



Electrophoretic deposition of graphene nanosheets: A suitable method for fabrication of silver-graphene counter electrode for dye-sensitized solar cell



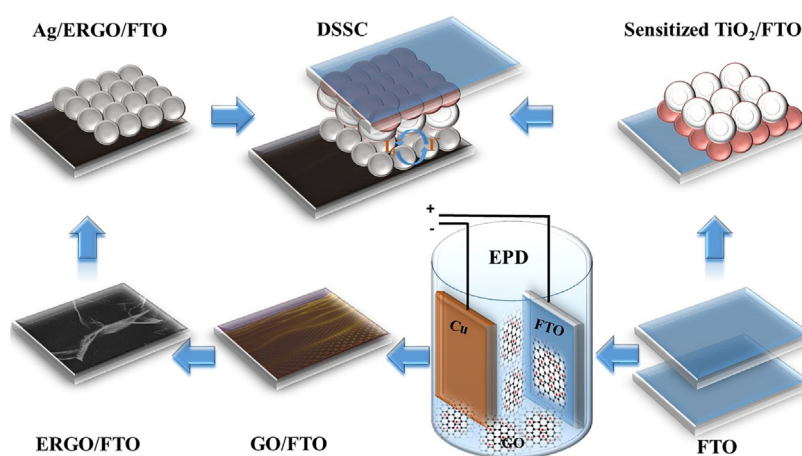
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HIGHLIGHTS

- GO nanosheets were deposited at the surface of FTO by electrophoretic technique.
- GO/FTO electrode were electrochemically reduced into ERGO/FTO.
- Silver nanoparticles were grown on ERGO/FTO electrode to produce Ag/ERGO/FTO.
- DSSC was fabricated out of TiO₂ photoanode and Ag/ERGO/FTO counter electrode.
- The DSSC performed suitable parameters ($V_{oc} = 0.8\text{ V}$, $j_{sc} = 29.04\text{ mA cm}^{-2}$ and $\eta = 4.24\%$).

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 21 September 2016
 Received in revised form 26 January 2017
 Accepted 3 February 2017
 Available online 5 February 2017

Keywords:

Dye-sensitized solar cell
 Graphene nanosheets
 Silver nanoparticles
 Electrophoretic deposition
 Iodine redox shuttle

ABSTRACT

Dye-sensitized solar cell (DSSC) was fabricated by a suitable counter electrode (CE) based on graphene nanosheets in order to facilitate electrochemical reduction of triiodide in organic medium. Graphene oxide (GO) nanosheets were deposited at fluorine doped tin oxide (FTO) glass substrate by electrophoretic deposition (EPD) technique as an easy short-time method. Then GO nanosheets were converted to electrochemically reduced graphene oxide (ERGO) nanosheets by chronoamperometry technique. Graphene nanosheet-based electrode was modified by silver nanoparticles and was characterized by field emission scanning electron microscopy (FE-SEM), energy dispersive spectroscopy (EDS), X-ray elemental mapping (MAP) and atomic force microscopy (AFM) studies. Also, the electrochemical behaviors of as-prepared electrodes were investigated by cyclic voltammetry method where electrodes fabricated by graphene-silver nanohybride (Ag/ERGO/FTO) presented the highest current density and the lowest peak-to-peak separation in comparison to bare FTO, GO/FTO and ERGO/FTO electrodes in iodine electrolyte medium. Moreover, electrochemical impedance spectroscopy (EIS) was applied to investigate the electrocatalytic capability of the modified electrode both in I₃⁻/I⁻ electrolyte and as a symmetrical dummy cell which represented relatively lower charge transfer resistance.

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Finally, the fabricated DSSC containing Ag/ERGO/FTO CE represented suitable photovoltaic characteristic parameters ($V_{oc} = 0.8 \text{ V}$, $j_{sc} = 29.04 \text{ mA cm}^{-2}$ and $\eta = 4.24\%$) under simulated light of AM1.5 G (1000 W m^{-2}) compared to Pt-based DSSC ($V_{oc} = 0.66 \text{ V}$, $j_{sc} = 19.54 \text{ mA cm}^{-2}$ and $\eta = 6.08\%$). Furthermore, the high conductivity of the cells was evaluated by EIS during the applying different biases.

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1. Introduction

Dye-sensitized solar cell (DSSC) is a liquid-junction photoelectrochemical cell which has attained scientist's attention due to its cost-effective, low temperature fabrication and respectable power conversion efficiency, mostly after the first high (almost 7%) efficiency, world break up of O'Regan and Grätzel [1,2]. The key components of a DSSC are a photoanode which is composed of a mesoporous film of a wide band gap semiconductor usually TiO_2 nanoparticles coated on a transparent conductive oxide (TCO) glass substrate and sensitized by dye molecules, a redox mediator (typically I_3^-/I^- in an aprotic electrolyte medium) and a counter electrode (CE). Among them, CE plays two important roles in the cell performance; it transfers electrons from the external circuit to the electrolyte solution and catalyzes the reduction of I_3^- , thereby facilitating sensitizer regeneration. However, the electrochemical activity of iodine species at the surface of TCO (e.g. fluorine doped tin oxide (FTO)) is quite sluggish and conventionally corrosion-resistant platinumized cathodes were utilized for this purpose. Since platinum is a precious metal with limited availability, many investigations have been achieved to increase the efficiency and the stability of the cell or diminish the overall cost of device fabrication especially in large scale by introducing new cathode materials.

