Electronic Supplementary Information for

Hierarchically micro/mesoporous activated graphene with large surface area for high sulfur loading in Li-S batteries

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Experimental Section

Synthesis of a-MEGO/S. a-MEGO powder was synthesized by KOH chemical activation of microwave exfoliated graphite oxide as described in our previous report.[1-3] Sulfur (Aldrich, with a purity of >99.995%) and a-MEGO were thoroughly mixed and grinded in a mortar with a weight ratio of 80:20. The mixture was then sealed in a glass container in vacuum and annealed in a tube furnace at 400 ºC for 5 h to yield a-MEGO/S composite.

Structural Characterization. A field emission scanning electron microscope (JEOL 6701F) operated at 10 kV were utilized to visualize the morphologies and sizes of the a-MEGO and a-MEGO/S composite. TEM, HRTEM, STEM, elemental compositions and mapping of the samples were obtained by JEOL 2011 coupled with an energy diffraction energy-dispersive X-ray spectroscopy (Phoenix) system. XRD patterns

were collected on a Rigaku D/max-2500 with Cu Kα radiation (λ = 1.54056Å) operated at 40 kV and 200 mA. Raman spectra were performed on a DXR from Thermo Scientific using a laser wavelength of 533 nm. TGA was conducted on a TG/DTA6300 instrument with a heating rate of 10 ºC /min under N_2 flow (50 mL min^{-1}). To investigate the BET surface area and the pore size, the nitrogen adsorption/desorption isotherms were obtained at 77.3 K with an Autosorb-1 specific surface area analyzer from Quantachrome. The theoretical sulfur content that a-MEGO can theoretically contain is calculated according to equations (1) and (2):

$$M_S = V \times M_G \times \rho \quad (1)$$

$$S\ wt\% = M_S/(M_S+M_G) \quad (2)$$

Where M_G is the mass of a-MEGO; V is the pore volume of a-MEGO per gram; ρ is the density of sulfur; M_S is the mass of sulfur that a-MEGO could theoretically contain, S wt% is the weight percentage of sulfur in a-MEGO/S composite. According to the above equations, the sulfur content is calculated to be 81%.

Electrochemical Measurements. To prepare the working electrode, a-MEGO/S composite, carbon black and poly(vinyl difluoride) were thoroughly mixed with a weight ration of 8:1:1. The mixed slurry was coated onto the Al foil (Goodfellow) and dried under 60 °C for 12 h. Li-S batteries were assembled in CR2032 coin cells with PP/PE/PP separator from Celgard and Li foil as the anode in an Ar glove box with oxygen and water content less than 1 ppm. 1 M lithium bis(trifluoromethane) sulfonamide (LITFSI) dissolved in 1,3-dioxolane/dimethoxymethane (DOL/DME, 1:1 by volume) (Zhangjiagang Guotai Huarong New Chemical Materials Co.,Ltd.) with

0.1 M $LiNO_3$ additive were employed as the electrolyte. Galvanostatic discharge/charge cycling of the assembled cells were performed on an Arbin BT2000 system between the voltage range of 1.8−2.7 V (vs Li^+/Li). CV was conducted on an Autolab PG302N workstation with a scan rate of 0.1 mV s^{-1}. All electrochemical measurements were carried out at 25 °C. The C-rate used was based on the theoretical capacity of S (1675 mA h g^{-1}).

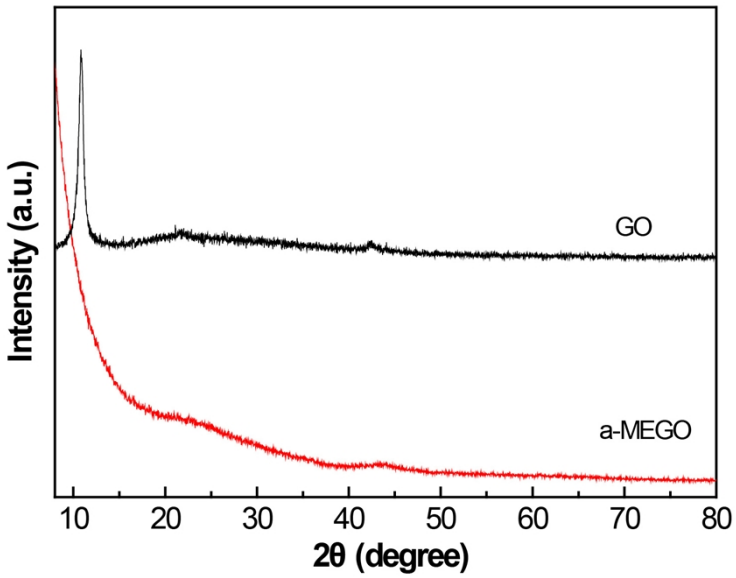

Fig. S1 XRD patterns of GO and a-MEGO.

