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High performance platinum nanofibers with interconnecting structure using in dye-sensitized solar cells



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ABSTRACT

Platinum nanofibers (PtNFs) with interconnecting structure are prepared by a simple electrospun method and novel film forming technique, which are used as the counter electrode (CE) materials for the dye-sensitized solar cells (DSSCs). Electron microscope images demonstrate that PtNFs are constituted by Pt nanoparticles. Cyclic voltammetry measurements indicate that PtNF CE has an excellent catalytic activity for the reduction of triiodide to iodide. DSSC based on PtNF CE achieves an enhanced photovoltaic conversion efficiency of 9.31% compared to that of DSSC based on Pt CE (7.32%) prepared by the thermal decomposition, owing to the three-dimensional interconnecting nanostructure has abundant catalytic surfaces, large contact area with the electrolyte, and lots of electronic transmission channels. DSSC efficiency based on PtNF CE remains 90.08% after continuous scanning 1000 s under an open system without any sealing in ambient atmosphere, and 83.19% of the efficiency is remained after 1000 h under a strictly packaging for the device application.

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1. Introduction

Dye-sensitized solar cell (DSSC) has attained momentous interests as a promising type of renewable energy device, with the most efficient model utilizing a mesoporous dye-sensitized titanium dioxide (TiO₂) photoanode, a counter electrode (CE), and an iodine based electrolyte between the two electrodes [1-4]. In a DSSC, the major task of CE is to speed up the reduction of triiodide (I₃) to iodide (I⁻) with electrons to complete the electric circuit, and a CE with a highly catalytic activity reduces the DSSC's internal series resistance, resulting in a high fill factor [5,6].

Despite vast efforts in the past decades to develop low platinum (Pt) composite catalysts [7,8] and Pt-free catalytic materials [9–16], Pt remains the optimal choice for the catalytic material in the DSSC. For example, Pt–Ru nanofiber alloys were prepared by a hydro-thermal method and employed as CEs for DSSCs, and the DSSC based on PtRu₃ alloy CE obtained 6.80% of the efficiency [17]. Recently, we prepared cubic Pt nano-crystals with slender tentacles by a hydrothermal method in the oleylamine, which had good electrocatalytic activity toward I_3 in DSSCs, and the CE achieved

9.24% of the photovoltaic conversion efficiencies, higher than that of the thermal decomposition Pt CE (7.26%) [18]. Therefore, design and synthesis of Pt catalyst with particular nanostructure is still a challenge to further improve its intrinsic activity [19–21].

Electrospinning is a facile and scalable method to produce nanofibers or ultrafine fibers [7,22,23]. Qiao and coworkers fabricated Pt-carbon nanofibers by using an electrospun method, and based on which the DSSC produced about 8% of the efficiency [7,24]. However, the pure Pt nanofibers (PtNFs) by the facile electrospun method were lack of enough attention and research. Herein, we prepared the pure PtNFs with an interconnecting nanostructure by the simple electrospun method. The PtNFs based CE had an excellent catalytic activity for the reduction of triiodide to iodide, and the DSSC with the PtNFs CE achieved an enhanced photovoltaic conversion efficiency of 9.31%, which was higher than that of the DSSC with the thermal decomposition Pt (TDPt) CE (7.32%).

2. Experimental

2.1. Materials

Chloroplatinic acid ($H_2PtCl_6 \cdot 6H_2O$), polyvinylpyrrolidone (PVP, Mw = 1,300,000), isopropyl alcohol, iodine, lithium iodide, lithium pechlorate, tetrabutyl ammonium iodide, 4-*tert*-butylpyridine,



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ganidine thiocyanate, and acetonitrile were purchased from Shanghai Chemical Agent Ltd., China (Analysis purity grade). Dye N719 was purchased from Dyesol, Australia. The above agents were used without further purification.

2.2. Electrospun preparation of PVP-H₂PtCl₆ fiber films

Chloroplatinic acid (with a quantity of 0.30×10^{-6} , or 0.60×10^{-6} , or 0.90×10^{-6} , or 1.20×10^{-6} mol) was severally dissolved into 1.0 mL isopropyl alcohol, then 100 mg PVP was added and stirred well to obtain the electrospun mixture. The electrospun mixture was offered by a stainless-steel syringe pipette needle with a rate of about 1.0 mL h⁻¹, carrying a potential of +15.0 kV. PVP-H₂PtCl₆ fibers were collected on a sheet of electrically grounded aluminum foil placed 15 cm below the syringe tip. The resultant fibrous films based on different quantity of H₂PtCl₆ were peeled off from the collector and then dried at room temperature under ambient atmosphere, and named as a, b, c, and d.

2.3. Preparation of PtNF CEs

The CE was prepared by following four steps: First, the four PVP-H₂PtCl₆ fiber films (a, b, c, and d) were severally cut into an appropriate size (0.5 cm \times 0.6 cm); Second, the cleaned F-doped tin oxide (FTO) substrate (1.5 cm \times 2 cm, 12 Ω sq⁻¹, NSG, Japan) was treated by water vapor to form a very thin water film; Third, the tailored PVP-H₂PtCl₆ fiber film was guickly adhered on the moisture FTO surface. As we all known, PVP and H₂PtCl₆ are water soluble, when pasting the PVP-H₂PtCl₆ fibers onto the surface of the humid FTO glass, the lower surface of the PVP-H₂PtCl₆ fibers is partly dissolved in the water, and then acts as the adhesion agent to enhance the adhesion between PVP-H₂PtCl₆ fibers and FTO substrate. Finally, the achieved samples were sintered at 450 °C for 30 min under air atmosphere, and named as PtNF-a, PtNF-b, PtNF-c, and PtNF-d, respectively. For comparison, a thermal decomposition Pt (TDPt) CE was employed, which was prepared by dipping H_2 PtCl₆ isopropanol solution with a concentration of 0.50 wt% onto the FTO surface, and then heated at 450 °C for 30 min.

