

Electronic Supplementary Information (ESI)

## Sulfur-graphene composite with molybdenum particles for stabilizing lithium-sulfur batteries

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### Experimental Details

**Synthesis of GO.** Graphite oxide suspension was synthesized as the previous literature.<sup>1</sup> Graphene oxide (GO) suspension was obtained by ultrasonically dispersing graphite oxide with probe sonicator and then dried in a vacuum oven at 60 °C.

**Preparation of the Thermally Exfoliated Reduced Graphene.** Thermally exfoliated reduced graphene was prepared by putting the dried GO in a quartz tube purged with argon gas, into a horizontal quartz tube furnace followed by annealing at 800 °C for 1 h.

**Preparation of S/G composite.** The composite of sulfur and graphene (S/G) was prepared by mechanically mixing sublimed sulfur and the thermally exfoliated graphene and then undergone a vacuum-assisted thermal treatment. In brief, the mixture of sulfur powder and graphene in a 50 ml flask with the mass ratio of 5:1 was wetted by a suitable amount of carbon disulfide. After drying naturally in the fume cupboard, the flask was vacuumized, sealed and kept under 160 °C for 6 h. Subsequently, the grey composite was transferred into a quartz boat within argon protection and maintained 2 h for the sake of removing the bulk sulfur on the outer surface of S/G.

**Determination of the sulfur content of S/G.** The sulfur content of the obtained S/G composite was determined by the following method. 20 mg of S/G was heated to 500 °C and remained for 1 h at argon protection with the increasing and decreasing temperature rate of 5 °C min<sup>-1</sup>. The final residual was weighed and calculated the graphene content in S/G using the mass ratio of the final residual to S/G (20 mg). Finally, 79.5% of the sulfur content was indirectly determined by graphene content.

**Materials characterization.** SEM images and EDS mapping were recorded by an FEI Nova NanoSEM 230. TEM observation was performed using a JEOL JEM-2010. Raman spectroscopy was performed on a Jobin Yvon Labram-010 micro-Raman system with a 632 nm laser excitation. XRD analyses were executed on a Siemens D500 diffractometer with a Cu K $\alpha$  source. XPS analysis was carried out on an ESCALAB 250Xi photoelectron spectrometer

### Preparation of S-G, S-G-5, S-G-10, SG-10 electrodes and Electrochemical Measurement.

S-G electrode was prepared by homogeneously mixing the slurry of S/G composite (80 wt%), super P carbon black (10 wt%), and polyvinylidene fluoride binder (PVDF, 10 wt%) dispersed in anhydrous N-methylpyrrolidone (NMP) solvent. The slurry was stirred overnight, coated uniformly on an aluminium foil using a doctor blade and then dried under vacuum at 60 °C for 12 h. The dried foil was pressed, cut into a circular disk with a diameter of 12 mm, and used as a cathode. S-G-5 and S-G-10 electrodes were prepared as preparing S-G electrode except for the extra addition of 5 wt% and 10 wt% of Mo powder. The preparation of SG-10 electrode is that S-G-10 electrode after 100 cycles was disassembled and rinsed several times in argon-filled glove box using the mixed electrolyte of DOL and DME (1:1 by volume). The electrolyte was 1.0 M lithium bis-trifluoromethanesulfonylimide (LiTFSI) in 1,3-dioxolane and 1,2-di-methoxyethane (DOL:DME, 1:1 v/v) with 0.1 M LiNO<sub>3</sub> additive. A 2032-type stainless steel coin cell was used to assemble a coin cell inside an Ar-filled glovebox (Mikvouna) using a lithium metal foil as the anode. Cyclic voltammogram (CV) was performed between 1.7 and 2.8 V at a scan rate of 0.1 mV s<sup>-1</sup>. Electrochemical impedance spectrum (EIS) was executed on a potentiostat galvanostat (Metrohm Autolab PGSTAT302N) over the frequency range from 100 kHz to 10 mHz with the amplitude of 5 mV. A LAND-CT2001A battery test system was used to perform the galvanstatic charge/discharge measurement at the current densities varied from 0.1 to 1 C (1 C equals to 1675 mAh g<sup>-1</sup>). The charge/discharge voltage range was 1.7-2.8V.

