



Review article

Advanced chemical strategies for lithium–sulfur batteries: A review

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Abstract

Lithium–sulfur (Li–S) battery has been considered as one of the most promising rechargeable batteries among various energy storage devices owing to the attractive ultrahigh theoretical capacity and low cost. However, the performance of Li–S batteries is still far from theoretical prediction because of the inherent insulation of sulfur, shuttling of soluble polysulfides, swelling of cathode volume and the formation of lithium dendrites. Significant efforts have been made to trap polysulfides via physical strategies using carbon based materials, but the interactions between polysulfides and carbon are so weak that the device performance is limited. Chemical strategies provide the relatively complemented routes for improving the batteries' electrochemical properties by introducing strong interactions between functional groups and lithium polysulfides. Therefore, this review mainly discusses the recent advances in chemical absorption for improving the performance of Li–S batteries by introducing functional groups (oxygen, nitrogen, and boron, etc.) and chemical additives (metal, polymers, etc.) to the carbon structures, and how these foreign guests immobilize the dissolved polysulfides.

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Keywords: Lithium–sulfur batteries; Chemical reaction; Lithium polysulfides; Functional groups; Additives

1. Introduction

The ever-increasing environment pollution and the decreasing fossil fuels force people to develop renewable energy, and it is wise to store and release the spare energy in certain forms whenever needed. Thus, the highly efficient energy storage system has attracted extensive interest in recent years, and various applications have been found in mobile devices, electric vehicles, and sustainable energy industry. Due to their light weight, high open circuit voltage, high capacity, and non-memory effect, lithium-ion batteries have been commercialized since 1990s by Sony corporation and dominated the market for portable electronic devices [1]. Although the performance of their cathode and anode materials has achieved the theoretical limits after twenty years of development, it still cannot meet the requirement of energy output.

Therefore, it is urgent to find new cathode and anode materials with higher energy density and excellent electrochemical performances.

Sulfur is one of the basic elements of the earth, and it is obtained extensively from nature. Moreover, sulfur is abundant, nontoxic, and environmentally benign, possessing great superiority compared with the limited and pollution-yielding fuel oil. More importantly, its ultrahigh theoretical capacity (1675 mAh g⁻¹) and theoretical energy density (2600 Wh kg⁻¹) distinguish it from other cathode materials [2–7]. Thus, the area of lithium–sulfur (Li–S) batteries developed robustly in the past few decades, and substantial achievement has been realized [8–12]. However, there are still some scientific and technical problems hindering Li–S batteries to be commercialized.

- (I) Both sulfur and lithium sulfides are intrinsically insulated, which impedes the transportation of electrons and ions.

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- (II) The volumes of cathode and anode materials change greatly during cycling process, bringing about the collapse of electrode structures.
- (III) Lithium sulfides, as the ultimate discharge products, are indissoluble in the electrolyte, and mostly deposit on the surface of the conductive framework.
- (IV) The intermediate discharge products lithium polysulfides (Li_2S_n , $4 < n < 8$) are soluble in the organic electrolyte, that will result in the loss of active materials and energy storage.

Among them, the dissolved lithium polysulfides will further diffuse to the electrolyte and form a membrane on the surface of the anode, leading to the crazing of the solid electrolyte interface film (SEI). The shuttle phenomenon gives rise to an irreversible loss of active materials, rapid capacity fading, low Coulombic efficiency, and short cycle longevity. Researchers have tried various approaches to solve this problem by virtues of delicately designed nanocarbon frameworks, such as porous carbon matrix, and so on [13–20]. They hope to take advantage of unique porous structure to prevent the dissolution, diffusion, and shuttling of polysulfides by physical encapsulation. For example, Zhang et al. designed the nested pore structure carbon with an ordered distribution of micropores and mesopores, which ensured an adequate accommodation for polysulfides to diffuse and reside evenly, and cycling performance of the device was greatly improved [16]. Park and his co-workers synthesized the honeycomb-like well-organized porous carbon nanosheets to trap lithium polysulfides [18]. Nevertheless, these physical pathways failed to immobilize polysulfides efficiently, and only physical encapsulation was not able to realize the practical application of Li–S batteries.

