

MoS₂-graphene hybrids as efficient counter electrodes in CdS quantum-dot sensitized solar cells



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ABSTRACT

MoS₂-graphene (M-G) hybrid was prepared by a hydrothermal method and used as counter electrode materials in CdS quantum dot-sensitized solar cells (QDSSCs). Under simulated solar light irradiation, the cell with M-G counter electrode has higher efficiency than that of Pt and bare MoS₂ counter electrode owing to the synergistic effects between MoS₂ and graphene. The electrochemical impedance spectroscopy (EIS) and Tafel-polarization measurements reveal that M-G counter electrodes show better electrocatalytic activity. Moreover, the fabricated cell shows high efficiency (6.23%) under weak light intensity (25 mW/cm²).

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1. Introduction

Quantum dot-sensitized solar cells (QDSSCs), which represent a type of next-generation solar cells, have been intensively investigated in the past few years due to the advantages of quantum dots, including their low cost, easy fabrication, size-dependence band gap, and multiple exciton generation possibilities [1–6]. In order to improve the cell performance, considerable efforts have been focused on optimizing the components including the photoanode [7–11], the quantum dots sensitizer [12–17], the electrolyte [18–20] and the counter electrode. Among which, the counter electrodes play an important role in QDSSCs including collecting electrons from external circuits and catalyzing the reduction of the oxidized species in the electrolytes. Some basic requirements for the ideal counter electrode are to possess high electrical conductivity and catalytic activity, large surface area, chemical durability and low cost [1]. Pt and Au are most used counter electrode materials in QDSSCs [21,22]. However, they are quickly poisoned by the polysulfide electrolyte, primarily because their surface would actively absorb sulphur atoms to inhibit their conductivity [2]. Therefore, numerous studies have been done in order to investigate new counter electrode materials for QDSSCs.

Recently, metal sulphides such as Cu₂S [23–25], CuS [26,27], NiS [28,29], PbS [30,31], CoS [32–36], FeS₂ [37] and copper zinc tin sulphide (CZTS) [38,39] have been intensively exploited. Among

these materials, Cu₂S counter electrodes prepared by depositing Cu₂S on brass foil exhibits high electrocatalytic activity in polysulfide reduction [40,41]. However, continual corrosion would happen to Cu₂S counter electrodes when they are exposed to the polysulfide electrolyte [42]. Therefore, highly effective and durable counter electrode materials are required.

MoS₂, composed of three atom layers with one Mo atomic layer sandwiched between two S atomic layers, has been proved to be a potential counter electrode material candidate for DSSCs, owing to the high conductivity and catalytic activity [43–46]. However, little work has been done on MoS₂ as counter electrode materials in QDSSCs [22]. Graphene, consisting of a unique sp² hybrid carbon, has been widely used to modify the counter electrodes of solar cells due to its large surface area and excellent electrical conductivity [23]. Based on the above considerations, a MoS₂-graphene (M-G) hybrid was prepared by a hydrothermal method and used as counter electrode materials in QDSSCs. Owing to the synergistic effects between MoS₂ and graphene, the M-G hybrid shows improved electrochemical properties. The fabricated cell with M-G counter electrode has higher efficiency than pure MoS₂ or Pt counter electrode and the cell shows high efficiency (6.23%) under weak light intensity (25 mW/cm²).

2. Experimental

2.1. Preparation of M-G counter electrode

The composite samples were synthesized by a routine hydrothermal method according to literature [47]. All the reagents

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were of analytical grade and used without further purification. Graphene oxide (GO) was synthesized from natural graphite powder (>99.8%, Alfa Aesar) by a modified Hummers' method. In a typical synthesis of MoS₂/graphene (M-G) hybrid, 1 mmol of Na₂MoO₄·2H₂O and 5 mmol of thiourea were dissolved in 60 mL of distilled water, and then 0.0135 g of the prepared GO were added into the solution. Next, the homogeneous solution was transferred into a 100 mL Teflon-lined autoclave and held at 210 °C for 24 h. The black precipitate was collected by filtrated and washed with deionized water for three times, followed by drying 12 h at 80 °C. The obtained samples were made into a slurry by using proper amounts of ethanol as solvent and polyethylene glycol (PEG, mole weight = 20000) as binder. Then, the slurry was coated onto a 2 cm × 1.5 cm F-doped SnO₂-coated glass (FTO glass) to form a film with a thickness about 15 μm by the doctor blade technique. The electrodes were calcined at 400 °C for 30 min.

2.2. Fabrication of CdS/ZnS QDSSCs

The Cluster shaped TiO₂ arrays were reported as before [48]. It was directly grown on FTO glass by hydrothermal method. In brief, 0.6 mL Titanium isopropoxide (TIP, 99.999%, Sigma Aldrich) was added into 5 M 30 mL hydrochloric acid, and then the TIP precursor was transferred to 50 mL Teflon-lined autoclave. The cleaned FTO (1.5 × 1.5 cm²) substrate was immersed in the precursor and the autoclave was soaked at 150 °C for 14 h. The obtained FTO samples were rinsed by deionized water and were dried by dryer. Then, they were immersed in HCl (30 mL, 6.67 M) again and post-treated at

150 °C for 7 h. Next, the samples were rinsed again and immersed in TiCl₄ (0.2 M) aqueous solution at 70 °C for 1.5 h for surface modification followed by annealing at 450 °C for 30 min.

