

Multiwalled carbon nanotube thin films prepared by aerosol deposition process for use as highly efficient Pt-free counter electrodes of dye-sensitized solar cells

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Received 6 February 2014; received in revised form 7 June 2014; accepted 11 June 2014

Available online 8 July 2014

Communicated by: Associate Editor Frank Nuesch

Abstract

In this study, transparent and catalytic multiwalled carbon nanotube (MWCNT) thin films with controlled thickness were fabricated using a simple and conventional multiple nozzle-based aerosol deposition process (ADP). MWCNTs were homogeneously dispersed in deionized water via surfactant treatment, and the MWCNT-dispersed aqueous solution was then aerosolized and deposited on fluorine-doped tin oxide glass substrates to form MWCNT thin films. The characteristics of MWCNTs, degree of MWCNT dispersion stability in the aqueous solution, and morphology and light transmittance of the prepared MWCNT thin films were systematically examined. Finally, the prepared MWCNT thin films were used as the counter electrodes (CEs) of dye-sensitized solar cells (DSSCs). For comparison purposes, the photovoltaic performance of DSSCs composed of MWCNT thin-film CEs was compared to that of reference DSSCs composed of conventional Pt thin-film CEs. The results showed that the power conversion efficiency of DSSCs composed of critical amount of MWCNTs coated on CEs was almost equal or slightly higher than that of conventional Pt-based DSSCs. Thus, it can be concluded that the ADP-assisted precisely controlled accumulation of transparent and catalytic MWCNT thin films on the CEs of DSSCs is a very promising approach for replacing the expensive Pt metal that is currently used in DSSC industries.

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Keywords: Multiwalled carbon nanotubes; Aerosol deposition process; Dye-sensitized solar cells; Counter electrodes

1. Introduction

Dye-sensitized solar cells (DSSCs) have been developed extensively because of the relatively low cost involved in their manufacturing processes (Grätzel, 2001, 2003; O'Regan and Grätzel, 1991). Among the various components of DSSCs, the counter electrode (CE) is one of the most important parts because it collects the photogenerated electrons from an external circuit and simultaneously

regenerates dye sensitizers by reducing the iodide electrolyte used (Halme et al., 2006; Murakami and Grätzel, 2008; Papageorgiou, 2004). In order to fabricate CEs for DSSCs, Pt is generally coated on the surface of fluorine-doped tin oxide (FTO) glass substrates because Pt has high electrochemical reactivity (Fang et al., 2004; Lin et al., 2010; Olsen et al., 2000). However, since Pt is a relatively expensive noble metal and dissolves slowly in corrosive iodide electrolytes, it is necessary to find an appropriate replacement of Pt as a comparable potential catalytic material for DSSCs. Furthermore, the CEs of DSSCs should be stable and transparent for various applications such as

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building-integrated photovoltaic cells (Heiniger et al., 2013; Jelle et al., 2012; Yoon et al., 2011) and tandem cells (Ahn et al., 2007; Dürr et al., 2004; Nattestad et al., 2008), as well as simultaneously have low electrical resistance and high electrocatalytic reactivity for effectively reducing iodide/tri-iodide (I^-/I_3^-) electrolytes.

In addition to being inexpensive, multiwalled carbon nanotubes (MWCNTs) have rapid electron transfer kinetics, a relatively large specific surface area, and high electrocatalytic activity; therefore, numerous research groups have employed MWCNTs as a replacement of Pt for fabricating the CEs of DSSCs (Anwar et al., 2013; Cha et al., 2010; Hsieh et al., 2011; Lee et al., 2009, 2010; Yan et al., 2013). Typically, MWCNTs are coated on FTO glass to be used as a CE for DSSCs. In order to fabricate MWCNT-coated FTO glass CEs, various techniques such as doctor blade (Cha et al., 2010; Hsieh et al., 2011; Lee et al., 2009; Li et al., 2010), screen printing (Anwar et al., 2013), spin coating (Fan et al., 2008; Jo et al., 2012), spray coating processes (Hsieh et al., 2011; Huang et al., 2012; Ramasamy et al., 2008), and hydrothermal deposition (Sirirotj et al., 2012) have been developed. However, these techniques suffer from some inherent problems in fabricating uniform MWCNT layers with precisely controlled thickness owing to the formation of highly agglomerated and entangled MWCNTs during the aforementioned coating processes. Forming a non-uniform MWCNT thin film on an FTO glass substrate as a CE can eventually deteriorate electron transfer in DSSCs. Therefore, a more versatile and reliable method is required to realize a uniform spatial distribution of MWCNTs on an FTO glass substrate for use in DSSCs.

