Angewandte Chemie
Eine Zeitschrift der Gesellschaft Deutscher Chemiker

Supporting Information

An Aligned and Laminated Nanostructured Carbon Hybrid Cathode for High-Performance Lithium–Sulfur Batteries**

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Supporting Information

Experimental section

Spinnable carbon nanotube (CNT) arrays were prepared by chemical vapor deposition in a tube furnace. The catalyst was prepared by depositing 1 nm Fe and 10 nm Al_2O_3 in sequence on a silicon wafer. Ethylene was used as the carbon source carried by a mixture of Ar and H_2 gases. The spinnable CNT array was synthesized at 740 ℃ for 10 min. The as-synthesized CNT array showed a height of ~200 μm.

To prepare CMK-3@S particles, 100 mg CMK-3 and 300 mg sulfur were mixed by milling in a mortar. The obtained mixture was then co-heated at 160 ℃, where the viscosity of sulfur would reach the lowest, so the sulfur can be imbibed into channels of CMK-3 by capillary forces. Then 100 mg of CMK-3@S composite particles was dispersed into 10 mL of ethanol, followed by an ultrasonic treatment for 15 min.

CNT sheets were drawn from a spinnable CNT array. Five layers of CNT sheets were paved on a Cu foil, followed by adding 0.04 mL of CMK-3@S/ethanol suspension. After solvent evaporation, another five CNT sheets were cross-paved, followed by dipping another 0.04 mL of the suspension. The process can be repeated to obtain a set sulfur load. Benefited from the light weight of CNT sheets, the cathode retained a high sulfur weight percentage of 71 wt % as confirmed by the thermogravimetric analysis (**Figure S8**). Also, for the hybrid cathodes with sulfur loads of 1, 5, 10 and 20 mg cm^{-2}, the weight percentages of the sulfur are the same of 71 wt %.

The lithium sulfur battery was assembled in a standard 2025-type coin cell with a lithium foil as the anode. The electrolyte consisted of 1 M lithium bis(trifluoro-methanesulfonyl)imide and 0.25 M $LiNO_3$ dissolved in a mixture solvent of 1,3-dioxolane and 1,2-dimethoxyethane (volume ratio of 1/1). The amount of electrolyte added was tuned by the sulfur weight in the cathode with a ratio of 40 μL mg^{-1} (S). The flexible lithium sulfur battery was assembled with PDMS as the packing material. A CMK@S/CNT hybrid cathode and a Li foil were separated by a commercial separator of polyvinylidene fluoride. After the injection of electrolyte, they were sealed and taken out of the glove box.

The galvanostatic charge and discharge tests were conducted in the voltage window of 1.7-2.6 V at room temperature by ARBIN electrochemical workstation (MSTAT-5V/10 mA/16 Ch). The cyclic voltammogram measurements were also performed on the ARBIN electrochemical workstation at a scanning rate of 0. 1 mV s^{-1}. The sulfur content was measured by thermogravimetric analysis. The structure of the electrode was characterized by scanning electron spectroscopy (SEM, Hitachi, 4800-1), transmission electron microscopy (TEM, JEOL, JEM-2100F) and X-ray diffraction (XRD, Bruker AXSD8).

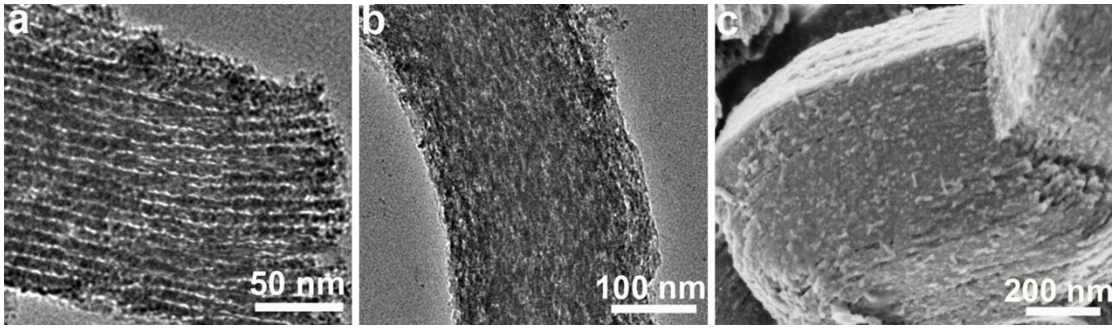

Figure S1. **a** and **b.** Transmission electron microscopy (TEM) images of CMK-3@S particles with CMK-3/S weight ratios of 1/2 and 1/4, respectively. **c.** Scanning electron microscopy (SEM) image of CMK-3@S particle with weight ratio of 1/4.

