Organic Electronics xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Investigation of graphene nanosheets as counter electrodes for efficient dye-sensitized solar cells

Chih-Hung Tsai *, Chih-Han Chen, Yu-Chen Hsiao, Ping-Yuan Chuang

Department of Opto-Electronic Engineering, National Dong Hwa University, Hualien 97401, Taiwan

A R T I C L E I N F O

Article history:
Received 15 October 2014
Received in revised form 4 November 2014
Accepted 21 November 2014
Available online xxxx

Keywords:
DSSCs
Counter electrode
Graphene nanosheets
Platinum

A B S T R A C T

In this study, graphene nanosheets (GNs) were used to fabricate novel counter electrodes (CEs) for dye-sensitized solar cells (DSSCs). The electrode properties of various CEs were comprehensively analyzed using scanning electron microscopy (SEM), atomic force microscopy (AFM), Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), energy dispersive X-ray spectrometry (EDS), and cyclic voltammetry (CV). DSSCs with various GN CEs were characterized using current density–voltage (*J–V*), incident photo-to-current conversion efficiency (IPCE), and electrochemical impedance spectroscopy (EIS) measurements. The results show that GN CEs sintered at 400 °C in a nitrogen atmosphere for 30 min yielded the optimal electrode properties and DSSC efficiency. This study also fabricated GN–Pt composite and GN–Pt stacked CEs for the DSSCs, and the influences of the CEs on the efficiency of the DSSCs were investigated. The results show that the GN–Pt stacked CEs yielded the optimal electrochemical catalytic properties and DSSC efficiency. The power conversion efficiency of the DSSCs based on GN–Pt stacked CEs yielded a 16.7% improvement compared with conventional Pt CEs.

1. Introduction

Energy shortage and environmental pollution are two major problems of the 21st century. Renewable energy refers to energy generated from inexhaustible natural resources that produce no pollutants when used. Among the various types of renewable energy, solar energy is advantageous because of its inexhaustibility, few geographical constraints, and absence of environmental pollutants, making it the most advantageous renewable energy resource. Dye-sensitized solar cells (DSSCs) were first reported by Professor Grätzel in 1991 [1]. The advantages of DSSCs include high efficiency, low cost, simple structure, and easy fabrication. Therefore, DSSCs have attracted considerable attention in the academic community [2–10].

A typical DSSC device structure includes a transparent conductive substrate, a TiO_2 nanoparticle thin film, a dye, an electrolyte, and a platinum (Pt) counter electrode (CE) [11–13]. The role of a DSSC CE is to catalyze the reduction of the I_3^- ions in the electrolyte that are produced during the regeneration of oxidized dyes, which enables the dyes to return from an excited state to a ground state [14]. Pt is typically used as a CE material because it yields highly efficient DSSCs [15–23]. However, as a noble metal, Pt is relatively expensive, which is a considerable obstacle for the large-scale application of DSSCs. Therefore, a highly efficient replacement material for Pt is a crucial focus of DSSC-related studies.

Graphene, a one-atom-thick hexagonal mesh of carbon atoms, is a novel carbon-based material that has received considerable attention [24]. In 2004, Geim and Novoselov successfully isolated graphene by using Scotch tape to peel off single sheets of graphene from graphite, thereby proving that graphene can exist independently under stable

* Corresponding author. Tel.: +886 3 8634199; fax: +886 3 8634180.
E-mail address: cht@mail.ndhu.edu.tw (C.-H. Tsai).

http://dx.doi.org/10.1016/j.orgel.2014.11.016
1566-1199/

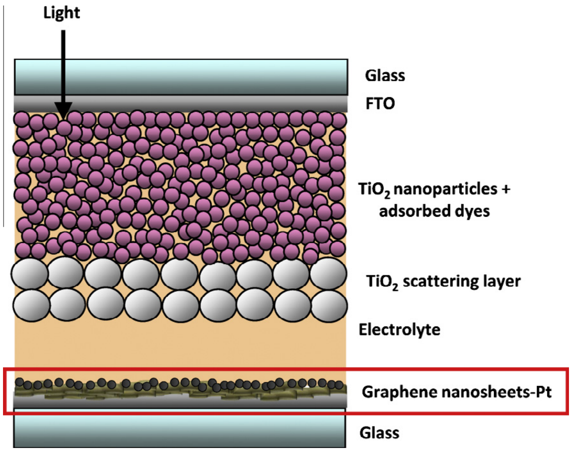

Fig. 1. The schematic device structure of a DSSC using the GN–Pt stacked counter electrode.

