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# Supporting Information

## High Sulfur Loading Lithium-Sulfur Batteries Based on

## Upper Current Collector Electrode with Lithium-ion

### **Conductive Polymer**

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#### **Supporting Information**

#### **Experimental Section**

#### Preparation of AI-S-VGCF electrode

The fabrication of the AI-S-VGCF electrode is quite facile. First, commercial sulfur powder (95 wt %) and poly(vinylidene fluoride) (PVDF, 5 wt %) binder were mixed and dispersed well in N-methyl-pyrrolidone (NMP), and then coated onto an AI foil substrate by using the doctor blade casting method to obtain AI-S electrode, vacuum drying overnight at 60 °C. Through restricting the amount of NMP solvent and the thickness of S layer, the AI-S electrodes with different areal sulfur loadings could be obtained easily. Second, the commercial VGCF (90 wt%), PEO (5 wt%) and carboxy methylated cellulose (CMC, 5 wt%) binders were mixed and dispersed well in deionized water, and then coated onto the AI-S electrode to obtain AI-S-VGCF electrode, vacuum drying at 60°C.

#### Characterization

The morphologies and element mapping of as-prepared materials were characterized by using a scanning electron microscope (SEM, HITACHI S-4800) equipped with energy dispersive X-ray spectroscopy (EDX) for elemental analysis. Transmission electron microscopy (TEM) images was tested on JEM-2100 (JEOL, Japan) at 200 kV. The laser Raman spectra were recorded at the resolution of 1 cm<sup>-1</sup> in back scattering (180°) configuration using 532 nm excitation. The XRD patterns were recorded by the Philips X'pert Pro Super X-ray diffract meter and Cu Kα radiation.

#### **Electrochemical measurements**

The CR2016-type coin cells were assembled using the prepared electrodes and lithium metal in an argonfilled glove box. The electrolyte was formed by adding lithium bis(trifluoromethanesulfonyl)imide (LiTFSI, 1 M) salt and lithium nitrate (LiNO<sub>3</sub>, 1 wt%) into the mixture of 1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME) at the volume ratio of 1:1. The galvanostatic charge-discharge experiments were measured at different current densities between 1.8 and 2.6 V (vs. Li<sup>+</sup>/Li) using the CT2001A cell test instrument (XINWEI Electronic Co.). Galvanostatic intermittent titration technique (GITT) profile was recorded by applying a constant current (50 mA g<sup>-1</sup>) for 10 min, followed by a resting step of 10 min.



Figure S1. (a) SEM image of the VGCF, (b) TEM image of the VGCF, (c) XRD pattern of the VGCF and

(d) Raman spectrum of the VGCF.



Figure S2. The rate performance of the high mass loading AI-S-VGCF electrode.



Figure S3. Element mapping of cross-section of AI-S-VGCF electrode after cycling 100 cycles (charge

state) (sulfur areal loading of  $\sim$  5 mg cm<sup>-2</sup>).



Figure S4. Element mapping of surface of AI-S-VGCF-PEOCMC electrode before cycling and after

cycling 100 cycles (charge state) (sulfur areal loading of  $\sim$  5 mg cm<sup>-2</sup>).



Figure S5. Element mapping of surface of AI-S-VGCF-CMC electrode before cycling and after cycling

100 cycles (charge state) (sulfur areal loading of  $\sim$  5 mg cm<sup>-2</sup>).



**Figure S6.** Cycling performance of Li/(AI-S-VGCF) cell without PEO (at the first cycle at the current density of 50 mA  $g^{-1}$  and after at the current density of 100 mA  $g^{-1}$ ).



Figure S7. Cycling performance of Li/(Al-S-VGCF) cell (at the first cycle at the current density of 50 mA  $g^{-1}$  and after at the current density of 100 mA  $g^{-1}$ ).