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## Enhancement of electrochemical properties by polysulfide trap in graphene-coated sulfur cathode on patterned current collector

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### **1. Preparation of patterned Al current collector**

Patterned Al current collectors were prepared using a standard photolithography technique and dry etching process.<sup>1,2</sup> Al foil (Good fellow, 99.0%) was coated with photo-resist (AZ9260) at 3000 rpm for 30 s by using a spin coater. To make open-hole pattern masks, the photo-resist films were exposed to a 365 nm UV intensity of 13 mW cm<sup>-2</sup> for 25 s and developed by MIF-300 solution for 3 min. Subsequently, the basin patterned Al current collectors were fabricated by a reactive-ion etching process in Cl<sub>2</sub> gas (350 W RF power, < 70 mTorr, and etching rates of 100 Å s<sup>-1</sup>). Then, the photo-resist polymer was removed in acetone by sonication. In order to make honeycomb patterns in the basin patterned Al current collector, a making process was repeated with honeycomb pattern mask. Finally, the patterned Al current collectors that have honeycomb patterns in the basin pattern were obtained.

### **2. Preparation of sulfur cathode on patterned Al current collector**

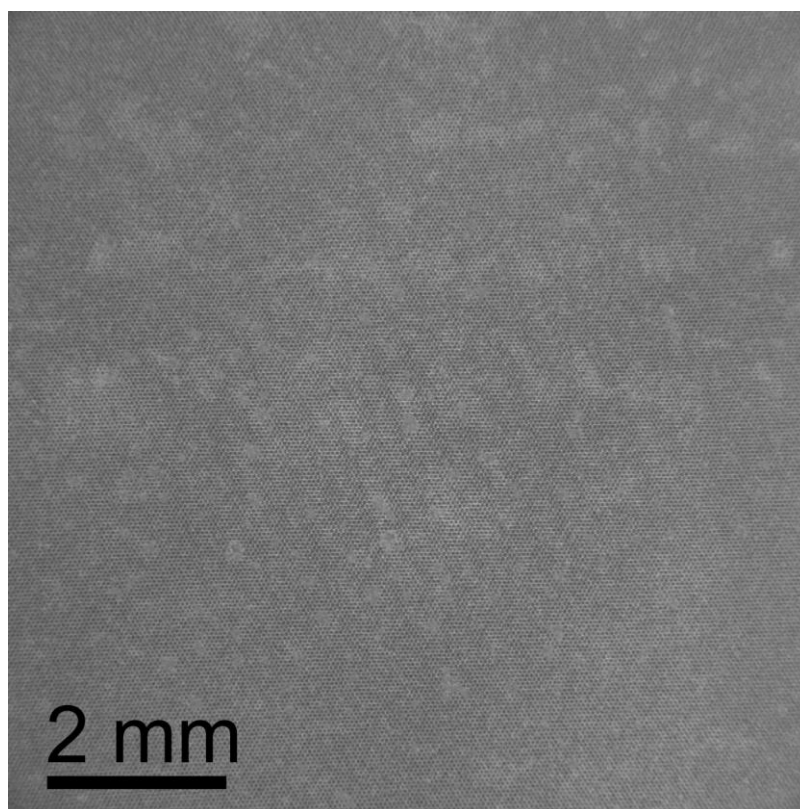
A slurry composed of sulfur, super P, and polyvinylidene fluoride (5:3:2 by weight) in n-methyl-2-pyrrolidone was coated on the prepared patterned Al foil by using the doctor-blade method. For comparison, an identical slurry was coated on a normal Al foil. The prepared sulfur cathodes were dried in an oven at 50 °C for 12 h.

### ***3. Chemical vapor deposition of graphene and covering of graphene on the sulfur cathode***

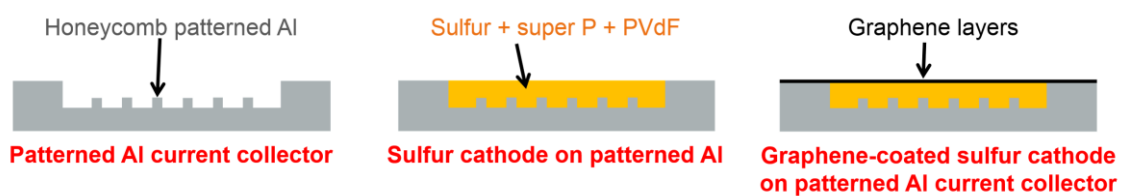
Graphene was deposited on a copper foil through chemical vapor deposition. The copper foil was heated at 1000 °C in a quartz tube for 1 h under a H<sub>2</sub> flow of 5 sccm, following which 35-sccm CH<sub>4</sub> was injected for 30 min to grow graphene. Finally, the furnace was rapidly cooled down to room temperature under H<sub>2</sub>. The top of the graphene layer on the copper foil was coated with poly(methyl methacrylate), and then, the copper foil was removed using ammonium persulfate. The floating graphene layers were stacked to form a triple-layered graphene film. These graphene layers were finally transferred onto the prepared sulfur cathode with the patterned Al foil.

