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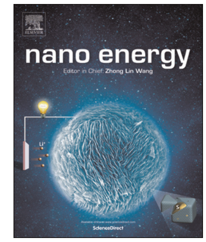

Porous carbonized graphene-embedded fungus film as an interlayer for superior Li-S batteries

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Abstract
Graphene-embedded carbon fiber (GFC) film has been fabricated by using filamentous fungus (*Aspergillus niger*) as carbonizable fibers to drive the graphene nanosheets to embed in the hyphae network system and then carbonizing at 700 °C. The molecular structure of the GFC film is primarily composed of aromatic components, and the film is doped by N (8.62%) and O (8.12%) elements. The conductivity of the final product reaches as high as 0.71 S cm^{-1}. The GFC film serves as the conductive interlayer to greatly improve the performance of Li-S batteries including capacity retention and rate capability. By inserting the GFC film, the battery can deliver a capacity of ~700 mAh g^{-1} after 300 cycles at 1 C. Even performed at 5 C, it is able to deliver a reversible capacity of more than 650 mAh g^{-1}. This research presents a facile and effective method for the fabrication of superior macroscopic carbon monolith, which holds great potential in other forms of electrochemical energy storage.
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Introduction

The development of sustainable electric systems (e.g., vehicles) has been an inconvertible trend all over the world, which significantly required advanced rechargeable batteries with high energy density and long cyclability [1-4]. Lithium-sulfur (Li-S) batteries as a promising alternative possess high theoretical capacity (1675 mA h g^{-1}) and energy density (2600 Wh Kg^{-1}) [5]. Because of many advantages of sulfur (e.g., abundant resource, low cost, harmless), Li-S batteries have been recognized as one of the most competitive choices for the next generation electrochemical energy storage [6]. Unfortunately, the current Li-S batteries cannot totally fulfill the expectations of researchers, which limits its commercialization. One main reason is the insulating nature of sulfur and its lithiumization compounds (or named polysulfides), which lowers the cycling performance and rate capability [7]. Another one is that the polysulfide is readily dissolved in liquid electrolyte, which can transport through the separator to the anode. This phenomenon is called "shuttle effect" and inevitably decreases the coulombic efficiency and cyclability of the batteries [7].

To circumvent the aforementioned challenges, lots of methods have been proposed, which mainly concentrates on cathodes improvement. Successful examples include infiltrating the sulfur into various carbonaceous materials (e.g., reduced graphene oxide, carbon nanotubes, porous carbon) [4,8-18], using metal-organic framework as host to storage the sulfur [19,20], and covering traditional cathodes with polymers [21,22]. These methods not only restrict the shuttle effect, but also withstand the sulfur volume change. Apart from the above methods, it is also reported that coating or covalently bonding the sulfur with conducting polymers is very effective to suppress the dissolution and transportation of polysulfides, resulting in enhanced cyclability [23,24]. The modifications of sulfur cathodes indeed make the future of Li-S batteries more promising, but their fabrications are relatively complex and expensive [5], which restricts the large-scale applications of Li-S batteries.

Most recently, modifying cell configuration through inserting a conductive interlayer shows unexpected potential on improving the cyclability and capacity of Li-S battery. The conductive interlayer blocks the migration of the dissolved lithium polysulfides and readily reduced the potential polarization [25]. Until now, carbon black, multi-walled carbon nanotubes, conducting polymers, reduced graphene oxide and metal have been used to construct the conductive interlayer [25-32]. However, the procedures of preparing the interlayer by these raw materials are still relatively time consuming and complex. To further enhance the potential of interlayer, biomass-derived carbonaceous materials have been designed and synthesized. Leaf, cassava and eggshell were used as feedstock to obtain the porous biomass-derived carbon [33-36]. For the sake of practical applications, the carbonaceous materials need be assembled into a film by binders and this complicated the process. In an alternative way, paper towel and filter paper were carbonized to biomass-derived film acting as conductive interlayer, which avoided the introduction of binders [37,38]. But the performance of the battery is still needed to be improved as compared with that using graphene or carbon nanotubes as interlayer.

In present research, we design a microorganism-based approach for fabricating conductive porous carbon film (GFC film) through using filamentous fungus as carbonizable binders to stabilize the graphene nanosheets within the film. The film precursor is formed by a simple vacuum filtration of the mixture of graphene nanosheets and fungus hyphae suspension, which costs less than 1 min and is able to be easily scaled up. Moreover, the usage of filamentous fungus to prepare the carbon film satisfies the green and sustainable purpose. Furthermore, the nature of fungus decides high-content N element in the final product. This is benefical to improving the performance of Li-S cells because the binding of sulfur-containing species at the N sites is more stable than binding on a carbon site [4]. The obtained film is expected to improve the performance of Li-S batteries when used as interlayer.

Experimental section

Materials

Graphene nanosheets powders were donated by The Sixth Element (Changzhou) Ltd. *Aspergillus niger* was provided by China Center for Type Culture Collection (CCTCC). Without special caution, the chemicals used were of analytical grade.

Culture of fungus

Potato medium was adopted to culture the fungus. Specifically, 200 g of potato was sliced and cooked in boiling water for 30 min. The potato was collected by gravity filtration using a gauze and the filtrate was put into a conical flask. Then 20 g of glucose, 0.05 g of $MgSO_4 \cdot 7H_2O$ and 0.1 g of KH_2PO_4 were added into the potato juice. The mixture solution (or potato medium) was treated at 115 °C under high pressure for 30 min. The fungus hyphae was inoculated into the potato medium. The culture temperature is 33 °C and the time is ~2 days.

Preparation of fungus hyphae dispersion

Fungus mycelium pellets were gradually formed during the solution culture of *Aspergillus niger*. The pellets were treated by a juice machine for 6 min, which became the dispersed hyphae. The hyphae were rinsed with deionized water for 5 times and collected by vacuum filtration. The hyphae were dispersed in mixture of water and ethanol (volume ratio 1:1) again to generate a suspension with a mass concentration of 10 mg mL^{-1}.

Synthesis of GFC film

1 g graphene nanosheets were dispersed in 50 mL 0.01 wt% of Tween aqueous solution for 150 min by ultrasonication (100 W). The graphene nanosheets dispersion was then poured into 400 mL of fungus hyphae suspension. The mixture was stirred vigorously for 10 min by electric blender with speed 1500 rounds per minute. To prepare the GFC film, 10 mL of black mixture was treated by vacuum

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