



Facile one-pot hydrothermal with subsequent carbonization preparation of hollow tin dioxide@carbon nanostructures as high-performance anode for lithium-ion batteries



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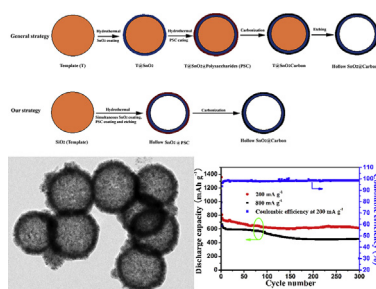
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HIGHLIGHTS

- Two kinds of hollow SnO₂@C nanostructures were prepared by a novel and facile route.
- The hollow structure and uniformly carbon coating endowed the composites superior physical buffer ability.
- The composites exhibited excellent electrochemical performances.

GRAPHICAL ABSTRACT



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ABSTRACT

To overcome the issues of fast capacity fading and poor rate capacity for tin dioxide (SnO₂) anode materials, an effective strategy is explored to fabricate nanostructured SnO₂@carbon composites (SnO₂@C) with large specific surface area, abundant cavity and confined dimension, considering the peculiar nanostructure to achieve increased contact interface between the active materials and the electrolyte, restricted agglomeration of SnO₂, buffered volume variation, enhanced structure stability, shortened diffusion distance of lithium-ion and electron and improved electronic conductivity. Herein, two kinds of hollow SnO₂@C nanostructures, namely hollow SnO₂@C spheres and SnO₂@C tubes are prepared by a simple approach. Comparing with general strategies involved templates, the preparation process in our strategy is greatly simplified, and the typical and tedious respective SnO₂ coating, polysaccharides coating and template removal are avoided, instead, the three processes are achieved simultaneously. It is believed that our strategy may pave the way for facile preparation of various microstructures of hollow SnO₂@C composites with relatively uniform size and to a wide variety of applications. More importantly, when tested as promising anode materials for lithium-ion batteries, both the two hollow SnO₂@C nanostructures exhibited superior electrochemical performance due to the synergistic effect of hollow nanostructure and relatively uniform carbon coating. The SnO₂@C tubes delivered a reversible capacity of 587.1 mAh g⁻¹ at 200 mA g⁻¹ after 100 cycles. More importantly, the hollow SnO₂@C spheres delivered a reversible capacity of 612.9 mAh g⁻¹ at 200 mA g⁻¹ even after 300 cycles.

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1. Introduction

Currently, as the promising candidates for anode materials, transition metal oxides with high capacities from 700 to 1000 mAh g⁻¹ have been widely investigated to replace the commercial graphite anode for lithium-ion batteries (LIBs) [1,2]. Among them, SnO₂ has attracted extensive attention in particular due to its high theoretical capacity (782 mAh g⁻¹), relatively safe working potential and environmental benignity [3,4]. Despite have so much merits, however, the practical application of the SnO₂ in LIBs is greatly impeded by their intrinsic drawbacks such as the poor conductivity, and huge volume change (more than 300%) during discharge/charge process, finally causing fast capacity fading upon an extended number of cycles [5]. To mitigate these problems, great efforts have been made by the researchers worldwide are focus on the exploration of strategies which can be used to improve the lithium storage and cycling performance of SnO₂-based anodes. One of the well-known methods is engineer peculiar nanostructured SnO₂, such as nanotubes [6], nanoboxes [7], nanosheets [8], nanowires [9], nanorods [10] and so on, because these nanostructures by virtue of larger surface area and high surface-to-volume ratio to effectively reduce the absolute volume variation of SnO₂ and enhance the lithium storage to a certain extent. Unfortunately, considerable capacity decay still happened due to the crack, aggregation and pulverization of nanoscale SnO₂ upon the long-term repeated expansion/contraction process (more than 50 cycles) [7,8].

