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Structure and electrochemical performance of graphene/porous carbon coated carbon nanotube composite for supercapacitors



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ABSTRACT

A composite of graphene/porous carbon coated carbon nanotube (denoted as RGO/CNT@AC) is synthesized. Microstructural characterization of the RGO/CNT@AC composite shows that the CNTs are uniformly coated by activated porous carbons, and the porous carbon coated CNTs are inserted among graphene sheets. Electrochemical investigations indicate that the specific capacitance of the RGO/CNT@AC composite is up to 147 F g^{-1} at 10 mV s^{-1} , an 56% improvement compared with that of graphene/pure CNT (denoted as RGO/CNT) composite. Furthermore, the cyclic voltammogram curves of the RGO/CNT@AC composite can keep a rectangular-like shape even at a scan rate of 5000 mV s^{-1} , showing a significantly better rate capability. The improvement in supercapacitive performance of the RGO/CNT@AC composite may be attributable to the contribution of porous carbon to specific capacitance, the effective inhibition the restacking of graphene sheets and the construction of more mutually connected electrolyte ions transport paths by the shortened porous carbon coated CNTs among graphene sheets. The superior specific capacitance and rate performance imply that the RGO/CNT@AC composite is promising for supercapacitors.

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1. Introduction

Due to its high specific surface area (SSA), high electron mobility and other excellent properties, graphene has shown great promising in supercapacitors [1–3]. However, the graphene sheets are easy to restack because of the strong van der Waals interactions between individual graphene sheets, leading to the decreasing of available surface in the electrode, as a result, a poorer supercapacitive performance [4].

Many works have proved that the introduction of carbon nanotubes (CNTs) among the graphene sheets can effectively impede the restacking of individual graphene sheets and facilitate the achievement of superior supercapacitive performance [5,6]. Unfortunately, due to the low SSA and undeveloped pore structure [7], the CNT introduced in graphene sheets has little contribution to the supercapacitor performance.

Therefore, we propose to introduce the activated porous carbon coated carbon nanotube (denoted as CNT@AC) instead of pure CNT among the graphene sheets, constructing a RGO/CNT@AC composite. Due to the high SSA and developed pore structure of activated porous carbon (AC), as well as the effective prevention on

the restacking of graphene layers by CNT@AC, the RGO/CNT@AC composite can be expected to exhibit superior supercapacitive performance.

2. Experimental

2.1. Sample preparation

Graphene oxide (GO) powders were prepared by a modified Hummers' method, which is detailedly described in the supporting information.

0.1 g nitric acid-treated CNT (20~40 nm in diameter, obtained from the Shenzhen Nanotech Co. Ltd. of China) with 2.5 g glucose was dissolved into 100 ml deionized water and ultrasonically dispersed for 3 h. Then, the resultant mixture was put into Teflon-sealed autoclave with a fill rate of 80%. After 15 h hydrothermal treatment at $190 \text{ }^\circ\text{C}$, the produced black powders were further carbonized in a tube furnace at $800 \text{ }^\circ\text{C}$ in argon atmosphere. After carbonization, the products were mixed with KOH with a weight ratio of 1:4 and activated at $800 \text{ }^\circ\text{C}$ for 1 h in argon atmosphere. The obtained sample was immersed in 10 wt% HCl solution, then washed to neutral with deionized water and dried at $100 \text{ }^\circ\text{C}$ for 12 h, thereby achieving the CNT@AC composite.

0.08 g GO powders was added into 50 ml deionized water and

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dispersed by ultrasonication for 1 h. Then, 0.06 g CNT@AC was added and ultrasonically dispersed for 2 h. Thereafter, 5 ml hydrazine hydrate was added to reduce the GO to the reduced graphene oxide (RGO). The mixture was stirred at room temperature over night, washed by deionized water until to neutral, and then dried at 60 °C for 12 h, eventually preparing the RGO/CNT@AC composite.

The RGO/CNT composite was also prepared by mixing the pristine CNT (without AC coating) and the RGO in the same way. For comparison, the CNTs with equal quantity were added in the RGO/CNT and the RGO/CNT@AC composites.

2.2. Characterization

The morphologies of the samples were observed using a Hitachi S-4800 field emission scanning-electron microscopy (SEM) at 15 kV. X-ray diffraction (XRD) patterns between 10 (2θ) and 60 (2θ) degrees were collected by Rigaku D/MAX-2500 powder diffractometer with Cu-K α radiation ($\lambda=0.154$ nm) operated at 40 kV and 200 mA. In addition, TEM observations and Gas adsorption/desorption test were also conducted, which are detailedly described in the supporting information.