In this study, we developed a multiple nozzle-based aerosol deposition process (ADP) for a MWCNT-dispersed aqueous solution, and subsequently employed the process for fabricating MWCNT-deposited thin films on FTO glass substrates for assembling CEs of DSSCs. Further, we systematically investigated the effect of fabricated MWCNT thin films on the photovoltaic performance of the resulting DSSCs. Various DSSCs containing MWCNT-deposited CEs were then compared with conventional DSSCs containing Pt-deposited CEs.

2. Experimental

2.1. Fabrication of MWCNT-dispersed aqueous solution

First, MWCNTs were purchased from CNT Co., Ltd (Korea) and used without further treatment. The MWCNTs were generated using a thermal CVD method, and they had the average diameter of ~20 nm and the length distribution of 1–25 μm . The purity of MWCNTs was larger than approximately 95% and the specific surface area was approximately 150–250 m^2/g . In order to prepare a homogeneously MWCNT-dispersed aqueous solution, the surfactant addition method was employed. Appropriate surfactant addition can homogeneously disperse

inherently hydrophobic MWCNTs in an aqueous solution by forming an interconnection between hydrophilic and hydrophobic surfaces. In the surfactant addition method, carboxy methyl cellulose (CMC, Sigma Aldrich, Mw: 700,000) was used as the surfactant (Cha et al., 2010; Imoto et al., 2003; Lee et al., 2009; Takahashi et al., 2006). CMC is a kind of surfactant with several hydroxyl groups, and it has good properties of stabilizing the stock, implying that the long CMC molecules wrap around the MWCNTs and allowing them to be homogeneously dispersed in water. A CMC-added MWCNT aqueous solution containing 0.1 wt% of MWCNTs and 0.15 wt% of CMC in deionized water was prepared by sonicating the solution for 1 h.

2.2. Aerosol deposition process (ADP) for preparing MWCNT-dispersed aqueous solution

MWCNT-deposited CEs were fabricated by depositing the aerosolized MWCNTs onto FTO glass substrates as shown in Fig. 1. The ADP system used for this approach

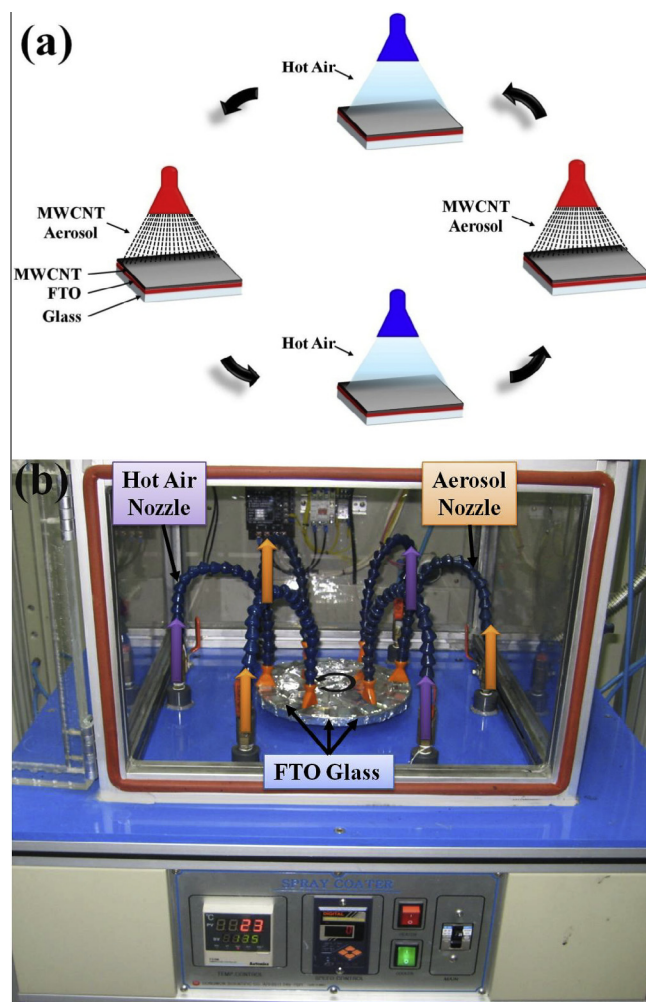


Fig. 1. (a) Schematic and (b) photograph of an aerosol deposition process system for fabricating MWCNT thin films on FTO glass substrates.

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