

Supplementary information for

A Hierarchical Carbon Nanotube-Loaded Glass-Filter Composite Paper Interlayer with Outstanding Electrolyte Uptake Properties for High-Performance Lithium-Sulphur Batteries

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Experimental Procedure

Interlayers Preparation: : The GF/CNT paper was fabricated by ultrasonication of the GF paper in a solution containing CNTs. The CNT solution was purchased from Nano Solution (with 3 wt. % CNTs dispersed in isopropyl alcohol, TMCD-300D-IPA). It was diluted to 0.3% using isopropyl alcohol (IPA, 99.9%, Dongwoo Fine-Chem). In order to obtain well-dispersed CNTs in IPA, the CNT solution was bar-sonicated (Sonics & Materials, VC 750) for 20 min. A glass filter (GF/C, Whatman) was immersed into the sonicated CNT solution, bath-sonicated for 240 min, and dried in a vacuum desiccator at room temperature. For comparison with the GF/CNT paper, we prepared a GF paper and a CNT paper. The GF papers were used as purchased from Whatman and the CNT papers were prepared by vacuum filtration with a 0.3 wt. % CNT solution. The thickness of the CNT paper was set to 260 μm , identical to that of the GF paper. All papers were prepared by cutting with a diameter of 16 mm.

Cell Assembly: 2032 coin-type Li–S cells with the three types of interlayers (CNT, GF, and the GF/CNT composite) or without an interlayer were assembled with a conventional sulfur cathode (70 wt. % sulfur as the active material, 20 wt. % carbon black as the conductive

agent, and 10 wt. % polyvinylidene fluoride as the binder), a Celgard separator, and a lithium-metal anode in an argon-filled glove box. The sulfur electrode was cut into a circular pellet with a diameter of 14 mm and was used as a cathode. The mass loading of the sulfur cathode was approximately 1.6 mg cm^{-2} . The electrolyte was a solution of 1.0 M lithium bis-trifluoromethanesulphonylimide (LiTFSI) in a mixed solvent of 1,2-dimethoxyethane (DME) and 1,3-dioxolane (DOL; 1:1, v/v) with 0.2 M lithium nitrate (LiNO_3) as an additive. A mixed solvent without lithium salts was used as the washing solution for the cycled samples.

Characterization techniques: The morphologies of the GF, CNT, and GF/CNT papers were characterized with a field-emission scanning electron microscope (FE-SEM, Nova230, FEI) before and after cycling, as well as by energy-dispersive X-ray spectroscopy (EDS) after cycling. The thickness of each interlayer was measured using a micrometer (MDC-25PJ, Mitutoyo). The electrolyte uptake property was determined via Eq. (1) with the weights of the interlayers before and after soaking them in a liquid electrolyte.

$$\text{Electrolyte uptake (\%)} = (W_f - W_i) / W_i \times 100, (1)$$

where W_i and W_f are the weights of the interlayer before and after soaking in the electrolyte.

All electrochemical measurements were carried out at room temperature. The subsequent experiments were performed in a voltage range of 1.5 ~ 2.8 V (vs. Li^+/Li^0) using a battery cycle tester (WBCS3000, WonATech). The rate capabilities were evaluated by discharging at various current densities (0.1 C, 0.2 C, 0.5 C, 1 C, and 2 C) after charging at a constant current of 0.1 C. The cycle performance was investigated at rates of 0.2 C and 1 C for 230 and 300 cycles. The specific capacities were calculated based on the weight of the active materials in the sulfur cathode. The 1 C rate was equivalent to 1675 mA g^{-1} . Electrochemical spectroscopy measurements were taken with a potentiostat (PHE200, Gamry Instruments) in

a frequency range of 0.1 MHz to 10 MHz with an AC voltage amplitude of 5 mV at the open-circuit voltage.

[Figure S1]

