CARBON 84 (2015) 383–389

Available at www.sciencedirect.com

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Graphene-based nanohybrid materials as the counter electrode for highly efficient quantum-dot-sensitized solar cells

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ARTICLE INFO

Article history:
Received 9 August 2014
Accepted 3 December 2014
Available online 9 December 2014

ABSTRACT

Dry plasma reduction is an excellent approach for easily and uniformly immobilizing Pt, Au and bimetallic AuPt nanoparticles (NPs) on a graphene nanoplatelets (GC)-coated layer under atmospheric pressure at a low temperature and without using any toxic reductants. The NPs with an average size of about 2 nm were stably and uniformly hybridized on the surface of reduced graphene nanoplatelets (RGC) after co-reduction of metal precursor ions and GC to metal atoms and RGC, respectively. Quantum-dot-sensitized solar cells exploiting AuNP/RGC, PtNP/RGC and bimetallic AuPtNP/RGC counter electrodes (CEs) exhibited power conversion efficiencies of 2.7%, 3.0% and 4.5%, respectively. The efficiencies are comparable to that of device with a conventional Au-sputtered CE (3.6%). The effect is ascribed to high electrochemical catalytic activity and high electrical conductivity of developed nanohybrid materials.

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1. Introduction

Owing to using low band gap semiconductors for light-trapping, quantum-dot-sensitized solar cells (QDSCs) have attracted much attention as the next generation of dye-sensitized solar cells (DSCs) [1]. Except for the effect of quantum confinement of the exciton in the absorber, the performance of a QDSC can be improved by optimizing the band alignment at the TiO_2/QD-sensitizer interface for efficient charge transfer, by increasing the light collection efficiency and by matching the absorption spectrum of the QD-sensitizer to the solar spectrum. These advantages of the QDSC can be realized by controlling the size of the QD and the band gap energy of the semiconductor QD-sensitizer. Indeed, the value of the band gap of II–VI group compound semiconductors and the mixed metal compounds can be varied with the chemical composition of the film in the range between 1.1 and 3.7 eV. Currently, QDSCs are very promising devices to replace conventional DSCs because the energy conversion efficiency can reach 44% based upon theoretical prediction [2]. Nevertheless, until now, the champion-device efficiency of 8.5% was shown by solar cells based on Sb_2S_3 and PbS QDs at a laboratory scale [3]. The development of the QDSCs is of crucial importance despite the fact that these devices currently yield low efficiencies.

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http://dx.doi.org/10.1016/j.carbon.2014.12.014
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The conversion efficiency and production cost are key issues in the photovoltaic (PV) technologies today for commercialization. So far, the development of low-cost and scalable counter electrodes (CEs) with high redox activity toward the sulfide/polysulfide couple remains an issue in fabrication of QDSCs [4]. Pt and Au have been common materials for the CEs in the QDSCs due to high catalytic activity, high conductivity and chemical stability [4,5]. However, they are known to be an expensive material with limited reserves. Thus, their large-scale application is too expensive to compete with conventional solid-state PV devices. Attention to date has focused on noble-metal-free materials as alternative types of efficient electrodes. The metal chalcogenides (CoS, CuS, CuS/CoS, Cu_2S, PbS, MoS_2) [6–9], carbon-based materials [10], and graphene-based hybrid materials (TiN/CNT–graphene, Cu_2S/graphene, Au/graphene, Mo-compound/CNT–graphene) [1,11–14] have been widely studied as low-cost cathode materials. However, the advantages of low-cost cathode materials must always be weighed against the drawbacks of their processing parameters, such as high processing temperature, chemical toxicity, and a long reduction time. Despite the many research studies on the subject, the development of low-cost and scalable CEs with high redox activity toward the sulfide/polysulfide couple remains one of the major challenges for QDSC technology to date.

This research addresses one of the key questions in developing a CE for highly efficient QDSCs. For this purpose, we developed an experimental approach for co-reduction of metal precursor ions and graphene oxide through dry plasma reduction (DPR) at near room temperature under atmospheric pressure [15]. The process provides uniform hybridization of well-dispersed metal-NPs on the surface of reduced graphene oxide. We expect that the metal-NP/reduced graphene oxide nanohybrids will achieve high conductivity as well as high catalytic activity toward regenerating the S^{2-}/S_x^{2-} redox couple.

