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High electro-catalytic counter electrode based on three-dimensional conductive grid for dye-sensitized solar cell

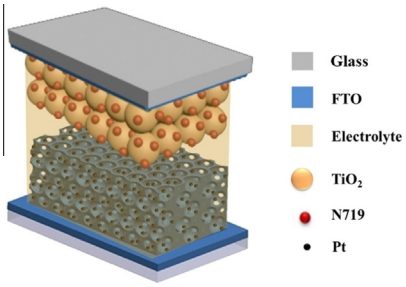
Min Wang [a], Yin Zhao [a], Shuai Yuan [a,*], Zhuyi Wang [a], Xin Ren [a], Meihong Zhang [a], Liyi Shi [a,*], Dongdong Li [b]

[a] Research Center of Nanoscience and Nanotechnology, Shanghai University, 99 Shangda Road, China
[b] Division of Energy & Environment Research, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201203, China

HIGHLIGHTS

- Three-dimensional conductive grid was constructed on the FTO glass.
- Pt with smaller size was evenly dispersed in the three-dimensional channels.
- Three-dimensional counter electrode improved the performance of DSSC.

GRAPHICAL ABSTRACT



Structure of DSSC based on 3D counter electrode

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ABSTRACT

Composite photoelectrode with Pt nanoparticles deposited on three-dimensional (3D) fluorine-doped tin oxide (FTO) conductive grid was fabricated as the counter electrode (CE) of dye sensitized solar cell (DSSC), which exhibits larger electrochemical active surface area (9.54 m^2/g) and lower charge transfer resistance (2.1 Ω) in iodide/triiodide (I^-/I_3^-) redox electrolyte than the conventional planar counter electrode. The cell efficiency of DSSC with 3D counter electrode was 8.7%, which was 14% higher than that of DSSC with planar counter electrode.

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1. Introduction

During the last decade, dye-sensitized solar cell (DSSC) has been extensively studied because of its simple and low-cost fabrication. So far, the best cell conversion efficiency is 12.3% [1]. In addition to the reduction of recombination loss in photoanode [2–4], the extensive absorption of visible light [5–7], the electro-catalytic ability of counter electrode (CE) [8,9] is also very important to increase the cell efficiency, because the regeneration rate of electrolyte in reduction state is decisive.

On the counter electrode of a DSSC, there are two reduction reactions: triiodide ions (I_3^-) are reduced to iodide ions (I^-) though getting two electrons, and iodine (I_2) is reduced to triiodide ions. The former one is the decisive step [10]. The CE works as an important part in improving the cell efficiency, which was studied to achieve high conductivity and excellent electro-catalytic properties [11].

In order to improve the performance of the cells, there are two main research directions occurred on CE. The first one is to explore

* Corresponding authors. Tel./fax: +86 21 66136082.
E-mail addresses: s.yuan@shu.edu.cn (S. Yuan), shiliyi@shu.edu.cn (L. Shi).

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new materials as catalyst, such as conducting polymers [11,12] carbon materials [13,14]. For example, Ahmad et al. prepared nanoporous poly(3,4-ethylenedioxythiophene) as CE [11]. Jo et al. constructed a mesoporous carbon–carbon nanotube composite as CE [13]. Compared with these new materials, however, Pt is still regarded as the best catalyst due to its good electro-catalytic performance and thermal stability. Therefore, it is also attractive to use the conventional Pt material to construct different nanostructures to increase the active sites and accelerate the reduction rate of I_3^-. For instance, Hsieh et al. electrodeposited Pt nanoflowers on conductive glass. The cell based on this CE exhibited high conversion efficiency [15]. Dao et al. sputtered Pt on the surface of polystyrene (PS) microspheres to fabricate CE with enlarged active area to improve the DSSC efficiency [16]. However, these methods improve the electro-catalytic activity by sacrificing more Pt. Since platinum is very expensive and has limited resources, our works pay more attention to enhance the electro-catalytic activity of Pt nanoparticles by exposing more active surface area without depositing more Pt.

The aim of this work is to construct a high electro-catalytic CE with Pt for DSSC. The strategy is to fabricate a three dimensional fluorine doped tin oxide (FTO) structure on FTO conductive glass, then introduce highly dispersed and smaller Pt nanoparticles in the pores of 3D conductive grid, which may provide more electro-catalytic sites and facilitate the transfer of electrons from CE to I_3^-.

