



Enhancing photovoltaic performances of dye-sensitized solar cells by multi-layered nanostructured titanium oxide photoelectrode

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Abstract

In the present study, a multi-layered nanostructured titanium oxide film has been fabricated as the photoelectrode for dye-sensitized solar cells (DSSCs). The multi-layered TiO₂ electrode has the advantages of easy fabrication, being compatible with current deposition process and readily modifiable by adjusting the process parameters. The application of TiO₂-MWCNTs (multiwalled carbon nanotube) as a nanocomposite overlayer reduces the inhomogeneity of dispersing carbon nanotubes (CNTs) throughout the whole photoanode, which largely simplifies the fabrication process and improves the performance of the as-prepared photoanodes. By using a multi-layered photoelectrode comprising a structure of TiO₂-MWCNT/Degussa P25-TiO₂/compact TiO₂/TCO glass, a 27% enhancement in conversion efficiency is realized in DSSC as compared with pristine P25-TiO₂ photoanode of identical thickness. The enhancement in efficiency brought by incorporation of MWCNTs, as revealed by electrochemical impedance spectroscopy (EIS), could be attributed to the excellent charge transport properties of CNTs, which led to efficient charge transfer within nanoporous TiO₂ networks. A compact TiO₂ blocking layer suppresses effectively the dark current, thus enhancing both J_{SC} and V_{OC} prominently.

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1. Introduction

Dye-sensitized solar cells (DSSCs) are one of the third-generation solar cell technologies for converting sunlight into electricity effectively and economically (Razykov et al., 2011). Currently, the conversion efficiency of DSSCs, a strikingly record of 12.3% has already been achieved by incorporating a Co(II/III) tris(bipyridyl)-based redox electrolyte in conjunction with a custom synthesized donor- π -bridge-acceptor zinc porphyrin dye as sensitizer (Yella et al., 2011). In principle, a substantial increase in DSSC efficiency can be achieved by employing a new sensitizer with a higher molar extinction coefficient and broader

spectral response, by employing an efficient photoelectrode with improved open-circuit voltage (V_{OC}), and/or by increasing the diffusion length of photoelectrons within the nanocrystalline oxide to maximize red light conversion (Law et al., 2006). In light of photoanode fabrication, strategies taken for enhancing cell performance include (i) manipulating the TiO₂ morphology for efficient light harvesting, (ii) promoting the electron transfer from the adsorbed dyes to the TiO₂ electrode, (iii) preventing the recapture of photo-injected electrons by electrolytes, and (iv) efficient mediator transport to the counter electrode through the liquid or solid electrolytes (Ito et al., 2005; Umeyama and Imahori, 2008; Xia and Yanagida, 2011).

It is a popular topic in DSSC-related studies which employs one-dimensional nanomaterials as a blocking layer (Lin et al., 2013) and/or scattering layers (Xia and

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Yanagida, 2011). By retarding the interfacial recombination or improving the light-harvesting of the as-synthesized photoanodes, the energy conversion efficiency of the DSSCs could be enhanced substantially. Carbon nanotubes (CNTs) resembling fullerenes had recently attracted much attention in organic devices (Ago et al., 1999). Owing to their excellent charge transport properties and chemical stability, CNTs have been employed to improve photovoltaic performance of DSSCs through their modifying TiO₂ films (Jang et al., 2004; Kongkanand et al., 2007; Brown et al., 2008; Ahn et al., 2013) or acting as TiO₂-CNT composite photoelectrodes (Kim et al., 2006; Lee et al., 2007, 2008; Muduli et al., 2009; Sawatsuk et al., 2009; Sun et al., 2011) and bilayered photoelectrodes (Lin et al., 2011). On the other hand, CNTs have also been tested as electrolyte additives (Lee et al., 2010) or as catalysts in counter electrodes (Roh et al., 2011; Anwar et al., 2013).

