Supporting Information for

A binder-free sulfur/carbon composite electrode prepared by sulfur sublimation method for Li-S batteries

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Experimental section

1. Preparation of sulfur/carbon composite electrodes

Commercial binder-free carbon paper called buckypaper (Buckeye Composites) was used as a support for sulfur and the current collector in this study. To prepare the composite electrode, 1.5 g of sulfur powder (Fisher Scientific) was uniformly loaded in a 20 mL beaker. A disc of carbon paper (~ 10 cm^2) was put on top of the beaker. The beaker was heated at about 200 °C on a hot plate. The whole setup was installed in a fume hood with constant air flow (55 ft/min) to enhance sulfur vapor infiltration. The sulfur powder was fully melted into liquid phase and sulfur vapor with white/yellow mixed color went into the carbon paper. Four deposition times, which are 2, 4, 8, and 15 minutes, were applied for making these electrodes which are designated as SE-2, SE-4, SE-8 and SE-15, respectively. Finally the prepared electrode was cut into ~ 1 cm^2 discs, each contains 1.9 mg carbon.

2. Preparation of liquid electrolytes

The blank electrolyte used in this study was prepared by dissolving lithium bis(trifluoromethane)sulfonimide (LiTFSI, 98%, Acros Organics) in a mixture of dimethoxy ethane (DME, 99+%, Acros Organics) and 1,3-dioxolane (DOL, 99.5%, Sigma Aldrich) (1:1,

v/v) by magnetic stirring to render 1.0 M LiTFSI solution. Another electrolyte containing lithium nitrate ($LiNO_3$) additive was prepared by dissolving an appropriate amount of $LiNO_3$ (99+%, Acros Organics) in the blank electrolyte to render 1.0 M LiTFSI/0.1 M $LiNO_3$ solution. The electrolytes were prepared in an Argon-filled glove box.

3. Morphological characterizations

The morphological characterizations of the electrodes were conducted with a JEOL JSM-7800F field emission scanning electron microscopy (SEM). X-Ray Diffraction (XRD) patterns were recorded by using Cu-Kα radiation at 50 kV with an X-ray diffractometer (D8 Discover A25, Bruker AXS). N_2 sorption/desorption measurement was carried out on a Quantachrome Autosorb iQ gas sorption analyzer, and the pore size distribution was calculated based on the NLDFT model assuming a slit-shape pore structure.

4. Cell assembly

CR2032 coin cells were used and assembled in the Argon-filled glove box to evaluate the electrochemical performance of as-prepared electrodes. To prepare the cells, 20 μL of the electrolyte was added into an electrode, and then a Celgard® 2400 separator was placed on top of the electrode. Another 20 μL of the electrolyte was added on the separator. Finally, the lithium metal anode was placed on the separator. For the control cell without active material, a piece of blank carbon paper was used as the electrode. The cell was crimped and taken out of the glove box for testing. Cells with the blank electrolyte were made for the measurement of cyclic voltammetry. Cells with $LiNO_3$ additive in the electrolyte were made for evaluating cycle life.

5. Electrochemical measurements

Cells were galvanostatically discharged to 1.7 V and charged to 2.8 V on an Arbin battery cycler with 5-minute rest time between cycles. All cells were tested immediately after they were made.

The *C*-rate used for cycling measurements was based on the mass of sulfur in the electrode (1*C* = 1,672 mA g^{-1}). The specific capacity values shown in this paper are calculated by dividing the capacities obtained by the mass of sulfur. Cyclic voltammetry was performed on a Bio-Logic VSP potentiostat between 1.5 V and 3.0 V at a scanning rate of 0.05 mV s^{-1}.

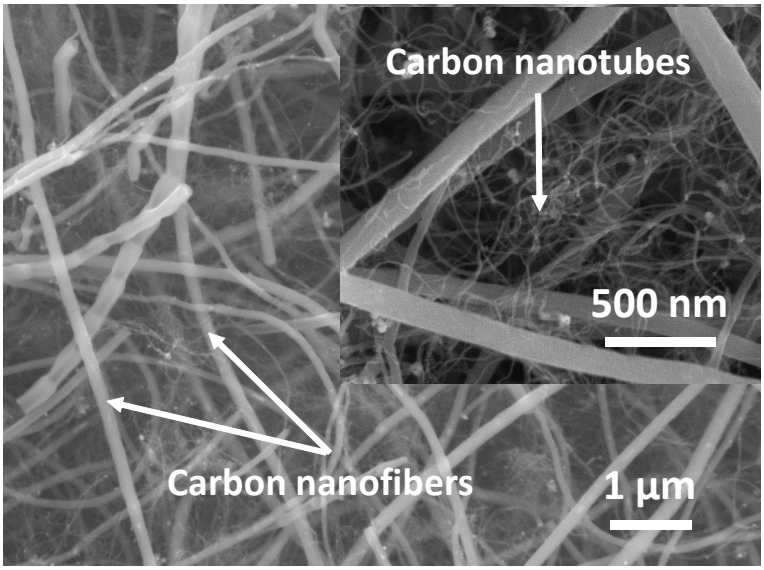

Fig. S1. SEM image of pristine carbon paper with inset magnified image to show carbon nanofibers and carbon nanotubes.

