

Full Length Article

In-situ grown CNTs modified SiO₂/C composites as anode with improved cycling stability and rate capability for lithium storage



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ARTICLE INFO

Article history:

Received 8 May 2017

Received in revised form

29 September 2017

Accepted 9 October 2017

Available online 16 October 2017

Keywords:

Lithium ion batteries

In-situ synthesis

Chemical vapor deposition

Carbon nanotubes

SiO₂/C/CNTs composites

ABSTRACT

Silica (SiO₂) is regarded as one of the most promising anode materials for lithium ion batteries owing to its high theoretical specific capacity, relatively low operation potentials, abundance, environmental benignity and low cost. However, the low intrinsic electrical conductivity and large volume change of SiO₂ during the discharge/charge cycles usually results in poor electrochemical performance. In this work, carbon nanotubes (CNTs) modified SiO₂/C composites have been fabricated through an in-situ chemical vapor deposition method. The results show that the electrical conductivity of the SiO₂/C/CNTs is visibly enhanced through a robust connection between the CNTs and SiO₂/C particles. Compared with the pristine SiO₂ and SiO₂/C composites, the SiO₂/C/CNTs composites display a high initial capacity of 1267.2 mA h g⁻¹. Besides, an excellent cycling stability with the capacity of 315.7 mA h g⁻¹ is achieved after 1000th cycles at a rate of 1 A g⁻¹. The significantly improved electrochemical properties of the SiO₂/C/CNTs composites are mainly attributed to the formation of three dimensional CNT networks in the SiO₂/C substrate, which can not only shorten the Li-ion diffusion path but also relieve the volume change during the lithium-ion insertion/extraction processes.

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1. Introduction

As one of the most promising secondary batteries, lithium ion batteries (LIBs) have been widely applied in many fields, such as portable electronic devices, electric vehicles, hybrid electric vehicles, and stationary storage, on account of their high energy density, long cycle life, no memory effect, and environmental benignity [1,2]. To meet the ever-increasing demand for the high energy density and long cycle life LIBs, developing new anode materials for various technological applications is urgently needed [3–5]. Compared with the traditional commercial graphite anode (372 mA h g⁻¹), silicon-based materials have been extensively exploited in recent years, due to their high theoretical capacity (4200 mA h g⁻¹) and safe operation voltage above lithium [6,7]. However, the large volume change (400%) during repeated insertion/extraction cycling processes and the complicated and time-consuming processes for the preparation of Si have severely hampered its widespread application [8–10]. Thus, it is crucial to

develop a new and well-designed anode material with high electrochemical performance.

Recently, similar to the Si materials, silica (SiO₂) has also been considered as a prospective high-energy anode material, because of its high theoretical specific capacity (1965 mA h g⁻¹) and a low plateau potential [11,12]. Of particular importance, as one of the most abundant materials in the Earth, SiO₂ is extremely low cost and low toxicity. Besides, it can be synthesized easier than Si [13]. Unfortunately, SiO₂ also suffers a large volume change in the processes of discharge/charge, resulting in severe pulverization and rapid capacity fading of the electrode [14]. In addition, SiO₂ exhibits a poor intrinsic electrical conductivity, which caused a poor rate performance [15]. Hence, to circumvent these problems, various nano structures, such as thin film [16], nanospheres [17], hollow nanocubes [18], nanobelts [19], nanotubes [20] have been designed to restrain the volume expansion meanwhile guaranteeing the rapid lithium ion transfer. As a consequence, their cycling stability has been significantly improved when compared with the bulk SiO₂ [16–20]. Moreover, to solve the issue of low rate capability, numerous SiO₂/carbon nanocomposites have been fabricated by template method, ball milling process, sol-gel approach and hydrothermal reaction [21–26]. Especially, integrating the SiO₂

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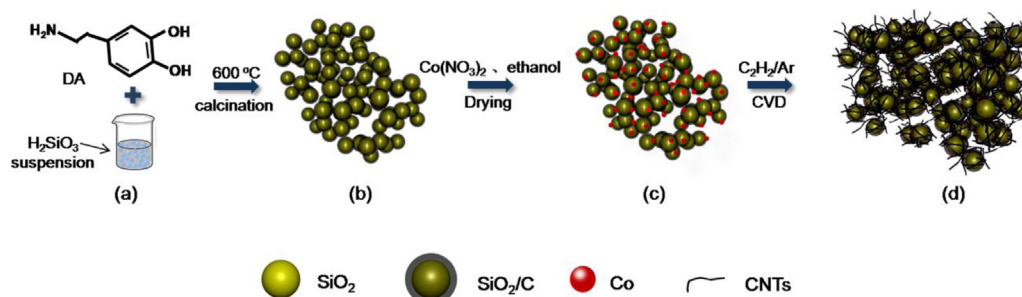


Fig. 1. Schematic illustration of the procedure for in situ catalytic synthesized SiO₂/C/CNTs composites: (a) mixture of dopamine and H₂SiO₃ suspension, (b) SiO₂/C composites, (c) Co catalyst on SiO₂/C composites, and (d) SiO₂/C/CNTs composites.