### ***4. Structural and electrochemical characterization***

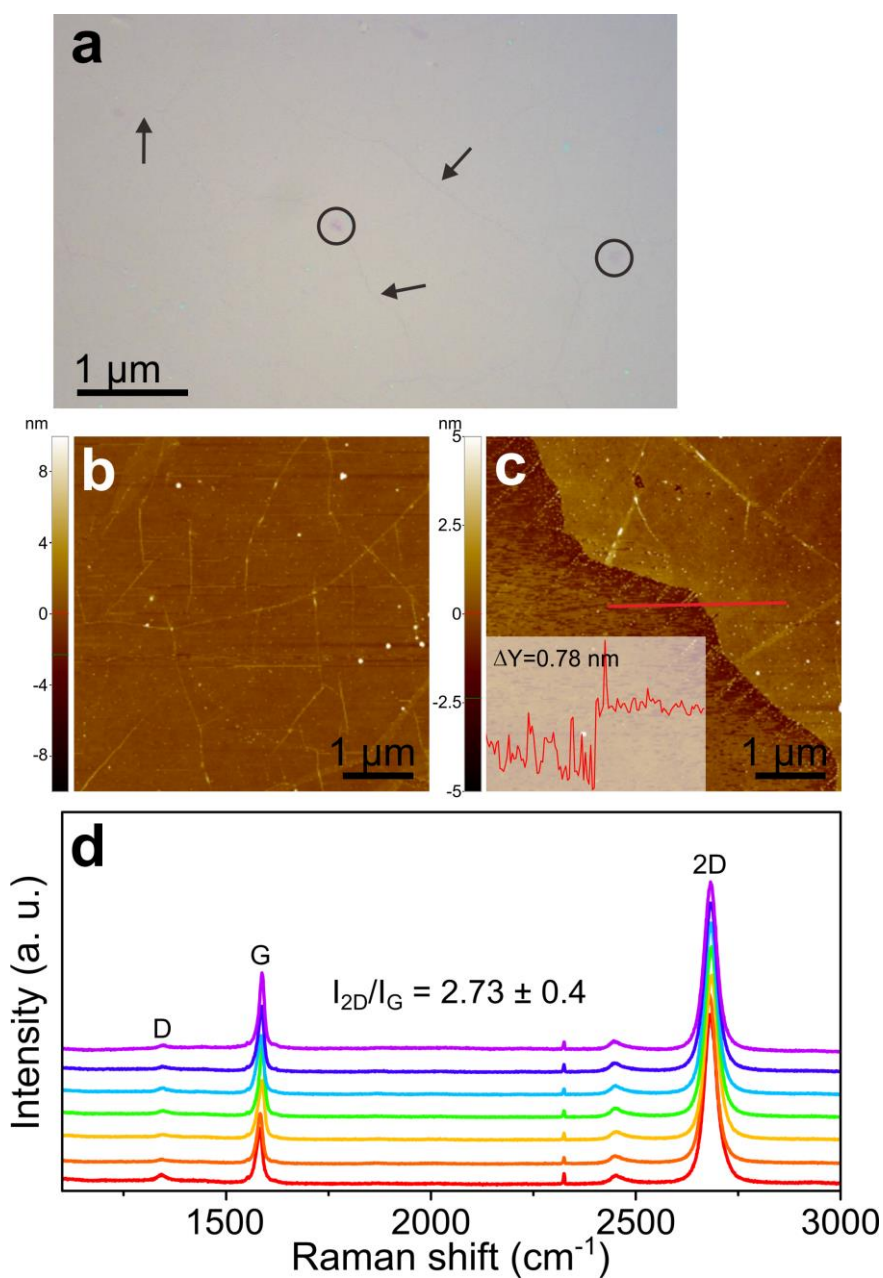
The morphology and energy-dispersive X-ray spectroscopy mappings of the sample were acquired using a field-emission scanning electron microscope (SUPRA 55VP, Carl Zeiss). Electrochemical tests were conducted using 2032-type coin cells with lithium foil as counter and reference electrodes, and 1 M lithium bis(trifluoromethanesulfone)imide (LiTFSI) in a mixture of 1,2-dimethoxyethane (DME) and 1,3-dioxolane (DOL) at 1:1 volume ratio with 0.1 M LiNO<sub>3</sub> as an electrolyte. The cells were galvanostatically charged and discharged in the potential range between 3.0 and 1.5 V (vs. Li<sup>+</sup>/Li) with a WBCS3000 cycler (WonATech, Korea) at 25 °C.



