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Cobalt telluride/reduced graphene oxide using as high performance counter electrode for dye-sensitized solar cells

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ABSTRACT

A cobalt telluride/reduced graphene oxide (CoTe/RGO) hybrid is synthesized by hydrothermal method and used as counter electrode in dye-sensitized solar cell (DSSC). The synthesized samples are characterized by field emission scanning electron microscopy and transmission electron microscope, which shows that CoTe/RGO has a larger specific surface area than CoTe. Cyclic voltammogram measurement indicates that CoTe/RGO electrode has larger current density and better electrocatalytic activity in the reduction of triiodidie than Pt electrode. Electrochemical impedance spectroscopy shows that CoTe/RGO electrode has lower charge-transfer resistance (2.94 $\Omega\,cm^2$). Owing to above advantages, the DSSC based on CoTe/RGO counter electrode achieves a high power conversion efficiency of 9.17%, outperforming that based on Pt counter electrode (8.17%). Therefore, inexpensive CoTe/RGO is a promising counter electrode to replace noble metal Pt.

1. Introduction

To comply with environment-friendly and low-carbon mainstream in the future, large amount of clean energy resources have emerged. Comparatively solar energy is a sustainable energy source available in our daily life and hence has received tremendous interest in recent years. Dye-sensitized solar cell (DSSC) is famous for low-cost production, preparation without difficulty, outstanding photovoltaic characteristic and environmental benignity [1–3]. Recently, Simon et al. reported a DSSC with power conversion efficiency (PCE) of 13% through the molecular engineering of porphyrin sensitizers and a Co(II/III) tris(bipyridyl)-based redox electrolyte [4]. As a key component, the role of counter electrode (CE) is in charge of the reduction of triiodide (I_3^-) to iodide (I^-) at CE/electrolyte interface. Hence, an efficient CE should have outstanding electrocatalytic activity, high electrical conductivity, and good stability [5]. So far, Pt CE is widely used in high-efficiency DSSCs due to its excellent conductivity and high catalytic activity toward I^-/I_3^- couple [6]. However, Pt CE used in DSSCs has some disadvantages. Pt is expensive and can be decomposed to PtI_4 or H_2PtI_6 by I^-/I_3^- redox couple [6], which is not conducive to the application of Pt CE in DSSCs.

In order to overcome these disadvantages, numerous attempts have been done to explore competent substitutes for Pt [5]. The substitutes reported so far include carbon materials [7–10], conducting polymers [11–14] and inorganic compounds such as nitride [15,16], carbide [17,18], etc. Metal chalcogenide is a prospective CE material in DSSCs owing to their high conductivity and excellent catalytic activity for the reduction of I_3^- [5]. Among them, the metal sulfides are the most researched CE materials [19–21]. Metal oxides and selenides also are used as CE materials in DSSCs [22–24]. However, the research for metal tellurides is few, although they have the potential to be a good counter electrode material in DSSCs [25]. Here, a cobalt telluride/reduced graphene oxide (CoTe/RGO) hybrid is synthesized by hydrothermal method and used as counter electrode in DSSC. The counter electrode shows low resistance and prominent catalytic activity for reduction of I_3^-. The DSSC based on CoTe/RGO counter electrode achieves a power conversion efficiency of 9.17%, which is higher than the DSSC with Pt electrode (8.17%).

2. Experimental

2.1. Preparation of CoTe CE

The feasible preparation procedure of cobalt telluride (CoTe) is similar with previous work [26]. Sodium tellurite (Na_2TeO_3, 0.005 mol) was mixed with cobalt sulfate ($CoSO_4$, 0.005 mol) in 60 mL distilled water. Then, 20 mL of hydrazine hydrate

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($N_2H_4{\cdot}H_2O$) was added under stirring. After stirring for 30 min, the mixture was transferred into a 100 ml Teflon-lined autoclave. After the hydrothermal reaction at 140 °C for 8 h, the CoTe was produced and the precipitation was centrifuged with a rate of 5000 rpm. The precipitation was washed with deionized water for several times. The black product was dispersed in ethanol to form a 0.04 g mL^{-1} ink. The black CoTe ink was spin-coated on the clean FTO glass substrate at a rate of 2000 rpm for 20 sec. After that, the FTO covered with CoTe was exposed to 150 °C for 2 h under a nitrogen atmosphere, thus a CoTe CE was obtained.

2.2. Preparation of CoTe/RGO CE

Graphene oxide was prepared by using a modified Hummers method [27–29]. Under ultrasonic vibrating, the prepared graphene oxide was dispersed in distilled water to form a graphene oxide (GO) solution (2mg mL^{-1}). Cobalt telluride/reduced graphene oxide (CoTe/RGO) hybrid and CE were prepared by using similar method as the above experimental procedures for CoTe except that 60 mL of distilled water was replaced with 60 mL GO solution.

2.3. Fabrication of TiO_2 photoanode

The TiO_2 film of photoanode comprises a compact TiO_2 block layer and mesoporous TiO_2 layer. The preparation of block layer and mesoporous layer are described in detail in our previous work [30,31]. In order to improve the surface state of the film, the film was immersed into a $TiCl_4$ solution (50 mM) for 25 min at 80 °C, and then annealed at 500 °C for 30 min. After above-mentioned treatment, the film was immersed into dye N719 ethanol solution (0.25 mM) for 24 h, thus a dye-sensitized photoanode was obtained.

