



Short communication

## Sulfur-embedded porous carbon nanofiber composites for high stability lithium-sulfur batteries



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## ABSTRACT

The porous carbon nanofibers-sulfur (PCNFs-S) composite electrode with micro-mesoporous (< 10 nm) and meso-macroporous (> 10 nm) is successfully designed and prepared via the electro-blown spinning technique and thermal treatment. The meso-macroporous structure in the PCNFs-S composite can provide high sulfur content up to 80 wt%, while micro-mesoporous affords efficient trapping of polysulfides and inhibits the “shuttle effect”. And the tailored porous fibrous network structure and carbon nanotubes (CNTs) in the composite facilitate the construction of a high electrical conductive pathway. Therefore, the lithium-sulfur (Li-S) batteries electrode exhibits high initial discharge capacities as high as 954 and 602.2 mAh g<sup>-1</sup> at 0.5 C and 2 C, respectively, and can obtain quite stable capacities of 795 and 601.5 mAh g<sup>-1</sup> after 350 cycles.

## 1. Introduction

Soaring demand for efficient and economic electric energy storage system has intensively promoted the development of rechargeable batteries with high energy density and long cycle life [1,2]. At present, the lithium-sulfur (Li-S) battery shows a great potential due to the use of sulfur as the electrochemical active material endowing the Li-S batteries with the high theoretical capacity of 1675 mAh g<sup>-1</sup> and the energy density of 2567 Wh kg<sup>-1</sup> [3,4]. Moreover, sulfur is one of the most abundant and nonpoisonous elements on the earth [5,6]. However, realizing commercialization of Li-S batteries is hindered by several difficulties. Firstly, the insulation of sulfur and the insolubility of the discharged products (Li<sub>2</sub>S, Li<sub>2</sub>S<sub>2</sub>) lead to the low sulfur utilization [7–8]. Secondly, the high solubility of the intermediate polysulfide (Li<sub>2</sub>S<sub>x</sub>, 4 ≤ X ≤ 8) results in the irreversible capacity fading and low coulombic efficiency [9–10]. Thirdly, large volumetric change (80%) during redox process causes the instability of the cathodes and shortens the cycle life [11].

To overcome the above problems, great efforts have been made to build carbon/sulfur composites, such as micro-mesoporous carbon, porous graphene, carbon nanotubes (CNTs), and the concrete electrochemical performances of these examples are shown in Table 1 [12]. In addition, porous carbon nanofibers (PCNFs) also can be considered as a kind of promising carbon material for Li-S batteries, which have been

expected to form a three-dimensional (3D) conductive network for the transmission of lithium ions owing to the high aspect ratio [13]. Lee et al. [14] reported a sulfur-carbon composite based on multichannel carbon nanofibers activated by potassium hydroxide (KOH) for Li-S batteries. The highly conductive porous network carbon matrix allowed Li-ion to transport fast and adsorbed polysulfides effectively. The specific capacity maintains above 920 mAh g<sup>-1</sup> after 300 cycles at 0.2 C, and the coulombic efficiency remains 99.5%. Wang et al. [15] prepared mesoporous CFs via the electrospinning technique by using resol as the carbon source and triblock copolymer pluronic F127 as the template. The mesopores of the carbon fibers (CFs), acted as the reaction room, not only reserved sulfur but also reduced the polysulfide shuttle. The ordered CFs-S composite electrode showed a stable capacity of 690 mAh g<sup>-1</sup> at 0.3 C after 350 cycles. Although the use of PCNFs has significantly improved the performance of Li-S batteries, the above complex preparation process of PCNFs limited the commercialization of Li-S batteries.

Herein, we report the preparation of sulfur-embedded porous carbon nanofibers as the cathode material for Li-S batteries via a simple electro-blown spinning technique of poly (acrylonitrile) (PAN)/polystyrene (PS) solution and following thermal treatment.

PAN is used as the carbon precursor due to its high carbon yield, good thermal stability, and spinnability. PS can be used as a pore-making agent to create void spaces in the fibers. The PCNFs contain

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**Table 1**  
Electrochemical performances of various porous carbon as sulfur cathodes for Li-S batteries.

Carbon electrodes	Discharge current rate	Initial discharge capacity (mAh g <sup>-1</sup> )	Ultimate discharge capacity (mAhg <sup>-1</sup> )	Cycle number	Ref.
Hierarchical porous carbon modified with ionic surfactants	0.1 C	1300	1116	100	[9]
3D N-doped graphene nanomesh foam	0.5 C	827	578	500	[10]
Caterpillar-like reconfigurable graphene	2 C	531	363	200	[11]
Mesoporous carbon structures	0.1 C	1390	840	100	[12]
Mesoporous carbon nanotube	0.1 C	776	528	100	[13]
<b>Porous carbon nanofiber</b>	<b>0.5 C</b>	<b>954</b>	<b>795</b>	<b>350</b>	<b>This work</b>
	<b>2 C</b>	<b>602.2</b>	<b>601.5</b>	<b>350</b>	

