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Graphene oxide as stable electrocatalytic substrate for solid-state bifacial dye-sensitized solar cells

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ABSTRACT

Bifacial dye sensitized solar cells (BDSSCs) are regarded as advanced photovoltaics for markedly increased light-harvesting efficiency and the ability of both-side incident illumination absorption. We present here the fabrication of graphene oxide electrocatalyst supported polyaniline (GO/PANI) composite film *via* an in-situ electrochemical polymerization method and used it as transparent counter electrode (CE) for solid-state BDSSCs. The GO provides a large number of active nucleation sites, resulting in the formation of continuous PANI films through strong covalent bonds, which significantly increase the specific surface area and catalytic activity of the CE and further accelerate the charge transfer in the corresponding BDSSC. Due to the improved electrochemical catalytic activity of the GO/PANI CE with GO mass ratio of 0.6 wt%, both-illuminated power conversion efficiency of 5.74% with short-circuit current density of 20.23 mA cm^{-2} and open circuit voltage of 0.63 V is obtained for the solid-state BDSSC, which is 155.6% of the conventional bare PANI based BDSSC under front-irradiation. Electrochemical and electron transport/recombination performances have also been demonstrated for this type of device.
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1. Introduction

Bifacial dye-sensitized solar cells (BDSSCs) have recently attracted the attention of several research groups [1–3] because of its higher light-harvesting efficiency (LHE) and unlimited incident angle regarding the ability of both-side incident illumination absorption. And this transparent property renders the BDSSCs a highly promising device for building-integrated photovoltaic systems (BIPS) and portable power generators in which the device must combine with transparent substrates. Accordingly, the design and fabrication of a transparent counter electrode (CE) with a high ability for reducing I_3^- to I^- to complete the circulation of electron transport is highly demanded.

Transparent thin-layer platinum (Pt) CEs have been fabricated through sputtering or thermal deposition [4]. However, the dissolution in electrolyte, high cost and the high reflectance of thin Pt films restrict the realization of a BDSSC module. Accordingly, numerous functional materials, including carbon-based nano-materials and conductive polymers have been proposed to replace Pt as alternative materials of CEs in BDSSCs. Polyaniline (PANI) is particularly suitable for the transparent CE of BDSSCs due to its low-cost, environmentally friendly processing [5] and complementary light absorption to N719 dyes, which maximize the use of rear illumination for BDSSCs [2,5]. Our previous works incorporated Ag_2S and black phosphorus quantum dots into transparent PANI CE, which result in significant enhancement in light harvesting of the BDSSCs [6–8]. However, the single use of PANI film in CE always generates associated problems, for example, the electrocatalytic activity and charge transfer ability are relatively low because of its organic semiconductor nature, and the long-chain structures also hamper the effective contact with the solid-state electrolyte used in the BDSSCs. One of the compromising solutions is the integration of polymers with carbon materials into

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composites.

Graphene oxide (GO), a two-dimensional all-sp^2-hybridized carbon electrocatalyst with carbon atoms bearing various oxygen functional groups (such as hydroxyl, epoxide and carbonyl) on their basal planes and edges, has received a rapidly growing research interest [9–13]. The functionalized GO exhibits a broad electrochemical window, together with a large surface-to-volume ratios and a superior chemical stability, renders them as attractive electrocatalytic materials for lithium ion batteries [14], electrochemical supercapacitor [15,16] and bioimaging [17]. Incorporating GO nanosheets into polymer could improve the electrochemically active surface area, electrocatalytic properties [18] and thermal stability [19–21].

Herein, composite polyaniline counter electrodes with different GO electrocatalyst mass ratios of 0.2–0.8 wt% (noted as GO(0.2–0.8)/PANI) are synthesized by in-situ polymerization (Scheme 1). The introduction of GO into PANI nanoparticles greatly enhance the specific surface area and electrocatalytic activity towards the I^-/I_3^- redox reaction, and thus significantly improve the performance and stability of the composite CE. The solid-state BDSSC based on GO(0.6)/PANI CE exhibits a both-illuminated power conversion efficiency (PCE) of 5.74% with short-circuit current density of 20.23 mA cm^{-2} and open circuit voltage of 0.63 V, which is 155.6% of the conventional bare PANI assembled BDSSC under front illumination.

