

**Electronic supplementary information for**

**Self-assembled layer-by-layer partially reduced graphene oxide-sulfur composites  
as lithium-sulfur battery cathode**

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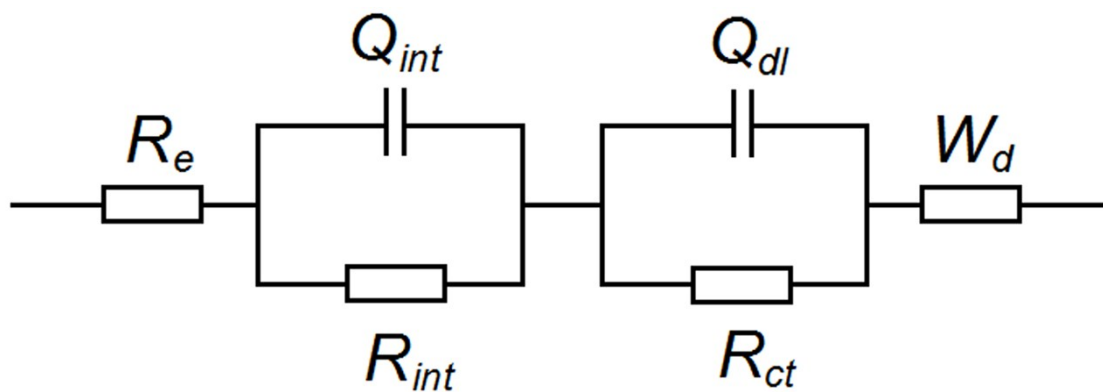


Figure S1. Equivalent circuit used in the simulation of impedance spectra. The fitting procedure, weighing modulus and circuit description codes are explained elsewhere.<sup>1</sup>

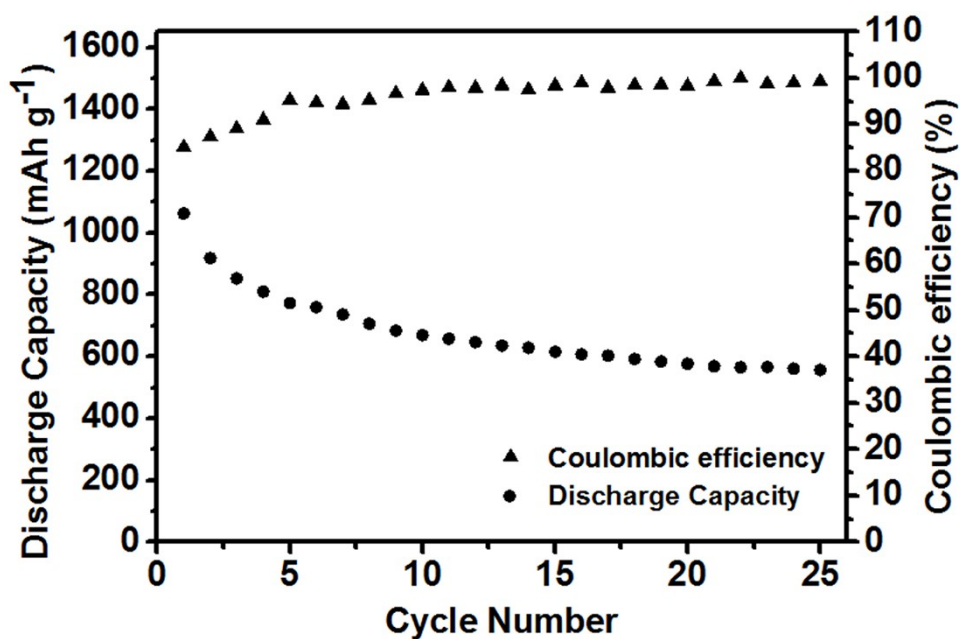


Figure S2. Cycling performance of prGO-240/S at 0.1 C.

