



Review article

The developments of SnO₂/graphene nanocomposites as anode materials for high performance lithium ion batteries: A reviewYuanfu Deng^{a, b, *}, Chengcheng Fang^c, Guohua Chen^{b, c, d, **}^a The Key Laboratory of Fuel Cell Technology of Guangdong Province, School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou, China^b Center for Green Products and Processing Technologies, Guangzhou HKUST Fok Ying Tung Research Institute, Guangzhou 511458, China^c Fok Ying Tung Graduate School, the Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China^d Department of Chemical and Biomolecular Engineering, the Hong Kong University of Science and Technology, Hong Kong, China

HIGHLIGHTS

- The advances of SnO₂/graphene composites as anode materials for LIBs are reviewed.
- Preparation methods and structural modifications of SnO₂/graphene are presented.
- Relationships of Li storage property, nanostructures and composition are discussed.
- Opportunities for future work are identified.

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ABSTRACT

With the increasing energy demands for electronic devices and electrical vehicles, anode materials for lithium ion batteries (LIBs) with high specific capacity, good cyclic and rate performances become one of the focal areas of research. Among the various anode materials, SnO₂/graphene nanocomposites have drawn extensive attentions due to their high theoretical specific capacities, low charge potential vs. Li/Li⁺ and environmental benignity. In this review, the advances, including the synthetic methods and structural optimizations, of the SnO₂/graphene nanocomposites as anode materials for LIBs have been reviewed in detail. By providing an in-depth discussion of SnO₂/graphene nanocomposites, we aim to demonstrate that the electrochemical performances of SnO₂/graphene nanocomposites could be significantly enhanced by rational modifications of morphology and crystal structures, chemical compositions and surface features. Though only focusing on SnO₂/graphene-based composites, the concepts and strategies should be referential to other metal oxide/graphene composites.

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1. Introduction

It is now universally accepted that the combustion of oil fuel is the major reason for modern cities' atmospheric contamination. And the decreasing amount of petroleum resources impels people to seek for other clean energy substitutes. Along with the development of renewable energy technology, such as solar, wind and

tidal energy, how to effectively store these kinds of energy becomes one of the major concerns. Since 1990s, lithium ion batteries have been developing rapidly, widely used in portable electronic devices [1]. Recent years, the expanding large-scale applications of LIBs in electric vehicles (EVs), plug-in hybrid electric vehicles (PHEVs) and large-scale energy storage urgently request a significant improvement in the lithium storage capacity to enhance their feasibility [2].

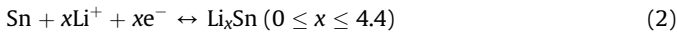
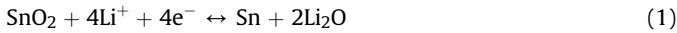
As one of the most important part of lithium ion batteries, anode materials have always been the focal area of research. Due to their high theoretical capacity, metal oxides have been studied intensively as anode materials for LIBs, such as Ti-based oxides (TiO₂ [3] and Li₄Ti₅O₁₂ [4]), Mn-based oxides (MnO [5], Mn₃O₄ [6], Mn₂O₃ [7] and MnO₂ [8]), Fe-based oxides (Fe₃O₄ [9] and Fe₂O₃ [10]), Co₃O₄ [11], NiO [12] and SnO₂ [13]. Among them, SnO₂ is believed to be

* Corresponding author. The Key Laboratory of Fuel Cell Technology of Guangdong Province, School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou, China.

** Corresponding author. Center for Green Products and Processing Technologies, Guangzhou HKUST Fok Ying Tung Research Institute, Guangzhou 511458, China.

E-mail addresses: chyfdeng@scut.edu.cn (Y. Deng), kechengh@ust.hk (G. Chen).

the most promising candidate, because of its low cost, environmental benignity and high theoretical specific capacity [13,14]. More importantly, SnO₂ has the lowest operating voltages (average discharge and charge voltages of 0.3 V and 0.5 V vs. Li/Li⁺, respectively) compared to the other transition metal oxides so that the energy density of full cells would be higher when SnO₂ is used as anode material [13,15]. For SnO₂-based electrode, the electrochemical processes involve the following two steps [15,16]:



For bulk SnO₂ materials, the first reaction is believed to be irreversible so that the theoretical capacity is about 780 mAh g⁻¹ [15,17,18]. While for nano-sized SnO₂ particles, it is reported that the first reaction becomes reversible/part reversible and the theoretical capacity can increase to the largest value of 1494 mAh g⁻¹ [16,19], which is about three times higher than that of the commercialized graphite (only 372 mAh g⁻¹). The second process is widely known to be reversible and the lithium can be repeatedly alloyed and dealloyed with Sn formed *in situ*. Fig. 1a and b shows the galvanostatic discharge/charge (G-D/C) profiles and the corresponding derivative capacity plots of micrometer-sized SnO₂ (10 μm), respectively, and Fig. 1c and d displayed the (G-D/C) profiles and cyclic voltagegrams (CVs) of the nanometer-sized SnO₂, respectively. As shown in Fig. 1c, the strong peak at 0.50 V and broad peak at 1.24 V during the charge process of the SnO₂ nanocrystal/nitrogen-doped reduced graphene oxide hybrid material (SnO₂ NC@N-RGO) correspond to the phase transition from Li_{4.4}Sn alloy and Li₂O to SnO₂, respectively [16]. This result suggested that the first reaction is reversible/part reversible in the nanometer-sized SnO₂. However, for micrometer-sized SnO₂, the