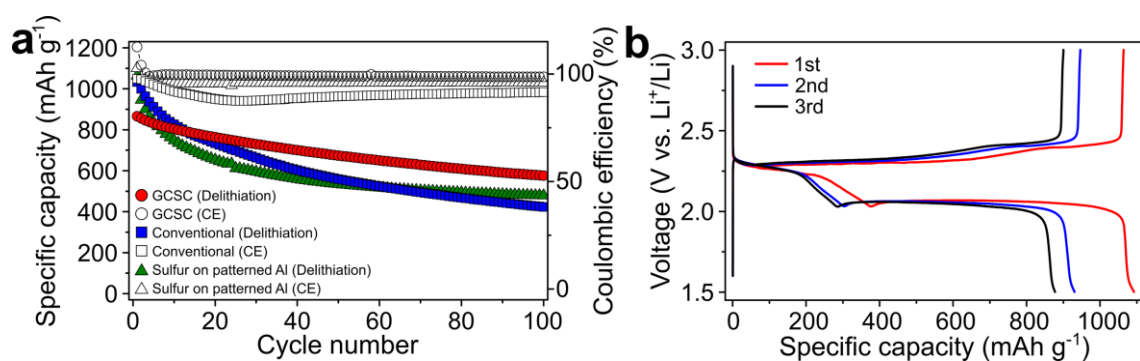
**Fig. S1.** Top-view SEM image of a honeycomb-shape-patterned Al current collector over a large area.



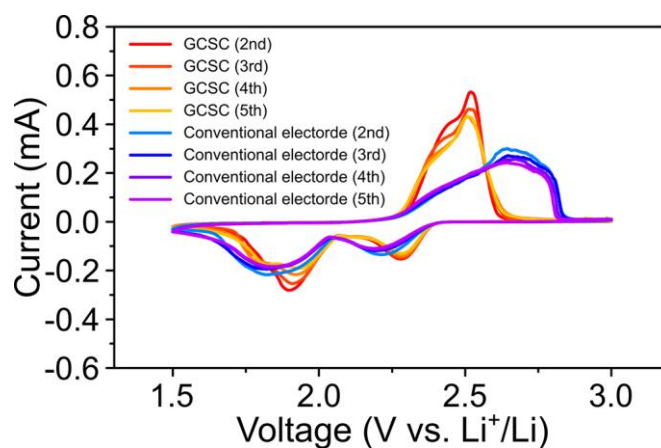
**Scheme S1** Cross-sectional view of patterned Al current collector, sulfur cathode on patterned Al and graphene-coated sulfur cathode on patterned Al current collector.



**Fig. S2** (a) Optical microscopy image of a graphene film transferred onto Si/SiO<sub>2</sub>. The arrows indicate graphene wrinkles, and circles present bilayer graphene. (b, c) AFM images of single-layer graphene transferred onto Si/SiO<sub>2</sub>. Inset of (c) shows the height profile along the red line. The AFM images were measured by a noncontact mode (Park System, XE-100). (d) Raman spectra of single-layer graphene transferred onto Si/SiO<sub>2</sub> substrates with a 514.5 nm excitation laser at different positions. The large 2D/G peak ratio and the negligible D peak intensities indicate that the graphene film is a high-quality single layer. Raman measurements were conducted using Invia Renishaw Raman spectrometer (RM 1000) with 514.5 nm laser.



**Fig. S3** (a) Cycle performance and Coulombic efficiency and (b) voltage profiles of sulfur cathode on patterned Al current collector without graphene coating at a rate of 0.2 C.



**Fig. S4** Cycle voltammograms of GCSC and conventional electrode at a scanning rate of 0.2 mV s<sup>-1</sup>.

### References for Electronic Supplementary Information

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2. S. Choi, T. -H. Kim, J. -In. Lee, J. Kim, H. -K. Song and S. Park, *ChemSusChem*, 2014, **7**, 3483.