Conducting polymers have been widely used in order to catalyze the reduction of I_3^- at the surface of CE in DSSCs. Polymers like polyaniline [3], polypyrrole [4] and poly(3,4-ethylenedioxythiophene) (PEDOT) [5] are highly desirable for scientists. Lately, carbon materials like graphene and carbon nanotube (CNT) have brought new area in the field of photovoltaic technology [6–8].

Graphene, an atom-thick two-dimensional (2D) carbon material is a good candidate among all carbon allotropes which can be used as transparent conducting electrodes due to its dominant electrical conductivity, high optical transparency and superb specific surface area. The remarkable thermal and electrical conductivity of graphene make it applicable as flexible conductor specially in organic photovoltaic cells [9]. Graphene, GO and composites based on them were widely used as different components in nanostructured solar cells [10–13].

Among various deposition techniques for the preparation of thin film of graphene like spin coating, chemical vapor deposition (CVD), drop evaporation and etc., electrophoretic deposition (EPD) has the applicability to prepare uniform and thickness-controlled layer film [14].

Although this zero-bandgap metal like material is highly conductive and stable, the enhanced performance of graphene in DSSC could be obtained by its nanocomposite with metal nanoparticles. Various strategies like vacuum deposition, evaporation and electrochemical deposition have been used to deposit different noble metal nanoparticles such as Pd [15] and Au [16] on graphene. Tjoa et al. proposed a facile photochemical method for synthesis of graphene-Pt nanoparticle composite for CE in order to catalyze the reduction of I_3^- to I^- in organic medium [17].

In order to enhance the electrical conductivity and the electrocatalytic properties of electrodes, silver nanoparticles have been found to be beneficial [18,19]. Also, the synergetic effect of Ag

nanoparticles and different conductive materials has been widely demonstrated [20,21]. Here, we introduce a nanohybrid composed of silver nanoparticles uniformly distributed at the surface of ERGO nanosheet film as a promising cathode material to facilitate the electrochemical activity and decrease the resistance of FTO toward the iodine species in DSSC.

2. Experimental

2.1. Materials

Commercial anatase titanium dioxide nanoparticles paste in two sizes (T/SP20 and R/SP300, Solaronix), cis-bis(isothiocyanato)bis(2,20-bipyridyl-4,40-dicarboxylato) ruthenium (II) bis-tetrabutyl ammonium (N719) (Dyesol), FTO transparent conductive glass (Solaronix), and Surlyn thermoplastic polymer (Solaronix) were purchased to construct solar cells. Also, graphite powder (Merck), sulfuric acid (98%, Merck), hydrochloric acid (37%, Merck), hydrogen peroxide (35%, Merck), potassium permanganate (99%, Merck), ethanol (99%, Fluka), acetone (99%, Fluka), silver nitrate (99%, Merck), lithium iodide (99%, Merck), iodine (I_2) (99%, Merck), lithium perchlorate (99.99% Sigma), DSSC-specific commercial iodine electrolyte in acetonitrile (Iodolyte HI-30, Solaronix), sodium nitrate (NaNO_3), potassium nitrate (KNO_3), acetonitrile (99.99% Merck), tert-butanol (Aldrich), ammonia aqueous solution (25%, Merck) were used as received without further purification. Moreover, all aqueous solutions were prepared by double distilled water.

2.2. Apparatus

Universal 320 centrifuge (Hettich, Germany) and 400W tip sonicate (Ultrasonic Technology, Iran) were used during the preparation of GO nanosheets. DC power supply (Sanjesh, Iran) was used to deposit GO nanosheets film on FTO electrode. AUTO-LAB 302N (Netherlands) electrochemical analyzer and PalmSense potentiostat-galvanostat (Netherlands) were applied for electrochemical deposition and investigation and electrochemical impedance spectroscopy (EIS), respectively. UV-vis spectrometer (2100 Beijing, China), Raman spectrometer (Senterra-Bruker), Fourier transform infrared spectroscopy (FTIR) (Vector 22, Bruker, Germany), field emission scanning electron microscopy (FE-SEM, Mira 3 XMU FE-SEM/EDS, TESCAN) and SEM (EVO 18, Carl Zeiss, Germany) and atomic force microscopy (Easy scan 2 Flex AFM, Nanosurf, Swiss) were used to characterize GO, ERGO and ERGO-based electrodes. The as-prepared DSSCs were tested under AM 1.5G solar simulator (Sharif Solar, Iran).

2.3. Methods

2.3.1. Preparation of graphene oxide nanosheets

GO was prepared by modified Hummers method [22]. Briefly, 0.7 g graphite powder was stirred with 115 mL sulfuric acid. 15 g potassium permanganate was slowly added to the mixture during an hour and the temperature was kept below 7°C . Later, the mixture was diluted by adding 250 mL water and 15 mL H_2O_2 (30%) to

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