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Manganese Dioxide/Cabon Nanotubes Composite with Optimized Microstructure via Room Temperature Solution Approach for High Performance Lithium-Ion Battery Anodes

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ABSTRACT

Manganese dioxide/cabon nanotubes (MnO_2/CNTs) composites with tunable density of MnO_2 nanosheets on the surface of CNTs were synthesized by a facile room temperature solution method. When applied in lithium-ion batteries (LIBs) as anode materials, the spacial density of MnO_2 nanosheets in the MnO_2/CNT composite was verified to be crucial for the lithium storage performance. The optimized composite with moderate spatial density of MnO_2 nanosheets delivered a reversible capacity of 903 mA h g^{-1} at the current rate of 0.24 A g^{-1}. Moreover, this composite exhibited a high stable capacity of 540 mA h g^{-1} even at a high current density of 2.4 A g^{-1} after 1500 cycles, demonstrating its potential for applications in LIBs with long cycling life and high power density. The enhanced electrochemical performance of the optimized composite was ascribed to sufficient space between the MnO_2 nanosheets on the CNTs, which not only allows the effective electrical contact between the CNT backbones and the conductive carbon but also accommodates the large volume changes upon repeated lithiation/delithiation.

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1. Introduction

Metal oxides that could reversibly react with lithium through multiple-electron transfer conversion reaction are considered promising high capacity anodes for next generation lithium-ion batteries [1–4]. Among the various metal oxides, binary manganese oxides are a group of versatile functional compounds for battery application due to their structural diversity, high abundance, and environmental friendliness [5–10]. Manganese dioxide (MnO_2), as one of the most stable forms of all manganese oxides compounds with high theoretical lithium storage capacity, has been widely used in primary alkaline batteries [11,12]. However, despite its successful application in primary batteries, the research on employing MnO_2 as a conversion-type anode material for lithium-ion battery remains a challenge [13,14].

Extensive earlier researches have been conducted to investigate the electrochemical performances of MnO_2, typically by engineering different nanostructures, such as nanowires, hollow urchins, and hollow spheres [15–19]. In general, these materials showed high initial discharge capacity, but poor rate capabilities and cycle lifespans were found. It was concluded that, like many other conversion type anodes, the electrochemical performances of MnO_2 were hindered by its intrinsic low electrical conductivity and large volume change accompanying the conversion reaction [20–22]. To improve the rate capability and cycle lifespan of MnO_2, Recent studies have been devoted to constructing composited microstructures of MnO_2 and conductive agents [23–28]. As a promising conductive additive, carbon nanotubes (CNTs) were used to incorporate with MnO_2 for enhancing its electrochemical performances [29–31]. Ajayan et al. reported a hard template method to synthesize coaxial MnO_2/CNT composite, which has a discharge capacity of ~500 mA h g^{-1} after 15 cycles [29]. Lu et al. prepared nanoflaky MnO_2/CNT composite through hydrothermal process; the discharge capacity remains about 600 mA h g^{-1} after 50 cycles at a current rate of 200 mA g^{-1} [30]. While the reported studies suggested that the poor stability of MnO_2 can be improved by incorporating CNT to form MnO_2/CNT composites, several

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issues have to be addressed to enable their wide commercial applications. For example, so far all reports on the MnO_2/CNT anode use relatively low current densities (e.g., below 500 mA g^{-1}) and limited cycling numbers (<100 cycles) in the charge-discharge measurements. Apparently, operation stability and high rating performance are two critical issues needing improvement. Furthermore, the electrochemical performances of composited electrode materials are closely reliant to their composition and microstructure [32,33]. Therefore, developing a facile method to tune the composition of MnO_2/CNT composite is also necessary for optimizing its performances.

In this work, we report a facile room temperature approach for preparing MnO_2/CNT composites with tunable composition. The synthesis process involves a redox reaction of divalent manganese salt with ammonium perfulfate on the surface of CNTs. Ethylene glycol (EG) was used as a dispersant and cosolvent to maintain the in-situ growth of MnO_2 nanosheets on CNTs. By regulating the concentration of the reaction agent, the spatial density of the MnO_2 nanosheets can be controlled. When applied in LIBs, the electrochemical performance of the composite was found to depend strongly on the spacial density of MnO_2 nanosheets, confirming the importance of composition adjustment. A optimized MnO_2/CNT sample was found to show a capacity of 903 mA h g^{-1} at the current rate of 0.24 A g^{-1}, and a capacity of ~540 mA h g^{-1} after 1500 cycles at a high current density of 2.4 A g^{-1}.

