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Original research

Enhanced electrochemical stability of carbon-coated antimony nanoparticles with sodium alginate binder for sodium-ion batteries[☆]Jianmin Feng^a, Liqun Wang^a, Dejun Li^{a,*}, Pengyi Lu^b, Feng Hou^b, Ji Liang^{c,*}^a Energy and Materials Engineering Center, Institute of Physics and Materials Science, Tianjin Normal University, No 393 Bin Shui West Road, Xiqing District, Tianjin 300387, China^b Key Laboratory of Advanced Ceramics and Machining Technology of Ministry of Education, School of Materials Science and Engineering, Tianjin University, Tianjin 300072, China^c Institute for Superconducting & Electronic Materials (ISEM), Australian Institute of Innovative Materials (AIIM), University of Wollongong, Innovation Campus, Squires way, NSW 2522, Australia

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ABSTRACT

The poor cycling stability of antimony during a repeated sodium ion insertion and desorption process is the key issue, which leads to an unsatisfactory application as an anode material in a sodium-ion battery. Addressed at this, we report a facile two-step method to coat antimony nanoparticles with an ultrathin carbon layer of few nanometers (denoted Sb@C NPs) for sodium-ion battery anode application. This carbon layer could buffer the volume change of antimony in the charge-discharge process and improve the battery cycle performance. Meanwhile, this carbon coating could also enhance the interfacial stability by firmly connecting the sodium alginate binders through its oxygen-rich surface. Benefitted from these advantages, an improved initial discharge capacity (788.5 mA h g⁻¹) and cycling stability capacity (553 mA h g⁻¹ after 50 times cycle) have been obtained in a battery using Sb@C NPs as anode materials at 50 mA g⁻¹.

1. Introduction

Sodium-ion batteries (SIBs) have attracted a rapidly increasing amount of interests recently, which has been put forward as a promising candidate to replace the currently state-of-the-art lithium-ion batteries (LIBs) as the next-generation energy storage system for the various renewable energy resources. Compared with LIBs, the energy storage mechanism of a SIB depends on the intercalation/deintercalation of Na ions in the electrode materials, which is similar to that of LIBs. However, the much larger size of Na ions (0.102 nm compared to 0.076 nm of Li ions) will not only lead to higher impedance of the intercalation/deintercalation process but also result in much larger volume change. Aiming at these issues, to explore electrode materials that are suitable for SIBs has become one of the major challenges in the development of SIBs. Different kinds of electrode materials have been developed, including carbon-based materials [1–3], titanium-based compounds [4–7], alloys of tin [8–11], antimony [12–16], phosphorus [17–19], organic compounds [20–22], and others. Theoretically, most of these materials have high sodium-ion storage capacity, however, they have one common obstacle, that is, the large volumetric change of the electrode materials along with the prolonged charge-discharge

process. This will deteriorate the cycle performance of SIBs from two main aspects, including the collapse of the active material particles and the detachment of the active material from the binders or conductive additives (e.g. carbon black or other substrates).

To address these challenges, a very effective strategy is to apply a secondary buffer phase/coating onto the active materials particles [23–26]. Carbon coating of materials could not only buffer the large volume variation of the materials during cycling, but also prevent the electrode materials from agglomerating and enhance the electronic conductivity, which have been used for LIBs. Now, carbon-based materials have also been one main popular secondary buffering material for SIBs, including carbon nanotube, graphene, Super P carbon, and porous carbon derived from metal organic frameworks [19,27–29].

Inspired by the previous works, the strategy of compositing active materials with high Na ion capacity and carbon coating will be promising to develop high performance SIB anode. Antimony is one of promising SIB anode materials, as it has high theoretical capacity of 660 mA h g⁻¹ and suitable discharge/charge platform (about 0.5–0.8 V vs. Na⁺/Na). Unavoidably, antimony-based anode also suffers from large volume change accompanying with sodiation-desodiation and leads to rapid failure of batteries. Recently, carbon compositing strategy