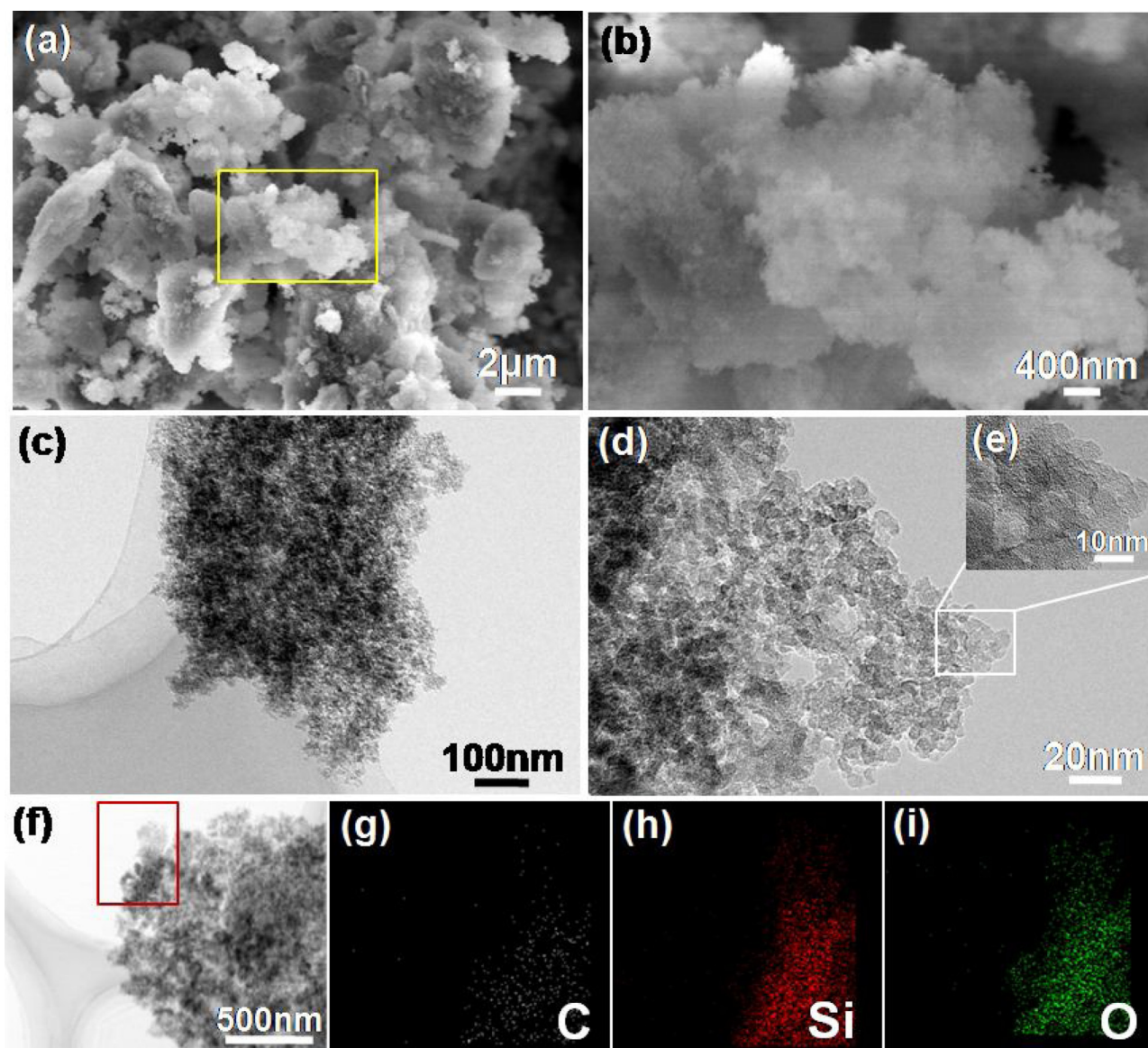


Fig. 2. (a) SEM image of the SiO₂/C composite, (b) magnified image of (a); (c, d) TEM images of the SiO₂/C composites, (d) is the magnified image of (c), and the inset (e) shows the average size of the SiO₂/C is approximately 5 nm; (f) TEM image for subsequent mapping of elements (red rectangle), (g), (h), and (i) EDX mapping distributions of C, Si, and O in the SiO₂/C composite. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

with carbon materials could not only improve the conductivity of SiO₂ but also buffer its volume changes [27]. Yuan et al. [28] prepared the SiO₂/C composites through a facile chemical synthesis method. The results showed that the composites had an excellent

electrochemical performance, due to the fact that the carbon materials could act as an electrical conducting pathway and mitigate the volume expansion. Therefore, the engineering of nanostructure and carbonaceous materials incorporated with SiO₂ can be

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