



Contents lists available at ScienceDirect

## Journal of Power Sources

journal homepage: [www.elsevier.com/locate/jpowsour](http://www.elsevier.com/locate/jpowsour)

## Explore the influence of coverage percentage of sulfur electrode on the cycle performance of lithium-sulfur batteries



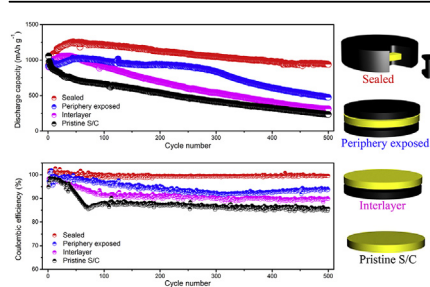
Zhong Li, Shaofeng Deng, Hejia Li, Hanzhong Ke<sup>\*\*</sup>, Danli Zeng, Yunfeng Zhang, Yubao Sun<sup>\*</sup>, Hansong Cheng<sup>\*\*\*</sup>

Sustainable Energy Laboratory, Faculty of Material Science and Chemistry, China University of Geosciences (Wuhan), 388 Lumo RD, Wuhan 430074, China

### HIGHLIGHTS

- The higher the surface coverage of sulfur the better the battery performance.
- 800 mAh g<sup>-1</sup> after 1000 cycles at 2 C with capacity retention over 77%.
- The sealed structure with narrow and irregular interstitials can host polysulfides.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Article history:

Received 15 December 2016

Received in revised form

11 February 2017

Accepted 18 February 2017

#### Keywords:

Lithium-sulfur battery

Polysulfides

Sealed electrode

Cycle performance

### ABSTRACT

We explore the influence of coverage percentage of the sulfur electrode on the cycle performance of lithium-sulfur batteries. Four sulfur electrodes covered with composite films made of carbon (BP2000), nano-Al<sub>2</sub>O<sub>3</sub> and polytetrafluoroethylene (PTFE) with different exposure are prepared and tested in lithium-sulfur batteries. The results display that both the capacity retention and coulombic efficiency are improved with the increase of surface coverage percentage among which the cell with the completely sealed sulfur electrode exhibits the best cycle performance. The discharge capacity of the sealed sulfur electrode is ca. 800 mAh g<sup>-1</sup> after 1000 cycles at 2 C with capacity retention over 77%. The ex-situ field emission scanning electron microscopy (FE-SEM), energy dispersive spectroscopy (EDS), Ultraviolet–Visible spectroscopy (UV) and nitrogen sorption measurements reveal that the sealed structure with narrow and irregular interstitials effectively suppresses the diffusion of soluble polysulfides out of the embraced environment with virtually no effect on lithium ion conduction. The present study demonstrates the effectiveness of the sealed structure for achieving robust electrochemical performance of lithium-sulfur batteries.

© 2017 Elsevier B.V. All rights reserved.

## 1. Introduction

The rapid development of advanced portable electronic devices and electric vehicles presents an increasing demand for batteries with high energy density and high power density. With a theoretical energy density as high as 2600 Wh kg<sup>-1</sup>, lithium-sulfur batteries are highly promising power devices and have been

\* Corresponding author.

\*\* Corresponding author.

\*\*\* Corresponding author.

E-mail addresses: [kehanz@163.com](mailto:kehanz@163.com) (H. Ke), [sunyubao@gmail.com](mailto:sunyubao@gmail.com) (Y. Sun), [chghs2@gmail.com](mailto:chghs2@gmail.com) (H. Cheng).

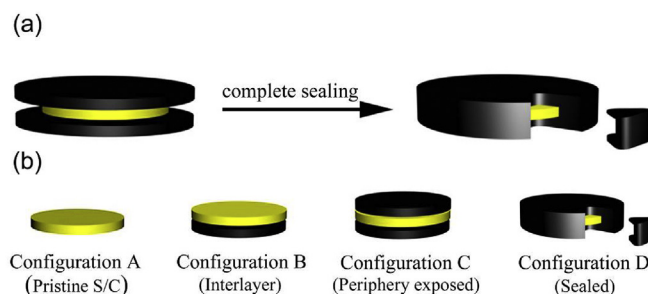
intensely studied [1,2]. Unfortunately, problems associated with the intrinsically low electronic conductivity ( $5.0 \times 10^{-30} \text{ S m}^{-1}$ ) of sulfur, which usually exists as a stable octatomic cyclic molecule in nature, as an electrode material, and polysulfide shuttling upon lithium insertion/desertion have prevented lithium-sulfur batteries from being broadly utilized in market place [3,4].

Electrochemically, electron transfer between an insulator and an electronic conductor can be significantly enhanced by either reducing the dimension of the insulator to a nano-size or converting the insulator-electrode interface to a liquid-electrode interface. Guo and co-workers successfully synthesized meta-stable small sulfur molecules of  $\text{S}_{2-4}$  in a confined space of a conductive microporous carbon matrix with 40 wt% loading of an active material and discovered that sulfur became electrochemically active in a carbonate-based electrolyte [5]. Alternatively, an ether based liquid electrolyte, e.g. LiTFSI in 1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME), is capable of reducing the substantial charge transfer resistance between carbon and sulfur by several orders of magnitude since the initially electrochemically reductive polysulfide anions become soluble in the ether-based electrolyte to enable the Faradic process to take place smoothly. However, a severe consequence arises from the soluble polysulfides that shuttle between electrodes through the porous Celgard separator and react with lithium metal, resulting in a loss of active materials and rapid capacity deterioration of the batteries.

