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Review

Recent advances in lithium–sulfur batteries

Lin Chen [a, b], Leon L. Shaw [a, b, *]

[a] Wanger Institute for Sustainable Energy Research, Illinois Institute of Technology, Illinois, USA
[b] Department of Mechanical, Materials and Aerospace Engineering, Illinois Institute of Technology, Illinois, USA

HIGHLIGHTS

- Li–S batteries have great potential as the next generation high capacity batteries.
- Nanostructured sulfur electrodes are essential to realize this potential.
- Anodes, electrolytes, additives, binders and separators also play critical roles.
- Cell configuration with novel components can result in breakthroughs.

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ABSTRACT

Lithium–sulfur (Li–S) batteries have attracted much attention lately because they have very high theoretical specific energy (2500 Wh kg^{-1}), five times higher than that of the commercial $LiCoO_2$/graphite batteries. As a result, they are strong contenders for next-generation energy storage in the areas of portable electronics, electric vehicles, and storage systems for renewable energy such as wind power and solar energy. However, poor cycling life and low capacity retention are main factors limiting their commercialization. To date, a large number of electrode and electrolyte materials to address these challenges have been investigated. In this review, we present the latest fundamental studies and technological development of various nanostructured cathode materials for Li–S batteries, including their preparation approaches, structure, morphology and battery performance. Furthermore, the development of other significant components of Li–S batteries including anodes, electrolytes, additives, binders and separators are also highlighted. Not only does the intention of our review article comprise the summary of recent advances in Li–S cells, but also we cover some of our proposals for engineering of Li–S cell configurations. These systematic discussion and proposed directions can enlighten ideas and offer avenues in the rational design of durable and high performance Li–S batteries in the near future.

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1. Introduction

Lithium-ion batteries (LIBs), one of common rechargeable power sources, have dominated in portable device markets for more than 20 years since their initial launch in 1990s. However, LIBs which nearly reach their theoretical capacity [1,2] and leave little room for further exploration cannot meet the current need for applications in systems such as electric vehicles that require large capacity and long battery cycle life [3]. With increasing demand and immense market potential [4], rechargeable batteries with superior energy density and low cost are thus always urgently searched in academia and industrial world [5–10].

Metallic lithium has a very high electronegativity while possessing the lowest density among all metals, leading to its highest specific capacity (3861 mAh g^{-1}) and thus has been considered to be the best candidate for rechargeable battery anodes [11]. Element sulfur has a theoretical capacity of 1673 mAh g^{-1} [12]. Thus, Li–S batteries can reach unparalleled gravimetric and volumetric energy densities of 2500 Wh kg^{-1} and 2800 Wh L^{-1}, respectively, assuming a complete reaction to Li_2S [13]. Also, they are cheaper compared to conventional LIBs owing to highly abundant sulfur storage in the earth. Further, the sulfur cathode can operate at a safer voltage range (1.5–2.5 V vs. Li/Li^+). Another advantage of sulfur is its non-toxicity. Undoubtedly, all of these advantages make Li–S cells an excellent alternative for energy storage and could play an important role in diversifying energy sources as well as utilizing

* Corresponding author. Wanger Institute for Sustainable Energy Research, Illinois Institute of Technology, Illinois, USA.
E-mail address: lshaw2@iit.edu (L.L. Shaw).

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renewable energy, thereby alleviating global warming and reducing the use of fossil fuels.

However, commercialization of Li–S batteries has been gravely hindered by several issues, such as the insulating nature of sulfur element (~5×10^{-30} S cm^{-1} at 25 °C) [14] and polysulfide dissolution causing active sulfur loss and rapid capacity fading. In order to address these challenges, many endeavors have been made to fabricate materials into nano-dimensions and nanostructures, which are considered as effective solutions to solve the problems owing to their more resistance to structural degradation by virtue of materials' nanoscale sizes [15]. Also, nanostructured materials with incorporation of carbon nanotubes and nanofibers can enhance the rate capability because the path for ionic and electronic conductivity is shortened. On basis of these thoughts, nanostructures can be very significant in circumventing some challenges of the Li–S system. Although nanostructured materials are not easy to prepare and many challenges still need to be overcome, researchers who are interested in Li–S systems have already made significant advancements. In spite of the tremendous progress, however, there are only several reports on lithium sulfur batteries with good capacity performance up to 1000 cycles [12,16], which is required by the US Department of Energy for electric vehicle grade battery systems [17]. Given the great potential of the Li–S system, a comprehensive review of the recent advances in the Li–S system is warranted.