2. Experimental details

2.1. Preparation of photoanode

Photoanodes were prepared on FTO glass substrates (purchased from Heptachroma Co. Ltd.). The FTO glass substrates were cleaned by sonication in acetone, ethanol and water, respectively. The FTO glass substrates were pretreated with 40 mM aqueous $TiCl_4$ solution at 70 °C for 30 min and then washed with water. A mesoporous TiO_2 layer with a size of 0.5 × 0.5 cm^2 were made by doctor-blading TiO_2 nanoparticle paste (purchased from Heptachroma Co. Ltd.) onto the pretreated FTO glass. The thickness of that layer was 13 μm measured with a profilometer (SGA-31). Mesoporous TiO_2 films on the FTO glass substrates were dried in an oven at 125 °C for 5 min before screen printing a scattering TiO_2 paste on them. The thickness of scattering layer was 10 μm. The resulting photoanodes were annealed in air at 500 °C for 30 min. After sintering, the electrodes were immersed in the 40 mM $TiCl_4$ solution again, followed by another heating process (500 °C, 30 min). When the temperature decreased to 80 °C, the electrodes were immersed in a dye bath containing 0.3 mM N719 in a mixture of acetonitrile and tert-butyl alcohol for 24 h. The films were rinsed thoroughly with dry ethanol, and dried by a stream of nitrogen.

2.2. Preparation of counter electrodes

2.2.1. Planar counter electrode (P-CE)

A mount of 15 μL Pt slurry (purchased from Heptachroma Co. Ltd.) was dipped on the FTO glass, dispersed in the area of 1.0 × 1.5 cm^2 uniformly. The amount of Pt slurry was 10 μL/cm^2. And then the Pt slurry was annealed in an air atmosphere at 450 °C for 30 min. The Pt precursor will turn into metallic Pt to work as a catalyst on the CE.

2.2.2. 3D counter electrode (3D-CE)

According to our previous work [3], the inversed opal three dimensional FTO conductive grid was prepared by impregnation of fluorine-doped tin dioxide precursor alcohol solution into a PS colloidal crystal template and removing the template by calcination at 500 °C for 30 min. Pt was attached to the three dimensional FTO conductive grid by the similar process as preparing planar counter electrode. The amount of Pt slurry was also fixed at 10 μL/cm^2.

2.3. Preparation of the DSSCs

Solar cells were assembled by sandwiching a 60 μm thick thermoplastic Surlyn frame (purchased from Heptachroma Co. Ltd.) between the photoanode and the CE with light pressure at 130 °C. Electrolyte was introduced through a hole in the counter electrode. The cell was sealed with Surlyn frame and a glass coverslip. The liquid electrolyte consisted of 0.1 M LiI, 0.05 M I_2, 0.5 M TBP (tert-butylpyridine), 0.6 M DMPII and 0.03 M GuSCN in acetonitrile. All chemical regents purchased from Aladdin.

2.4. Characterizations

Surface morphologies of 3D-CE were investigated using scanning electron microscopy (SEM, Hitachi S-4800) and transmission electron microscopy (TEM, JEOL JEM-2100F). The transmittance spectra measurements were performed with Shimadzu UV-3600 UV–vis spectrophotometer in the wavelength range 400–800 nm. The amount of Pt on the CE was detected by the ICP-AES method (ICAP 6300).

2.5. Measurements of electrochemical properties

Cyclic voltammetry (CV, CHI660D) was measured in a three-electrode system contained 0.1 M $LiClO_4$, 10 mM LiI and 1 mM I_2 in an acetonitrile solution. The CE worked as a working electrode. A Pt foil served as a counter electrode. The Ag/Ag^+ couple was used as a reference electrode. The effective electrochemical surface area (ECSA) of Pt was evaluated by the CV measurement in a 0.5 M H_2SO_4 solution at a scan rate of 500 mV s^{-1} with a standard three-electrode cell. A Pt foil worked as the counter electrode with a saturated calomel electrode (SCE) as the reference. The electrochemical impedance spectroscopy (EIS, PARSTAT 2273 Advanced Electrochemical system) of DSSC was performed in symmetrical cells. Two identical electrodes were assembled and sealed with a Surlyn frame (60 μm). The frequency ranged from 0.1 Hz to 10^6 Hz, with an alternative potential at 5 mV. The EIS data was fitted with Z-view simulation software.

The photocurrent density–voltage curves of the devices were measured under one sun AM-1.5 G irradiation with Oriel Sol3A Class AAA Solar Simulators, which was calibrated with a silicon-based reference cell. The incident photon-to-current conversion efficiencies (IPCE) of the corresponding devices were recorded with QEX10 Quantum Efficiency Measurement System.

3. Results and discussion

3.1. Structure and component of counter electrodes

The ordered structures of the PS colloidal crystal template and as-prepared 3D FTO conductive grid were revealed by SEM images. Fig. 1a shows the hexagonal close-packed PS spheres (560 nm) patterned on FTO substrate. In the ideal 3D face-centered cubic (FCC) model, one sphere is surrounded by twelve spheres, and 74% volume is occupied by the spheres, and the left 26% void volume can facilitate the infiltration of FTO precursor [3]. As a result, the corresponding replication of 3D FTO conductive grid, has an ordered structure similar to that of the original templates (Fig. 1b). The average diameter of the opal macropores is about 380 nm, which means the calcination-induced shrinkage is 32%. According to Fig. 1b, the sphere-like cells of 3D ordered macroporous FTO conductive grid

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