To date, substantial efforts have been made to suppress the mobility of the soluble species by either physical containment or chemical anchoring. The major techniques include (1) adsorbing or encapsulating sulfur with carbons of various forms [6–14], metal complexes [15–32], conductive polymers [33–40], and a variety of functional binders [41–46]; (2) introducing a functional film or interlayer between the separator and sulfur cathode to suppress the diffusion of the soluble polysulfides [47–66]; (3) assembling carbon films and sulfur electrodes to form a multilayered structure [67–69].

While a sulfur electrode can be stabilized and its cycling performance can be improved via a variety of methods mentioned above, there have been formidable challenges to fabricate lithium-sulfur batteries that are sufficiently stable and practical enough to produce in an industrial scale. In particular, several studies demonstrated that the lifespan of a lithium-sulfur battery can be considerably extended by merely placing an interlayer in between a sulfur electrode and a Celgard separator [57–60]. The interlayer covers nearly half of the surface of sulfur electrode. The percentage of the sulfur electrode coverage was found to profoundly affect the cycling performance of the battery. Recently, a multilayer sulfur electrode structure was demonstrated to serve as an effective barrier to suppress the diffusion of the polysulfides [67–69]. However, as the periphery of the multilayer sulfur electrode was still in direct contact with the liquid electrolyte, the exposure of the sulfur electrode to the electrolyte in the battery still gave rise to soluble active species, which subsequently readily diffused to the anode side and caused capacity deterioration. Manthiram et al. recently reported a core-shell configuration by placing a carbon O-ring between two pieces of carbon papers with the sulfur embedded inside [70]. They demonstrate that the core-shell structure could restrict the diffusion of polysulfide species out of the shell by both dynamic and static battery measurements.

Herein, we design a sulfur electrode with 100% coverage by sealing the pristine sulfur electrode between two sheets of composite films made of BP2000, nano- $\text{Al}_2\text{O}_3$  and PTFE with the mass ratio of 2:4:4 as shown in Scheme 1a. A small amount of carbon (BP2000) serves as a current collector with a marginal electrochemical double layer capacitance (EDLC). The nano-sized  $\text{Al}_2\text{O}_3$  particles reduce the interstitial space among the aggregates and



**Scheme 1.** (a) Schematic illustration of the sealed sulfur electrode. (b) Schematic illustration of the sulfur cathodes with four different configurations.

thus suppress the diffusion of the polysulfides. The binder (PTFE) was used to enhance the mechanical strength to ensure the integrity of the sealed structure. For comparison, a pristine sulfur electrode (configuration A in Scheme 1b), the pristine sulfur electrode covered with a nano- $\text{Al}_2\text{O}_3$  incorporated composite film as an interlayer (configuration B in Scheme 1b) and the pristine sulfur electrode sandwiched in between two pieces of the nano- $\text{Al}_2\text{O}_3$  incorporated composite films with periphery exposure (configuration C in Scheme 1b) were also investigated. A long-term battery test at high C-rate for the sealed sulfur electrode (configuration D in Scheme 1b) was conducted to verify the reliability and robustness of the designated configuration. Finally, ex-situ FE-SEM, EDS, Ultraviolet–Visible spectroscopy and nitrogen sorption measurements were performed to analyze the mechanism of polysulfides confinement.

## 2. Experimental

### 2.1. Materials

Acetylene black and BP 2000 were bought from Shenzhen Kejingstar Technology Ltd. Nano- $\text{Al}_2\text{O}_3$ , carbon disulfide ( $\text{CS}_2$ ), sublimed sulfur, 1-methyl-2-pyrrolidinone (NMP), 60 wt% polytetrafluoroethylene (PTFE) emulsion and electrolyte (1 M LiTFSI/DOL&DME, 1 wt%  $\text{LiNO}_3$ ) were purchased from Sino Pharm Co. Ltd.

### 2.2. Preparation of the pristine sulfur electrode

The S/acetylene black cathode material for the pristine sulfur electrode was prepared using an impregnation method. Briefly, 0.6 g of sublimed sulfur was dissolved in 20 mL of  $\text{CS}_2$  to form a transparent solution. Subsequently, 0.4 g of acetylene black was added into the solution followed by stirring and sonication to obtain a homogeneous slurry.  $\text{CS}_2$  in the slurry was then slowly vaporized via heating and stirring to obtain a well dispersed S/acetylene black cathode material. The obtained S/acetylene black, acetylene black and PTFE with the mass ratio of 7:2:1 were weighed and collected in a glass beaker. The mixture was blended and pressed with a roller to form a thin film, which was dried at  $60^\circ\text{C}$  under vacuum for 24 h and cut into circular disks with a diameter of 8 mm for cell assembly. The sulfur loading on each disk was about  $1.5 \text{ mg cm}^{-2}$ .

### 2.3. Preparation of the nano- $\text{Al}_2\text{O}_3$ incorporated composite film and the sealed sulfur electrode

The nano- $\text{Al}_2\text{O}_3$  incorporated composite film was prepared by mixing BP 2000, nano- $\text{Al}_2\text{O}_3$  and PTFE with the mass ratio of 2:4:4 followed by rolling. The prepared nano- $\text{Al}_2\text{O}_3$  incorporated composite film was cut into circular disks with diameters of 16 mm and

Download English Version:

<https://daneshyari.com/en/article/5149558>

Download Persian Version:

<https://daneshyari.com/article/5149558>

[Daneshyari.com](https://daneshyari.com)