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A Novel Fabrication for Manganese Monoxide/Reduced Graphene Oxide Nanocomposite as High Performance Anode of Lithium Ion Battery

P. Xia^{a,1}, H.B. Lin^{a,1}, W.Q. Tu^a, X.Q. Chen^a, X. Cai^a, X.W. Zheng^a, M.Q. Xu^{a,b}, W.S. Li^{a,b,*}

^a School of Chemistry and Environment, South China Normal University, Guangzhou 510006, China
^b Engineering Research Center of MTEES (Ministry of Education), Research Center of BMET (Guangdong Province), Engineering Lab. of OFMHEB (Guangdong Province), Key Lab. of ETESPG(GHEI), and Innovative Platform for ITBMD (Guangzhou Municipality), South China Normal University, Guangzhou 510006, China

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1. Introduction

Lithium ion battery has received intensive attention due to the need of high energy density power sources for large-scale applications such as in electric vehicles [1-3]. One of the interests is to find alternatives for currently commercial graphite, which has a theoretical specific capacity of only 372 mAh g⁻¹ [4]. Transition metal oxides are the potential candidates because of their high theoretical specific capacity (over 1000 mAh g⁻¹), environmental friendliness, free of lithium dendrite and low cost [5–7]. Among various transition metal oxides, manganese monoxide (MnO) is most attractive due to the abundance of manganese source and the low operating potential (about 0.5V and 1.2V vs. Li/Li⁺ for discharge and charge, respectively) [8–11]. Unfortunately, manganese oxides have poor electronic conductivity and drastic volume change during lithium intercalation and deintercalation process, leading to its poor performances as anode of lithium ion battery in terms of rate capability and cyclic stability [12-14].

¹ These authors contributed equally to this work.

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ABSTRACT

In this paper, we propose a novel fabrication for manganese monoxide and reduced graphene oxide nanocomposite (MnO/rGO), in which microemulsion is introduced to form a 3D architecture consisting of MnO nanoparticles embedded in reduced graphene oxide network. Physical characterizations from SEM, TEM, HRTEM, XPS, Raman, and XRD, indicate that MnO particles of about 230 nm are formed and uniformly embedded in rGO. Charge and discharge tests demonstrate that the resulting MnO/rGO exhibits excellent performances as anode of lithium ion battery, delivering a reversible capacity of as high as 776 mAh g⁻¹ at 1000 mA g⁻¹ after 155 cycles and rate capacity of 306 mAh g⁻¹ at 6000 mA g⁻¹ when it is evaluated in a half cell with lithium as the counter electrode.

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Much effort has been made to solve these problems in the past decade. The most widely adopted approach is to synthesize nanosized MnO. Nanoparticles provide larger surface area for lithium intercalation and deintercalation and shorten the transport path of lithium ion, leading to the improved rate capability of the oxide, compared to microparticles [15,16]. However, this approach is detrimental to the cyclic stability of the oxide because nanoparticles increase the contact area between the oxide and electrolyte, which accelerates the decomposition of electrolyte and the structural destruction of the oxide.

Recently, much attention has been paid to the fabrication for the composites of MnO with conductive carbon materials [13,17,18]. The carbon not only provides electronically conductive network but also acts as a buffer for the volume change during lithium intercalation and deintercalation, effectively improving cyclic stability and rate capability of the oxide. Graphene, a two-dimensional carbon sheet with honeycomb structure, is one of most attractive carbon materials for fabricating MnO/carbon composite, due to its superior conductivity, structure flexibility, large surface area, and chemical stability [19–21]. However, currently available fabrication methods of MnO/graphene are complicate. For example, hydrazine hydrate was needed to reduce graphene oxide [9]; MnO₂ was used as the precursor of MnO [22]; two steps combined with hydrothermal method and heat treatment were needed to synthesize the hybrid [18].







^{*} Corresponding author at: School of Chemistry and Environment, South China Normal University, Guangzhou 510006, China. Tel.: +86 20 39310256; fax: +86 20 39310256.

E-mail address: liwsh@scnu.edu.cn (W.S. Li).

In this work, we proposed a novel fabrication for MnO/graphene nanocomposite, in which microemulsion was introduced with MnCO₃ and graphene oxide (GO) as precursors of MnO and graphene, respectively. The microemulsion ensures the formation and the uniform embedding of MnO nanoparticles that are formed simply from thermal decomposition of MnCO₃, onto the reduced graphene oxide (rGO) that is formed under hydrogen atmosphere. Charge/discharge tests demonstrate that this fabrication yields excellent cyclic stability and rate capability of the resulting composite (MnO/rGO), compared with those that have been reported in literature.

2. Experimental

2.1. Preparation

MnO/rGO was synthesized by a simple precipitation reaction in microemulsion and a following thermal treatment in 10% H₂/Ar atmosphere. MnCO₃/GO precursor was synthesized in a microemulsion. Typically, 4 g cetyltrimethyl ammonium bromide

(CTAB) was dissolved in a mixture of 100 mL cyclohexane, 5 mL nbutyl alcohol and 5 mL 0.8 M NH₄HCO₃ aqueous solution. The mixture was stirred until it became transparent. Quantitative graphene oxide, which was obtained from graphite flakes (Nanjing JCNANO Technology Co., Ltd.) by modified Hummers' method [23], was uniformly dispersed in the mixture under ultrasonication for 30 min, then 5 mL 0.4 M MnSO₄ was dropped into the continuously stirred solution to produce a milky solution. MnCO₃/GO was obtained by filtering, washing several times with ethanol and distilled water and drying in a vacuum at 80 °C. The resulting MnCO₃/GO was sintered at 650 °C for 10 h with a rate of 3 °C min⁻¹ under 10% H₂/Ar dynamic atmosphere to get the final product MnO/rGO. MnO without graphene was also synthesized under the same conditions for a comparison.

2.2. Characterization

The morphology of the samples was analyzed by scanning electron microscopy (SEM, JEOL JSM-6380), transmission electron microscopy (TEM, JEOL JEM-2100HR) and high-resolution TEM



Fig. 1. Schematical formation process of MnO/rGO nanocomposite.

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