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SnO_2 nanospheres among GO and SWNTs networks as anode for enhanced lithium storage performances☆

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ABSTRACT

Conducting supporters of purified single-walled carbon nanotubes (SWNTs) and graphene oxide (GO) were used to confine pomegranate-structured SnO_2 nanospheres for forming SnO_2-GO-SWNT composites. As anode material for lithium ion batteries (LIBs), this composite exhibits a stable and large reversible capacity together with an excellent rate capability. In addition, an analysis of the AC impedance spectroscopy has been used to confirm the enhanced mechanism for LIB performance. The improved electrochemical performance should be ascribed greatly to the reinforced synergistic effects between GO and SWNT networks, and their enhanced contribution of the conductivity. These results indicate that this composite has potential for utilization in high-rate and durable LIBs.

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1. Introduction

To meet the increasing demand of high-power lithium-ion batteries (LIBs), metal oxides as anodes with high specific capacities have attracted great attention [1,2]. Especially, SnO_2 has been considered as a potential anode material because of its high theoretical capacity of 782 mAh/g, which is much larger than that of 372 mAh/g for commercial anode of graphite [3–6]. However, similar to the other metal oxide anodes, SnO_2 anodes with low intrinsic electric conductivity and large volume changes during cycling lead to the poor stability of repetitive cycling. To overcome above mentioned limitations is important to improve the electrochemical performance of SnO_2 anodes [7–9].

Typically, electrodes were fabricated from mixtures of active materials, conductive agent of particle-shape of carbon black and polymeric binders. In this case, ion and electron transport networks were constructed only based on the point-to-point contact. The as-formed conductive networks may be destroyed quickly because of the large volume change of metal oxides during cycling. The simplest strategy is to replace the particle-shape conductive agent with carbon nanotubes (CNTs) or graphene [10]. Conducting agent of CNTs can only effectively improve their conductance based on the so-called line-to-line contact, but not effectively cushion their huge volume changes and further retain the electrode integrity. On the other hand, graphene was recently considered as a potential alternative based on surface-to-surface contact due to its high surface area, chemical tolerance and structural flexibility [11]. However, there is only a fast electron transfer within a single graphene nanosheet, while the poor electronic contacts present in active material agglomerates coated in graphene sheet. Thus, the utilization of CNTs combined with physically separated graphene is promising to accommodate the poor conductivity and volume change of metal oxides. Many kinds of anode materials consisting of CNTs, graphene and active materials have been prepared and used in LIBs to take advantage of their superior electrical conductivity and mechanical flexibility, such as TiO_2, Fe_3O_4 and MoS_2, and so forth [12–14]. Especially, Zhang et al. synthesized an anode material of SnO_2-graphene-CNT mixture for LIBs, which delivered enhanced LIB performance [15]. Therefore, the research of metal oxide anodes mixed with CNTs and /or graphene for LIBs are far from complete.

In our previous work, the purified single-walled carbon nanotubes (SWNTs) as webs of curved nanotubes can form strong intertwined entanglements with a network structure, and further afford an obviously enhanced conductivity for their composites [16,17]. Meanwhile, the MWNTs with noodle structured networks cannot effectively build up the high conducted supporters for further improve their LIB performance. The other reports also support this point of view for the SWNTs [17,18]. More importantly, using

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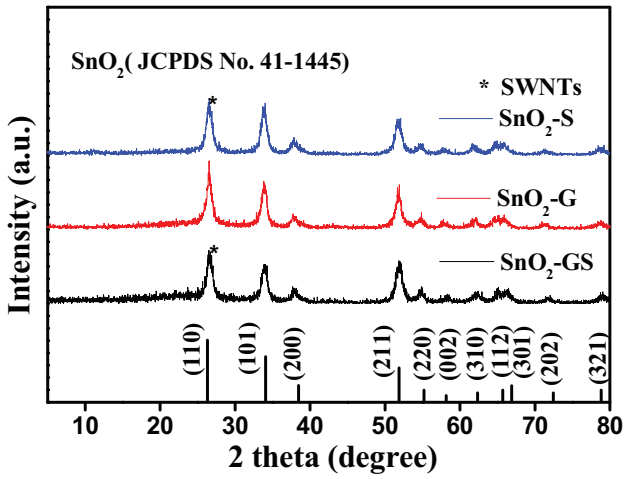

Fig. 1. XRD patterns of SnO_2-GS, SnO_2-G and SnO_2-S composites.

of both SWNTs and graphene oxide (GO) for enhancing their LIB performance has been rarely reported. Therefore, to obtain SnO_2-graphene-SWNT mixtures and investigate their enhanced LIB performance is needed.

In this work, pomegranate-structured SnO_2 nanospheres have been synthesized by a simple hydrothermal reaction and mixed with SWNTs and GO for forming composites of SnO_2-GO-SWNTs (denoted as SnO_2-GS). Directly used as electrodes via mixing with binder, the SnO_2-GS electrodes can deliver an obviously better electrochemical performance compared to the SnO_2-GO and SnO_2-SWNTs samples. In addition, an analysis of the AC impedance spectroscopy confirms that such configuration can effectively enhance the conductance of electrodes and further improve their LIB performance.

