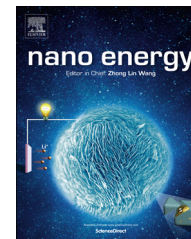


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COMMUNICATION

3D Cu-doped CoS porous nanosheet films as superior counterelectrodes for quantum dot-sensitized solar cells

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Abstract

Q3 Q2 Quantum-dot sensitized solar cells (QDSCs), as a promising type of new generation photovoltaic devices, require an efficient and inexpensive counterelectrode (CE) for their commercial application. In this work, novel noble-metal-free Cu-doped CoS (Cu-CoS) CEs are designed for QDSCs. The hierarchical three-dimensional (3D) Cu-CoS porous nanosheet films on fluorine doped tin oxide (FTO) glass are prepared by a simple hydrothermal reaction followed by a cation exchange reaction process. As an alternative to Pt electrode, the 3D structure of porous Cu-CoS nanosheets with high reflectivity can provide a great large number of active catalytic sites and easy accessibility toward S_n^{2-}/S^{2-} electrolyte solution, leading to high electrocatalytic activity. Theoretical calculations are further performed to demonstrate the superior catalytic activity for the Cu-CoS CE. As a result, the QDSC with an optimized Cu-CoS CE exhibits a high photovoltaic conversion efficiency of 6.1% under 100 mW cm^{-2} irradiation, which is much higher than that of the references Pt and bare CoS CEs. Furthermore, this QDSC device shows long-term stability, which makes it as a very promising and low cost effective material for QDSCs. © 2015 Elsevier Ltd. All rights reserved.

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Introduction

Quantum-dot-sensitized solar cells (QDSCs) are regarded as a promising alternative to dye-sensitized solar cells (DSCs), due to their low cost, acceptable photovoltaic conversion efficiency, tunable band gaps, and high molar extinction coefficient [1-4]. Similar to the cell structure of DSCs, QDSCs consist of a quantum-dot-sensitized semiconductor photoelectrode, a redox polysulfide electrolyte solution, and a noble-metal counterelectrode (CE). Although extensive efforts are focused on a quantum-dot light absorber, the reported conversion efficiencies of QDSCs are still far lower than those of DSCs. One of the major hurdles is the poor catalytic activity of counterelectrodes (CEs), decreasing the energy conversion efficiency in QDSCs [5,6]. Although Pt or Au is widely used as a standard CE due to its good interface between FTO substrates and metal catalysts, it can chemisorb sulfur compounds in polysulfide electrolyte solution, thereby causing a high overpotential and thus inefficient catalysis for redox polysulfide. Therefore, it is quite urgent and crucial to search for alternatives to Pt electrode.

Up to now, many kinds of materials have been widely investigated as CEs in DSCs, such as carbon, conductive polymers, nitrides, carbides, transition-metal sulfide and their composites [7-20]. With regard to QDSCs, chalcogenides have emerged as potential CEs for QDSCs, mainly due to their unique properties such as abundance, low-cost, high electrical conductivity and excellent electrocatalytic activity [21-24]. Copper sulfide and cobalt sulfide have been demonstrated to be efficient catalysts to replace Pt-group metal catalysts in high efficiency QDSCs [25-32]. However, it is known that the Cu_2S CE based on brass foil suffers from mechanical instability due to the continual corrosion reaction between the brass and the electrolyte solution [26,33]. Furthermore, these prepared sulfide CEs are still unsatisfactory for their utilization and their studies are inadequate. Therefore, it is urgent to develop an innovative strategy to fabricate highly effective catalysts of CEs and further improve the catalytic activity. Based on that, the coupling CoS with CuS might be a promising CE for QDSCs.

One promising method for improving the catalytic performance is to employ a doping strategy. It has been demonstrated that heteroatom doping is an effective way to regulate the electron structure and improve the electrochemical activity of the pristine precursors [34,35]. With proper control of doping amount, their catalytic activity can be greatly enhanced. Moreover, two-dimensional (2D) porous nanostructured materials have received broad interests due to their large surface-to-volume ratio, tunable transport properties, and unique surface chemistry which can lead to the effective interaction between the CEs and electrolyte solution and then increase the charge transport efficiency [36,37]. Accordingly, inspired by the positive synergistic effects, it is expected that 3D porous nanosheet film as a CE can provide superior electrocatalytic performance for QDSCs.

In this work, we report a low cost and low-temperature processable Cu-CoS composite CE for high efficiency QDSCs. Hierarchical 3D CoS nanosheet films are first synthesized by hydrothermal reaction and then converted to Cu-doped CoS nanoporous nanosheets through a cation exchange process. The

Cu doping into CoS nanosheets leads to favorable a 3D porous structure, which could improve the electron-transfer capability relative to CoS alone. Benefiting from the superior catalytic performance, the 3D Cu-CoS film exhibits superior electrocatalytic activity with high stability in QDSCs.

Experimental section

Preparation of counterelectrodes

All of the chemical reagents were supplied by Sigma-Aldrich and used without further purification. In a typical synthesis of CoS, a piece of cleaned FTO conductive glass with a size of 20 mm × 25 mm × 3 mm was placed against the wall of a 80 mL Teflon-lined stainless steel autoclave, which had been fed with $\text{Co}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$ (0.4 mmol), L-cysteine (1.6 mmol) and 60 mL of distilled water and stirred for 30 min. Then the autoclave was sealed and maintained at 200 °C for 10 h. After cooling down to room temperature, the sample was washed with water and ethanol, and dried at 60 °C for 2 h under vacuum. Finally, CoS film was deposited on the FTO glass. With regard to the Cu-CoS composites, $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ (5 mmol) and 60 mL H_2O were first introduced into an autoclave, and the FTO glass coated by the CoS film was placed into the autoclave. The autoclave was sealed and underwent different reaction times at 120 °C. Cu-CoS films with different amounts of Cu could be obtained by adjusting the reaction times ranging from 1 h, 2 h, and 5 h. For comparison purpose, Pt counterelectrode was prepared by thermal decomposition of hexachloroplatinic acid at 390 °C for 0.5 h.

Fabrication of QDSCs

TiO_2 photoanode was constructed by a compact layer, TiO_2 layer and scattering layers. First, the compact layer was coated on the FTO glass by spray pyrolysis at 500 °C [38,39]. Thereafter, the mesoporous TiO_2 layer (Solaronix ~5 μm) and scattering layer (WERZ-O, Dyesol, ~3 μm) were screen-printed onto the substrate. The electrode was heated at 450 °C for 30 min to form TiO_2 photoanode [10,32]. A post-treatment of the dried TiO_2 film with an aqueous solution of TiCl_4 (0.05 M) was then carried out. With regards to the sensitizer, $\text{CdSe}_{0.45}\text{Te}_{0.55}$ alloyed QDs with a size of around 5 nm were prepared following a previously reported procedure (Figure S1 in Supporting information) [40-42]. The obtained TiO_2 electrode was then sensitized with the $\text{CdSe}_{0.45}\text{Te}_{0.55}$ QD sensitizers by pipetting directly on the electrode surface. After the deposition, the QD absorbed TiO_2 film was coated with ZnS layer to suppress the charge recombination at the QD sensitizer/electrolyte interface. The prepared CE and the QD-sensitized photoelectrode with an active area of 0.16 cm² were sandwiched between a hot-melt ionomer film (Surllyn, 60 mm) under heating. The polysulfide electrolyte solution composed of Na_2S (1 M), S (1 M) and 0.1 M NaOH aqueous solution was prepared and injected through the pre-drilled holes of the CE, and each hole was sealed by using a small piece of Surllyn and a microscope cover glass.

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