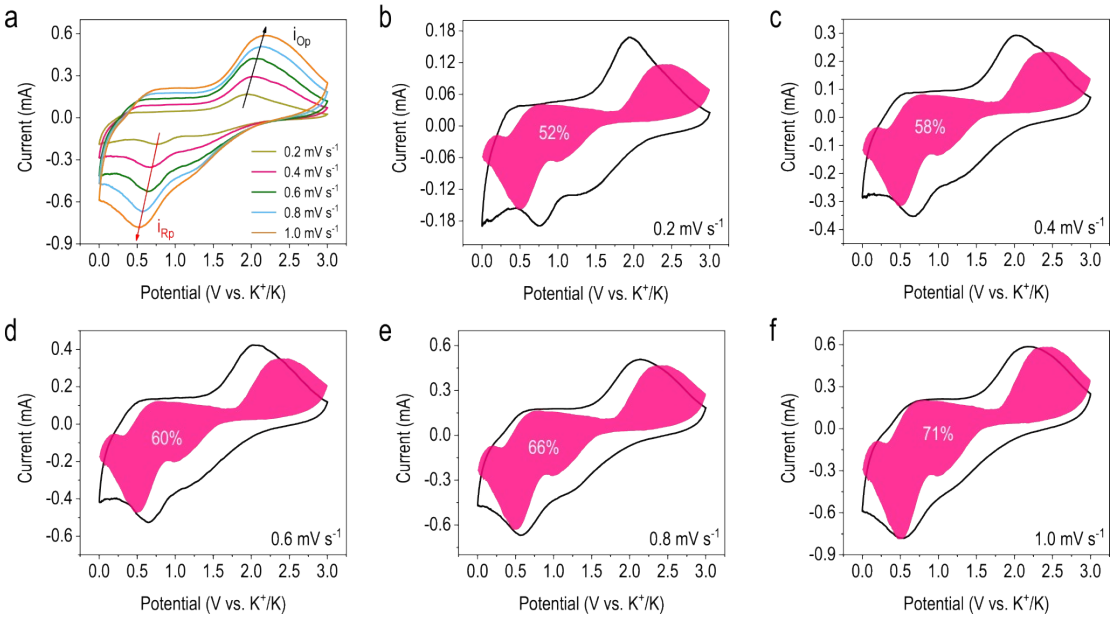


**Fig. S13.** (a) CV curves of C-S-450 at varying scan rates of 0.2-1.0 mV $s^{-1}$. (b-f) Contribution of capacitive process at varying scan rates: (b) 0.2 mV $s^{-1}$. (c) 0.4 mV $s^{-1}$. (d) 0.6 mV $s^{-1}$. (e) 0.8 mV $s^{-1}$. (f) 1.0 mV $s^{-1}$.

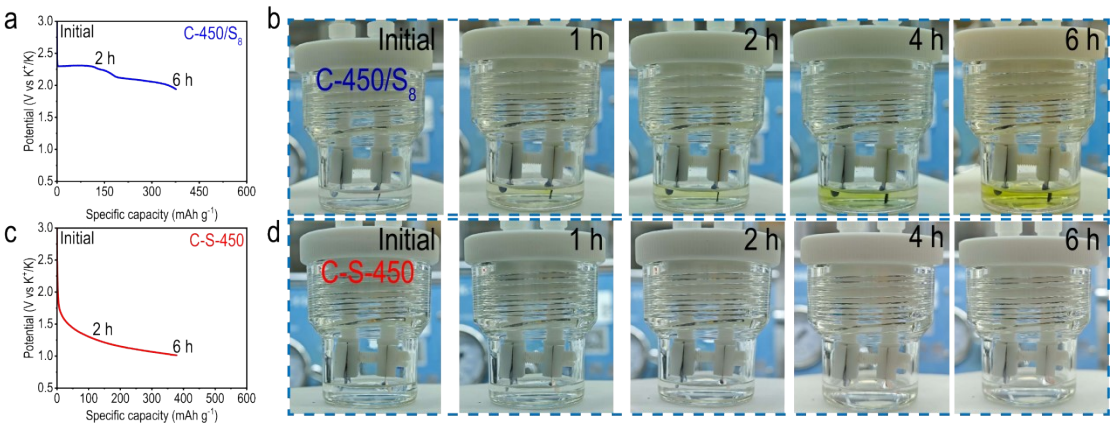


**Fig. S14.** (a, c) Discharge profiles of C-450/$S_8$ (a) and C-S-450 (c). (b, d) Optical photos of beaker cells based on C-450/$S_8$ (b) and C-S-450 (d). The K metal was used as counter electrode.

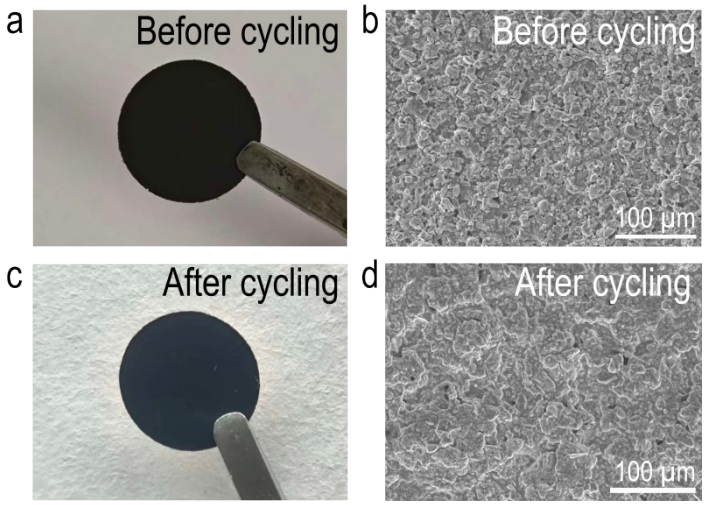


**Fig. S15.** (a-b) The optical photo (a) and SEM image (b) of C-S-450 electrode before cycling. (c-d) The optical photo (c) and SEM image (d) of C-S-450 electrode after cycling more than 150 cycles at 0.9 A $g^{-1}$.

**Notes and References**


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