2. Experimental section

2.1. Synthesis of nanohybrids materials and CEs

Three solutions containing 10 mM $H_2PtCl_6{\cdot}xH_2O$ (⩾37.5% Pt basic, Sigma–Aldrich) and iso-propyl alcohol (IPA) (99.5%, Sigma–Aldrich), 10 mM $HAuCl_4{\cdot}3H_2O$ (Aldrich) in IPA and a solution in a mixed precursor of 10 mM $H_2PtCl_6{\cdot}xH_2O$ in IPA and 10 mM $HAuCl_4{\cdot}3H_2O$ in IPA with a volume ratio of 1:1 were prepared. Next, 3 μL of the precursor was dropped onto a graphene nanoplatelets-grade C 750 m^2/g (GC) CE, which was prepared separately in a previous study [16], and the solvent was allowed to evaporate at 70 °C for 15 min. Then, the specimens were reduced using Ar plasma under atmospheric pressure at a power level of 150 W, a gas flow rate of 5 lpm, a treatment time of 15 min, and a substrate moving speed of 5 mm/s. This procedure was performed with the different precursors.

2.2. Preparation of working electrodes

The working electrodes were prepared as described in our previous study [11]. Briefly, a 50 nm-thick ZnO buffer film was sputtered onto FTO glass (~8 Ω/□, Pilkington, USA) and was then immersed in an aqueous solution containing $Zn(NO_3)_2{\cdot}6H_2O$ (0.01 M) and NH_4OH (0.5 M) for 12 h at 95 °C. Sensibilization of ZnO nanowire (NW) electrodes with CdS was conducted using the ion-layer adsorption and reaction (SILAR) method. For this purpose, the electrodes were dipped into an aqueous solution of $CdSO_4$ (200 mM) for 30 s, rinsed with deionized water for 30 s, dipped in an aqueous solution of Na_2S (200 mM) for 30 s, and finally rinsed with de-ionized water for 30 s. Then, CdSe QDs were deposited in situ on CdS/ZnO NWs using chemical bath deposition from an aqueous solution containing of 2.5 mM of $Cd(CH_3COO)_2$ (as Cd ion source), 2.5 mM of Na_2SeSO_3 (as Se ion source), and 45 mM of NH_4OH (as complexing agent) for 3 h at 95 °C. The quantum dot depositions were carried out in a home-built bath equipped with a Teflon-bladed electric stirrer and a Teflon substrate holder. Bath temperature was adjusted to the desired temperature level using a thermostat. To achieve a suitable loading of CdSe on the CdS/ZnO-NWs, the procedure was repeated three times. The experimental details of the fabrication of working electrodes are described in previous study [11].

2.3. Assembly of QDSCs

The assembly of the QDSCs was carried out as described in our previous study [11]. Briefly, the working electrode and CE were assembled into a sandwich type cell, which was sealed with a thermobonding polymer (Surlyn, DuPont) of 60 μm thickness at 120 °C for 5 min. A drop of the electrolyte composed of Na_2S (0.5 M), S (2 M), KCl (0.2 M) in methanol/water (the volume ratio of 7:3) was injected in each hole (0.8 mm) located in the back side of the working electrode. Two holes were sealed with Surlyn layer.

2.4. Characterization and measurements

The morphologies of nanohybrid materials were characterized by TEM measurement (JEM-2100F, Joel, Japan). The chemical states of the nanohybrids were analyzed by energy-dispersive X-ray spectroscopy (EDS) (JEM-2100F, Joel, Japan). The reduction of GC to reduced GC (RGC) induced by the DPR process was systematically studied using X-ray photoelectron, Raman spectroscopy, and thermal gravimetric analysis (TGA) in our previous work [16].

The photocurrent density–voltage characteristics of the cells were measured under a simulated air mass 1.5 G solar spectrum. The constant light illumination was adjusted to 100 mW cm^{-2} using a national renewable energy laboratory (NREL)-certified silicon reference cell equipped with a KG-5 filter. An active area of 0.25 cm^2 was accurately defined using a mask placed in front of the cell [11]. Electrochemical impedance spectrum (EIS) analysis of the counter electrodes was carried out in a symmetric cell configuration using an Ivium potentiostat. The frequency range was from 100 kHz to 100 mHz with a modulation amplitude of 5 mV at a 0 V bias voltage [15,16]. EIS spectra were fitted using the Z-view software package.

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