



Multi-interfacial polyaniline-graphene/platinum counter electrodes for dye-sensitized solar cells



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ABSTRACT

Pursuit of multi-interfacial counter electrodes (CEs) for enhanced triiodide (I_3^-) reduction reaction has been a persistent objective for dye-sensitized solar cells (DSSCs). Here we report the synthesis of multilayer CEs consisting of positively charged polyaniline-graphene (PANI-G) complex and negatively charged platinum (Pt) nanoparticles. The $(\text{PANI-G/Pt})_n$ (n represents the bilayer number) multilayer displays multi-interfaces for I_3^- reduction and charge transfer. Moreover, the complexation between PANI and G can markedly accelerate the electron migration from G to PANI. The DSSC with $(\text{PANI-10wt}\%G/\text{Pt})_9$ electrode yields an impressive power conversion efficiency of 7.45% under simulated air mass 1.5 global sunlight. The promising efficiency along with cost-effectiveness and scalable materials demonstrates the multi-interfacial CEs to be good candidates for robust DSSCs.

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1. Introduction

Nowadays, dye-sensitized solar cells (DSSCs) have become one of the most promising photovoltaic devices due to their superiorities in cost-effectiveness, easy fabrication, environmental friendliness, and relatively high conversion efficiency [1–3]. Since the first prototype described by O'Regan and Grätzel in 1991 [1], a power conversion efficiency as high as 13% has been achieved on DSSCs [4]. Generally, a classical DSSC device comprises of three key components: dye-sensitized photoanode, a redox electrolyte, and a counter electrode (CE). The CE is a prerequisite to collect electrons from external circuit and to reduce triiodides (I_3^-) into iodide (I^-) species [5]. The maximum open-circuit voltage (V_{oc}) is determined by the difference between quasi Fermi energy of electrons in an anode and redox potential energy of electrolyte. Although commonly used Pt electrode exhibits excellent catalytic performance, the high expense of Pt species has restricted the commercialization of DSSCs. Therefore, it seems to be great significance to search for Pt-free or at least low-Pt electrodes.

Significant achievements have been made in previous studies on CE materials, such as Pt alloys [6–8], Pt-free alloys [9,10],

carbonaceous materials [11,12], and conducting polymers [13,14]. Among various CE candidates, carbonaceous materials and conducting polymers have applicable prospects as alternative CEs. Conducting polymers, in particular polyaniline, attract more and more research interests due to their excellent electrocatalytic abilities and extremely low cost, meanwhile they have good conductivity and environmental stability [15]. Aiming at improving the electron-migration ability, conducting polymer-carbonaceous complex materials (single wall carbon nanotube [16], multiwalled carbon nanotube [17], and graphene [18]) have been synthesized by a reflux method and employed as CE materials. In our previous work, we develop an avenue of assembling CEs with enormous interfacial area and rapid charge-transfer by a layer-by-layer self-assembly technique from polyaniline-carbon nanotube complex and graphene oxide [16]. However, graphene oxide has relatively low catalytic and conductive property, which reducing the power conversion efficiency of the DSSCs.

In search for other efficient and multi-interfacial CEs with enhanced catalytic activity and charge-transfer capability, here we have successfully synthesized polyaniline-graphene (PANI-G) complex by a reflux technique and homogeneous Pt hydrosol via reducing the chloroplatinic acid with sodium citrate. The multi-interfacial $(\text{PANI-G/Pt})_n$ electrodes are fabricated by a layer-by-layer self-assembly strategy utilizing the electrostatic interactions. Results reveal that the both PANI-G and Pt nanoparticles have acceleration effects on the electrocatalytic activity and therefore

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photovoltaic performances of solar cells. An impressive efficiency of 7.45% is recorded on the solar cells with (PANI-10wt%G/Pt)₉ electrode.

2. Experimental

2.1. Reflux synthesis of aniline-G complexes

The aniline-G complexes were synthesized by a reflux process similar to our previous work [16]. In details, three aniline-G mixtures at G dosages of 4, 8, and 10wt% were sealed in a three-neck flask filled with high-purity N₂ gas. In dark, the mixtures were refluxed for 6 h at 184 °C to obtain the target aniline-G complexes, which were subsequently stored in a dark and cold atmosphere.

2.2. Synthesis of PANi-G complexes

50 ml of aqueous solution consisting of HCl (1 M) and (NH₄)₂S₂O₈ (APS, 0.5 wt%) was dipped into 50 ml of a mixture consisting of HCl (1 M) and aniline-G (0.325 M) at 0 °C within 2 h. After reaction at 4 °C for 10 h, the resultant product was successively rinsed with 1 M HCl aqueous solution and deionized water to remove unreacted aniline monomers and oligomers. Finally, the PANi-G complexes were vacuumly desiccated at 50 °C for 24 h and then dissolved in deionized water to prepare a solution with concentration of around 1 mg ml⁻¹.

2.3. Reflux synthesis of Pt-hydrosol

A mixture consisting of 1 mL of chloroplatinic acid aqueous (0.4 mM) and 15 ml polyvinylpyrrolidone aqueous (10 mg ml⁻¹) was added into a sealed three-neck flask. Then, the mixture was heated to 100 °C with oil bath under magnetic stirring to make reactant dissolved thoroughly, subsequently adding 3 ml sodium citrate aqueous (10 mM) into the above reagent and keeping this temperature for 30 min.

