Supporting Information

Highly stable performance of lithium-sulfurized polyacrylonitrile battery using a lean ether-based electrolyte

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1.Experimental Section

Materials Synthesis

The SPAN are prepared as reported [1]: the weight ratio of sulfur and PAN was 4:1, whereas the mixture was carried out via ball-milling at 500 rpm for 6 h, then heated at 155 °C for 2 h and at 300 °C for 6 h under a continuous N_2-gas flow. The content of sulfur in the synthesized SPAN composite is 42% (Carbon sulfur analyzer). The SPAN was used for preparing electrode as follows: the slurry was prepared by mixing SPAN, acetylene black, and CMC in a mass ratio of 7:2:1, and stirred with deionized water, then the slurry was coated onto aluminum foil using doctor blade and dried under vacuum, then punched into 10 mm disks as the working electrode. The typical SPAN loading is about 6-7 $mg \cdot cm^{-2}$.

The different kinds of electrolytes were prepared: the C0 is 1 M $LiPF_6$ in ethylene carbonate/dimethyl carbonate (EC/DMC/EMC, volume ratio 1:1:1); E0 is 1 M LITFSI in 1, 2-dimethoxyethane and 1,3-dioxolane (DME/DOL, volume ratio 1:1); EL0 is 1 M LITFSI in DME/DOL, 1:1 with 0.2 M $LiNO_3$; EL1 is 1 M LITFSI in FEC/DME/DOL, 1:4.5:4.5 with 0.2 M $LiNO_3$; EL3 is 1 M LITFSI in FEC/DME/DOL, 3:3.5:3.5 with 0.2 M $LiNO_3$; EL5 is 1 M LITFSI in FEC/DME/DOL, 2:1:1 with maximum solubility of 0.15 M $LiNO_3$, which all obtained from fusai corporation and used without further drying.

Materials Characterizations

Morphological was obtained by using scanning electron microscope (SEM) and digital camera. The distribution of element was determined by energy dispersive X-ray spectroscopy (EDX).

Electrochemical measurement

The CR2032-type coin cells were assembled in an Ar-filled glove and test using LAND-CT2001A testers. The Li-SPAN battery is conducted with lean electrolyte (7 $\mu l/mg_{span}$) and 4

mAh cm^{-2}, in which, SPAN is used as cathode and lithium metal foil as anode, while Celgard 2400 used as the separator. AC impedance of symmetric lithium cells were measured over the frequency range from 1 Hz to 0.1 MHz. All the electrochemical measurements were carried out in a dry atmosphere at 25 °C.

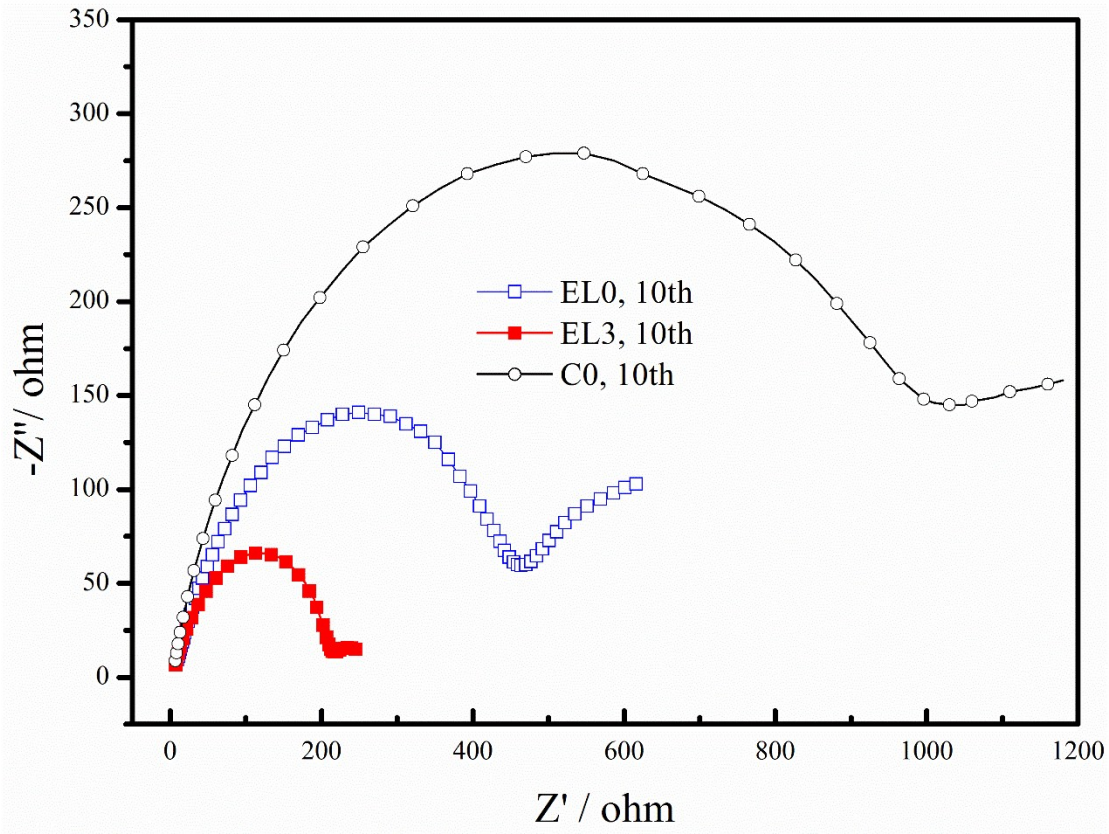

Fig S1. Electrochemical impedance spectroscopy of Li/Li symmetrical cell after 10 cycles.

