Graphene and reduced graphene oxide for high energy density Li-S battery

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Abstract

Lithium-sulfur (Li-S) battery delivers a significantly higher theoretical energy density (2567 Wh/kg) compared to state-of-the-art lithium-ion batteries (LIBs). However, many problems, e.g. low conductivity of sulfur, volume change and diffusion of soluble polysulfides, need to be addressed before the Li-S batteries can find practical use. We have systematically studied the use of graphene and reduced graphene oxide (rGO) to solve the above problems [1-4].

Firstly, we have prepared a rGO-sulfur (G-S) hybrid materials with sulfur nanocrystals anchored on interconnected fibrous graphene by a facile one-pot strategy using a sulfur/carbon disulfide/alcohol mixed solution (Figure 1). Such G-S hybrids exhibit a highly porous network structure constructed by fibrous graphene, and can be cut and pressed into pellets to be directly used as Li-S battery cathodes without using metal current collectors, binders, and conductive additives. The porous network and sulfur nanocrystals enable rapid ion transport and short Li^{+} diffusion distance, the interconnected fibrous graphene provides highly conductive electron transport pathways, and the oxygen-containing (mainly hydroxyl/epoxide) groups show strong binding with polysulfides, preventing their dissolution into the electrolyte based on first-principles calculations. As a result, the G-S hybrids show a high capacity, an excellent high-rate performance, and a long life over 100 cycles.

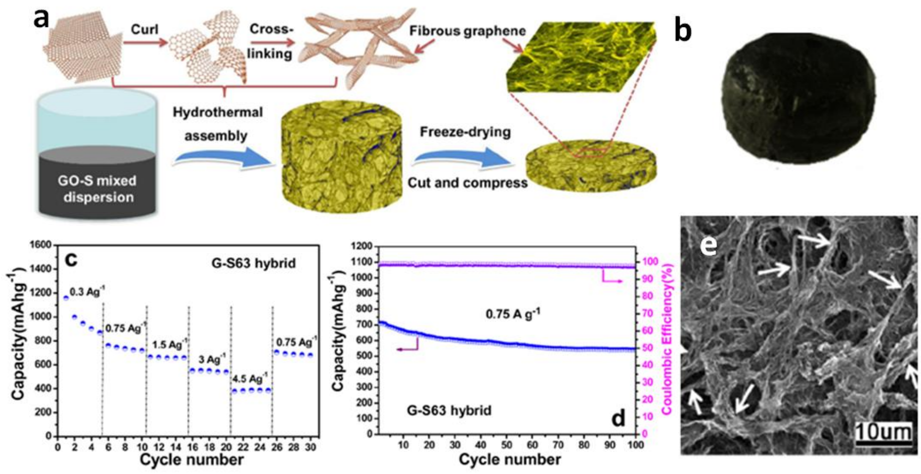

Figure 1. The preparation process (a), morphology (b, e) and performance (c, d) of the rGO-S hybrid electrode

Secondly, we have proposed a very simple but effective strategy for obtaining high-performance Li–S batteries through the use of a unique sulfur electrode that consists of two graphene membranes as current collector (GCC) and separator (G-separator) (Figure 2). In comparison to an Al-foil current collector and commercial separator, the GCC and G-separator efficiently decrease the contact impedance of the current collector, the active material, and the electrolyte. The electrode with two graphene membranes can provide rapid ion- and electron-transport paths, accommodate sulfur volumetric expansion and store and reuse migrating polysulfides to alleviate the shuttling effect. The light weight of the GCC can also contribute to a higher energy density of Li–S batteries. In addition, the sulfur cathode was directly mixed with carbon black without confining sulfur in special carbonaceous matrixes or using polymer coatings, which simplifies the electrode preparation process. The fabrication of large-area GCC and G-separators was also demonstrated, which indicates that this sulfur electrode design can be scaled for industrial manufacture.