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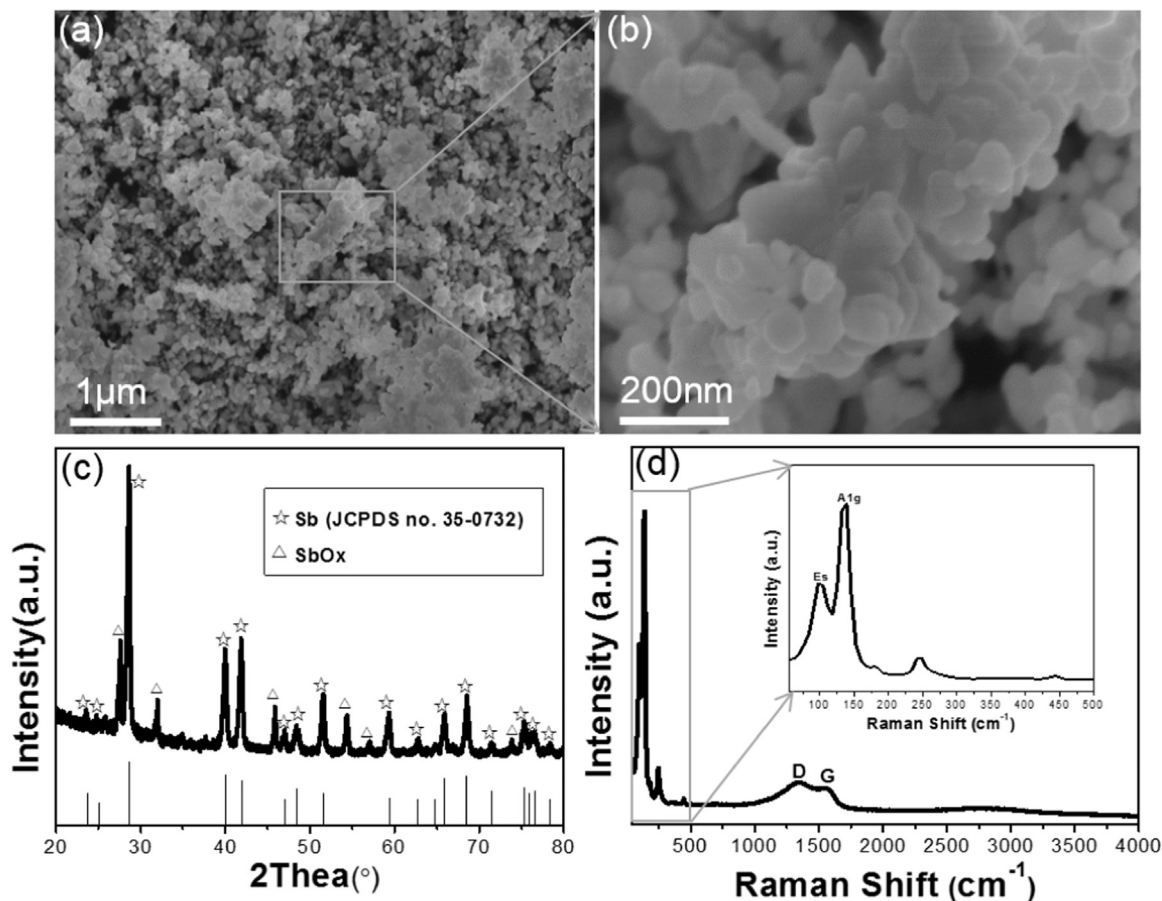


Fig. 1. Sb@C NP SEM images (a and b), XRD pattern (c), and Raman spectrum (d).

has been adopted to improve the cycle performance of antimony-based anode materials. Different kinds of carbon/antimony structures have been prepared and explored as SIB anode materials, such as antimony/nitrogen-doping porous carbon [30,31] and antimony/graphene [32,33]. However, in these materials, the content of carbon-based buffer materials is high, sometimes even over 50 wt%. Consequently, the Na ion storage capacity of these carbon-based buffer materials is often much lower than that is expected on antimony-based materials. In addition, the reported preparation process of such carbon-based buffer matrix is often quite complex with multiple steps. Hence, to facilitate the preparation of a thinner carbon-based buffer layer on the antimony-based materials not only will be useful to improve their capacity but also will be positive to reduce cost.

With such a purpose, antimony nanoparticles with nanometer-thick carbon coating layer (Sb@C NPs) was prepared through a facile two-step method of liquid-phase reduction from SbCl₃ and following hydrothermal carbon-coating using glucose as carbon precursor. The carbon coating layer is beneficial to the interface binding between the sodium alginate binder and the active materials, which has enhanced the electrode's cycle performance. The carbon content is only 1.5 wt%, which is much smaller than the values of previous works and thus does not significantly compromise the capacity of the materials. Sodium alginate has been utilized as the binder, which effectively improved the battery performance of the Sb@C NPs to achieve a higher capacity and better stability of 533 mA h g⁻¹ at a current of 50 mA g⁻¹.

2. Experimental section

The preparation of Sb NPs was conducted in a three-neck flask heated in an oil bath. In a typical synthesis, 45 mL of Diethylene glycol was added in the flask, then 0.7 g of Polyvinylpyrrolidone (PVP) (MW

= 40,000) was added and dissolved under mechanical stirring. The solution was heated up to 180 °C, after which, 10 mmol of SbCl₃ was added. A freshly prepared solution of NaBH₄ (0.872 g in 8 mL of diethylene glycol) was then injected into the solution with stirring at 0.5 mL min⁻¹ by a syringe pump.

The carbon coating process was carried out through a hydrothermal carbon-coating process. 0.4 g Sb NPs was dispersed in 40 mL aqueous solution containing 0.0125 M glucose, under sonication. Then the solution was transferred into a 50 mL Teflon-lined stainless-steel autoclave, sealed, maintained at 180 °C for 12 h, and cooled down naturally. The resulted brown solution was centrifuged and the brown solid product was collected, washed with distilled water and ethanol, and then freeze dried for 72 h. Finally, the dry brown product was put in a tube furnace and kept at 500 °C for 3 h in a 200 sccm argon flow to carbonize the carbon coating.

The morphology and structure of Sb@C NPs was characterized by a scanning electron microscopy (SEM, TDCLS-8010, Hitachi Japan), transmission electron microscopy (TEM, Tecnai G2 F20, FEI, Holland), X-ray diffraction (XRD, D8 Advance, Bruker, German), Raman spectroscopy (Horiba Jobin Yvon, LabRAM HR800, 17 mW, 514 nm, He-Ne laser), and X-ray photoelectronic spectroscopy (XPS, Perkin-Elmer, PHI-1600), respectively. The carbon content of the products was evaluated by thermal gravimetric analysis and differential scanning calorimeter (TGA and DSC, TGA/DSC1/1100, METTLER, Germany) in an air flow (15 sccm) at a heating rate of 10 °C min⁻¹ using 15 mg samples.

The as-prepared Sb@C NPs were used as anode electrode materials and assembled into SIBs for electrochemical measurements. Specifically, the Sb@C NPs were mixed with acetylene black powders (conducting additives) and sodium alginate (binders) at a mass ratio of 85:10:5 for these three components. The mixture was then dipped in N-methyl-2-pyrrolidone and water, then mechanically grinded to form a

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