2.4. Assembly of DSSC

A dye-sensitized solar cell (DSSC) was assembled by injecting liquid electrolyte into the gap between the dye-sensitized TiO_2 photoanode and the CE. After injecting, a cyanoacrylate adhesive was served as sealant to avoid electrolyte leakage. The redox electrolyte contained 0.5 M sodium iodide, 0.1 M tetrabutyl ammonium iodide, 0.5 M 4-tert-butylpyridine, and 0.05 M iodine in acetonitrile solution.

2.5. Measurements

The surface morphologies of CoTe and CoTe/RGO were investigated with a field emission scanning electron microscopy (FESEM, SU8010, HITACHI) and transmission electron microscope (TEM, JEM-2100, Japan). The specific surface area of sample was determined by ASAP2020HD88. The compositions of prepared samples were determined by energy dispersive spectroscopy. The phase and structure of products were characterized with an X-ray diffractometer (XRD, Smart Lab 3 kW, Rigaku, Japan). Photovoltaic tests were carried out by measuring the current-voltage (J-V) characteristic curves on a Keithley 2400 source meter under a simulated solar light irradiation with intensity of 100 mW cm^{-2} (AM 1.5) from an AAA solar simulator (Newport-94043A, USA) equipped with a Xe lamp (450 W). The cyclic voltammetry (CV) curves were obtained from an electrochemical workstation (CHI 660E, China). The testing system included a saturated Ag/AgCl reference electrode, the prepared sample as working electrode, a platinum sheet counter electrode, and an acetonitrile electrolyte containing 0.1 M $LiClO_4$, 10 mM LiI and 1 mM I_2. Electrochemical impedance spectroscopy and Tafel polarization curves were carried out on the same workstation (Zennium/IM6, Zahner, Germany) by applying an AC voltage of 5 mV amplitude and at 0 V bias in the frequency range between 100 mHz and 100 kHz at room temperature.

3. Results and discussion

3.1. Structure and morphology

Fig. 1 (a) shows the X-ray diffraction (XRD) patterns of the as-prepared CoTe, CoTe/RGO samples and the standard CoTe. The peak positions as-prepared CoTe, CoTe/RGO powder can be well indexed to the standard CoTe (PDF#34-0420). On the other hand, it can be clearly observed that the incorporation of reduced graphene oxide results in the weakening of the diffraction intensity for CoTe/RGO sample. Compared the as-prepared samples with the standard sample, no other obvious diffraction peaks appear, which indicates that the as-prepared samples have high purity. The chemical compositions of as-prepared CoTe was determined by energy dispersive spectroscopy (EDS) spectrum and shown in Fig. 1 (b). The results display that the atomic ratios of Co:Te is 0.96. The measured atomic ratios are close to the stoichiometry of CoTe, which is consistent with the analysis of XRD.

Field emission scanning electron microscopy (FESEM) and transmission electron microscope (TEM) images of the as-prepared CoTe and CoTe/RGO are shown in Fig. 2. As shown in Fig. 2 (a) and (c), it can observe that the as-prepared CoTe shows a one-dimensional rod configuration. Interestingly, when RGO is incorporated into CoTe, the rod configuration is changed into wire configuration, from Fig. 2 (b) and (d). The wire-like CoTe/RGO

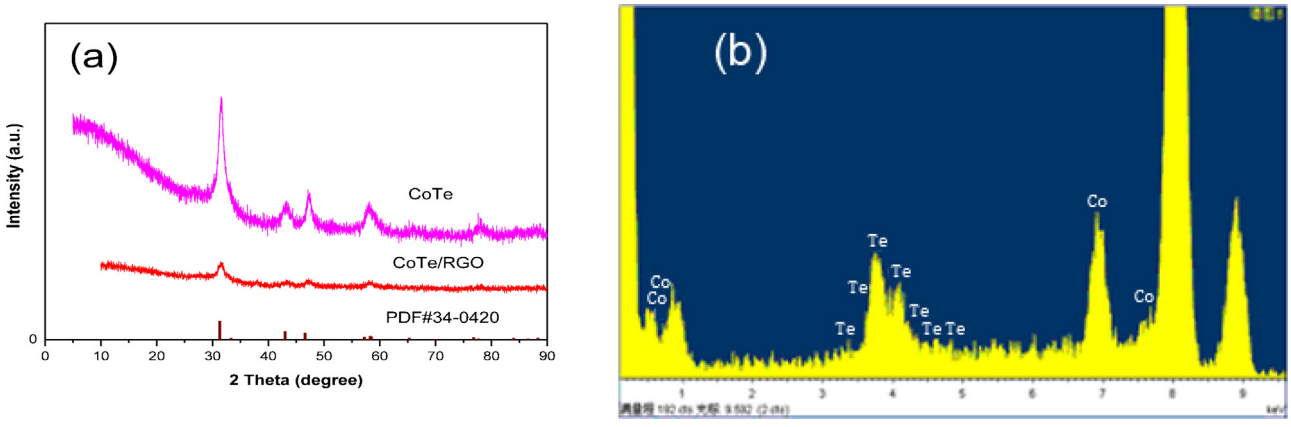

Fig. 1. (a) XRD and (b) EDS patterns of cobalt telluride.