broad peak at ~1.24 V is not observed in the second cyclic curve, suggesting this reaction is irreversible [20]. In addition, detailed lithium storage mechanism of an ordered mesoporous SnO₂ electrode material by Synchrotron X-ray diffraction and absorption spectroscopy also reveals that some portion of Li₂O decomposes upon delithiation when the discharge voltage up to 2.0 V and the resulting oxygen reacts with Sn to form the SnO_x phase along with dealloying of Li_xSn (Fig. 2) [21].

Commercial use of SnO₂-based anode materials are still largely hampered by three major issues: (1) large initial irreversible capacity loss because of the continual formation of a very thick solid electrolyte interphase (SEI) during cycling [16–19]. (2) Large volume change during charge/discharge process causes severe internal stress, resulting in electrode pulverization and losing electrical contact with current collector so that capacity fading is inevitable during extended cycles and cycling performance becomes poor [21–23]. The *in-situ* high resolution transmission electron microscope (HRTEM) images (Fig. 3) of a nanoscale electrochemical device consisting of a single SnO₂ nanowire as an anode, an ionic liquid-based electrolyte (ILE) and a cathode of LiCoO₂ particles demonstrated the obvious morphology change and volume expansion of the SnO₂ nanowires [22]. (3) X-ray diffraction (XRD) [24] and HRTEM (Fig. S1) [25] studies confirmed that the small and active Sn particles aggregated into larger and inactive Sn clusters during cycling.

To solve these existing problems, three major strategies have been adopted. The first one is to synthesize various unique nanostructured tin oxide materials, such as SnO₂ nanoparticles [18,25,26], nanowires [22,27] and nanospheres [13,28]. Nano-sized materials can not only shorten the diffusion length for electrons and lithium ions, but also buffer the large volume effect [25,29,30]. The second one is to fabricate porous or hollow structured SnO₂, which could provide enough empty space to alleviate pulverization

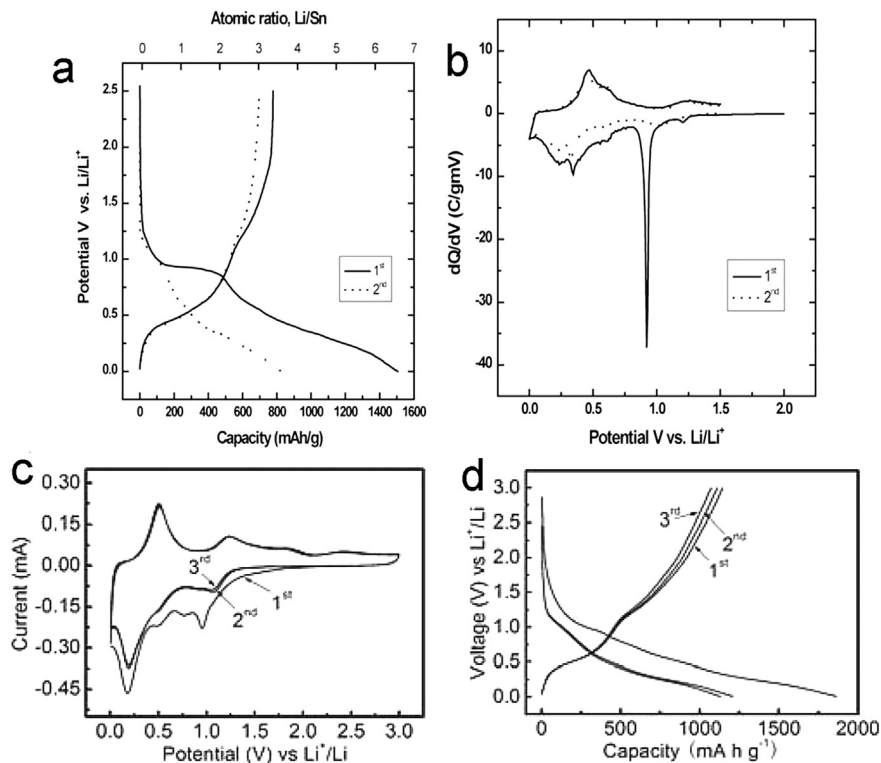


Fig. 1. (a) and (b) The first and second charge/discharge curves and their derivative capacity plots of 10 μm SnO₂ [20]. Copyright 2009, Elsevier. And (c) and (d) CV curves and galvanostatic charge/discharge profiles of the first three cycles of the SnO₂ NC@N-RGO electrode [16]. Copyright 2013, Wiley-VCH.

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