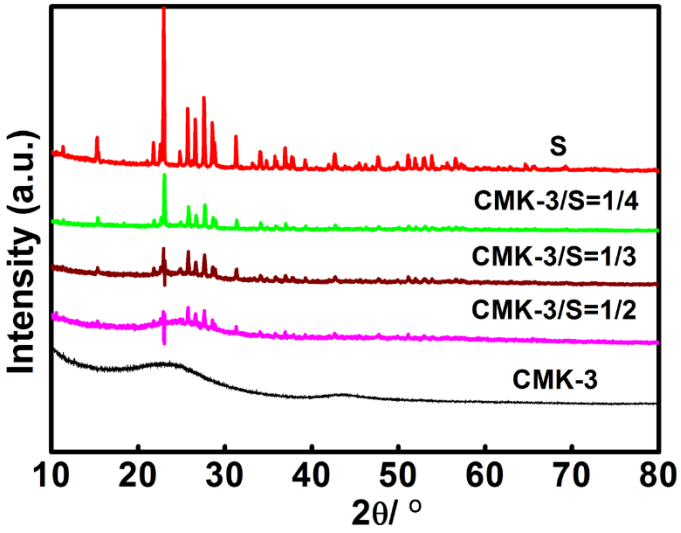

Figure S2. X-ray diffraction patterns of crystal sulfur, CMK-3 particles, CMK-3@S particles with CMK-3/S weight ratios of 1/2, 1/3 and 1/4.

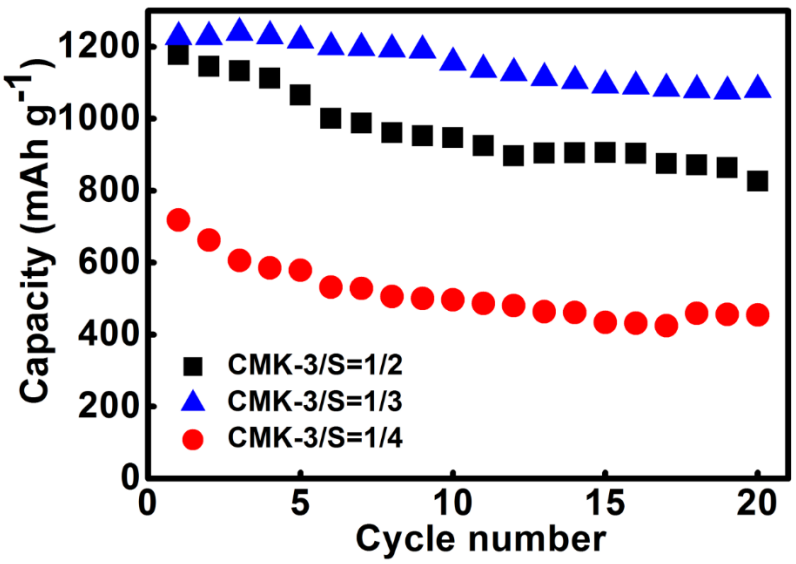

Figure S3. Electrochemical performance of the hybrid cathode with different weight ratios of CMK-3/S at 0.1 C.