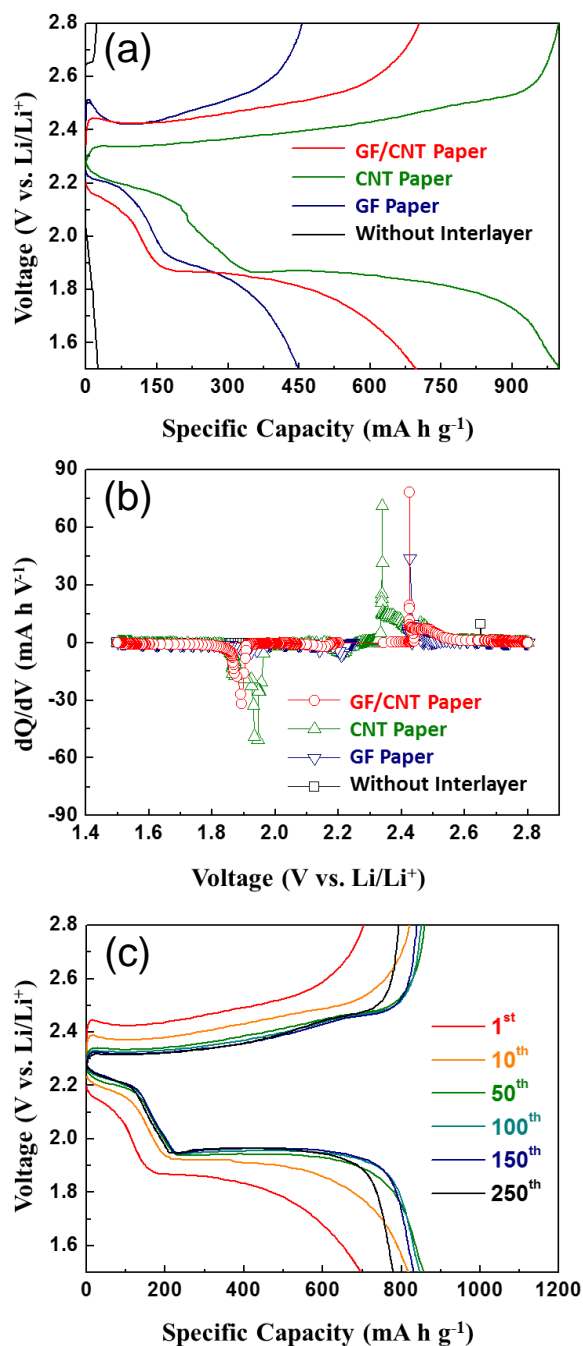


Fig. S1 Electrochemical properties of Li-S cells with different configurations: (a) Initial galvanostatic charge-discharge profiles of cells with different configurations at 1 C. (b) Differential capacity (dQ/dV) versus potential (V) profiles of cells with different configurations at the first cycle. (c) Galvanostatic charge-discharge profiles of cells with a GF/CNT paper interlayer at 1 C for different cycles within a potential window of 1.5 ~ 2.8 V vs. Li⁺/Li⁰.

[Figure S2]

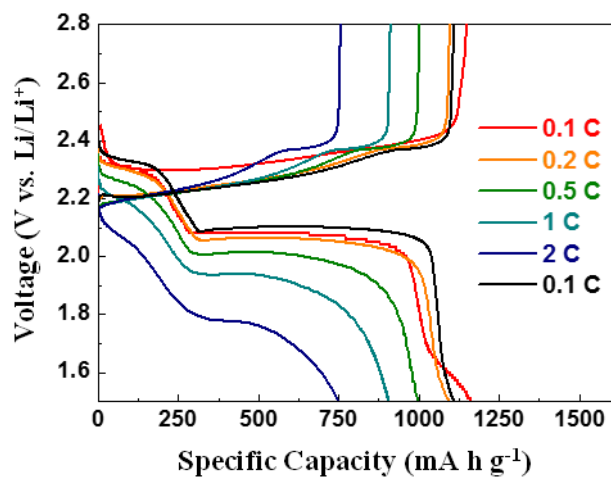


Fig. S2 Galvanostatic charge–discharge profiles of Li–S cells with a GF/CNT paper interlayer at different C rates within a potential window of 1.5 ~ 2.8 V vs. Li⁺/Li⁰.

[Figure S3]

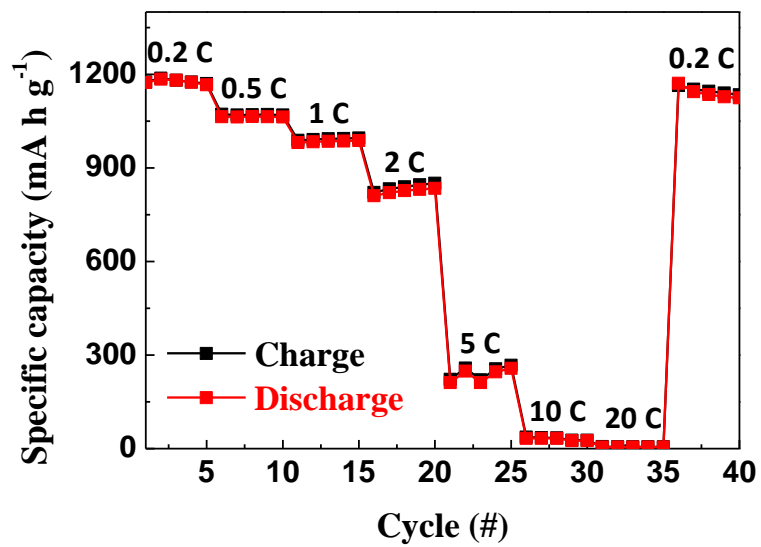


Fig. S3 Rate performances of cells with the GF/CNT paper interlayer at different C-rates within a potential window of 1.5 ~ 2.8 V vs. Li^+/Li^0 . (The C-rate is based on the theoretical capacity of sulphur, i.e., 1675 mA h g^{-1} .)

