

1/22

energy storage materials



materials



Mechanochemical construction of fibrous stacked Li_2S cathode with high activity and low tortuosity for anode-free Li-S batteries

Chao Ding^a, Wenqiang Wang^{a,*}, Yifan Zhang^a, Jianlong Ding^a, Xiaoyu Hou^a, Zhichao Jia^a, Gengchao Wang^{a,*}, Chunzhong Li^b

^a Shanghai Engineering Research Center of Hierarchical Nanomaterials, Shanghai Key Laboratory of Advanced Polymeric Materials, School of Materials Science and Engineering, East China University of Science and Technology, Shanghai 200237, China

^b Shanghai Engineering Research Center of Hierarchical Nanomaterials, School of Chemical Engineering, East China University of Science and Technology, Shanghai 200237, China

ARTICLE INFO

Keywords:

Anode-free configuration
Lithium-sulfur batteries
Lithium sulfide
Mechanochemistry
Fibrous stacking

ABSTRACT

Anode-free configuration is expected to leverage the high specific energy of Li-S batteries. However, the insulation and slow electrochemical reaction kinetics of Li_2S , inefficient migration of ions in highly tortuous pores inside the cathode and uneven lithium deposition/stripping on bare current collector result in poor performance of anode-free lithium sulfur batteries (AFLSBs). Herein, we propose a construction strategy of fibrous stacked Li_2S cathode with high activity and low tortuosity. To synthesize fibrous cathode material with Li_2S nano coating, the mechanochemical method was employed and in-situ conversion of sulfur and Li_3N was achieved on the surface of poly(1-aminoanthraquinone) (PAAQ) modified carbon nanotubes. Furthermore, the fibrous stacked cathode was constructed through a reciprocating stamping technique. The electrochemical reaction of Li_2S can be significantly accelerated by its nano-size and PAAQ redox mediator. Meanwhile, quasi-2D network stacked structure of fibrous Li_2S cathode forms a perfect conductive framework with low tortuosity to improve the charge transfer efficiency. In addition, Li_3N residue in the cathode can shuttle to anode and form a dense Li_3N -rich SEI, thus stabilize the deposition and stripping of lithium. The above advantages lead to a significant improvement in the rate performance and cycle stability for AFLSBs.

1. Introduction

In recent years, the rapid development of low altitude economy, electric vehicles and smart grids has inspired people to develop energy storage devices with higher energy density [1–3]. The anode-free lithium sulfur battery (AFLSB) composed of lithium sulfide (Li_2S) cathode and bare current collector has the potential to leverage the high specific energy of Li-S batteries [4–6]. However, AFLSB is a full cell with $N/P = 1$ (equivalent to thin lithium sheets with stoichiometric sulfur matching) [7], it is different from the previously reported "Li-S batteries" based on thick lithium sheets which are actually half batteries with sulfur or Li_2S cathodes. This will cause positive feedback coupling between the reversibility of Li_2S cathode and the reversibility of lithium deposition/stripping on the anode [8,9], making the AFLSB system highly unstable with a rapid capacity decay in just a few cycles. For Li_2S cathode with practical loading, the reversibility is limited by insulation, shuttle effect and slow redox kinetics of Li_2S [10,11], as well as the

inefficient migration of ions in highly tortuous pores formed by the disordered stacking of Li_2S particles, which will further deteriorate the reversibility of anode. Therefore, in order to construct AFLSBs with ideal electrochemical performance, it's of vital importance to consider efficient charge transfer of Li_2S intrinsic and stacking structures in the cathode.

In terms of the intrinsic properties of Li_2S , schemes such as nano-sizing [12–14] and highly active catalysts [15,16] have been proven effective to reduce the high activation overpotential and improve the utilization of active materials. From the perspective of cathode structure, constructing a continuous conductive framework [17], designing cathode with nano pores, high porosity [18] and low tortuosity can significantly optimize the internal charge transfer efficiency, along with the alleviation of cathode damage caused by the volume change between sulfur and Li_2S [19–21]. Although the above works provide effective strategies for modifying Li_2S cathode, rational cathode modification must also consider the electrochemical stability of the anode in

* Corresponding authors.

E-mail addresses: wangwenqiang@ecust.edu.cn (W. Wang), gengchaow@ecust.edu.cn (G. Wang).

<https://doi.org/10.1016/j.ensm.2025.104354>

Received 27 March 2025; Received in revised form 14 May 2025; Accepted 27 May 2025

Available online 31 May 2025

2405-8297/© 2025 Elsevier B.V. All rights are reserved, including those for text and data mining, AI training, and similar technologies.