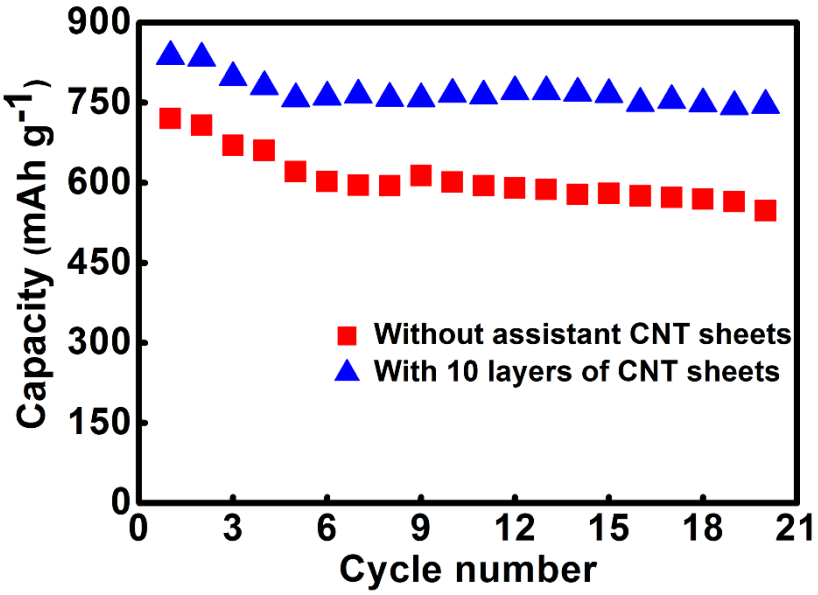

Figure S4. Cyclic performances with the CMK-3/sulfur weight ratio of 1/3 at 0.2 C.

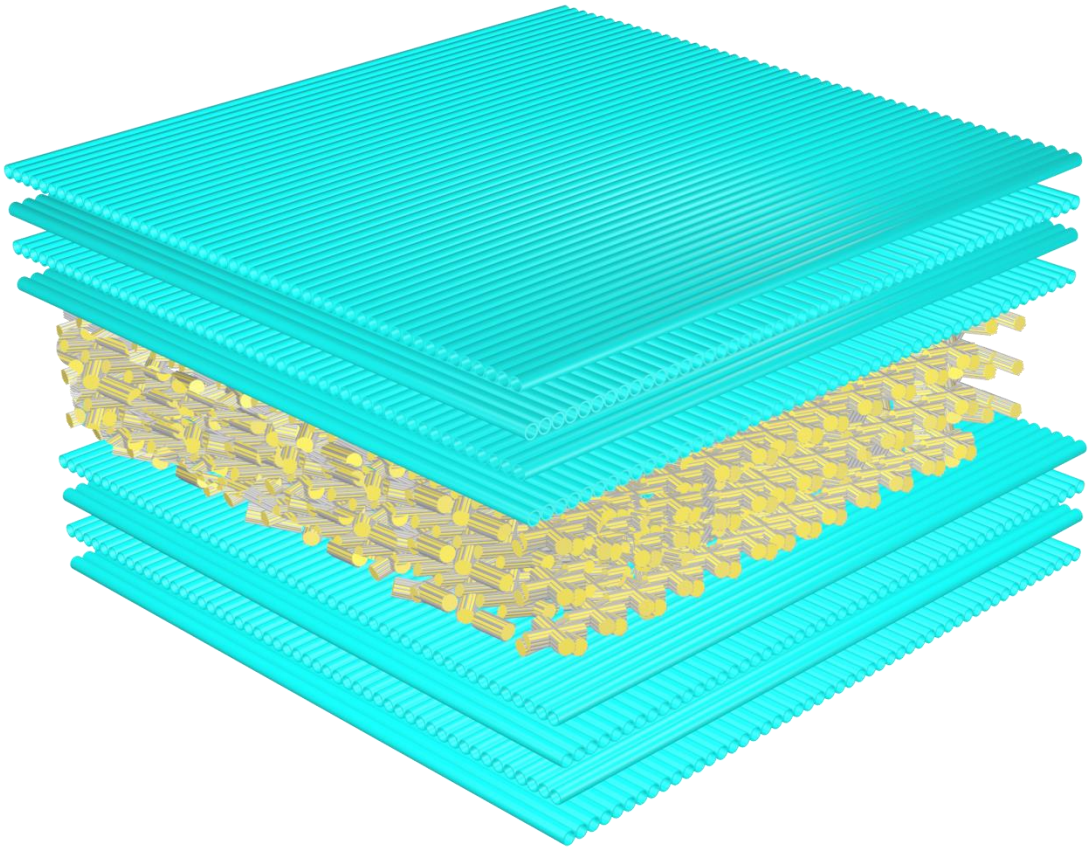

Figure S5. Schematic illustration to the hybrid cathode without the laminated structure.