[Figure S4]

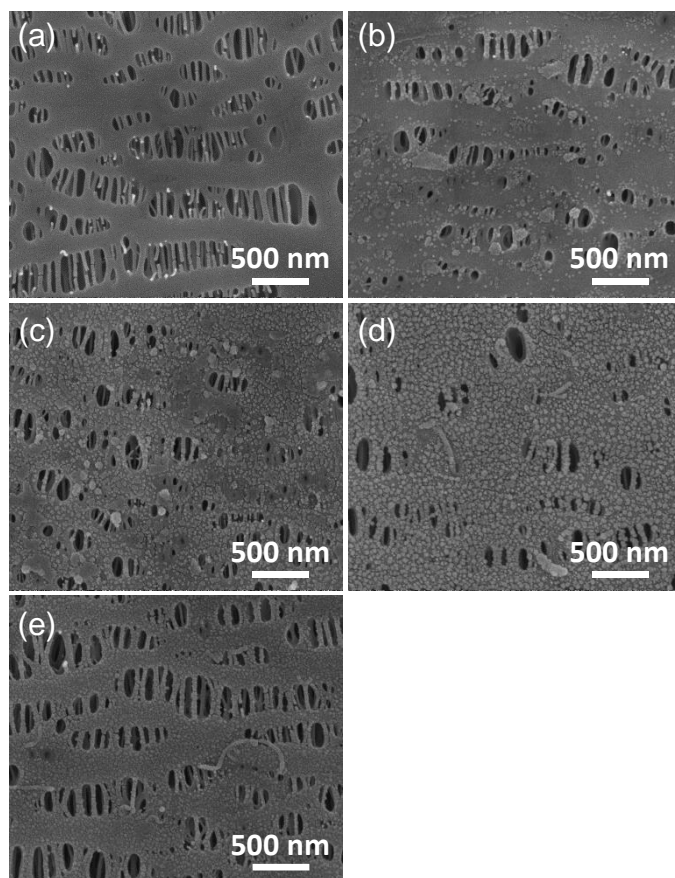


Fig. S4 SEM image of (a) a pristine Celgard separator and *ex-situ* SEM images of the separators used for (b) the pristine cell and for the cells with (c) the GF (d) the CNT and (e) the GF/CNT paper interlayers after 230 cycles at 0.2 C. These images were captured at the cathode side of the separator.

[Figure S5]

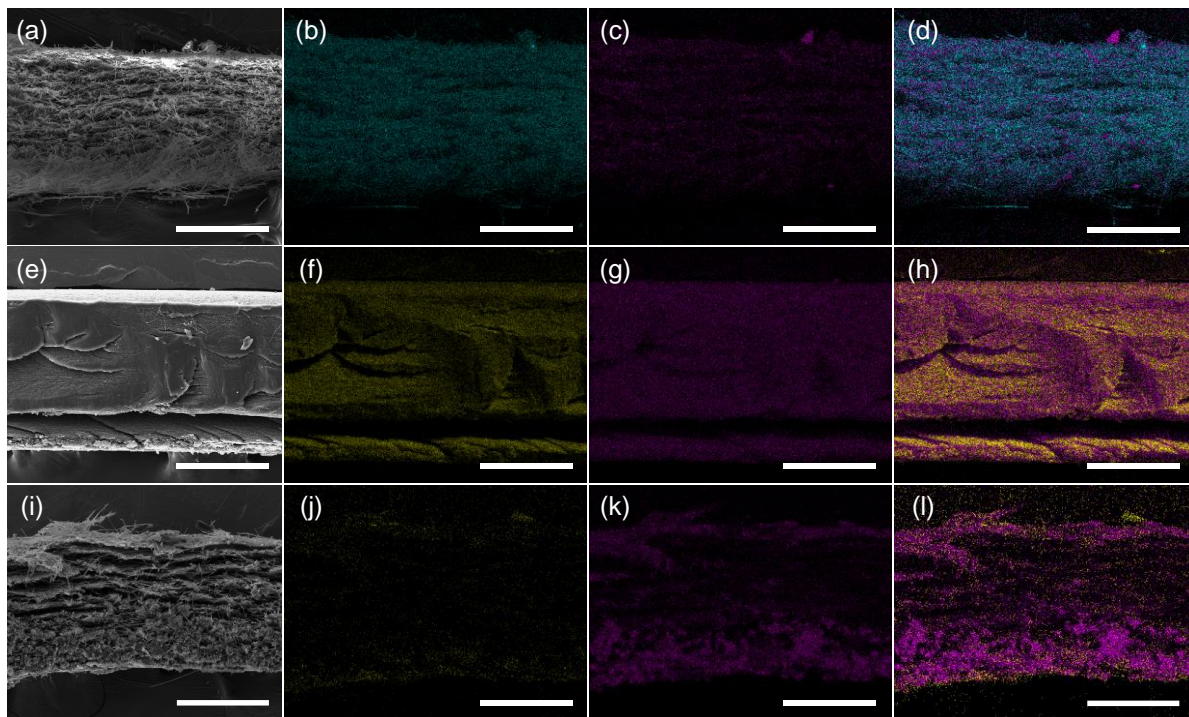


Fig. S5 *ex-situ* SEM cross-sectional images and the corresponding EDS dot mapping results of the backbone materials (silicon: cyan, carbon: yellow), sulphur (magenta), and overlay elements on the cross-section of (a-d) the GF, (e-h) the CNT, and (i-l) the GF/CNT paper interlayers measured after 230 cycles at 0.2 C. The scale bars represent a distance of 200 μm.