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A transparent cobalt sulfide/reduced graphene oxide nanostructure counter electrode for high efficient dye-sensitized solar cells

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ABSTRACT

A transparent nanostructured cobalt sulfide/reduced graphene oxide (CoS/rGO) hybrid film is prepared by a simple hydrothermal method in alkaline solution and is used as counter electrode (CE) for dye-sensitized solar cell (DSSC). The surface morphology and micro structures of film are characterized by field emission scanning electron microscopy and Raman spectroscopy. The electrochemical properties of the CEs are measured by cyclic voltammetry, electrochemical impedance spectroscopy and Tafel tests. The photovoltaic performance of the DSSCs with CoS/rGO CEs is investigated. Owing to good transmission conductivity and electrocatalytic activity for CoS/rGO film, the DSSC based on CoS/rGO CE with the thickness of 72 nm achieves power conversion efficiency (PCE) of 9.82% and 8.38% under white light irradiation of 100 mW cm^{-2} (AM 1.5 G) from the front and back illumination, which are higher than the PCE of DSSC with Pt CE. The results indicate that CoS/rGO hybrid film is a low-cost, Pt-free and high efficient CE for DSSCs.

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1. Introduction

In renewable energy, solar energy resource is very rich and clear, which has attracted great attention in the whole world [1]. And photovoltaic device is an important way to the using of solar energy. Science the first report of Grätzel in 1991 [2], dye-sensitized solar cells (DSSCs) had been great development due to low cost, easy fabrication, and high light-to-electric conversion efficiency [3–5]. Counter electrode (CE) is an integral part of DSSC and the function of CEs is to collect the electrons from external circuit and catalytic the reaction of I_3^- to I^-. Platinum (Pt) is a common material for CE because of its encouraging electro-catalytic activity and conductivity. However, Pt is a noble material and it is easily corroded by I_3^-/I^- electrolyte [6]. This prompted scientists to explore new materials to replace Pt, such as carbon materials [7], transition-metal carbide [8], -oxide [9], -nitride [10], -sulfide [11], -selenide [12], conductive polymers [13,14], and hybrid materials [15,16].

Among many transition-metal compounds, cobalt sulfide (CoS) is a super star to be the CE material with good catalytic activity. However the grain boundaries and defects between the CoS nanoparticles make the conductivity of CoS not very well. To further improve the electrocatalytic activity and conductivity of CoS CE, some other materials especially graphene were introduced to be CE [17,18]. Graphene is a special kind of carbon materials and usually used to be CE material bacause of its exceptional surface area, high optical transparency, and exceptional conductivity [19–21]. Some studies showed that the CoS and graphene hybrid CE not only increased the active sites of graphene for reducing I_3^- but also improved the conductivity of CE [22]. Gao et al. [23] used a facile hydrothermal method to synthesize CoS_2-graphene composites CE, which obtained power conversion efficiency (PCE) of 6.55%, compared with that of Pt CE (6.20%). This method was simple and easy to control the size and distribution of CoS_2 onto graphene composites. However, the problems of π-π stack of graphene sheets and low conductivity existed. In 2015, Sun et al. [24] successfully synthesized CoS_2-reduced graphene oxide (rGO) composite (CSG) by the layer by layer assembly method and thermal decomposition process. the DSSCs with CSG CEs exhibited an excellent PCE of 5.43%, which was comparable to that of DSSCs with Pt CE (5.84%). Zhu et al. [25] presented a facile electrochemical strategy to fabricate the rGO-CoS CEs, and DSSCs with this CE would obtain a higher PCE (8.34%) than that of DSSCs with Pt CEs

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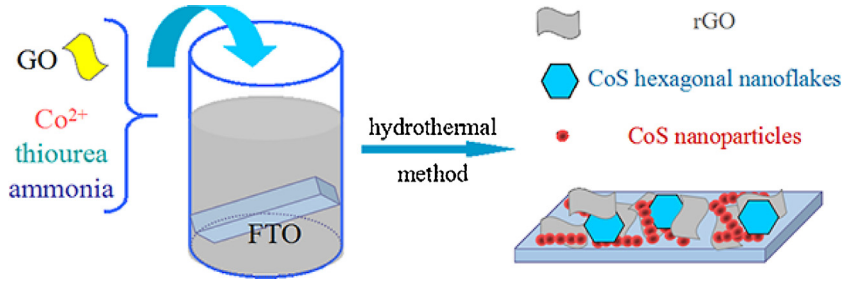

Scheme 1. Fabrication a CoS/rGO CE by a simple hydrothermal method.

(7.50%). There are many methods to prepared CoS CE, for example, electrodeposition [26,27], two-step approach including chemical bath deposition and a pyrolysis treatment [28]. Here, we use an improved hydrothermal method with a certain amount of ammonia solution to prepare a transparent CoS/rGO hybrid film on FTO glass. This thin film can increase the conductivity and electrochemical performance of CE. The DSSC using this CoS/rGO CE possesses high short-circuit current density (18.903 mA cm^{-2}) and excellent PCE (9.82%), which is higher than that of DSSC with Pt CE (8.38%).