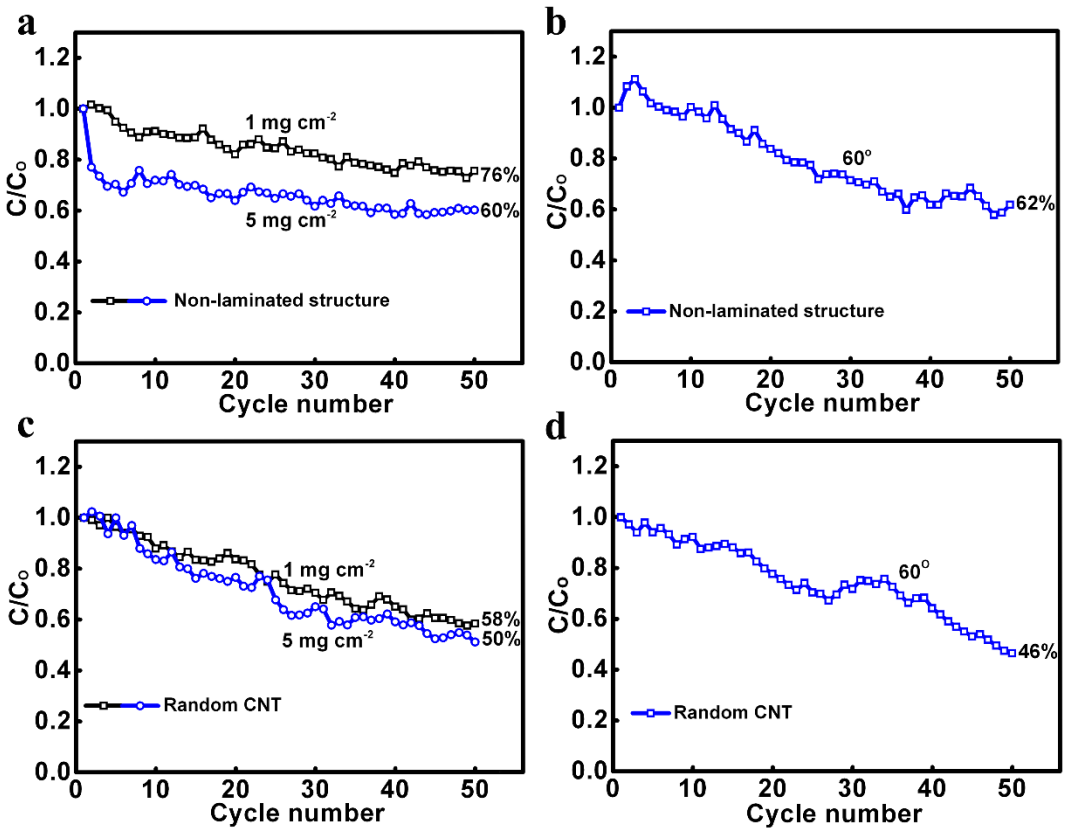

Figure S6. a. Capacity retentions for the batteries without the laminated structure at 0.1 C. **b.** Capacity retention for the battery without the laminated structure at a bent state at 1 C. **c.** Capacity retentions for the batteries using random CNTs at 0.1 C. **d.** Capacity retention for the battery using random CNTs at a bent state at 1 C. The sulfur loads for hybrid cathodes in b and d are the same of 1 mg cm^{-2}.