2. Experimental

2.1. Materials synthesis

All chemicals in this work were of analytical grade and used as received. GO was synthesized by the oxidation of natural flake graphite power using a modified Hummers method [16]. The SWNTs were produced by an arc-discharge method according to our previous report [17]. Pomegranate-structured SnO_2 nanospheres were prepared via a simple hydrothermal reaction. 10 mmol sodium stannate (Na_2SnO_3) was dissolved in 50 mL of 1 M glucose aqueous solutions. The clear solution was obtained through ultrasonic dispersion for 1 h before being transferred to a Telflon-lined stainless steel autoclave heated at 180 °C for 4 h. The precipitates were harvested by centrifugation and washed thoroughly with deionized water and ethanol several times, respectively. After drying at 80 °C overnight, SnO_2 powders were obtained at 550 °C for 4 h with a heating rate of 2 °C/min under air. Then, 15 mg of SWNTs was dispersed into a 30 mL GO (0.5 mg/mL) aqueous suspension by sonication for 12 h. Afterward, 70 mg of SnO_2 powders was added into above suspension through ultrasonic dispersion for 2 h. The SnO_2-GS anodes were obtained after filtrated and dried at 80 °C under vacuum. SnO_2 anode with GO as a conductive agent composites (SnO_2-G) and with SWNTs (SnO_2-S) composites were harvested in the same way.

2.2. Materials characterization

The samples were characterized by X-ray diffraction (XRD, RIGAKU SCXmini), scanning electron microscope (SEM, Hitachi SU8010), transmission electron microscope (TEM, Tecnai G2 F20).

2.3. Electrochemical measurements

The electrochemical behaviors were carried out via CR2025 coin-type cells assembled in a dry argon-filled glove box. The test cell consisted of working electrode (about 1.2–1.5 mg/cm^2) and lithium sheet which were separated by a Celgard 2300 membrane and electrolyte of 1 M $LiPF_6$ in EC:EMC:DMC (1:1:1 in volume). The working electrodes were prepared by mixing 90 wt% composites material (SnO_2-GS), and 10 wt% polymer binder (Carboxymethylcellulose, Na-CMC). The electrodes were dried at 80 °C overnight in a vacuum. These electrochemical properties are all calculated based on the overall mass of composites material. The cells were cycled by LAND2001A at room temperature. Cyclic voltammetry curves (CVs) were carried out on a CHI660D

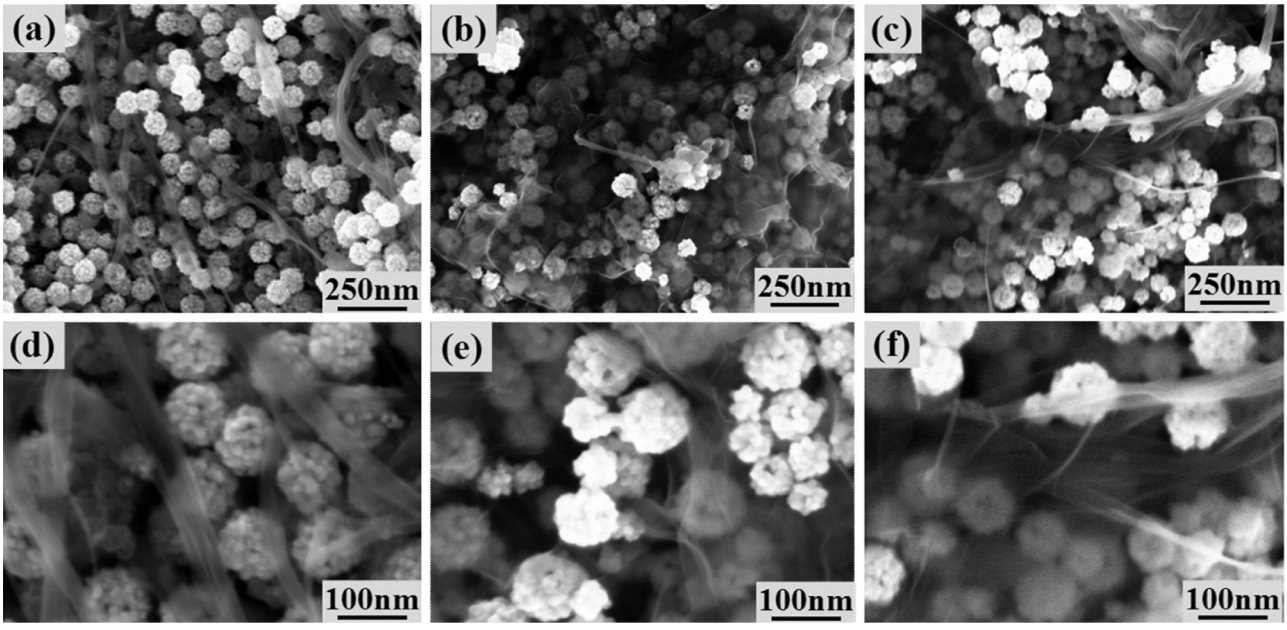

Fig. 2. SEM images of the (a,d) SnO_2-S, (b,e) SnO_2-G and (c,f) SnO_2-GS.

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