



Transparent molybdenum sulfide decorated polyaniline complex counter electrodes for efficient bifacial dye-sensitized solar cells



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ARTICLE INFO

Article history:

Received 13 January 2017

Received in revised form 27 February 2017

Accepted 19 March 2017

Keywords:

Bifacial dye-sensitized solar cells

Transparent counter electrode

Polyaniline-molybdenum sulfide complex

Electrocatalyst

ABSTRACT

Exploration of cost-effective and transparent counter electrodes (CEs) with high electrocatalytic activity has been a persistent objective of bifacial dye-sensitized solar cells (DSSCs) development. Here, with an aim of accelerating charge transfer and increasing the active sites of a transparent CE, molybdenum sulfide (MoS₂) decorated aniline complexes are synthesized by a reflux technique and subsequently in-situ polymerized for transparent polyaniline (PANI)-MoS₂ complex CEs for efficient bifacial DSSCs. The preliminary results indicate that the electrocatalytic activity toward I₃⁻ reduction of PANi-MoS₂ complex CE is dramatically enhanced due to the fast charge transfer between PANi (N atoms) and MoS₂ (Mo atoms) by the metal (dπ)-nitrogen (pπ) antibonding interaction. Owing to the high optical transparency, electrocatalytic reduction toward I₃⁻ species, superior charge-transfer ability for I⁻/I₃⁻ redox couples, the bifacial DSSCs based on PANi-6 wt% MoS₂ complexes CE yield a maximum power conversion efficiency of 7.99% from front irradiation, 3.40% from rear irradiation and 9.71% from both irradiation, which are higher than front, rear and both efficiencies of 6.37%, 1.78% and 7.50% for DSSC employing PANi CE, respectively. The high optical transparency and electrocatalytic activity along with simple preparation, relatively low cost and scalability demonstrate the potential use of PANi-MoS₂ complex as a robust CE in bifacial DSSCs.

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1. Introduction

Dye-sensitized solar cells (DSSCs), as the third-generation solar cell, have emerged as one of the most promising solution to global energy consumption and environmental problems owing to their low cost, environmental friendliness, easy fabrication and relatively high power conversion efficiency (O'Regan and Grätzel, 1991; Mathew et al., 2014). A standard DSSCs device consists of three components: a dye-sensitized nanocrystalline semiconductor photo-anode, an electrolyte containing triiodide/iodide (I⁻/I₃⁻) redox couple and a counter electrode (CE). The CE, as a crucial component in DSSCs, acts as a key role of rapidly collecting electrons from external circuit and catalyzing the reduction of triiodide to iodide at the CE/electrolyte interface. Platinum (Pt) is commonly used as the CE catalytic material owing to its excellent catalytic activity for the reduction of triiodide and high electrical conductivity (C.-C. Wang et al., 2013; Tang et al., 2013). However, the high cost and limited reserves of Pt CE have restricted the commercial application of DSSCs. It is very necessary to exploit other cost-effective and high electrocatalytic CE materials to replace the

traditional Pt. In recent years, some alternative CE materials are investigated, such as carbonaceous materials (H. Ren et al., 2015; G.-Q. Wang et al., 2013), selenides (Duan et al., 2014b), alloy compound (Zhang et al., 2015a,b; Chen et al., 2014; He et al., 2014a,b,c, 2015, 2016; Tang et al., 2015), conducting polymers (Yu et al., 2016; Lin et al., 2015), and polymer/carbonaceous composites (Zhang et al., 2015a,b; He et al., 2014d,e,f,g), which exhibits excellent electrocatalytic activity for I₃⁻ reduction and high electrical conductivity for electrons transfer.

