

Counter electrodes from polyaniline–carbon nanotube complex/graphene oxide multilayers for dye-sensitized solar cell application



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ABSTRACT

With an aim of accelerating charge transport and increasing interface area for triiodide electroreduction reaction, positively charged polyaniline–single wall carbon nanotube PANi–SWCNT complex, chemically polymerized from aniline–single wall carbon nanotube complex, is combined with negatively charged graphene oxide (GO) by a layer-by-layer self-assembly technique. Covalent bond between PANi and SWCNT dramatically diminish the charges-transfer resistance within the complex, which is confirmed by electrical and electrochemical characterizations. The multilayer architecture of resultant (PANi–SWCNT/GO)_n (*n* represents bilayer number) counter electrode provides enormous interface for triiodide electrocatalysis. An impressive power conversion efficiency of 6.88% is recorded from the DSSC with (PANi–4 wt% SWCNT/GO)₅ multilayer counter electrode. These results are far from optimal but the preliminary photovoltaic performances make the strategy promising in efficient DSSC applications.

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1. Introduction

Nowadays, low-carbon economy has been attracting growing interests for the sustainable development of human society. However, fossil fuels such as coal, oil, and gas have occupied most proportion of energy market, leading to environmental destruction and energy exhaustion. Considering the merits on relatively low cost, easy fabrication, zero emission, and high efficiency, dye-sensitized solar cell (DSSC) [1–3], an electrochemical device converting solar energy to electricity, is a preferred candidate. During the work of a DSSC, only electricity is generated with no other emission. To date, a light-to-electric power conversion efficiency as high as 12.3% has been recorded from a typical DSSC device [4]. However, the high cost of Pt counter electrode is still a burden for commercial application of DSSCs. Therefore, it is a prerequisite to find alternative counter electrode materials honored by low cost with no sacrifice of conversion efficiency [5–8].

Carbon materials (i.e. graphene, graphene oxide, carbon nanotube) and electron-conducting polymers (i.e. polyaniline, polypyrrole) are good candidates for low-expenses counter

electrodes [9–12]. The highlight of carbon materials is their excellent electron-conduction, whereas conducting polymers are regarded because of their favorable electrocatalytic performances. Combination of carbon materials and conducting polymers collects electron-conduction and electrocatalytic behaviors on their composites. Traditional approaches in fabricating carbon/conducting polymer composite counter electrodes are chemical or electrochemical codeposition or blending [13–15]. The task of an efficient counter electrode is to rapidly electroreduce triiodides into monoiodides under the function of refluxed electrons. However, the above mentioned methods are expected to provide large interfacial resistance between carbon and conducting polymer owing to a physical contact.

With an aim of significantly enhance the charge-transfer from FTO layer to carbon/conducting polymer composite, and between carbon and conducting polymer, here we reported the reflux synthesis of aniline–single wall carbon nanotube complex and layer-by-layer (LbL) self-assembly of the resultant polyaniline–single wall carbon nanotube (PANi–SWCNT) with oppositely charged graphene oxide (GO). Covalent bonding between PANi and SWCNT is promising in accelerating charge-transfer within them, however, the interfaces between PANi–SWCNT complex and GO provides enormous space for electrocatalytic reaction of triiodides. Results indicated that a power conversion efficiency of 6.88% was recorded from the DSSC using (PANi–4 wt% SWCNT/GO)₅ multilayer counter electrode.

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2. Experimental

2.1. Reflux synthesis of aniline – SWCNT complexes

The aniline-SWCNT complexes were synthesized by a reflux process. In details, three aniline – SWCNT mixtures with SWCNT dosages of 1, 2, and 4 wt% were sealed in a three-neck flask filled with high-purity N₂ gas. In dark, the mixtures were refluxed for 3 h at 184 °C to obtain homogeneous aniline – SWCNT complexes, which were subsequently stored in a dark and cold atmosphere.

2.2. Synthesis of PANi – SWCNT complexes

50 mL of aqueous solution consisting of HCl (1 M) and (NH₄)₂S₂O₈ was dipped into 50 mL of a mixture consisting of HCl (1 M) and aniline – SWCNT (0.325 M) at 0 °C within 2 h. After reaction at 4 °C for 10 h, the as-synthesized complexes were successively rinsed with 1 M HCl aqueous solution to remove unreacted aniline monomers, oligomers, and remnant salts. Finally, the PANi – SWCNT complexes were vacuumly desiccated at 50 °C for 24 h and then dissolved in deionized water to prepare a solution with concentration of around 1 g·L⁻¹.

2.3. Assembly of (PANi – SWCNT/GO)_n ultrathin films

(PANi – SWCNT/GO)_n multilayer CEs were fabricated on FTO glass substrates (sheet resistance 12 Ω·cm⁻², purchased from Hartford Glass Co., USA). Prior to assembly, the substrates were first cleaned with deionized water and anhydrous ethanol. FTO substrates were immersed in a piranha solution [7/3 (v/v) of H₂SO₄/H₂O₂ at room temperature] for 1 h. To assemble CE, the pre-treated FTO glass was immersed in PANi – SWCNT aqueous solution for 5 min, rinsed with deionized water for 1 min and dried by N₂ gas stream, then immersed in GO aqueous solution (7 ~ 10 mg·mL⁻¹) for 5 min, then rinsed again by deionized water for 1 min and dried by N₂ gas stream. By repeating the previous procedures, we can obtain FTO supported (PANi – SWCNT/GO)_n multilayer CE (n represents the deposition cycle).