In order to fabricate CdS/ZnS QDSSCs, A successive ionic layer adsorption and reaction (SILAR) method was used to deposit CdS and ZnS onto the prepared TiO₂ films [49]. The TiO₂ films were dipped in 0.1 M Cd(NO₃)₂ ethanol solution for 2 min, rinsed with ethanol and dried with dryer, then dipped in 0.1 M Na₂S water-methanol (1:1 vol ratio) solution for 2 min, then rinsed and dried. This is one SILAR cycle. Five cycles were repeated per photoanode. After CdS deposition, the preparation of protective ZnS layer was also applied via SILAR. The same process was repeated again with 0.1 M Zn (NO₃)₂ aqueous solution and 0.1 M Na₂S water-methanol (1:1 vol ratio) solution for 5 min/dip to form ZnS layer. The process was repeated 3 times. It was then annealed in the air at 300 °C for 30 min again.

The sandwich-like QDSSC device was fabricated using the CdS/ZnS TiO₂ photoanodes, M-G (or MoS₂) counter electrodes and the polysulfide electrolyte. The two electrodes with each area of 0.12 cm² were placed face to face. Then the electrolyte solution composing 0.5 M Na₂S, 0.125 M S, 0.2 M KCl water-methanol (1:1 vol ratio) was introduced into the space between the two electrodes by glass capillarity.

2.3. Characterizations

Powder X-ray diffraction (XRD) measurements were recorded ranging from 5 to 80° at room temperature on a Siemens D5005

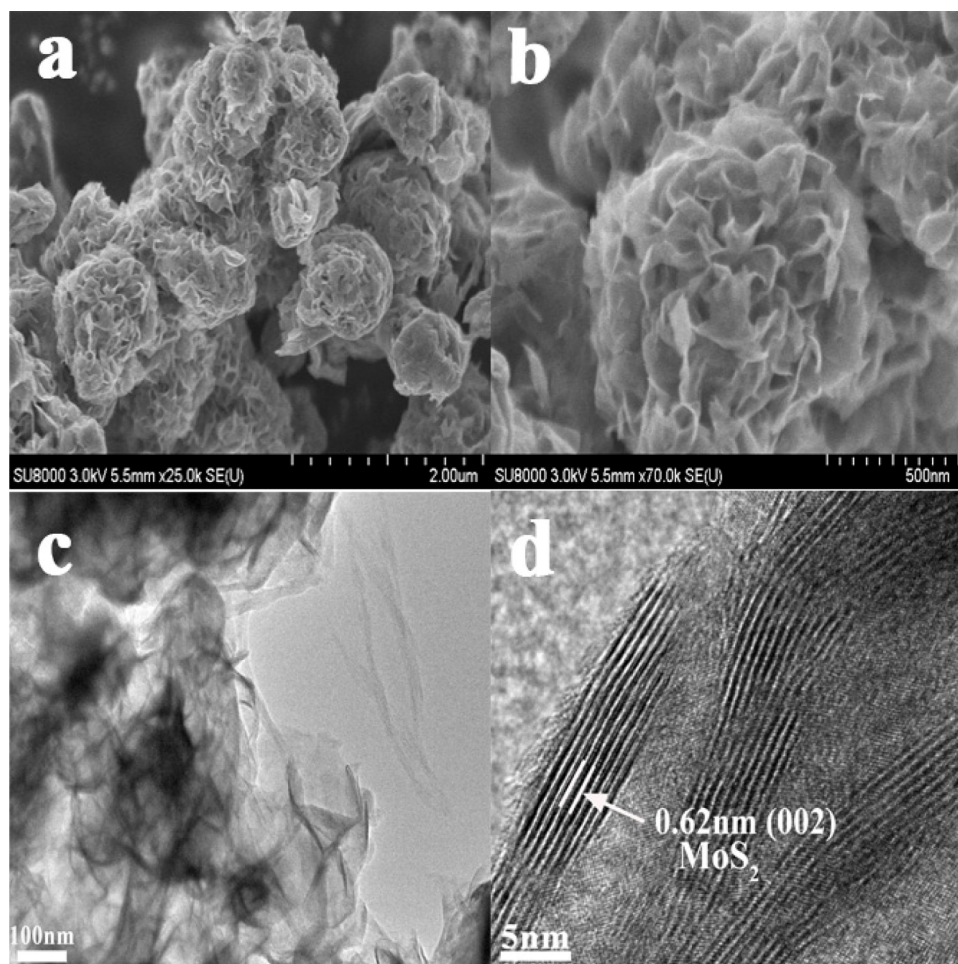


Fig. 1. (a,b) SEM images of M-G composite. (c) TEM and (d) HRTEM images of M-G composite.

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