Among various alternative CE materials, conducting polyaniline (PANI) has been considered as one of the most preferred CE materials due to its easy fabrication, low cost, environmental stability, high electrocatalytic activity and transparency. Especially for the high electrocatalytic activity and transparency of PANi, it has been used as transparent CE for bifacial DSSCs (Tai et al., 2011; J.H. Wu et al., 2014). Bifacial DSSCs based on transparent CEs can be irradiated from either front or rear side, which has been introduced by Grätzel and co-workers to help increasing light harvesting efficiency and reducing the cost of solar-electric conversion (Ito et al., 2008), and a bifacial DSSC collects sunlight from either of its two sides, facilitating practical applications (Bisquert, 2008). PANi has been deposited on FTO by Zhao et al. as a transparent CE for bifacial DSSC, yielding front and rear-irradiated power

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conversion efficiency of 6.54% and 4.26%, respectively (Tai et al., 2011). In order to compensate for the excessive loss of incident light within the dye-sensitized photo-anode film and enhance dye molecules excitation, our group has pioneered illuminated the DSSCs from both TiO₂ photo-anode and transparent PANi CE, getting an impressive efficiency of 8.35%, which is increased by 24.6% compared with the front efficiency (J.H. Wu et al., 2014). With an aim of enhancing the electron-transfer and electrocatalytic activity of transparent PANi CE for bifacial DSSCs application, we have synthesized PANi-multiwalled carbon nanotube (MWCNT) complex CE, achieving a power conversion efficiency of 9.24% when the bifacial DSSCs based on PANi-8 wt% MWCNT complex CE for both illumination (Zhang et al., 2015a,b). However, MWCNT has the drawback of limited active sites toward I⁻/I₃⁻ redox reaction and therefore undesirability electrocatalytic activity (Wan et al., 2011).

To the best of our knowledge, molybdenum disulfide (MoS₂) has high transparent and large surface area to absorb electrolyte, providing a large amount of active sites for I₃⁻ reduction and benefiting electrons migration, comparable to that of Pt material (Hu et al., 2015). In search for other robust PANi-based composite transparent CEs, here aniline monomers-MoS₂ complex has been synthesized by a reflux process, subsequently deposited on FTO by in-situ polymerization method to form PANi-MoS₂ complex transparent CE. The electrocatalytic activity of PANi-MoS₂ complex is significantly enhanced compared with that of pure PANi, which may be caused by the increased active sites of MoS₂ and rapidly charge-transfer dynamics between PANi (N atoms) and MoS₂ (Mo atoms) via the metal (d π)-nitrogen (p π) antibonding interaction. The PANi growing along MoS₂ not only hinders the agglomeration of PANi and MoS₂ nanoparticles, increasing the active sites for I₃⁻ reduction, but also generates a conducting matrix to accelerate electron transfer between MoS₂ nanoparticles and along PANi chains. Owing to the highest transparent and electrocatalytic activity, a potential power conversion efficiency of 9.71% is achieved from the bifacial DSSC based on PANi-6 wt% MoS₂ complex CE for both illumination, which is higher than 7.50% of bifacial DSSC employing PANi CE and DSSCs illuminated from only front or rear side.

2. Experimental

2.1. Synthesis of MoS₂ nanoparticles

Ammonium thiomolybdate (ATTM) precursor was prepared according to the progress reported in the previous literature (Chai et al., 2007). The typical synthesis of MoS₂ nanoparticles was as follows: 0.525 g of ATTM was added into 20 mL of deionized water at pH ~ 10 under stirring for 3 h and then 0.275 g of HONH₃-Cl was added. The resulting mixture solution was stirred for 30 min and then transferred into a 30 mL Teflon stainless steel autoclave for hydrothermal reaction at 240 °C for 24 h. The as-synthesized powder was washed successively with deionized water and ethanol, finally dried at 60 °C for 24 h under vacuum.

2.2. Reflux synthesis of aniline-MoS₂ complexes

Aniline-MoS₂ complexes with MoS₂ dosage of 1, 2, 4, 6, 8 and 10 wt% were synthesized by a reflux process. Detailedly, MoS₂ powder with various dosages was added into 10 mL of aniline monomer, the resulting mixtures were stirred for 30 min and then transferred into a three-neck flask for reflux at 184 °C for 6 h under nitrogen atmosphere in dark. The obtained aniline-MoS₂ complexes were placed in a brown bottle at 4 °C for later use.