Different from the physical encapsulation, chemical adsorption displays a great potential in immobilizing polysulfides. Various chemical bonding approaches have been employed, in which, the functional groups and additives are introduced into carbon matrix to capture polysulfide species and prevent shuttle effect [21–26]. In this review, we show the recent advances in regard to chemical interactions between polysulfides and the functional groups or additives in Li–S batteries. Firstly, we introduce the

electrochemical transportation fundamentals of Li–S cells, and then various types of functional groups and additives that utilize chemical interactions to anchor polysulfides by different sulfur hosts were discussed and analyzed. Finally, we summarize and present the challenges and prospect of Li–S batteries.

2. Fundamental of Li–S batteries charge/discharge

The conventional Li–S batteries are constituted by a cathode of sulfur, an anode of lithium metal, and an indispensable ether-based electrolyte. In principle, the sulfur existing as ring-like octatomic molecules (S_8) will be reduced to Li_2S as the final discharge product, and oxidized to sulfur reversibly when the battery was charged. The whole reaction can be represented as $\text{S}_8 + 16 \text{Li} = 8 \text{Li}_2\text{S}$. However, the actual discharge and charge processes are exceedingly complex, accompanied with many multiple side reactions simultaneously [27–32].

A schematic illustration describing the working mechanism of Li–S batteries during the cycling test and a typical galvanostatic charge/discharge profile is shown in Fig. 1. The discharge process has two or three reduction stages depending on the composition of electrolyte [30,33,34]. The first stage, a rapid dynamics reaction, takes up about a quarter of the profile, corresponding to the reduction of S_8 to Li_2S_4 at about 2.4 V [32,35]. The resulted lithium polysulfides will dissolve and diffuse into the organic electrolyte. With the discharge process going on, these high-order polysulfides will be reduced to low-order polysulfides (Li_2S_n , $2 < n < 4$) and Li_2S . The second stage accounts for another quarter of the whole discharge process with a total discharge capacity of 1316 mAh g^{-1} and a plateau voltage of less than 2.1 V. However, the last stage in the discharge profile matching with the further reduction of Li_2S_2 to Li_2S , is not exhibited in the cyclic voltammetry (CV) curve, which is attributed to the sloped shape and the voltage difference between the previous two stages.

The dissolution and diffusion of lithium polysulfides have a serious influence on the electrochemical performance of Li–S batteries due to the following reasons [36–39]. (i) Causing the loss of active materials during cycling tests. A critical issue with respect to the polysulfides is their dissolution in organic

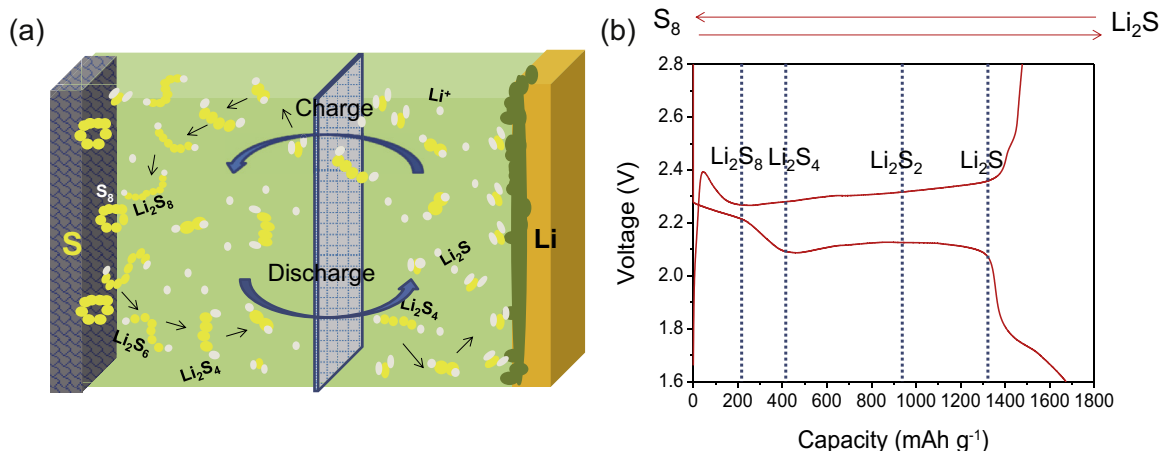


Fig. 1. (a) A schematic illustration of the redox reactions in Li–S batteries. (b) Galvanostatic charge/discharge profiles and the typical chemicals in each stage.

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