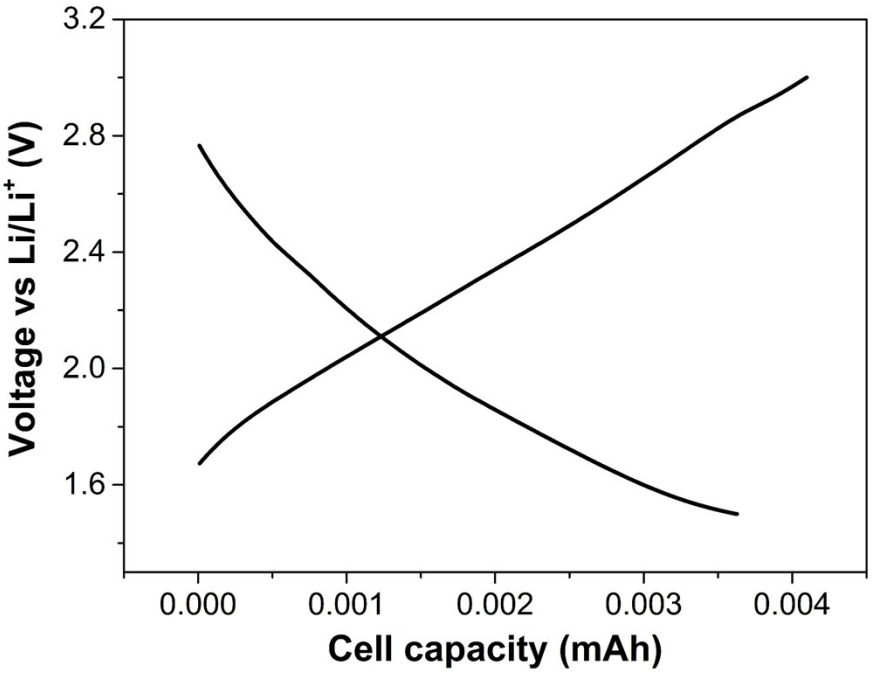

Fig. S2. Voltage profile for a control cell without active material to determine the capacity contribution from the carbon paper.

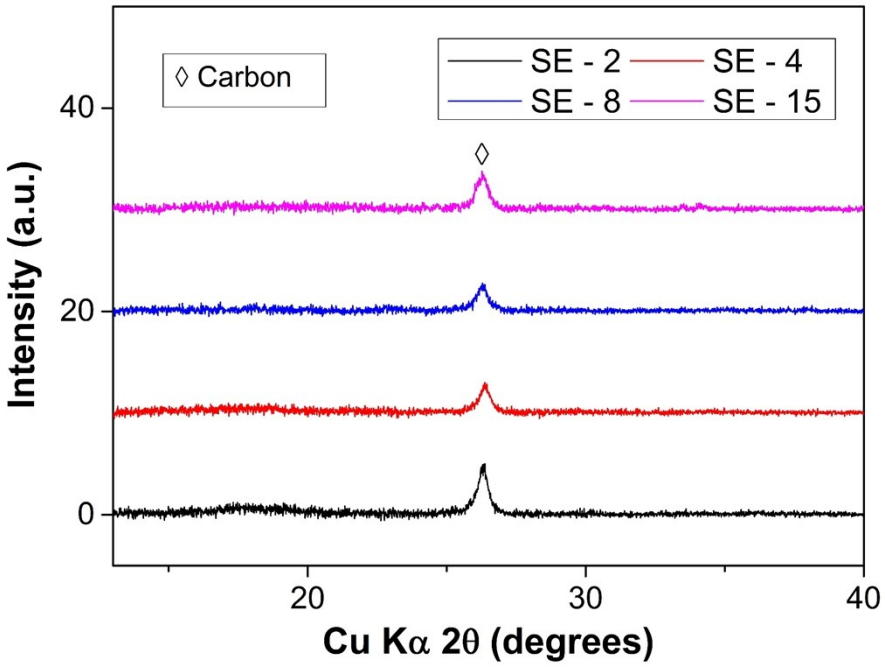

Fig. S3. XRD patterns of the composite electrodes SE-2, SE-4, SE-8, and SE-15 after one cycle.