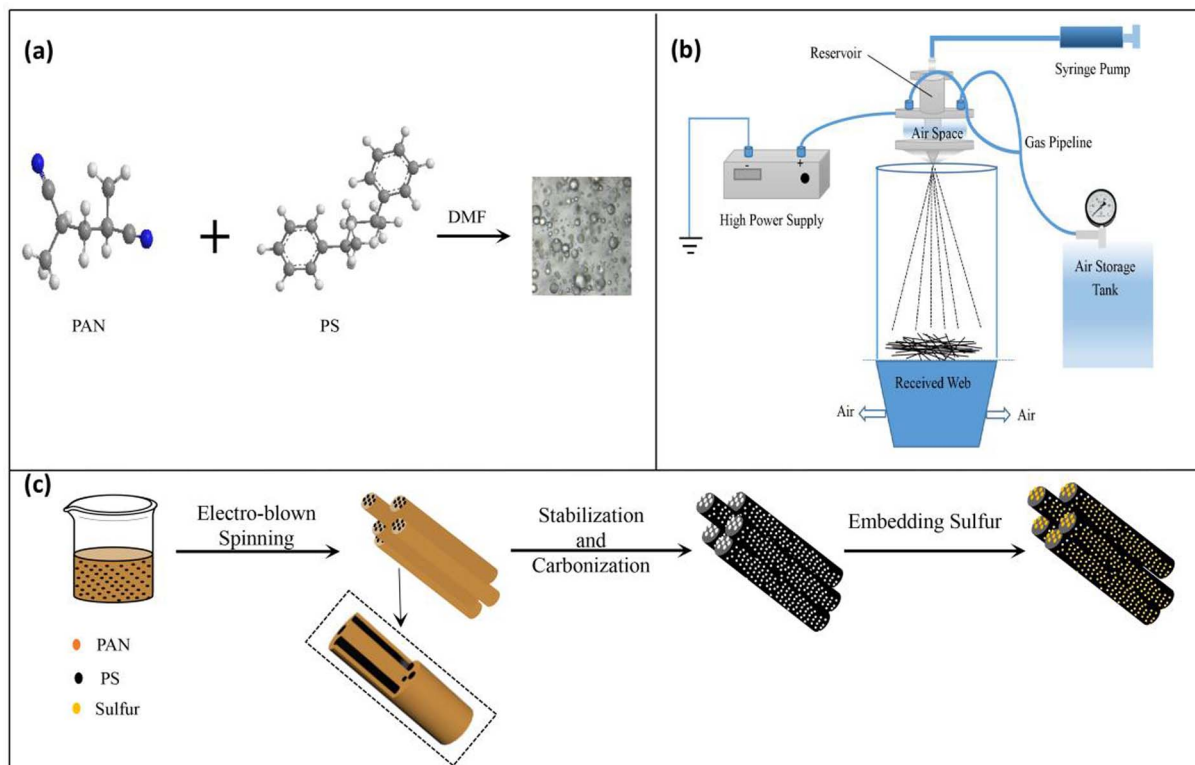
large amount of mesoporous and macroporous originating from the phase separated of incompatible PAN/PS in the fiber and the thermal decomposition of the PS. Finally, the sulfur was incorporated into the PCNFs by its sublimation. The cathode material performed outstanding cycle stability and high coulombic efficiency.

## 2. Experimental

PAN ( $M_w = 150$  kDa, Aldrich Co. Ltd.) and PS ( $M_w = 250$  kDa, Aldrich Co. Ltd.) with the weight ratio of 2:1 were dissolved in N,N-dimethylformamide (DMF) (Tianjin Kermel Co. Ltd., China) co-solvent by the magnetic stirring, and the microphase separation of PAN and PS happened during the dissolving process as shown in Fig. 1(a). The PAN/PS bicomponent fiber membrane was prepared by electro-blown spinning (EBS), as shown in Fig. 1(b). Then, the fiber membrane was stabilized at 260 °C for 1 h in the air atmosphere and carbonated at 1000 °C for 1 h in nitrogen atmosphere to form the PCNFs. The carbon yield of the bicomponent fiber is calculated using the following formula:

$$P_a(\%) = \frac{m_{\text{carbon}}}{m_{\text{PAN}}}$$

Where,  $P_a$  is the actual carbon yield,  $m_{\text{carbon}}$  is the mass of the PCNFs,  $m_{\text{PAN}}$  is the mass of PAN in PAN/PS, and the actual carbon yield is calculated to be 50.34%. Finally, porous carbon nanofibers-sulfur composite (PCNFs-S) was fabricated by blending the sulfur and PCNFs uniformly at the mass ratio of 4:1, and being heated at 155 °C for 24 h. The preparation process of the PCNFs-S composite shown in Fig. 1(c). The surface morphology and the crystalline structure of PCNFs and PCNFs-S were examined with a field emission scanning electron microscope (FE-SEM, S-4800, Hitachico., Japan), X-ray diffraction (XRD, AXS D8, Bruker), and X-ray diffraction spectroscopy (XPS) (K-al phaX, Thermo Fisherco, USA). The thermal property was conducted under nitrogen atmosphere with a heating rate of 10 °C min<sup>-1</sup> via thermal gravimetric analysis (TGA, NSK, TG/DTG 6350). The electrochemical performance of PCNFs-S composite cathode was analyzed using a CR2340-type coin cells. The electrode was fabricated by mixing active material (PCNFs-S, 70 wt%), carbon nanotubes (CNTs, 10 wt%) conducting additive (super P, 10 wt%), and binding (PVDF, 10 wt%) in N-methylpyrrolidinone solvent. The composite slurry was uniformly scraped onto the coating carbon aluminum foil and dried at 50 °C under the vacuum for 12 h. The mass loading of the active material was averagely between 1.0 mg in the single cathode, and the ratio of



**Fig. 1.** (a) The microphase separation of PAN and PS during the dissolving process. (b) The schematic diagram of the home-made electro-blown spinning (EBS) device. (c) Preparation process of the PCNFs-S composite.

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