2.3. Preparation of PANi-MoS₂ complex CEs

The pretreated FTO conductive glass was put in a culture dish with the conductive side up under 0 °C, 20 mL of aqueous solution containing 1 M of HCl and 0.325 M of aniline-MoS₂ complex was added into the culture dish. Then 20 mL of aqueous solution containing 1 M of HCl and 0.125 M of (NH₄)₂S₂O₈ (APS) was slowly added into the above solution by drop within 2 h, and continue reaction at 4 °C for 24 h. The resultant FTO supported PANi-MoS₂ complex CEs were immersed in 0.1 M of HCl aqueous solution for 5 min and then washed with deionized water. Finally, the PANi-MoS₂ complex CEs were dried at 60 °C for 24 h under vacuum. As a comparison, pure PANi CE was also synthesized under the same conditions.

2.4. Assembly of DSSCs

TiO₂ photo-anode film with a thickness of 10 μ m was synthesized according to the progress reported in our previous work (Duan et al., 2014a). The resultant photo-anode was immersed into a ethanol solution with 0.5 mM of N719 dye ([cis-di(thiocyanato)-N,N'-bis(2,2'-bipyridyl-4-carboxylic acid)-4-tetrabutylammonium carboxylate]) for sensitizing at ambient for 24 h. The DSSCs were assembled with a sandwich type by combing the dye-sensitized TiO₂ photo-anode, a redox electrolyte (consisting of 100 mM of tetraethylammonium 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-butyl-pyridine in 50 ml acetonitrile) and PANi-MoS₂ complex CEs. The redox electrolyte was also applicable to electrochemical impedance spectroscopy (EIS) and Tafel polarization measurements.

2.5. Characterizations

The UV-vis absorption spectra were measured on a UV-3200 spectrophotometer by diluting the samples in toluene solvent. The fluorescence emission spectra were collected using a conventional setup at excitation wavelengths of 515 nm by a Fluorolog3-P spectrophotometer. The morphologies of the PANi or PANi-MoS₂ complex CEs were observed by a scanning electron microscope (SEM, S4800) and transmission electron microscope (TEM, JEM2010, JEOL). Fourier transform infrared spectrometry (FTIR) spectra of the CEs were recorded on a Perkin Elmer spectrum (1760 FTIR spectrometer). The optical transparency of the CEs was also measured on the UV-3200 spectrophotometer. X-ray powder diffraction (XRD) with Cu K α radiation was applied to investigate the crystallographic structure of the as-prepared products.

All electrochemical measurements were carried out by a CHI660E workstation at ambient. Cyclic voltammetry (CV) curves were measured in acetonitrile solution consisting of 0.01 M of I₂, 0.05 M of LiI and 0.5 M of LiClO₄ at scan rates of 20, 30, 40, 50, and 60 mV s⁻¹ in the potential range from -0.6 V to 1.6 V with a three electrodes system, which containing PANi-MoS₂ complex CEs working electrode, Ag/AgCl reference electrode and Pt sheet counter electrode. Tafel polarization curves were recorded at a scan rate of 10 mV s⁻¹ in a potential window of -1 to 1 V with a CE|electrolyte|CE symmetrical dummy cell.

The photocurrent-voltage (*J-V*) characteristics of DSSCs were carried out at a scan rate of 100 mV s⁻¹ in a potential range from 0 to 1 V under 100 mW cm⁻² irradiation (AM 1.5G) equipped with CHF-XM-500W solar simulator (Beijing Trusttech Co., Ltd) on CHI660E electrochemical workstation at atmosphere. A FZ-A type radiometer (Beijing Normal University Photoelectric Instrument Factory) was used to calibrate the intensity of incident light. In order to control the experimental error for all photovoltaic parameters within \pm 5%, each *J-V* curve was repeated at least five times.

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