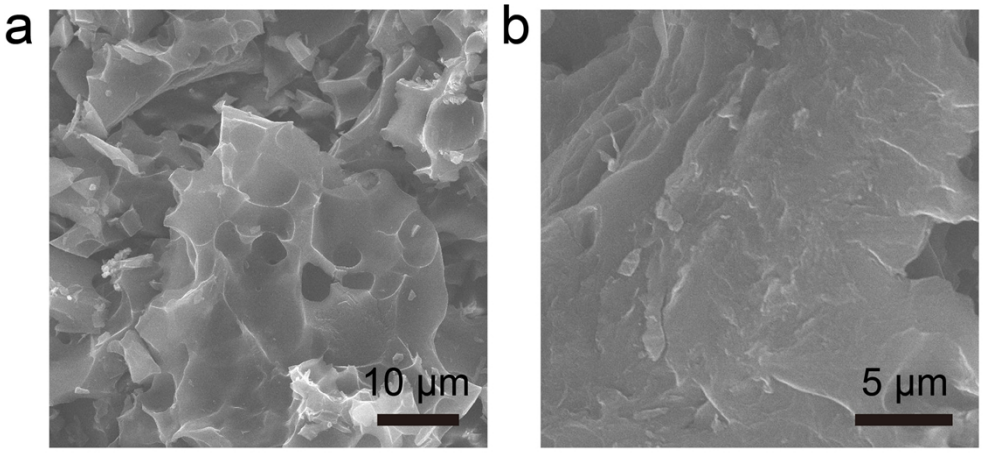

Fig. S2 (a) and (b) SEM images of MEGO.

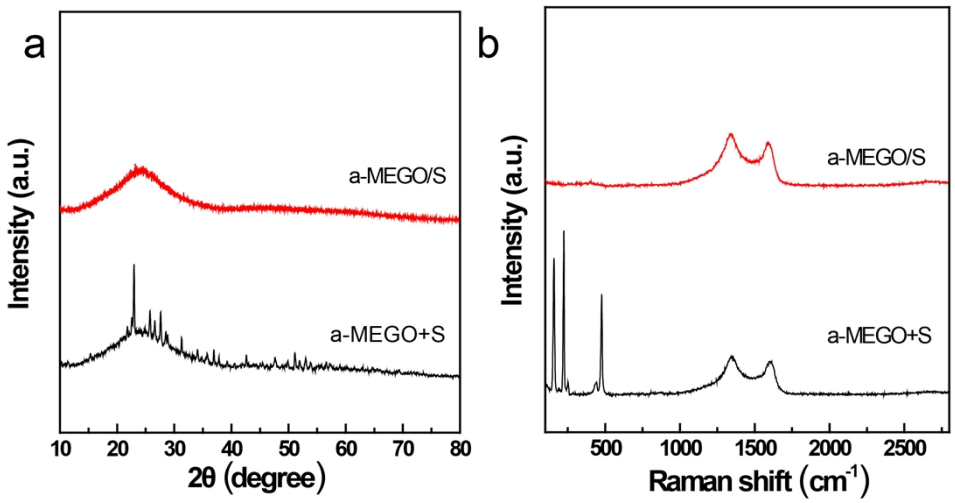

Fig. S3 (a) XRD patterns and (b) Raman spectra of a-MEGO+S mixture and a-MEGO/S composite.