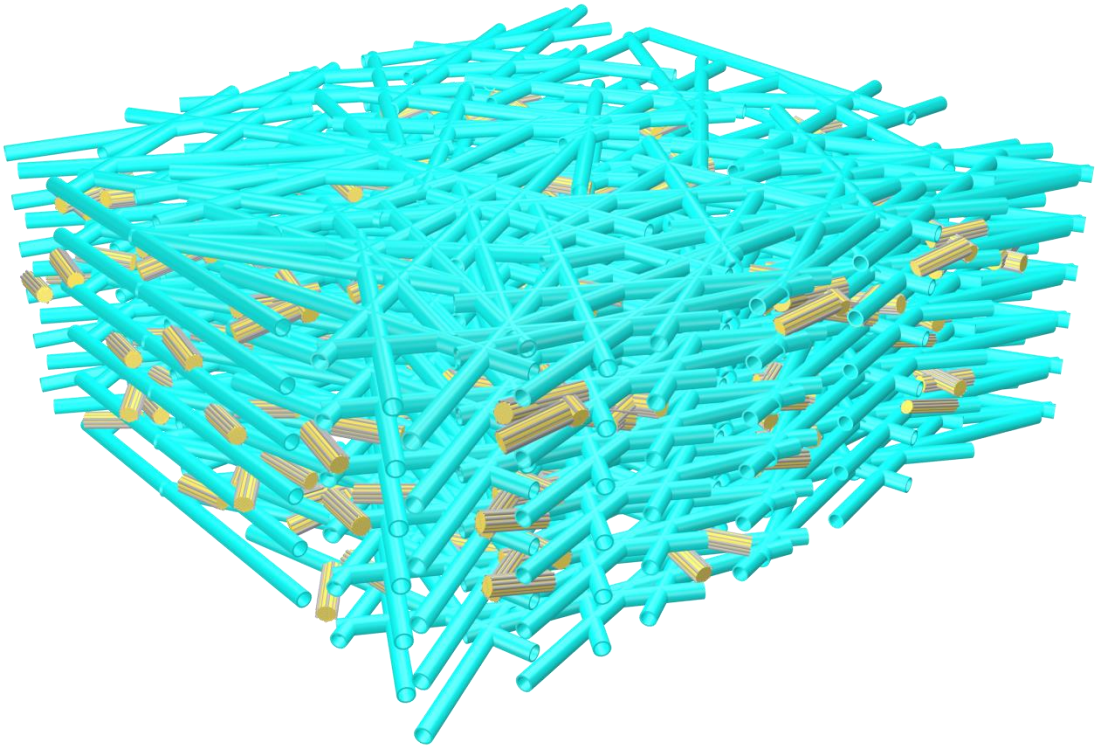

Figure S7. Schematic illustration to the hybrid cathode based on random CNTs.

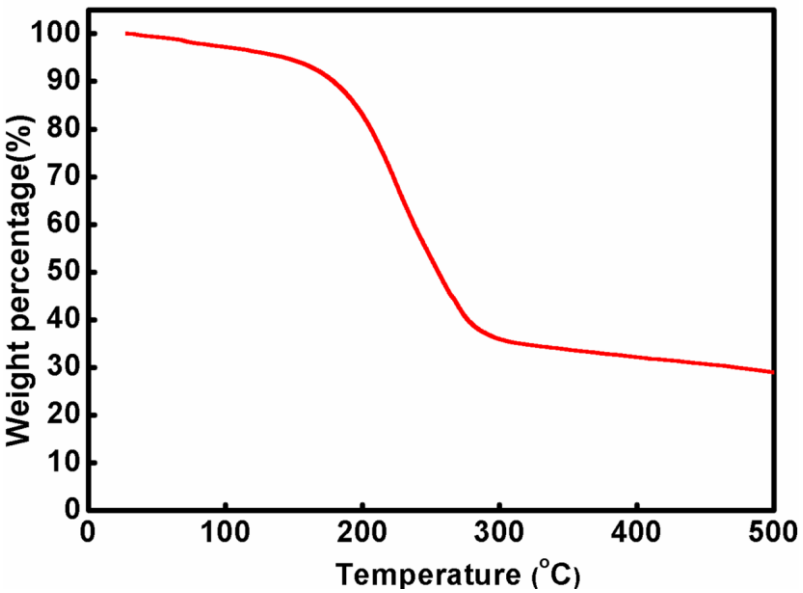

Figure S8. Thermogravimetric analysis of the hybrid cathode.