For solid-state DSSCs, a compact TiO₂ layer deposited by spray pyrolysis on a transparent conducting oxide (TCO) substrate was employed to avoid direct contact between the hole-transport medium layer and SnO₂, which would short-circuit the cell (Bach et al., 1998; Kruger et al., 2001). In a DSSC using I⁻/I₃⁻ as the redox couple, the macroscopic position in the cell at which carrier recombination occurs has been found to be predominantly near the substrate supporting the titania film (Zhu et al., 2002). Further investigation shows that under illumination, charge recombination occurred near the TCO substrate; while in the dark, photoelectrons from the TiO₂ electrode were captured by I₃⁻ at the surface of the nanocrystalline TiO₂ electrode (Ito et al., 2005; Cameron et al., 2005). In order to obtain high V_{OC}, preventing the recapture of photoinjected electrons by I₃⁻ is very critical. Previous investigations disclosed several effective strategies for achieving this goal, including fabrication of core-shell structured electrodes (Diamant et al., 2004; Chappel et al., 2002) and/or application of a TiO₂ blocking layer (BL) onto the surface of a TCO substrate of photoelectrodes (Wu et al., 2010; Hore and Kern, 2005; Cameron and Peter, 2003; Hart et al., 2006). By increasing the shunt resistance to prevent the back transfer of electrons from the TCO to the electrolyte, the TiO₂ blocking layer inhibits electron loss at the TCO/electrolyte interface, thus improving substantially the fill factor (Hore and Kern, 2005; Hart et al., 2006; Ferber et al., 1998; Yoo et al., 2010). In addition, the thin TiO₂ layer on the TCO was found to enhance adhesion between the bulk nanocrystalline TiO₂ film and the TCO (Chen et al., 2009).

A bilayer photoelectrode, as proposed by Lin et al. (2011), comprising a TiO₂-multi-walled CNT (TiO₂-MWCNT) nanocomposite secondary layer, leads to an improvement in photovoltaic performance as compared with a traditional electrode. By utilizing a multi-layered TiO₂ electrode constructed by the layer-by-layer deposition using three different kinds of TiO₂ pastes with average diameters of 9, 20 and 300 nm, respectively, the overall power conversion efficiency of the DSSC devices improves

substantially owing to the enhanced light harvest efficiency caused by light scattering (Lee et al., 2009; Dai et al., 2012). For preparation of the TiO₂-CNT composite, the homogeneity of CNTs dispersing within the TiO₂ paste is of major concern. However, owing to the hydrophobic nature of the CNT surface, it is challenging to maintain uniform dispersion of CNTs throughout the entire thick films, especially for those made by the layer-by-layer deposition technique. To overcome this drawback, DSSCs with enhanced efficiency are fabricated by depositing simply a single layer of TiO₂-MWCNT nanocomposite onto the mesoporous TiO₂ film, whose thickness has been optimized by spin-coated deposition. It would largely simplify the deposition procedure, reduce effectively the failure conditions caused by thick TiO₂-MWCNT composite films. The multi-layered TiO₂ electrode also has the advantages of easy fabrication, being compatible with current deposition process and readily modifiable by adjusting the process parameters.

In this study, the major parameters that could affect the photovoltaic performance of the multi-layered TiO₂-MWCNT composite photoanode were evaluated, including the thickness of TiO₂ films, the amount of MWCNTs incorporated, and the role of the TiO₂ BL. In addition, the photoelectrochemical characteristics of DSSCs were described and discussed.

2. Experimental

2.1. Synthesis of multiwalled carbon nanotubes

In this study, MWCNTs were synthesized by catalytic pyrolysis with a home-made quartz tubular reactor of 2-in. diameter. The supported catalyst, Co-Mo/MgO, was prepared by incipient wetness impregnation followed by the procedures reported by Kibria et al. (2004) with Co/Mo = 3/2, and metal loading = 5 wt.%. Approximately 0.5 g of catalyst was uniformly dispersed onto an alumina boat in the central region of a horizontal quartz tube reactor. There, the catalyst was activated at 800 °C for 1 h under 50 sccm H₂ flow to prevent the oxidation of Co-Mo/MgO and to enhance the adhesion of metal layers (Lee et al., 1999).

After that, MWCNTs were then grown on the substrate at 800 °C in a mixture of ethylene, ammonia and argon at a flow rate of 10, 20 and 100 sccm, respectively for 15 min. Pristine MWCNTs are hydrophobic, and are difficult to be dispersed uniformly in a TiO₂ matrix (Lee et al., 2007). To solve this problem, the as-grown MWCNTs were carefully removed from the substrate and refluxed with 6 M HNO₃ at 120 °C for 3 h. to remove the metal catalyst impurities and to generate carboxylic groups (Pimanpang et al., 2009). Field emission scanning electron microscopy (FESEM) was conducted using a JEOL JSM-7401F with an operating voltage of 4.0 kV. The bushy purified MWCNT samples, as shown in the SEM micrograph in Fig. 1(a), are very long with quite uniform diameter. A Raman spectrometer (Nanofinder 30R, Tokyo Instru-

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