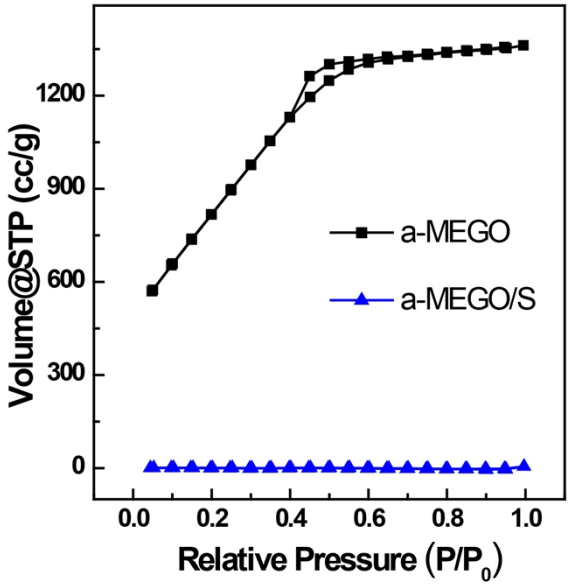

Fig. S4 N_2 adsorption/desorption isotherms of a-MEGO and a-MEGO/S composite.

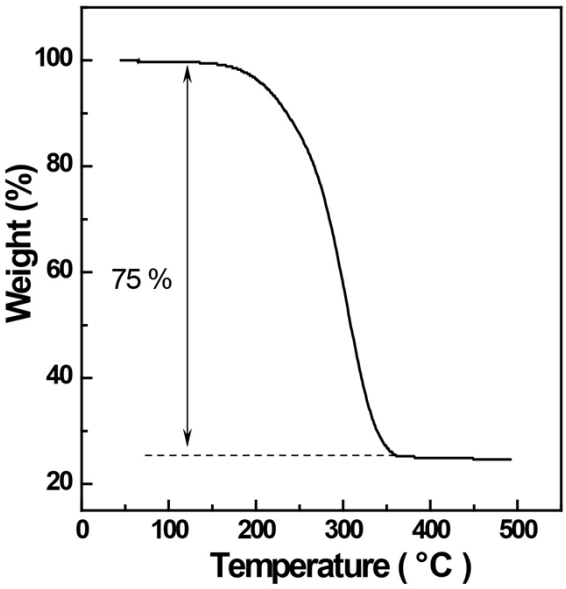

Fig. S5 TGA curve of a-MEGO/S composite. The weight loss of 75% is corresponding to sulfur content.

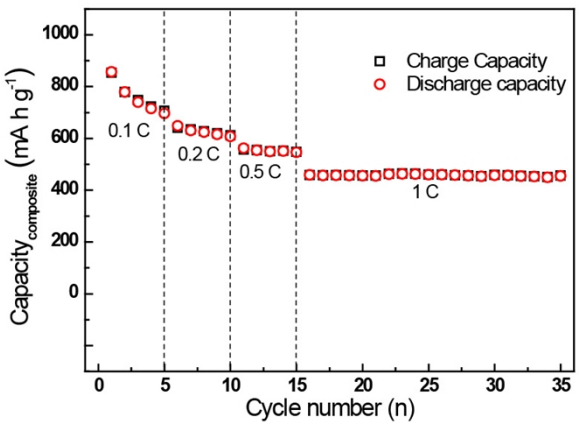

Fig. S6 Rate capabilities of a-MEGO/S composite under different current densities.

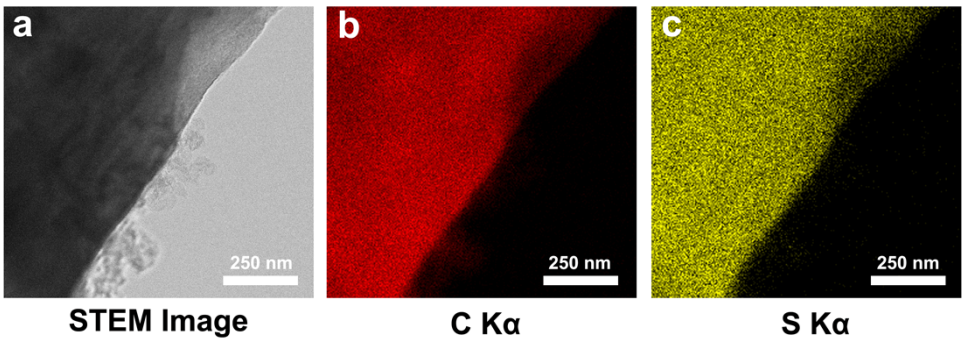

Fig. S7 (a) Bright field STEM image and the corresponding (b) C and (c) S EDX elemental mappings of a-MEGO/S composite after 200 cycles at 1C.

References

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