2.3. Preparation of electrode and electrochemical measurement

The electrochemical investigations were carried out in a symmetrical two-electrode cell with a cellulose separator. The working electrodes were prepared as follows [8]: A slurry consisting of 80 wt% active substance, 10 wt% carbon black and 10 wt% polytetrafluoroethylene (PTFE) binder was smeared into nickel foam and dried in vacuum at 120 °C for 10 h. The active area of the electrode is 1 cm². A 6 M KOH solution was used as electrolyte. Cyclic voltammograms (CVs) were collected on CHI650D electrochemical workstation in a potential range of 0~1 V. The scan speeds for all CV tests were in the range of 10~5000 mV s⁻¹.

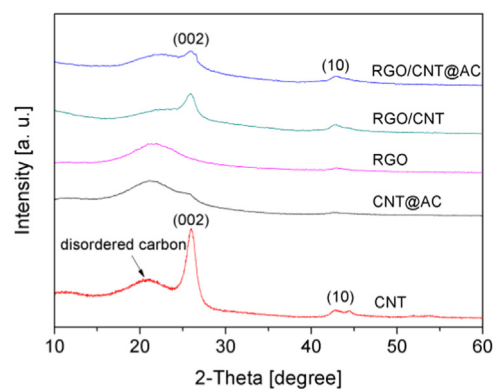


Fig. 2. XRD patterns of CNT, CNT@AC, RGO, RGO/CNT, and RGO/CNT@AC.

3. Results and discussion

Representative SEM images of CNT, CNT@AC, RGO/CNT and RGO/CNT@AC samples are shown in Fig. 1. It can be deduced from Fig. 1a and b that the AC is uniformly coated on the CNT surface, suggesting the achievement of the CNT@AC composite. An approximately 100 nm in diameter of CNT@AC indicates that the coating thickness of AC on the CNT is about 30 nm. And the structure of AC is mainly composed of disordered carbons (Fig. S1). In Fig. 1c and d, it can be observed that CNTs and CNT@ACs have been inserted among graphene sheets, indicating the successful preparation of RGO/CNT and RGO/CNT@AC composites. Obviously, the introduced CNT and CNT@AC can effectively prevent the restacking of graphene layers.

Fig. 2 shows the XRD patterns of CNT, CNT@AC, RGO, RGO/CNT and RGO/CNT@AC samples. A strong peak at $2\theta=26^\circ$ (002), two weak peaks at $2\theta=43^\circ$ and 44.5° (10), and a broad peak at $2\theta=22^\circ$, respectively, can be observed in the CNT diffraction pattern (Fig. 2). Comparatively, these peaks in the CNT@AC diffraction pattern become significantly weak, implying successful coating of activated porous carbon on the CNT surface. In addition, the XRD

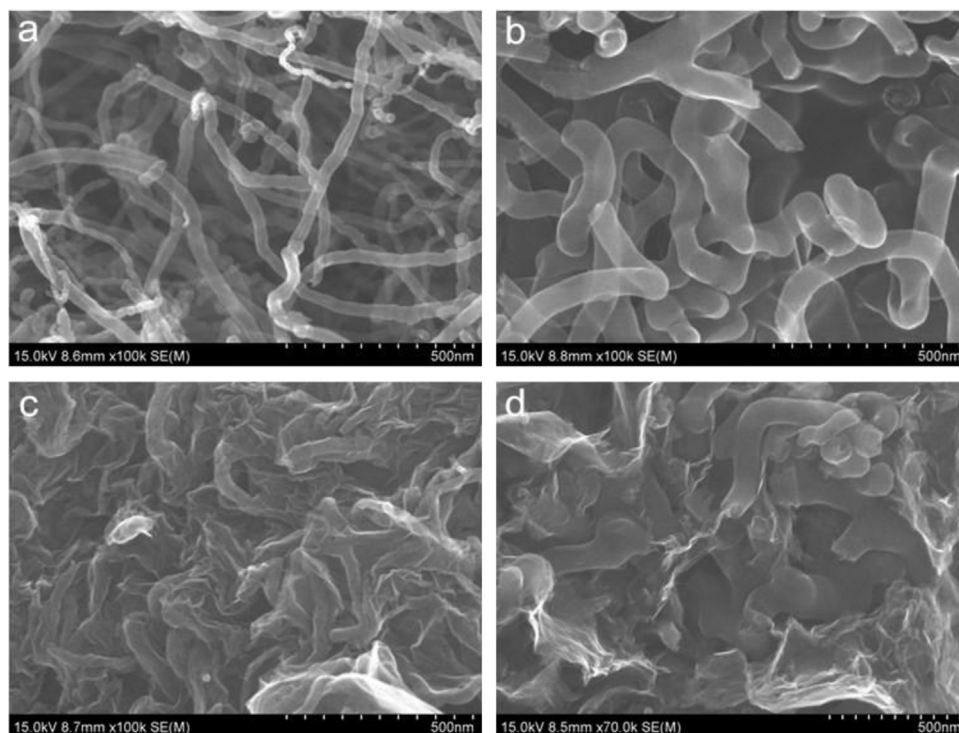


Fig. 1. Representative SEM images: (a) CNT; (b) CNT@AC; (c) RGO/CNT and (d) RGO/CNT@AC.

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