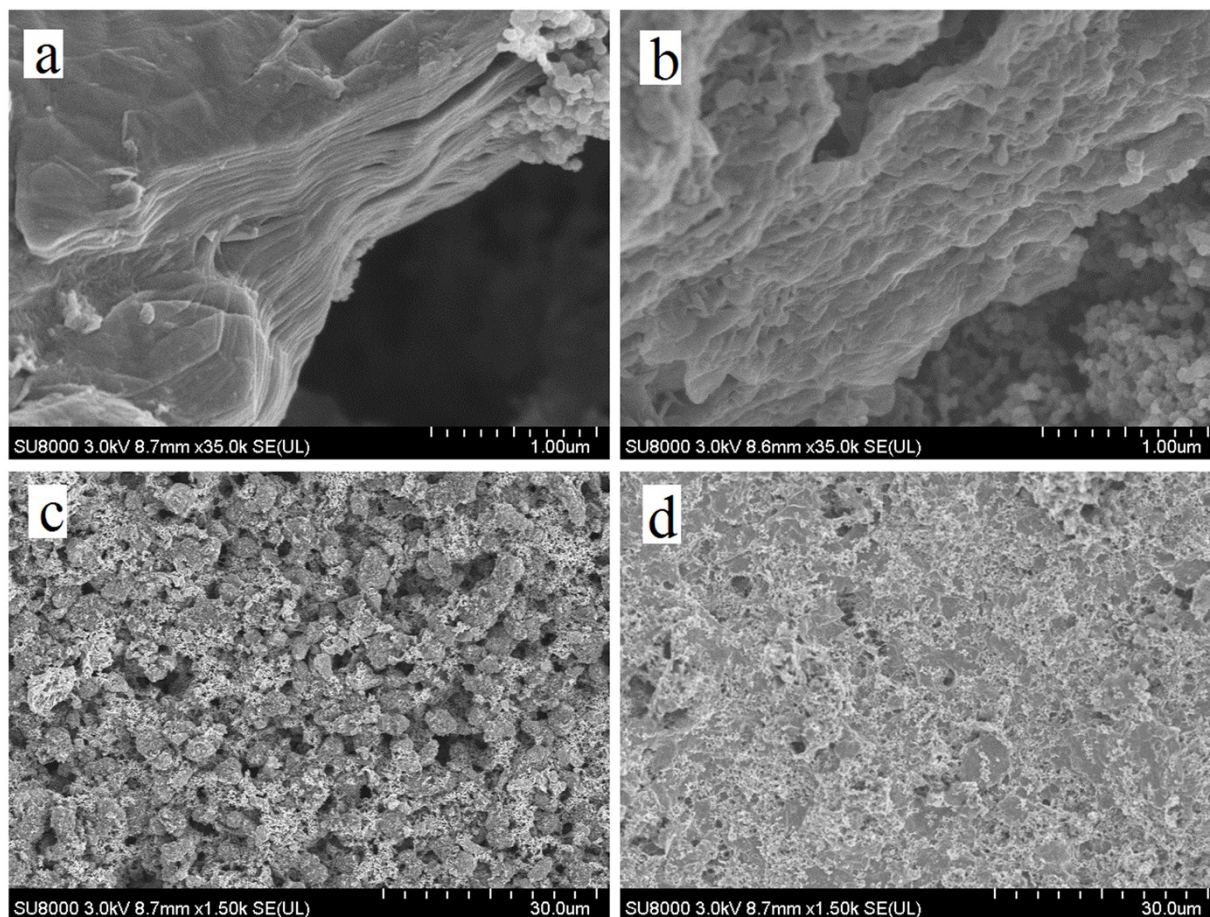


Figure S3. SEM characterization of prGO-120/S composite cathode. (a, c) Before cycling. (b, d) After 100 cycles. Scale bar,  $1\mu m$ (a, b),  $30\mu m$ (c, d)

Table S1. C/O ratios of prGO/S composites by XPS analysis.

Samples	pristine GO/S	prGO-30/S	prGO-60/S	prGO-120/S
C/O ratio	3.66	8.51	9.45	10.35

Graphene oxide, which has C/O ratio of 2/1, is none conductive. It usually has a conductivity of less than a micro S/m. To reduce graphene oxide to C/O ratio above 6 is required to make graphene oxide conductive by partially restoring the aromatic graphene structure. The conductivity of graphene oxide really depends on the degree of oxidation. In this article, partially reduced prGO-120/S provides higher conductivity, thus its electrochemical properties is better as shown in Fig. 8.

Table S2. Coulombic efficiency of prGO/S composites.

Coulombic efficiency	prGO-30/S	prGO-60/S	prGO-120/S	prGO-120/S /prGO	prGO-120/S/prGO with prGO interlayer
The 1st cycle	83.4%	87.4%	89.25%	93.6%	98.6%
The 2nd cycle	82.8%	85.5%	88.6%	89.6%	99.3%
The 10th cycle	88.7%	90.5%	92.6%	98.0%	98.8%
The 50th cycle	89.4%	91.3%	94.6%	98.7%	99.3%

During the first ten cycles, the instable coulombic efficiency corresponds to the surface side-reactions of the prGO/S composites. After 50 cycles, the coulombic efficiency of prGO-30/S and prGO-60/S composites decrease due to the relative irregular layer-by-layer structure compared with prGO-120/S composite. The coulombic efficiency was improved by prGO coating; however, it was still unstable in the first ten cycles. After adopting the prGO interlayer, the coulombic efficiency is around 99% from the first cycle indicating that the soluble intermediate polysulfides shuttle was prevented from the first charge-discharge process.

## Reference

1. Chakraborty Banerjee, P.; Singh Raman, R. K., *Electrochim. Acta*. 2011, 56, 3790-3798.