Several reviews of Li–S batteries have been published recently [13,18–24]. However, most of these reviews emphasize the recent progress in cathode research, whereas the advancements and challenges associated with anodes, electrolytes, additives, binders, and separators are little mentioned [13,18–23] or partially discussed [24]. In this review, we will cover all of these topics to provide a comprehensive overview of the challenges and advancements in various components of Li–S batteries. We will first discuss the working process of Li–S rechargeable batteries. We will then present the latest fundamental studies and technological development of various nanostructured cathode materials for Li–S batteries, including their preparation approaches, structure, morphology and battery performance. The development of other significant components of Li–S batteries such as anodes, electrolytes, additives, binders and separators will be subsequently summarized. Finally, based on the experiment exploration conducted to date, we will discuss the significant lessons and propose future directions that can be focused on. Not only does the intention of our review article include the comprehensive summary of recent advances in Li–S cells, but also we cover our ideas for some designs of Li–S cell configurations. We hope that the systematic review and proposed avenues can enlighten discussion and provide directions for developing durable and high performance Li–S batteries in the near future.

2. Lithium–sulfur battery operation and challenges

2.1. Operation and SEI formation

A lithium–sulfur battery encompasses three major components: a cathode, an anode and a non-aqueous electrolyte. The anode and cathode are typically separated by a porous separator soaked with the non-aqueous electrolyte, allowing ions but preventing electrons to pass through to avoid short circuit. Fig. 1 shows a schematic representation of the working process of a Li–S battery. The anode (negative electrode) and cathode (positive electrode) are connected to an external circuit. Upon discharging, Li ions from the anode diffuse to the cathode, while the electrons move from the anode through the external circuit to the cathode. It is known that sulfur in the cathode exists in the form of a large molecule, i.e., a

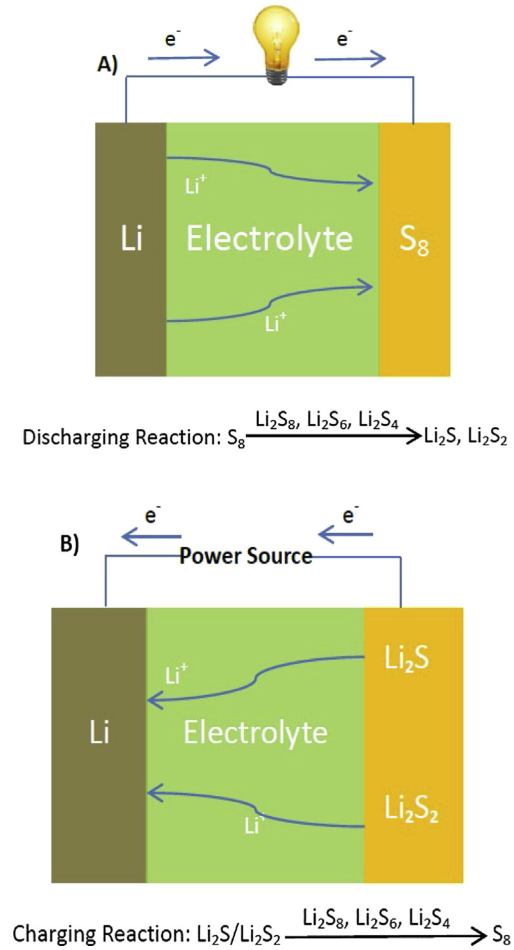

Fig. 1. Schematic presentation of a lithium–sulfur battery with the anode (metallic lithium) and cathode (sulfur-containing material), separated by a non-aqueous liquid electrolyte. A) Discharging process and B) charging process.

cyclooctasulfur S_8; thus, sulfur cannot turn into Li_2S in one-step reaction. Instead, when lithium ions from the anode react with the sulfur cathode, long-chain lithium polysulfides (Li_2S_x, $4 \leq x \leq 8$) [25,26], which are soluble intermediate products, are generated at the initial stage. These long-chain polysulfides will turn into insoluble Li_2S_2 and finally Li_2S with further discharging [27]. During this discharge process, negatively charged polysulfides dissolving in the liquid organic electrolyte also undergo shuttle moving driven by chemical potential and concentration difference between the cathode and the anode. Mikhaylik et al. [28] have reported a quantitative study of this shuttle phenomenon in the Li–S system. Their work shows that discharge curves of the Li–S cell at room temperature constitute two voltage plateaus, which are 2.3–2.4 V and ~2.1 V, corresponding to accepting 0.5 electron per sulfur atom with lithium tetrasulfide (Li_2S_4) as the product and accepting an additional electron per sulfur atom with lithium sulfide (Li_2S) and disulfide (Li_2S_2) mixture as the products, respectively, consistent with other reports [29,30].

Charging is a reverse process of discharging. In this process, external power source forces electrons to move from the cathode to the anode through an external circuit, while lithium ions diffuse through the separator back to the anode as shown in the schematic of Fig. 1b.

In addition to the key electrochemical reactions during charging and discharging described above, other processes could take place at the same time to impact the Li–S battery performance. These

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