Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Electronic supplementary information

Ultra-lightweight PANiNF/MWCNT-functionalized Separators with Synergistic Suppression of Polysulfide Migration for Li-S Batteries with Pure Sulfur Cathodes

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Experimental

Polyaniline Nanofibers (PANiNF) Synthesis

The PANiNF was prepared by a non-arduous and large-scale interfacial reaction.[1-3] The interfacial reaction process was carried out by adding 0.08 mol of ammonium persulfate (98%, ACROS) to hydrochloric acid (37.2%, Fisher Scientific). The dopant acid solution was subsequently introduced into chloroform (99.5 %, Sigma) containing a 0.32 mol of distilled aniline monomers (99.5 %, Sigma). After 24 h of reaction, the PANiNFs were collected and washed with deionized water for five times. The fibrous morphology of obtained PANiNF is shown in the insert of Fig. 1(b) which is in agreement with the previous reports.[1-3]

Ultra-lightweight Functionalized Celgard polypropylene (PP) Separator Preparation

The PANiNF (2.5 mg) and the MWCNT (2.5 mg) (Nanolab, Inc.) were dispersed in 300 mL of isopropyl alcohol. After being ultra-sonicated for 30 min, the PANiNF/MWCNT suspension was vacuum-filtered through a commercial Celgard 2500 membrane, followed by drying in a vacuum oven for 24 h at 50°C as the PANiNF/MWCNT-functionalized separator. The MWCNT-functionalized separator was prepared by the same procedure. The suspension had 5.0 mg

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MWCNT. The diameter, weight, and thickness of the coatings are, respectively, 1.9 cm, *c.a.* 0.01 mg cm^{-2}, and about 8 µm (Fig. S1 and S2).

Electrochemical and Microstructural Analyses

Electrochemical tests were carried out with CR2032-type coin cells. The Li-S cells were assembled with pure sulfur cathode, PANiNF/MWCNT-functionalized separator or the MWCNT-functionalized separator, Li foils (0.38 mm x 23 mm, Sigma) and nickel foam spacers. The pure sulfur cathode was prepared by mixing the precipitated sulfur with 25 wt. % Super P carbon and 15 wt. % polyvinylidenefluoride binder. The diameter and the active-material loadings of the sulfur cathodes are, respectively, 1.2 cm and 1.5 ~ 1.3 mg cm^{-2}. The electrolyte contained 1.85 M lithium trifluoromethane sulfonate and 0.1 M lithium nitrate in a mixed 1,2-dimethoxyethane and 1,3-dioxolane solution (1:1 vol). The sample cells employing the (PANiNF/)MWCNT-functionalized separators and the control cells employing the Celgard separators were controlled to have the same amount of electrolyte.

Discharge/charge processes, cycling performance, and Q_H/Q_L analysis were evaluated with an Arbin cycler between 1.8 and 2.8 V. Cyclic voltammetry (CV) measurements were executed with a potentiostat (VoltaLab PGZ 402, Radiometer Analytical) between 1.8 and 2.8 V at a 0.1 mV s^{-1} scanning rate. The electrochemical impedance spectroscopy (EIS) of the fresh cells was recorded with a computer-interfaced impedance analyzer (SI 1260 and SI 1287, Solarton) in the frequency range of 1 MHz to 100 mHz. The self-discharge behavior of the cell was monitored by measuring the first discharge capacity of the cell (at fully charged stage of 2.8 V) at C/5 cycling rate after a 12-hour rest. Microstructure change and elemental analysis of the functionalized separators before cycling and after 100 cycles (at the charged state of 2.8 V) were carried out with a field emission scanning electron microscope (FE-SEM, Quanta 650, FEI) with energy dispersive X-ray (EDX) spectrometers.

Supporting figures

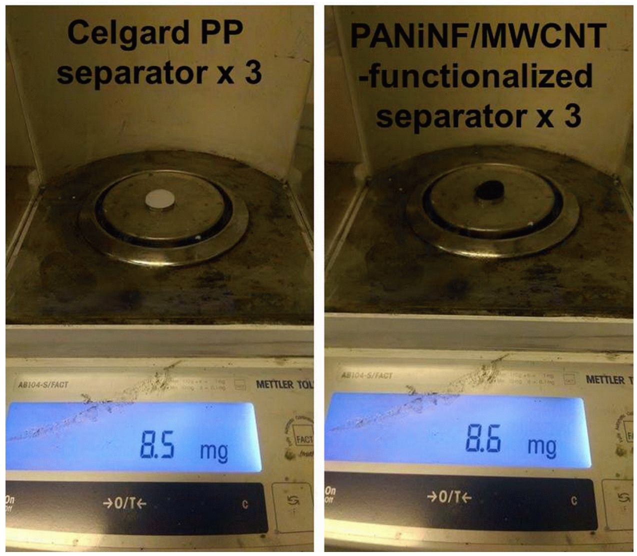

Fig. S1 The weights of the bare commercial Celgard separators and the PANiNF/MWCNT-functionalized separators.