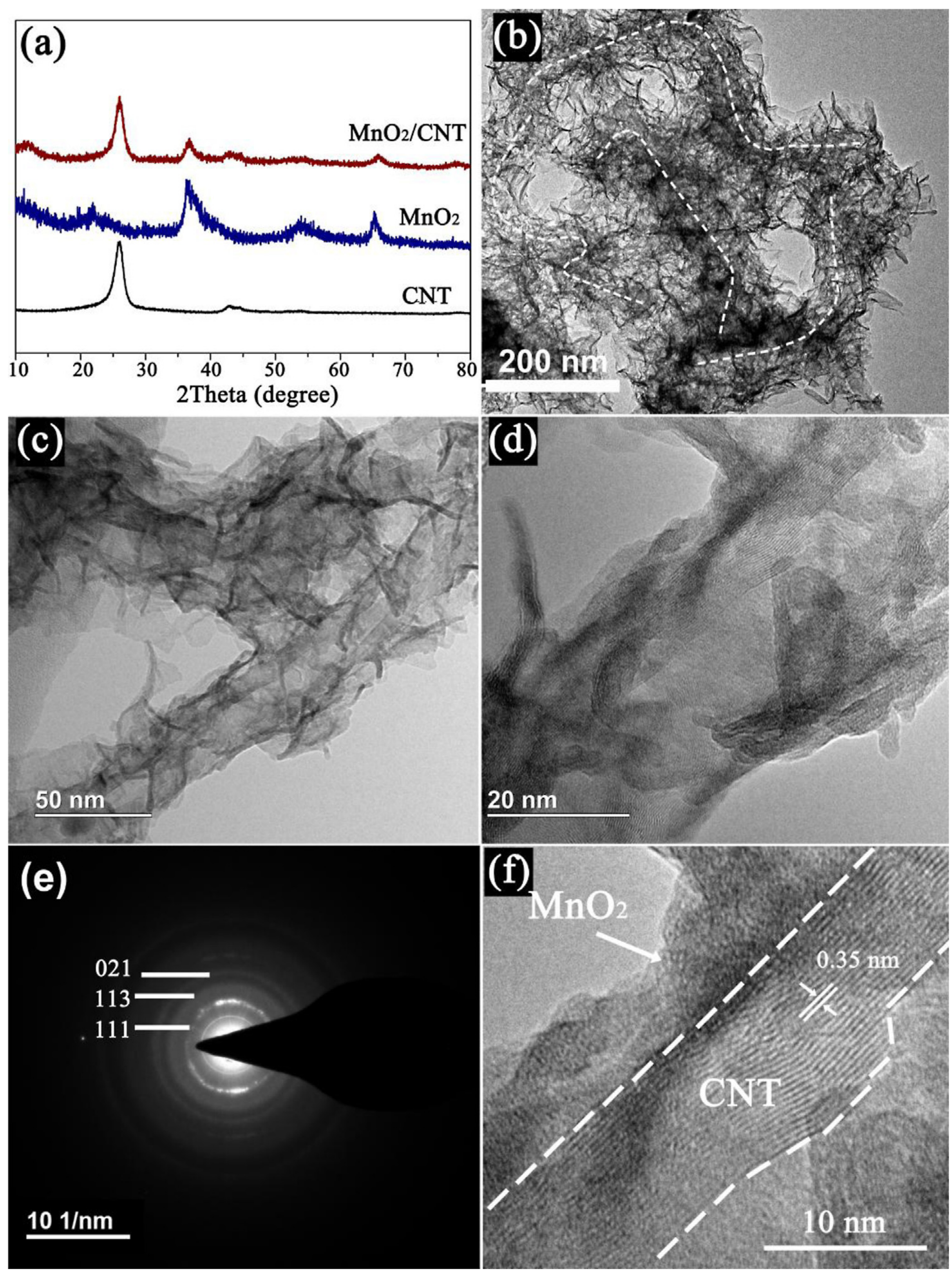

Fig. 1. a) XRD patterns of CNTs (black line), MnO_2 nanosheets (blue line), and MnO_2/CNT composite (red line). b,c,d) TEM images of MnO_2/CNT composite. e) Electron diffraction pattern of the MnO_2 nanosheets in the composite. f) High resolution TEM image of the MnO_2 nanosheets on the surface of CNT. The MnO_2/CNT composite here corresponding to MnO_2/CNT-2 sample. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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