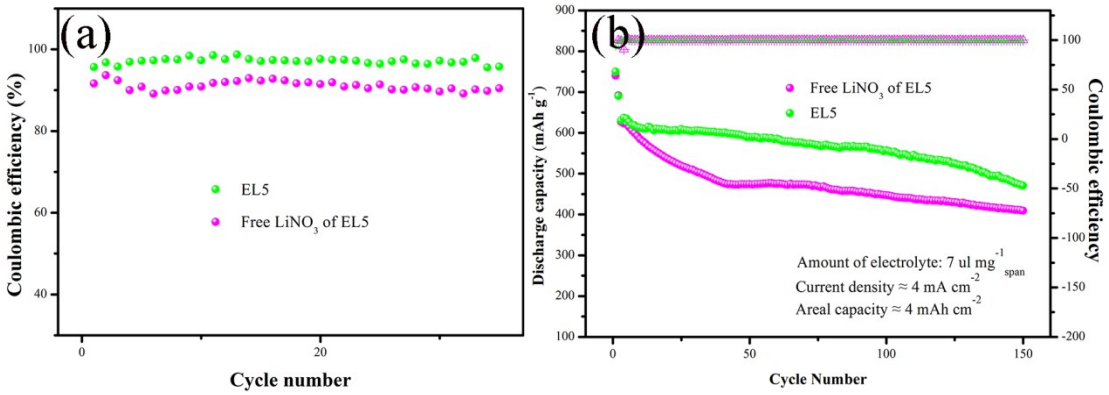

Fig S2. (a) Coulombic efficiency (CEs) of Li−Cu half cells (b) Electrochemical performance of Li-SPAN cells with EL5 and free $LiNO_3$ of EL5 at 1 C.

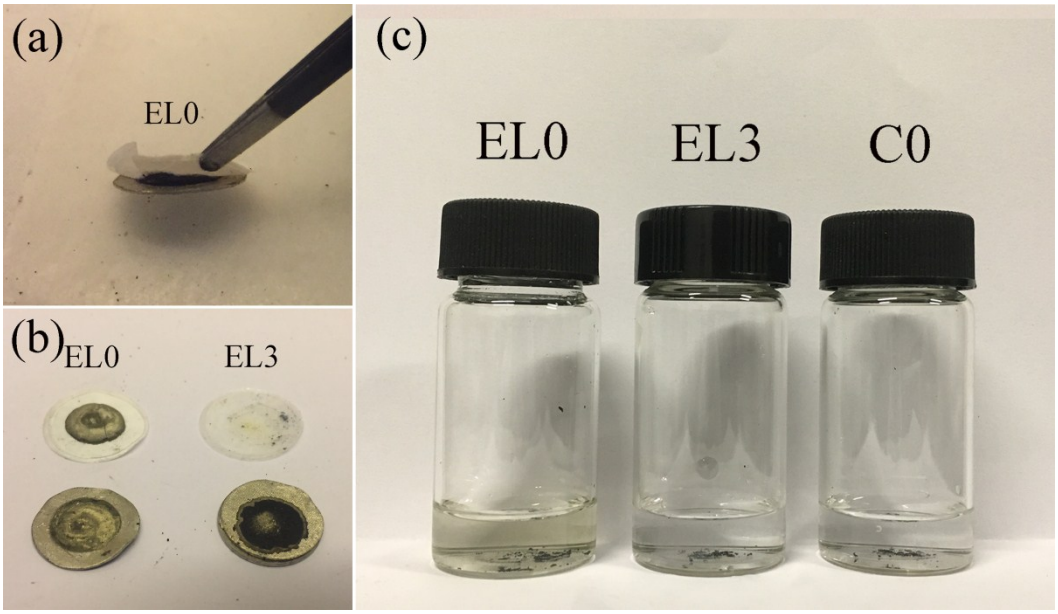

Fig. S3. (a, b) the photograph of lithium and separator in EL0 after several cycles, (c) the color of electrolyte when the disassembled electrode immersed in it.

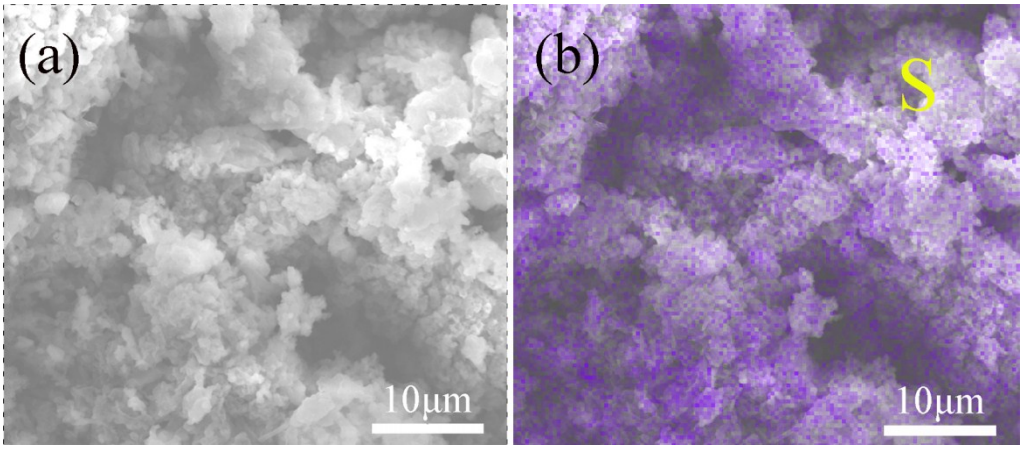

Fig. S4. (a, b) surface morphology and EDX of mossy lithium on the Celgard 2400 separator after several cycles with EL0 electrolyte.

Reference

[1] Y. Shuai, Z. P. Zhang, K. H. Chen, J. Lou, Y. Wang, Chemical Communications, 2019, DOI: 10.1039/C8CC09372E