conditions [25]. For this discovery, they were awarded the 2010 Nobel Prize in Physics. Graphene possesses unique material properties, such as high mechanical strength (approximately 1100 GPa), high thermal conductivity (approximately 5000 W m^{-1} K^{-1}), and high carrier mobility (200,000 cm^2 V^{-1} s^{-1}) [26]. These exceptional properties have attracted the attention of the scientific community, and graphene has become the focus of recent studies. Several research teams have also used graphene to fabricate DSSC CEs. Zhang et al. dispersed graphene nanosheets (GNs) in solutions containing terpineol and ethyl cellulose ethoce (EC) and screen-printed the GN solution onto the surface of fluorine-doped tin oxide (FTO). They analyzed the influence of the annealing temperature on DSSC devices and reported a conversion efficiency of 2.94% [27]. Roy-Mayhew et al. used functionalized graphene sheets as DSSC CEs and determined that increased oxygen-containing functional groups increased the surface catalytic activities of graphene [28]. Wan et al. fabricated graphene thin films on various substrates by using low-cost room-temperature solution processes, and determined that graphene can be applied to DSSC, supercapacitors, fuel cells, and chemical sensor devices [29]. Kavon et al. fabricated graphene sheets on an FTO substrate for use as a DSSC CE and achieved satisfactory catalytic activity [30].

This study used GNs to fabricate novel CEs, including GN, GN–Pt composite, and GN–Pt stacked CEs for DSSCs. The electrode properties of the various CEs were analyzed using scanning electron microscopy (SEM), atomic force microscopy (AFM), Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), energy dispersive X-ray spectrometry (EDS), and cyclic voltammetry (CV). The various GN CEs were investigated to identify the correlation between the CE fabrication conditions and DSSC characteristics and performance. The results show that the GN–Pt stacked CEs exhibited optimal electrochemical catalytic properties and DSSC efficiency. A device efficiency of 8.54% was obtained using the GN–Pt stacked CEs, yielding a 16.7% improvement compared with conventional Pt CEs.

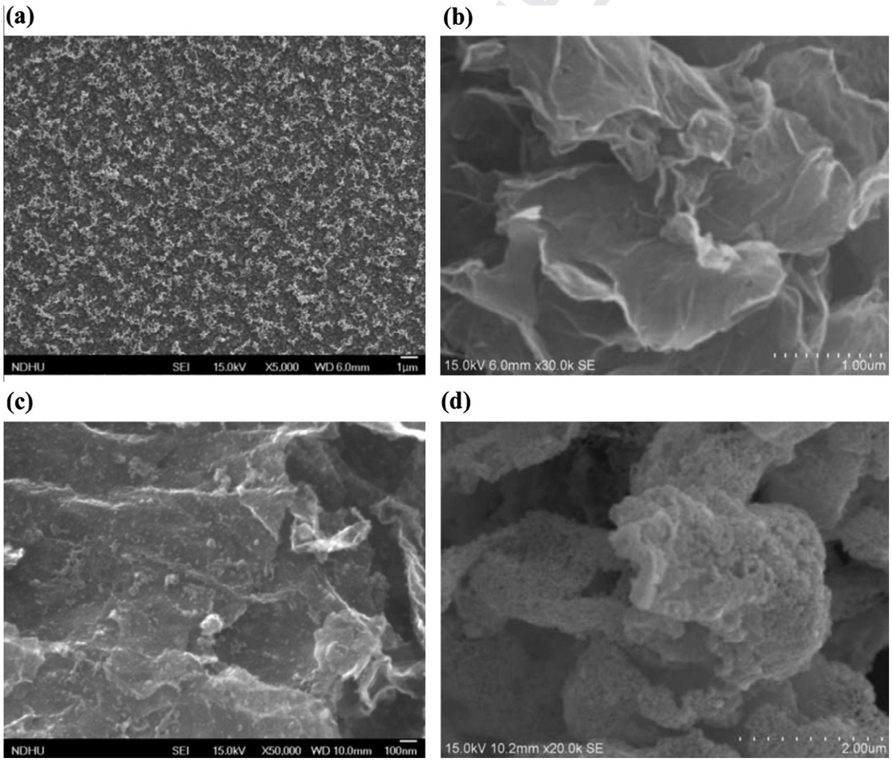

Fig. 2. SEM images of (a) Pt, (b) GN, (c) GN–Pt composite, and (d) GN–Pt stacked counter electrodes.