Delightedly, compositing nanostructured SnO₂ with carbon, such as carbon coating on the surface of nanostructured SnO₂ or SnO₂ embedded into various carbon matrices, have been proven to be an effective strategy to significantly improve the cycling performance of SnO₂ anode [11–18]. The flexible, mechanical elastic and electronic conductive carbon component can effectively buffer the huge volume change of SnO₂ and enhance the electronic conductivity of whole electrode, resulting in improved cycling performance and rate capability. In particular, hollow SnO₂@C (core@shell) nanostructures have been extensively researched as the promising anode materials for LIBs due to their inherent advantages. The inner cavity of the hollow structures can provide a larger free space for accommodating the volume change of SnO₂ and buffering the mechanical strain formed during repeated volume expansion/contraction process. The larger specific surface area and relatively thin shell of the hollow structures can ensure a high contact interface between the active materials and the electrolyte and a shortened diffusion distance for lithium ion. It is noteworthy that the flexible carbon coating on the surface of hollow SnO₂ nanostructures also play an extremely important role in improvement of SnO₂ electrochemical properties, because the carbon can not only alleviate the volume change, prevent SnO₂ from aggregation and pulverization, and ensure the implementation of electrode integrity during prolonged cycling process, but also enhance electronic conductivity of overall electrode and further facilitate electron and ion transport throughout the electrode. Thus, these features of the hollow SnO₂@C nanostructures bring SnO₂-based anode materials significant improvement in cycling performance and rate capability [19–23]. However, until now the typical strategy for preparation of hollow SnO₂@C nanostructures involved the use of various templates, generally contains preparation of templates, respective SnO₂ coating, carbon coating and templates removal [24,25]. The tedious and time-consuming preparation procedures may hinder them to be used in large-scale applications [26].

In this work, inspired by the aforementioned studies, we rationally designed and prepared two kinds of nanostructures, namely hollow SnO₂@carbon nanospheres (denoted as hollow SnO₂@C spheres) with uniform size distribution and SnO₂@carbon

nanotubes (denoted as SnO₂@C tubes), by a facile strategy of one-pot hydrothermal with subsequent carbonization for the first time, the fabrication process as shown in the schematic illustration (Fig. 1). Comparing with general strategies, the preparation process in our strategy is greatly simplified, and the tedious respective SnO₂ coating, polysaccharides coating and template removal are avoided, instead, the three processes are achieved simultaneously. In addition, the preparation of SiO₂ templates is simple. This strategy may pave the way for facile preparation of hollow metal oxide@C nanospheres or nanotubes with relatively uniform size distribution. The fully synergistic effect of hollow nanostructures and uniform carbon coating endowed the hollow SnO₂@C spheres and SnO₂@C tubes with superior electrochemical properties as promising anode for LIBs. When tested as anode materials for LIBs, the SnO₂@C tubes and hollow SnO₂@C spheres delivered a reversible capacity of 587.1 and 612.9 mAh g⁻¹ at 200 mA g⁻¹ after 100 and 300 cycles, respectively. Excellent rate capacities for the two SnO₂@C composites were also obtained.

2. Experimental section

2.1. Preparation of SiO₂ templates

The SiO₂ nanospheres with uniform size distribution were synthesized by using a Stöber method. Typically, 10 mL of deionized water and 3 mL of ammonium hydroxide (25%) were mixed with 74 mL of ethanol. After stirring for 30 min, 6 mL of tetraethylorthosilicate (TEOS) was added dropwise into the mixed solution under constant stirring, and then stirred another 6 h. Then, the white precipitate was collected, washed with ethanol and deionized water to get monodispersed SiO₂ nanospheres. The SiO₂ nanowires were prepared by according to previous method but some modification [27]. Typically, 0.246 g of triblock copolymer pluronic F127 (EO100PO70EO100, EO = ethylene oxide, PO = propylene) and 0.51 g of Cetyltrimethylammoniumbromide (CTAB) were dissolved in 48.3 ml of deionized water and then stirred for 30 min. After that, 8 mL of hydrochloric acid (HCl) was added into the solution under constant stirring. After stirred for 5 min, 0.85 mL of TEOS was added dropwise into the solution. Then, the mixture was allowed to stand under static condition at room temperature for about 20 h after about 20 s of stirring. Finally, the SiO₂ nanowires were obtained by collecting and deionized water wash of white precipitate.

2.2. Preparation of hollow SnO₂@Carbon nanospheres (hollow SnO₂@C spheres)

Typically, 100 mg of as-prepared SiO₂ nanospheres were dispersed into 60 mL of deionized water by ultrasound for 1 h. Then, 1.2 g of glucose was added into the suspension and stirred for 10 min. After that, 0.2 g of tin (II) chloride dehydrate (SnCl₂·2H₂O) was added into the suspension under stirring. After stirring for 30 min, ca. 0.044 g of ammonium fluoride (NH₄F) was added into the mixed suspension and then stirred another 30 min. After that, this mixed suspension was transferred into a Teflon-lined stainless steel autoclave, and then placed in an oven at 180 °C for 48 h. After cooled down to room temperature, the hollow SnO₂@Ploy-saccharides (PSC) nanospheres were collected by centrifugation, washed with deionized water and ethanol thoroughly, and dried in an oven at 60 °C overnight. Finally, the hollow SnO₂@C spheres composite was obtained by the dark brown SnO₂@PSC carbonization at a temperature of 500 °C for 3 h with a ramping rate of 0.5 °C min⁻¹ under argon atmosphere.

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