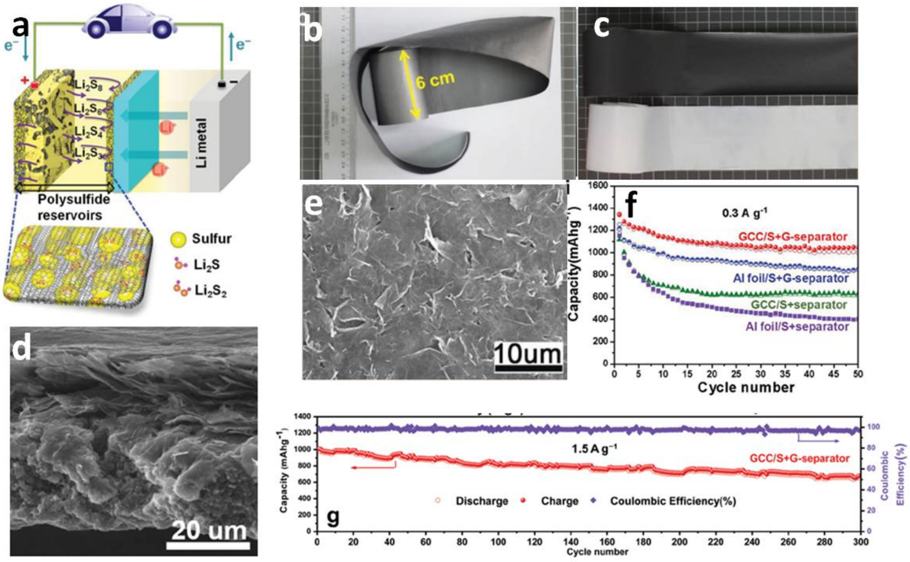

Figure 2. Schematic of a Li-S battery with GCC and G-separator (a), morphology of GCC (b,d) and G-separator (c,e), and performance of Li-S battery (f, g).

Thirdly, we have used the density functional theory calculations to investigate the interaction between graphene/rGO (graphene with residual groups) and polysulfides, which is the key to understand the role of rGO and graphene in the Li-S batteries. It was found that the interaction between polysulfides/-ions and rGO is much stronger than graphene (Figure 3). The hydroxyl groups on the graphene surface can induce an asymmetrical charge distribution on the two end sulfur atoms of a S_3 cluster, resulting in larger polarization and consequently stronger electrostatic interaction between a S_3 cluster and the HO-graphene.

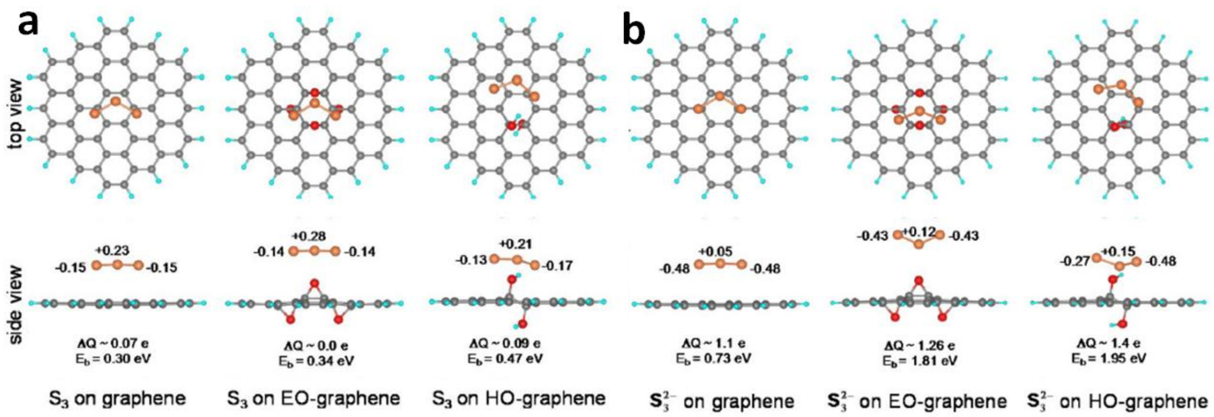

Figure 3. DFT calculation results on a neutral S_3 cluster (a) and S_3^{2-} polysulfides (b) on graphene, EO-graphene and HO-graphene surface. The calculated charge population for each sulfur atom, the binding energy (E_b), and corresponding charge transfer (ΔQ) are shown below the top and side views.

References

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