2. Experiment section

2.1. Fabrication of GO/PANI CEs

Homogenous composites of GO/PANI with different GO mass ratio were synthesized through in situ polymerization of aniline monomer in the presence of GO suspension. Firstly, the aniline (2.278 mL) was added into 50 mL GO suspended HCl solution and the mixture of aniline and GO was obtained after ultra sonication for 15 min. Then another 50 mL 0.5 M HCl solution containing 25 mM ammonium persulfate (APS, 98% purity, TCI) was added under vigorous stirring at 0 °C, followed by immersing a piece of FTO substrate which is cleaned subsequently in acetone, deionized (DI) water and ethyl alcohol by ultrasonicator for 15 min. After being kept for 20 min around 5 °C, the obtained GO/PANI films were washed repeatedly with DI water and 1 M HCl to remove the large aggregations and residual reagents. Finally, the GO/PANI composite films were immersed in 1 M HCl for 4 h and dried at 80 °C in vacuum for 30 min. The doping amount of GO varied as 0.2, 0.4, 0.6 and 0.8 wt%, and signed as GO(0.2)/PANI, GO(0.4)/PANI, GO(0.6)/PANI and GO(0.8)/PANI. For comparison, bare PANI was also synthesized through the above mentioned process without the presence of GO suspension.

2.2. Assembly of solid-state BDSSCs

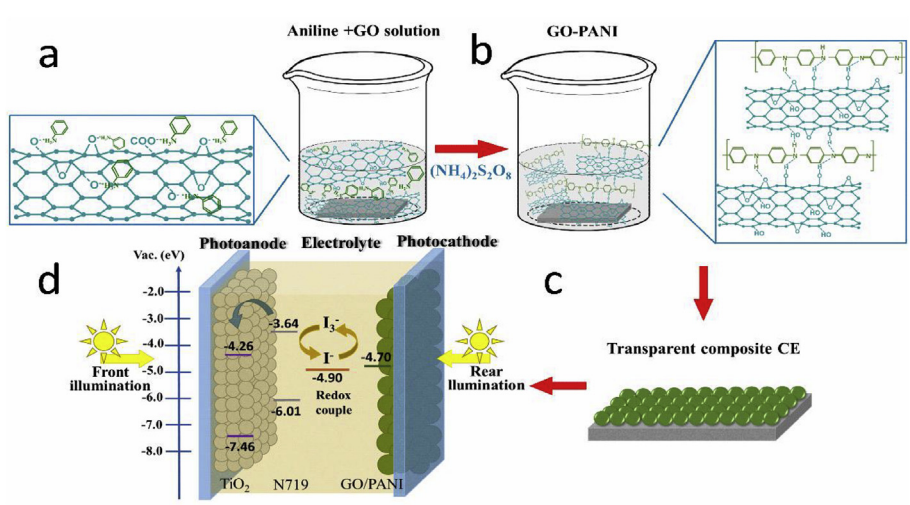
The N719 dyed TiO_2 photoanode and the agarose polymer electrolyte were prepared according to the details in previous reports [22,23]. Then the polymer electrolyte solution was dropped on the dyed TiO_2 film and when the polymer electrolyte became viscous in an oven at 75 °C, the obtained GO/PANI CE was placed on the TiO_2/agarose electrolyte surface to form a sandwich structure. Then the assembled device was dried at 65 °C for another 1.5 h–2.5 h to remove the solvent and create a solid-state BDSSC [24,25].

2.3. Material characterization

The morphology of GO/PANI surface were investigated by a Field Emission Scanning Electron Microscope (FESEM, Nova Nano SEM 230, Czech). Fourier Transform Infrared (FTIR, Thermo Nicolet Corporation, Nexus 670, America) spectra, X-ray Photoelectron Spectroscopy (XPS, ESCALAB 250Xi, America) and X-Ray Diffraction were carried out to confirm the bonding reaction between GO and PANI and chemical composition. Ultraviolet–visible spectrophotometer (UV–Vis, Hitachi, Japan) was used to measure the absorption of the GO/PANI CEs. The BET surface areas of the counter electrodes were measured with Micromeritics ASAP 2010 using N_2 gas at liquid-nitrogen temperature, and their pore size distribution was determined by the BJH method.

2.4. Electrochemical characterization

Cyclic voltammograms (CV) was tested with a three-electrode configuration using the PGSTAT302 N Electrochemical Workstation (Metrohm, AUT86802, Switzerland), in which an acetonitrile (ACN, 99.0%, SCR) was used as the electrolyte containing 0.1 M $LiClO_4$, 10 mM LiI and 1.0 mM I_2 at a scan rate of 5–80 mV s^{-1}. An



Scheme 1. The route for fabricating transparent GO/PANI composite CE: (a) monomer aniline pre-adsorbed on GO sheet and following (b) polymerization of monomer aniline on the GO basal planes and edges; (c) the prepared transparent GO/PANI composite CE; (d) schematic diagram of the BDSSC assembled with the GO(0.6)/PANI CE and the energy level diagram of the components.

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