Supplemental Information for:

Graphene-Enveloped Sulfur in a One Pot Reaction: a Cathode with Good Coulombic Efficiency and High Practical Sulfur Content

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Preparation of GSC - In a typical procedure graphene oxide (100 mg) prepared by the modified Hummer's method was sonicated in 20 mL of 5mM NaOH for 1 hr. In a separate container, sulfur (99.5% Alfa Aesar, 500 mg) was mixed with sodium sulfide nonahydrate (99.99+% Aldrich, 1g) in 100 mL 10% NaOH solution at 60°C to form a deep orange solution. These solutions were then mixed together and stirred for 4 hrs, and a stoichiometric amount of 5% HCl solution was added to lower the pH to 2. The GSC composite was obtained after filtration and drying.

Electrochemical Analysis – Positive electrodes were prepared from GSC (90 wt%), and polyvinylidene fluoride binder (PVdF) binder (10 wt%). The cathode material contained 78 wt% sulfur active mass. The cathode material was well dispersed in cyclopentanone by agitation and sonication, and was slurry-cast onto a carbon coated aluminum current collector (Exopack Advanced Coatings). The positive electrode was tested in 2325 coin cells using an electrolyte comprised of 1M LiTFSI in a mixed solvent of 1,3-dioxolane (DOL) and tetraethyleneglycol dimethyl ether (TEGDME) with a volume ratio of 1:1. Lithium metal foil was used as the counter electrode. A typical cell contained a cathode loading of approximately 1.2 mg of sulfur.

The batteries were cycled between 1.5 and 3 V using an Arbin battery cycler at room temperature. The discharge/charge rate C/5 (334 mA h g^{-1}) corresponds to a current density of 0.4 mA cm^{-2}.

Measurement of Sulfur Content. Chemical analysis of sulphur in the graphene-sulfur composite was carried out in duplicate at the Galbraith Labs (Tennessee), giving a mass fraction of 87 ± 1%. Thermogravimetric analysis of the same composite was performed on a TA Instruments SDT Q600 in a nitrogen atmosphere at a heating rate of 10°C/min from room temperature to 500°C, and showed the same sulphur content (see **Fig. S1**, below**).**

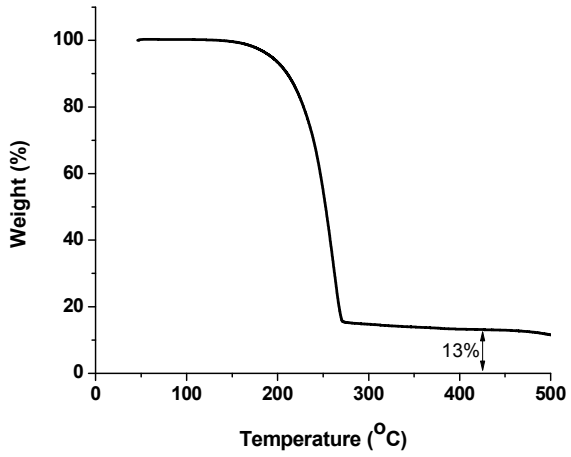

Figure S1. TGA curve (N2 atmosphere) of the graphene-sulfur composite prepared as described in the experimental, showing weight loss of 87% corresponding to the loss of sulphur.

Impedance Spectroscopy of the full cell. The full cell impedance was performed at open-circuit voltage using a BioLogic VMP3 instrument. The AC impedance was measured over a frequency range of 200 kHz to 100 mHz. Two cells (containing the same electrolyte as was used in the cycling studies, with a lithium anode) were studied: one contained the GSC composite as the

cathode and the other contained graphene oxide synthesized by the modified Hummer's method as the cathode. The circuit used for the AC impedance fitting is shown below: R_b is the bulk resistance in the cell; R_s and C_s are the resistance and capacitance of the interface layer formed on the surface of the electrodes; R_{ct} and C_{dl} are the faradic charge transfer resistance and its relative double-layer capacitance; W is the Warburg impedance.

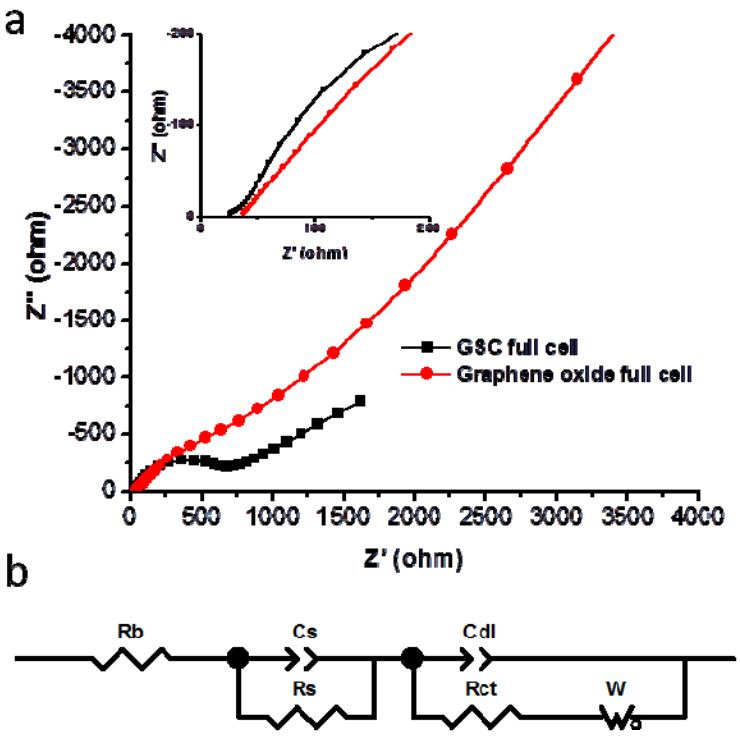

Figure S2. (a) The AC impedance plot of the GSC composite full cell (black) and the graphene oxide full cell (red), inset: zoom-in of high frequency region of impedance plot; (b) The equivalent electrical circuit of (a).