2.4. Assembly of multi-interfacial (PANI-G/Pt)_n CEs

(PANI-G/Pt)_n multilayer CEs were fabricated on FTO glass substrates (sheet resistance 12 Ω sq⁻¹, purchased from Hartford Glass Co., USA). Prior to assembly, the substrates were carefully cleaned with deionized water and anhydrous ethanol. FTO substrates were subsequently immersed in a piranha solution [7/3 (v/v) of H₂SO₄/H₂O₂ at room temperature] for 1 h. To assemble CE, the pretreated FTO glass was immersed in PANi-G aqueous

solution for 5 min, rinsed with deionized water for 1 min and dried by N₂ gas stream, then immersed in Pt-hydrosol for 5 min, then rinsed again by deionized water for 1 min and dried by N₂ gas stream. By repeating the previous procedures, we can obtain FTO supported (PANI-G/Pt)_n multilayer CE (*n* represents the deposition cycle or bilayer number). The growth process was determined by recording the UV-vis absorption spectra of the multilayer CEs with a Mapada 3200 UV-visible spectrometer. The bare FTO glass substrate was used as a benchmark for each UV-vis spectrum. The errors of the absorption peak were controlled within ± 5%.

2.5. Assembly of DSSCs

FTO-coated glass substrates were cleaned by ultrasonication with acetone, ethanol and deionized water. A layer of TiO₂ nanocrystal anode film with a thickness of 10 μm and active area of 0.16 cm² was prepared by coating the TiO₂ colloid using a screen printing technique, and sintered in air at 450 °C for 30 min. Afterward, the photoanodes were sensitized with 0.5 mM N719 [cis-di(thiocyanato)-N,N'-bis(2,2-bipyridyl-4-carboxylic acid-4-tetrabutylammonium carboxylate, purchased from Dyesol LTD, Australia) in ethanol solution for 24 h at room temperature. Then the DSSC was fabricated by sandwiching a sensitized photoanode and a multilayer CE injected with redox electrolyte. A redox electrolyte consisted of 100 mM of tetraethylammonium iodide, 100 mM of tetramethylammonium iodide, 100 mM of tetrabutylammonium iodide, 100 mM of NaI, 100 mM of KI, 100 mM of LiI, 50 mM of I₂, and 500 mM of 4-tert-butylpyridine in 50 ml acetonitrile.

2.6. Electrochemical characterizations

The electrochemical properties of FTO supported (PANI-G/Pt)_n multilayer CEs were characterized by a traditional three-electrode method on a conventional CHI660E Electrochemical Workstation (CHI660E, Shanghai Chenhua Device Company, China) comprising an Ag/AgCl reference electrode, a CE of platinum sheet, and a working electrode of FTO glass supported (PANI-G/Pt)_n multilayer. The cyclic voltammetry (CV) curves were recorded at room temperature from -0.8 to +1.8 V and back to -0.8 V in a supporting electrolyte consisting of 50 mM LiI, 10 mM I₂, and 500 mM LiClO₄ in acetonitrile degassed using nitrogen before the measurement. The active area of the CE could be determined by measuring the area below the liquid electrolyte. Electrochemical impedance spectroscopy (EIS) measurements on the symmetric dummy cells consisting two identical CEs were also carried out on the CHI660E Electrochemical Workstation in a frequency range of 0.01 Hz-

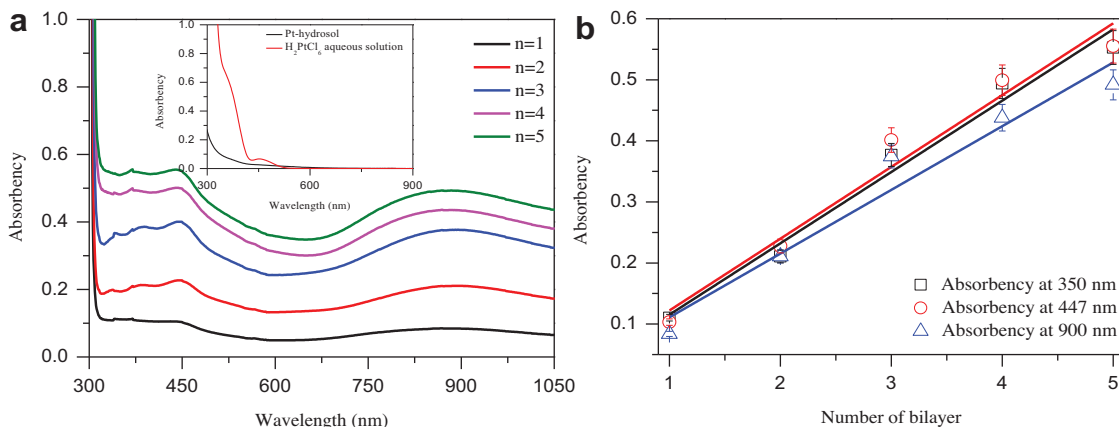


Fig. 1. (a) UV-vis absorption spectra of (PANI-10wt%G/Pt)_n (*n* = 1, 3, 5, 7, and 9) multilayers. (b) Linear plots of the absorbance intensity of PANi as a function of bilayer number. The inset in (a) represents the UV-vis absorption spectra of H₂PtCl₆ and Pt-hydrosol.

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