2.4. Electrochemical characterizations

The electrochemical performances were recorded on a conventional CHI660E setup comprising an Ag/AgCl reference electrode, a CE of platinum sheet, and a working electrode of FTO glass supported (PANi – SWCNT/GO)_n multilayer. The cyclic voltammetry (CV) curves were recorded from -0.8 to +1.6 V and back to -0.8 V. Before the measurement, the supporting electrolyte

consisting of 50 mM LiI, 10 mM I₂, and 500 mM LiClO₄ in acetonitrile was degassed using nitrogen for 10 min. Electrochemical impedance spectroscopy (EIS) measurements were also carried out on the CHI660E Electrochemical Workstation in a frequency range of 0.01 Hz ~ 10⁵ kHz and an ac amplitude of 10 mV at room temperature. The resultant impedance spectra were analyzed using the Z-view software. Tafel polarization curves were recorded on the same Workstation by assembling symmetric cell consisting of FTO – (PANi – SWCNT/GO)_n|redox electrolyte|FTO – (PANi – SWCNT/GO)_n.

2.5. Assembly of DSSCs

A layer of TiO₂ nanocrystal anode film with a thickness of 10 μm and active area of 0.09 cm² was prepared by coating the TiO₂ colloid using a screen printing technique, followed by sintering in air at 450 °C for 30 min. Subsequently, the TiO₂ film was soaked in a 0.5 mM N719 [cis-di(thiocyanato)-N,N'-bis(2,2'-bipyridyl)-4-carboxylic acid-4-tetrabutylammonium carboxylate, purchased from Dyesol LTD, Australia] ethanol solution for 24 h to uptake N719 dye for the fabrication of dye-sensitized TiO₂ photoanode. The DSSC was fabricated by combining dye-sensitized FTO-TiO₂ photoanode and multilayer CE injected with liquid electrolyte.

2.6. Photovoltaic measurements

The photocurrent-voltage (*J-V*) curves of the assembled DSSCs were recorded on an Electrochemical Workstation (CHI600E) under irradiation of a simulated solar light from a 100 W xenon-mercury arc lamp (CHF-XM-500 W, Beijing Trusttech Co., Ltd) in ambient atmosphere. The incident light intensity was calibrated using a FZ-A type radiometer from Beijing Normal University Photoelectric Instrument Factory to control it at 100 mW·cm⁻² (AM 1.5). Each DSSC device was measured five times to eliminate experimental error and a compromise *J-V* curve was employed.

2.7. Other characterizations

Ultraviolet-visible (UV-vis) absorption spectra were recorded with a Mapada 3200 UV-Visible spectrometer.

3. Results and discussion

Self-assembly of the multilayers has a dependence on adsorption time, as shown in Fig. 1a. The bands at 447 and 900 nm are assigned to π → localized polaron band of PANi [16,17],

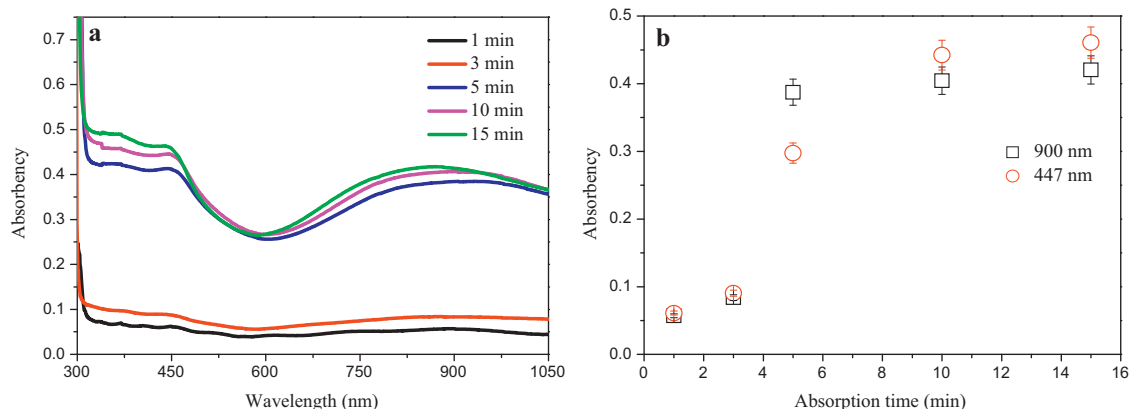


Fig. 1. (a) UV-vis spectra of (PANi – SWCNT/GO)₅ with various deposition times. (b) Plots of intensity of PANi absorbance as a function of deposition time. The SWCNT dosage is 4 wt%.

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