



Preparation and electrochemical investigation of polyaniline nanowires for high performance supercapacitor

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ABSTRACT

A high performance polyaniline (PANI) nanowires electrode was prepared by electrodeposition of aniline on porous tubular carbon (PTC). The resulting composites PTC/PANI with a vine-like shape exhibited remarkable electrochemical performance, including a high specific capacitance (477.7 F g^{-1} at 1 A g^{-1}) and an excellent cycling stability (91% capability retention after 5000 cycles). Such excellent performance was attributed to the synergistic effects between PTC and PANI. PTC with well-defined macropores and interconnected meso-/micropores structure can facilitate the transfer of ions and electrons and can also effectively avoid PANI agglomeration; the nanostructured PANI can provide pseudocapacitance. Moreover, the good contact between PANI and PTC, which was constructed by electrodeposition, ensures a low contact resistance and reinforces structural stability of the composite electrode.

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1. Introduction

Given the increasing energy storage requirements of the electrically driven modern world, supercapacitors have attracted increasing attention [1,2]. Polyaniline (PANI) has been considered as a promising supercapacitor electrode material because of its low cost, easy of synthesis, redox reversibility, and high specific pseudocapacitance associated with multiple redox state [3,4]. However, the swelling/shrinkage of PANI during the insertion/extraction process of ions leads to the structural destruction and instability, which greatly reduce the reversibility and cycle life of PANI-based electrodes. Moreover, the self-agglomeration and underutilization of PANI also hinder its practical application [5]. To alleviate these problems, many researchers have combined PANI with some substrates. Compared with other chemical techniques, electrodeposition to synthesis PANI is more simple and effective to control the morphology of PANI. Zhang et al. [7] have synthesized PANI nanowires using Ti plate as template by electrodeposition. PANI nanorod was also successfully arrayed on graphene paper by electropolymerization and the resulted graphene-PANI paper exhibited an excellent electrochemical performance [3]. Compared with the above used substrates, such as Ti plate and graphene paper, porous tubular carbon (PTC) with a larger specific surface area would be an ideal substrate to effectively avoid PANI agglomeration.

In this work, the PANI nanowires were successfully electrodeposited on tubular-liked PTC. With well-defined macropores and interconnected meso-/micropores structure, PTC can facilitate the transfer of ions/electrons and effectively avoid PANI agglomeration. By electrodeposition method, the structural stability of PANI can be reinforced and the PANI will have a high utilization rate. Therefore, the specific capacitance, rate performance, and cycle life of PANI-based electrode can be considerably improved.

2. Experimental

2.1. Preparation of PTC

PTC was prepared by the following strategies [8]: the coal tar pitch and sawdust were mixed at a mass ratio of 2:1 at $150 \text{ }^\circ\text{C}$. Subsequently, the mixture was pyrolyzed at $500 \text{ }^\circ\text{C}$ for 2 h under a N_2 atmosphere. The produced material was mixed with KOH at a mass ratio of 1:3. Then the samples were carbonized at $800 \text{ }^\circ\text{C}$ for 2 h. After washed with 1 M HCl and distilled water several times, the PTC was obtained.

2.2. Preparation of PTC/PANI electrodes

The electrodeposition of PANI was performed in a standard three-electrode system. The working electrode was prepared by mixing the PTC with acetylene black and polytetrafluoroethylene at a mass ratio of 8:1:1, and then coated and pressed on a nickel

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foam. The saturated calomel electrode and platinum plate were used as reference electrode and counter electrode, respectively. The electrolyte was an aqueous solution of 0.5 M H₂SO₄ and 0.05 M aniline. Electrodeposition was performed at a constant potential of 1 V. The deposition of aniline was kept running for 6, 9, 12, and 15 min and the corresponding mass of the PANI were 4.3, 4.5, 6.1, and 7.1 mg, respectively. The as-prepared samples were denoted as PTC/PANI-*x*, in which *x* refers to the deposition time (6, 9, 12, or 15).

2.3. Characterization methods

Field emission scanning electron microscopy (SEM) was performed using a JEOL JSM-7001F. X-ray photoelectron spectroscopy (XPS) measurement was performed on an ESCALAB 250. Fourier transform infrared (FTIR) spectra were recorded on a TENSOR 27 FTIR spectrometer. X-ray diffraction (XRD) patterns was recorded on D/max-2500. All the electrochemical measurements were performed in a standard three-electrode system using an electrochemical workstation (CHI660D) in 1 M H₂SO₄ electrolyte.

3. Results and discussion

The SEM image of PTC (Fig. 1a) shows that the PTC is composed of interconnected microtube bundle. The numerous microtube with a diameter $\sim 10\ \mu\text{m}$ can act as ion-buffering reservoir to minimize the diffusion of ions. Moreover, such unique structure would provide enough space for anchoring active materials. Fig. 1b–f show that the PANI nanowires grow on the substrate PTC in different degree. Only a small number of nanowires on the surface of PTC when the deposition time is 6 min (Fig. 1b), indicating the growth process of PANI has not completed fully. When the deposition time is 9 min (Fig. 1c and d), the nanowires of PANI distributed on the exterior and interior of PTC formed a multilevel network structure, which resemble the collection of “vine”. Such structure can provide much channels for electrons transport. The combina-

tion between the PANI and PTC caused by electrodeposition ensures the low contact resistance and mechanical stability [9]. With the increase of deposition time, the PANI nanowires become denser and coarser than before (Fig. 1e and f). The more PANI can provide more active sites but it also can block the porous structure of the PTC.