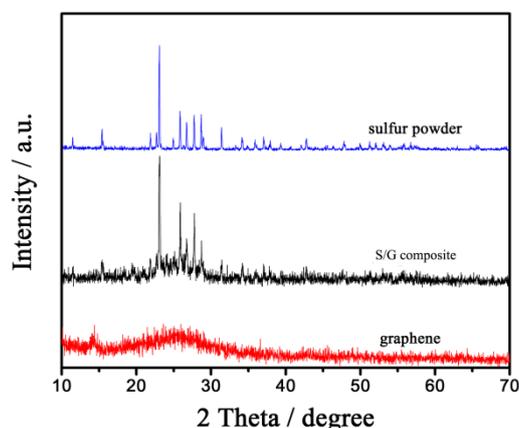


Figure S1. Sulfur powder, S/G composite and graphene

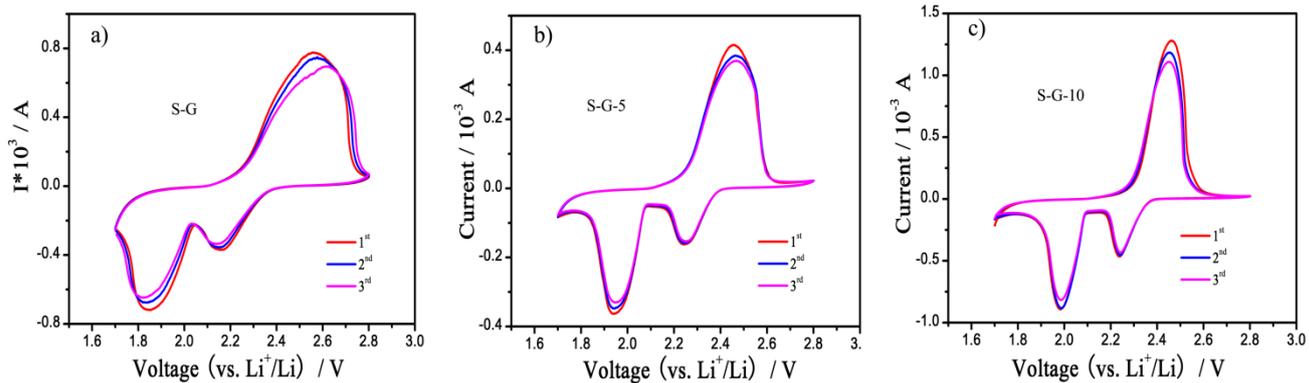


Figure S2. CV curves of S-G (a), S-G-5 (b) and S-G-10 electrodes (c) at  $0.1 \text{ mV s}^{-1}$  scanning rate.

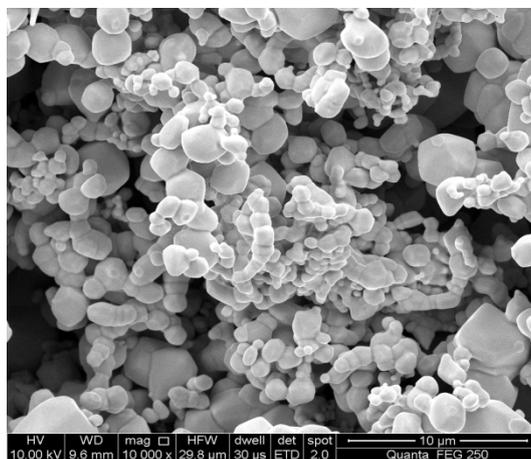


Figure S3. SEM of Mo powder

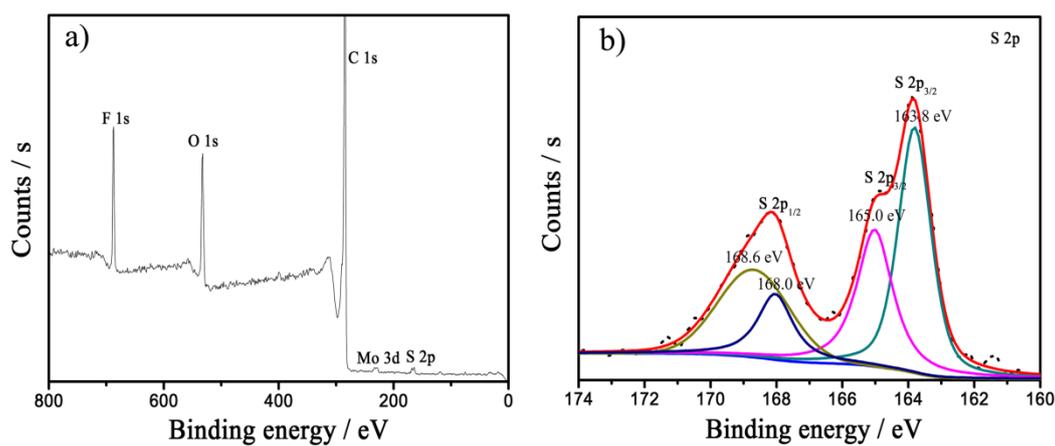


Figure S4. XPS of survey (a) and S 2p spectra of SG-10 electrode (b).

**Table S1 Discharge capacity and capacity retention of S-G, S-G-5 and S-G-10 electrodes**

| <b>Discharge capacity</b> | <b>1<sup>st</sup> cycle</b> | <b>50<sup>th</sup> cycle</b> | <b>100<sup>th</sup> cycle</b> | <b>Capacity retention</b> |
|---------------------------|-----------------------------|------------------------------|-------------------------------|---------------------------|
| <b>S-G</b>                | 973.5                       | 440.5                        | 379.4                         | 38.9%                     |
| <b>S-G-5</b>              | 1142.8                      | 583.2                        | 563.2                         | 49.2%                     |
| <b>S-G-10</b>             | 863.7                       | 687.7                        | 620                           | 71.8%                     |

1. Z. L. Ma, X. B. Huang, S. Dou, J. H. Wu, S. Y. Wang, *J. Phys. Chem. C* **2014**, *118*, 17231-17239.