2. Material and methods

2.1. Materials

The expanded graphite (~300 μm flakes) was purchased from Qingdao Graphite Company, Qingdao, Shandong, China. The potassium permanganate ($KMnO_4$), concentrated sulfuric acid (H_2SO_4 98 wt% aqueous solution), phosphoric acid (H_3PO_4), 30% hydrogen peroxide (H_2O_2), 30% hydrochloric acid (HCl), cobalt chloride hexahydrate ($CoCl_2{\cdot}6H_2O$), thiourea, ammonia solution, tetra-n-butyl titanate, oleic acid, cyclohexane, octadecylamine, toluene, titanium tetrachloride ($TiCl_4$), absolute ethyl alcohol, acetonitrile, tetraethyl ammonium iodide, tetrabutyl ammonium iodide, tetramethyl ammonium iodide, 4-tert-butyl pyridine (TBP), sodium iodide (NaI), potassium iodide (KI), lithium iodide (LiI), lithium perchlorate ($LiClO_4$) and iodine (I_2) were all A. R. grade and purchased from Sinopharm Chemical Reagent Co. Ltd, China. N719 dye (Cis-di(isothiocyanato)-bis-(2, 2′-bipyridyl-4, 4′-dicarboxy-lato) ruthenium (II) bis-tetrabutylammonium) was obtained from Dye sol.

2.2. Preparation of counter electrodes

The graphene oxide (GO) was prepared by improved Hummer's method [29]. A mixed acid of concentrated H_2SO_4 and H_3PO_4 (360:40 mL) was slowly added to the mixture of expanded graphite and $KMnO_4$ (3:18 g) in an ice bath. Then the mixture was heated to 50 °C and stirred 12 h. After the temperature down to room temperature, the mixture was added to 400 mL of ice water, and dropwised added 30% H_2O_2 until the solution was yellow. The yellow solution was centrifuged by 200 mL 30% HCl aqueous solution and deionized water until the pH value of solution equaled to seven. The obtained solid was dried at 60 °C, getting 4.5 g graphite oxide. The graphite oxide aqueous solutions of different concentrations were treated by ultra-sonic oscillation for about 10 h, and the GO aqueous suspension was obtained.

CoS/rGO CE was fabricated by a simple hydrothermal method and is shown in Scheme 1. In general process, the GO aqueous solution (55 mL) containing 0.1 mmol $CoCl_2{\cdot}6H_2O$ and 0.15 mmol thiourea was added 15 mL ammonia solution and under vigorous stirring for 10 min. Then the mixture was transferred into a Teflon liner of 100 mL capacity and a piece of FTO glass with FTO layer downward was loaded into this liner at a certain angle. After that, the liner was sealed in a stainless steel autoclave and maintained at 150 °C for 12 h. When the temperature of stainless steel autoclave cooled to room temperature naturally, the FTO glass was took out and washed with deionized water and the CoS/rGO counter electrode was prepared. According to the different concentration of GO aqueous solution, these CEs were denominated as CoS/rGO_x (x = concentration of GO aqueous solution, mg mL^{-1}) CEs, respectively. For comparison, rGO CE was prepared only by 55 ml 0.10 mg mL^{-1} GO aqueous solution and 15 mL ammonia solution.

2.3. Fabrication of DSSCs

A DSSC was fabricated with a dye sensitized TiO_2 photoanode, a CE and a redox electrolyte injecting into the gap between the

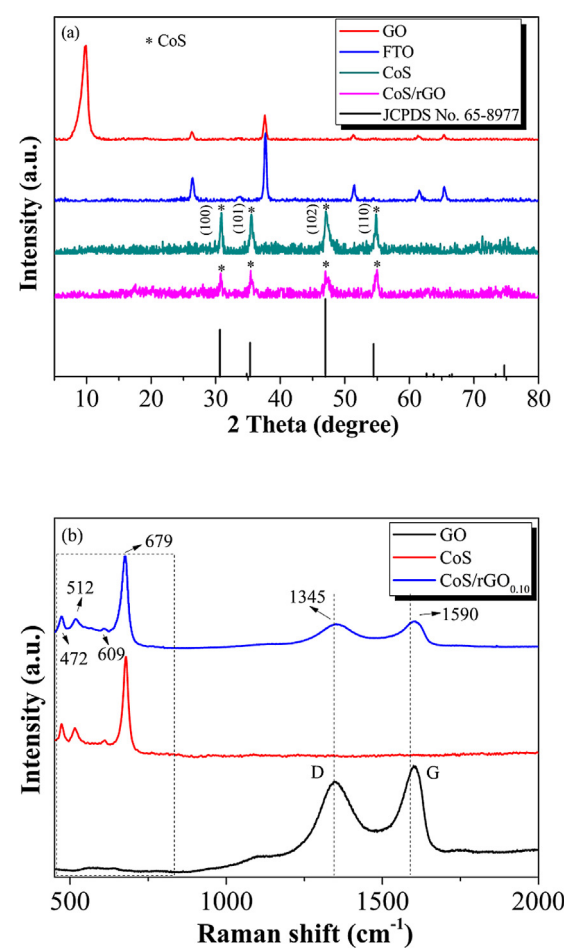

Fig. 1. (a) XRD of GO on FTO, FTO, CoS and CoS/$rGO_{0.1}$ samples and (b) Raman spectra of GO, CoS and CoS/$rGO_{0.10}$ CEs.

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