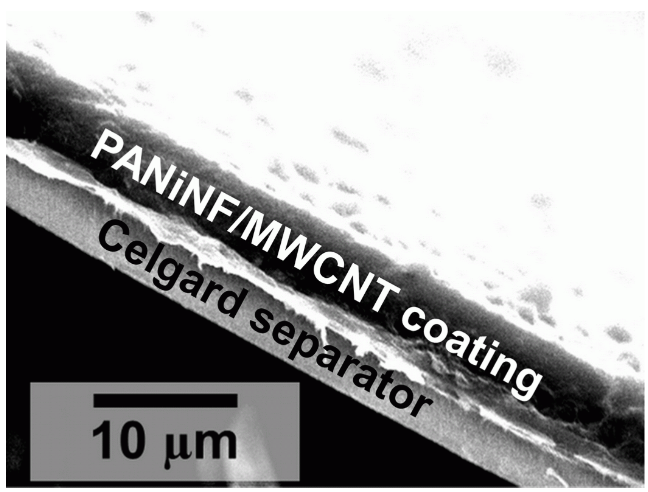

Fig. S2 Thickness of the PANiNF/MWCNT coating.

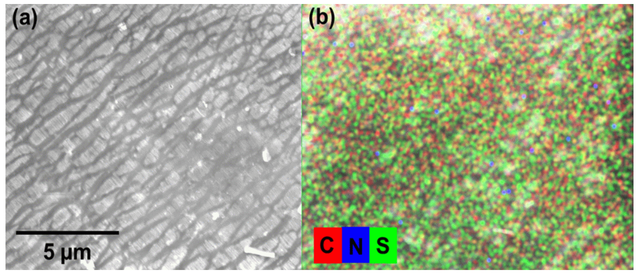

Fig. S3 SEM images of the bare Celgard separator (a) after 100 cycles. (b) Elemental mapping result of (a): carbon signals (red); nitrogen signals (blue); sulfur signals (green).

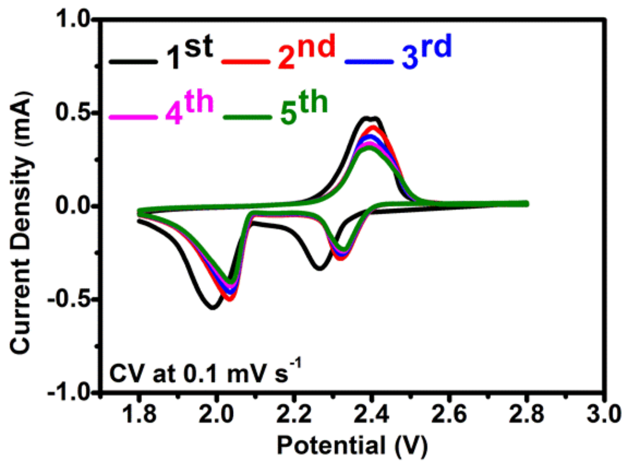

Fig. S4 CVs of the Li-S cells with the bare Celgard separators.

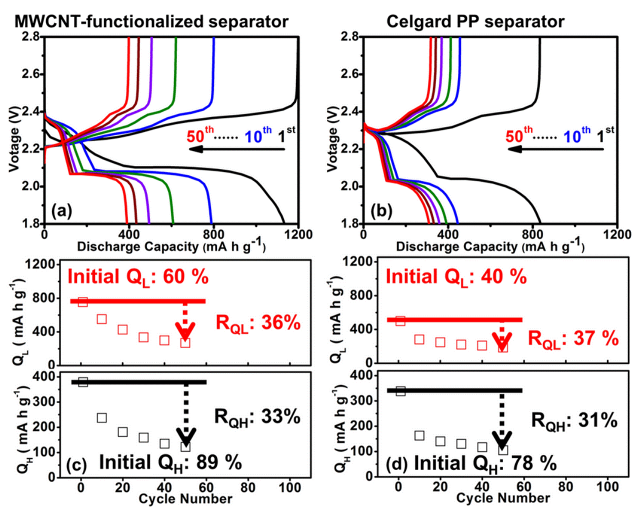

Fig. S5 Discharge/charge curves and reversible Q_H/Q_L of Li-S cells employing (a,c) MWCNT-functionalized separators and (b,d) Celgard PP separators at C/5 rate.

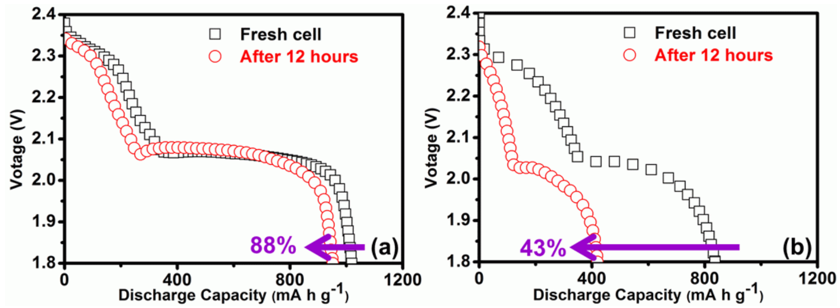

Fig. S6 Self-discharge behavior of Li-S cells (a) with and (b) without the ultra-lightweight PANiNF/MWCNT-functionalized separator with 12 hours of resting.