XRD pattern of PTC (Fig. 2a) shows two peaks at 24.1° and 44.6°, corresponding to (002) and (101) diffraction planes of PTC, respectively. As for PTC/PANI-9, the peaks at around 18.5° and 21.1° are assigned to the (020) and (200) crystalline planes of PANI [10], indicating the successful deposition of PANI. Other three peaks at 45.0°, 52.2°, and 76.7° can be ascribed to the diffraction of (111), (200), and (220) planes of the nickel foam, respectively [11].

Fig. 2b shows the FTIR spectra of PTC and PTC/PANI-9. A strong characteristic peak appeared at 3400 cm⁻¹ is corresponded to the stretching vibration of O–H or N–H bond [12]. In the spectrum of PTC, strong absorption bands at around 1642, 1395, and 1098 cm⁻¹ can be assigned to the skeletal vibration of C=C, C–O, and O–H deformation vibration, respectively. The peak at 1642 cm⁻¹ for PTC was shifted to a lower wavenumber 1620 cm⁻¹ for PTC/PANI-9, due to the C=C stretching vibration of the quinonoid ring. As for PTC/PANI-9, the band at 1307 cm⁻¹ is corresponded to the stretching vibration of C–N in a secondary aromatic amine. The band at 1148 cm⁻¹ is assigned to the aromatic C–H beading mode [6]. The N1s spectrum can be divided into three peaks at 399.3, 400.7, and 402.1 eV (Fig. 2c). The peaks located at 399.3 and 400.7 eV are associated with the quinoid imine (=N–) and benzenoid amine (–NH–), respectively. The peak at 402.1 eV is due to the interaction between N⁺ and protons introduced by the acid dopant [13].

In Fig. 3a, the CV curve of PTC exhibits a quasi-rectangular shape, indicating the characteristic of the double-layer capacitor, and the area of the curve is the smallest among all the curves. All of the CV curves for PTC/PANI-*x* have two couples of redox peaks, which originating from the redox reactions of PANI. The pair peaks

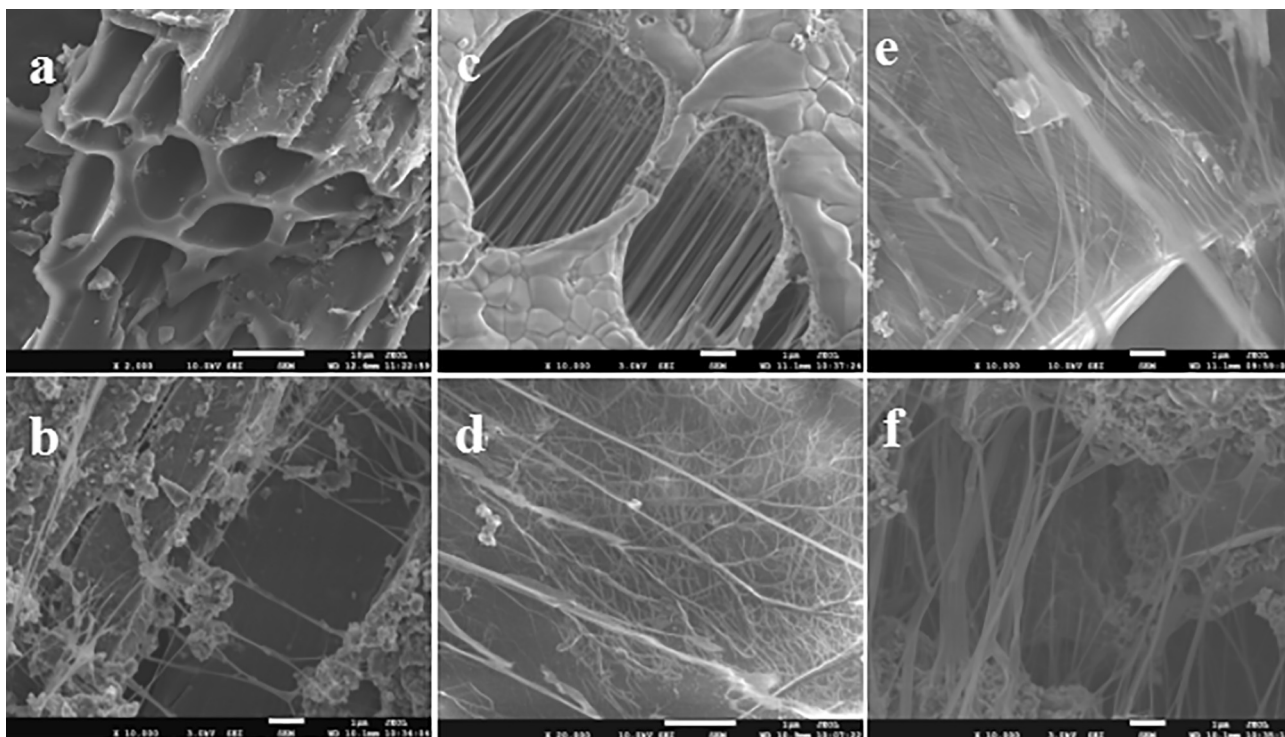


Fig. 1. SEM images of (a) PTC and PTC/PANI-*x* with different deposition time: (b) 6 min, (c, d) 9 min, (e) 12 min, and (f) 15 min.

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