2.4. Assembly of DSSCs

TiO₂ photoanodes were prepared by a blade-coating method according to our previous works [3,18]. Then TiO₂ photoanodes were sensitized by immersing them into a 0.3 mM N719 dye of ethanolic solution for 24 h. After that the DSSC was assembled by sandwiching redox electrolyte between dye-sensitized TiO₂ photoanode and FTO supported TDPt or PtNF CEs under an open system without any sealing. The redox electrolyte composed of 0.10 M lithium iodide, 0.10 M iodine, 0.60 M tetrabutyl ammonium iodide, 0.50 M 4-*tert*-butyl-pyridine, and 0.10 M ganidine thiocyanate in acetonitrile.

2.5. Characterizations and measurements

The surface morphologies of the PtNF CEs were observed using a field emission scanning electron microscopy (FESEM, JEOL-JSM-6701F) operating at 10 kV and a transmission electron microscope (TEM, JEOL-JSM-2100) operating at 200 kV. The specific surface area was measured by Brunauer-Emmet-Teller (BET) nitrogen adsorption-desorption (Micromeritic ASAP 2020 Instrument). Cyclic voltammograms (CVs) were carried out with a three-electrode configuration in an acetonitrile solution consisting of 0.05 M lithium iodide, 0.01 M iodine, and 0.05 M lithium pechlorate, using a computer-controlled potentiostat (Autolab Type III), the resultant CE acted as the working electrode, a Pt-foil as counter

electrode, and a Pt-wire as reference electrode. Electrochemical impedance spectroscopy (EIS) for the symmetrical CEs (CE//electrolyte//CE) was performed conducted by using a Zahner Zennium electrochemical workstation at a constant temperature of 20 °C in ambient atmosphere under a dark condition, and the impedance data covered a frequency range of 0.1–10⁵ Hz with 5 mV of amplitude and 0 V bias potential. Using a Z-view program, the fitted EIS parameters were obtained. The incident monochromatic photon-to-current conversion efficiency (IPCE) curves of DSSCs were carried out with a Zennium CIMPS-pcs2 (Zahner) system established with the tunable light source (TLS03). The photocurrent density-voltage characteristic of the DSSC was carried out using a CHI660D under an air mass (AM) of 1.5 G simulated sunlight using a solar simulator (CEL-S500, Beijing Ceaulight Science and Technology Ltd., China). The active cell area and the incident light intensity were 0.30 cm² and 100 mW cm⁻², respectively. Impedance, photocurrent density, and open-circuit potential as a function of time for the DSSC held at 0 V forward bias was measured using the CHI660D system under AM1.5 simulated sun light. In which the frequency for the Impedance-Time test was set at 1000 Hz.

3. Results and discussion

Fig. 1a-d show the FESEM images of PtNFs based on different quantity of H₂PtCl₆·6H₂O. PtNFs are established by many Pt nanoparticles interconnecting together with each other. The forming processes of this interconnecting nanostructure might be caused by two factors: first, the electrospinning PVP-H₂PtCl₆ fibers are made up of cross structure during the collecting process on the aluminum foil; second, the water vapor step to form the PVP-H₂PtCl₆ NF film adhered on the moisture FTO surface will bring about partial dissolution of the PVP-H₂PtCl₆ fibers, therefore forming the interconnecting nanostructure after the sintering process. The electrospun mixture with low concentration of Pt source leads to form a small number of interconnected Pt nanoparticles (Fig. 1a). The more quantity of Pt source results in the easier forming three-dimensional (3D) nanofiber structure with more interconnected Pt nanoparticles (Fig. 1b-d). Fig. S1 shows FESEM image of the coralline-like TDPt CE with a great numbers of openings on FTO glass substrate, in which the openings could reduce its active surface area. Moreover, TEM images demonstrate that PtNFs have interconnecting morphology and Pt nanoparticles size of about 10-40 nm (Fig. 2). The composition of PtNFs was analyzed using a powder X-ray diffraction (XRD, BRUKER D8-ADVANCE) (Fig. S2). The three main XRD peaks-(111), (200), and (220)—are located for the PtNFs [18,25,26].

Fig. 3 compares CV of CE materials to evaluate the catalytic activity of electrodes. Two typical pairs of peaks in the iodine-based solution are observed for all the CEs, the relative negative pair is assigned to the redox reaction of $I_3^- + 2e^- \leftrightarrow 3I^-$, which is the effective redox reaction in the DSSC [3,27]. The positive pair is attributed to the redox reaction of $3I_2 + 2e^- \leftrightarrow 2I_3^-$, which has little effect on the performance of the DSSC. The peak current densities from the relative negative pair show an order of PtNF-c > PtNFd > TDPt > PtNF-b > PtNF-a, indicating the same order of the catalytic activity for the CEs [28,29]. The peak-to-peak separation (E_{nn}) is depended on the rate of electron transfer and the porosity of electrode [30]. The E_{pp} decreases in the order of TDPt (805 mV) > PtNF-a (734 mV) > PtNF-b (671 mV) > PtNFd (644 mV) > PtNF-c (612 mV) (Table 1), indicating the PtNF-c CE shows the lowest overpotential loss. The largest electrocatalytic activity of the PtNF-c CE is probably due to its 3D interconnecting nanostructure, which results in abundant catalytic surfaces, large contact area with the electrolyte, and a large number of electronic transmission channels, as demonstrated in FESEM and TEM results.

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