

THE REGULATING ROLE OF CARBON MATERIALS IN RECHARGEABLE LITHIUM-SULFUR BATTERIES

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Introduction

The ever-increasing demands for batteries with high energy densities to power the portable electronics with increased power consumption and to advance vehicle electrification and grid energy storage have propelled lithium battery technology to a position of tremendous importance. Lithium-sulfur (Li-S) batteries are currently being explored intensely due to their high theoretical energy density and low cost.¹ However, their practical reliability has been greatly hindered by the insulating nature of sulfur, shuttling issue of polysulfides, as well as limited sulfur content and areal sulfur loadings in sulfur cathodes.² Carbon materials, known with many appealing properties, are investigated intensely for improving the performance of Li-S batteries. It is worth recognizing that carbon materials are not active lithium storage material but are more like a regulator: they do not electrochemically react with lithium ions and electrons, but serve to regulate the sulfur redox conversion reactions,³ contributing to substantially improved electrochemical reactions. Here, we explore the regulating role of carbon nanotubes, graphene and carbon fibers in Li-S batteries from the perspectives of fundamental electrochemistry, materials design and electrode architecture. Exceptional electrochemical performance has been demonstrated by high-sulfur loaded electrodes with high sulfur utilization and good cycling stability.

Results and Discussion

Based on the structural versatility of graphene, we developed an all-graphene cathode configuration, with highly porous graphene using as the sulfur host, highly conductive graphene as the current collector, and partially oxygenated graphene as a polysulfide-adsorption layer. This unique cathode structural design enables both high initial gravimetric specific capacity and areal specific capacity, together with excellent cycling stability (**Figure 1a**). Moreover, we theoretically indicated that the electronic conduction efficiency of the sulfur host nanomaterial plays a crucial role in determining the sulfur content, and a highly efficient single-wall carbon nanotube conductive network was constructed, allowing an unprecedentedly high sulfur content up to 95 wt%. High areal capacities and good cycling stabilities are demonstrated with high areal sulfur loadings (**Figure 1b**). We further explored the use of a 3D hollow carbon fiber current collector with interlinked electron and ion channels for achieving ultra-high sulfur loadings, resulting in a high areal capacity of 23.32 mAh cm⁻², which is valuable for the fundamental research and possible commercialization of Li-S batteries (**Figure 1c**).

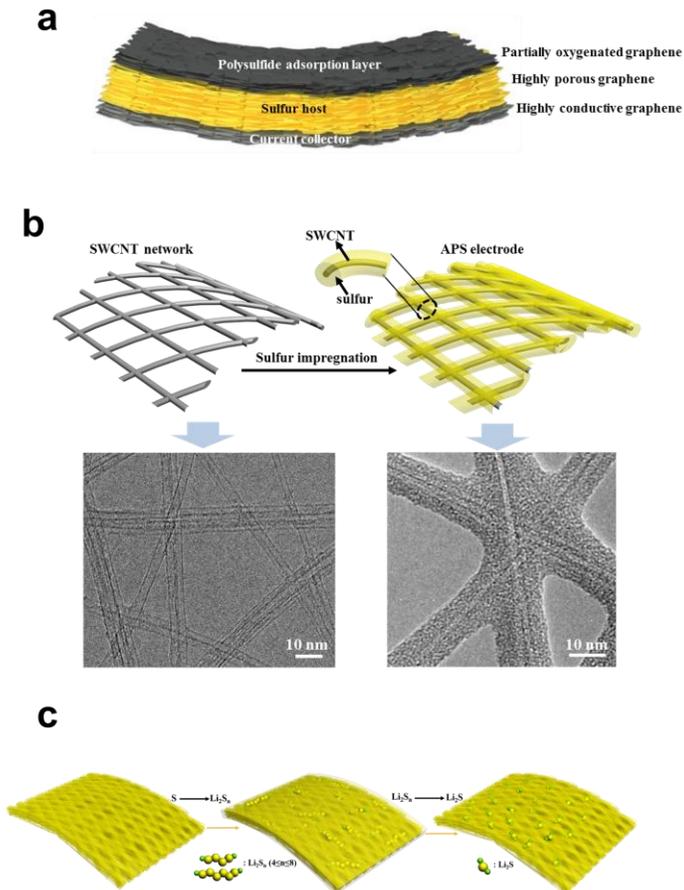


Figure 1. (a) Schematic of the all-graphene cathode structural design. (b) Illustration of the SWCNT network and the almost pure sulfur electrode (95 wt%). (c) Illustration of sulfur redox reactions within 3D interconnected hollow carbon fiber foam

Conclusions

The demand for high-energy batteries has been triggered by the rapid development of Li-S batteries. Carbon materials, mostly acting as regulators, have shown great effectiveness in building better Li-S batteries due to their excellent structural, electrical, thermal and mechanical properties. Here, the regulating role of carbon materials has been comprehensively demonstrated from many perspectives. Intriguing performance in terms of high energy density and good cycling stabilities has been achieved, showing the promise of revolutionizing Li-S battery technology for practical applications.

References

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