

Hydroxyl solvents prompted interwoven morphological deposition of iron sulfide nanoparticles as an effective counter electrode for quantum dot sensitized Solar cell



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ABSTRACT

A low-cost, stable and efficient FeS thin film counter electrode (CE) was fabricated for quantum dot-sensitized solar cells (QDSSCs) by a facile chemical bath deposition method using different acidic hydroxyl solvents (ethanol, isopropanol, and tert-butanol) and triethanolamine. The choice of solvents greatly affected the surface morphology and thickness of FeS CEs. Apart from surface morphology, the increase in the atomic percentage of sulfur in tert-butanol and triethanolamine-mediated FeS also plays a crucial role in increasing the electrocatalytic activity of the CE. The QDSSC's power conversion efficiency was 2.15%, which is attributed to the high fill factor (FF) of 0.443. The obtained PCE is far better than the previously reported value of 1.76% for FeS in QDSSCs. Excellent stability of the photoelectric performance of the FeS CE is also demonstrated. The enhanced electrochemical performance of the interwoven FeS networks can be attributed to their unique morphology, which possesses a shorter diffusion length of the ions and easier transportation of the electrons.

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1. Introduction

The global population is rapidly increasing and demand is continuously increasing for renewable and abundant energy sources. Solar cells have drawn tremendous attention because of the inexhaustible and easily available utilization of solar energy. Interest in energy harvesting applications, particularly quantum dot-sensitized solar cells (QDSSCs), has resulted in ample research reports worldwide. QDSSCs' intriguing properties include hot electron injection, tunable band gaps, large intrinsic dipole moments, high molar extinction coefficients, low cost, and facile fabrication processes [1–4]. The principle and structure of QDSSCs is similar to that of dye-sensitized solar cells (DSSCs). The photo degradation and instability of DSSCs opens the way for third-generation photon harvesters such as quantum dots (QDs) with enhanced efficiency and device stability advantages over DSSCs [5].

Generally, a QDSSC consists of mesoporous oxides, including titanium dioxide (TiO₂) and zinc oxide (ZnO), polysulfide electrolyte containing S_n²⁻/S²⁻ redox couples, and a counter electrode

(CE) [6]. The electrons flow through an external circuit to a counter electrode (CE), where oxidized redox species generated during the hole-scavenging process are subsequently reduced by the electrons [7]. The CE in a QDSSC is a critical component where the photo excited electrons are injected from sensitizers into TiO₂ nanocrystallites. The photovoltaic performance of QDSSCs is still poorer than that of DSSCs owing to the fast charge recombination at the TiO₂/electrolyte and QD/electrolyte interfaces. To date, various semiconductor QD sensitizers have been widely studied for QDSSCs, such as CdS [8], PbS [9], CdSe [10–12], and CuInS₂ [13,14]. Among these, CdS/CdSe-based QDSSCs are imperative widely used in aspects of power conversion efficiency (PCE) and cost effectiveness.

The widely used platinum (Pt) CE in DSSCs is not suitable for QDSSCs due to its strong chemisorption with sulfur ions, which results in a poisoning effect and poor photovoltaic performance [15]. Consequently, a large number of QDSSC CE materials for polysulfide electrolyte have been investigated, such as CuS [16–18], CoS [19,20], PbS [21], CuInS₂ [22], and carbon [23,24]. However, there are very few studies on low-cost earth-abundant iron-based sulfides for QDSSCs. In addition, iron is 1000 times cheaper than Pt. Hence, the improved PCE of FeS in solar cells has significant potential to replace the Pt CE in QDSSCs. The limiting factor for achieving high efficiency is the high dark current, which leads to

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small open circuit voltages ($<0.2\text{ V}$) caused by FeS phase impurities and large density surface states originating from thermodynamically unstable particles mostly terminated with sulfur dimers. Moreover, charge transfer to the oxidized redox polysulfide species (S_n^{2-}) on a CE is considered to be a major hurdle to attaining high fill factor and conversion efficiency. A complication in the FeS-based CE arises in the coexistence of a sulfur dimer and its instability in air due to surface decomposition, which results in lower PCEs of solar cells (less than $\sim 2\%$), much lower than the PCEs obtained with solar cells based on other chalcogenides such as CdTe, CIGS, and CZTS [25–27]. Recently, Chen et al. reported the preparation of iron sulfide counter electrode for QDSSCs using carbon steel as substrate through conventional Chemical bath deposition (CBD) process resulted PCE of 1.76% [28]. The same group has used electrochemically deposited FeS for QDSSCs, which gives satisfactory performance in PCE [29]. Possibly the structural oriented growth could involve in determining the PCE of photovoltaic solar cell devices [30]. In addition, Geng et al. reported the synergetic effect of FeS with Ni foam produced highest PCE of $>4\%$ [31]. However, there is no conventional process suggesting the morphological oriented growth deposition of FeS on FTO substrate. Hence, we deemed it worthwhile to investigate interwoven morphological FeS-based QDSSCs for their low cost and easily available iron and sulfur sources.

To date, no study has been done on iron sulfide CEs based on a triethanolamine base and weakly acidic hydroxyl solvents (WAHS) such as ethanol, isopropanol, and *t*-butanol (Fig. 1). We systematically studied iron sulfide CEs using different combinations of these hydroxyl solvents to assess their feasibility as electrocatalysts and to elucidate their electrochemical behavior and influence on solar cell performance. The expected effect from this structure is a high open-circuit voltage due to its regular interwoven structural order, which reduces its potential loss. In addition, the efficient electron/hole pair dissociation can result in a higher fill factor. Under optimum fabrication conditions, the obtained FeS QDSSC cells exhibited a PCE of 2.15% under 1 sun illumination conditions, as well as a short-circuit current of 9.10 mA/cm^2 , open-circuit voltage of 533 mV, and fill factor of 44.3%.

2. Experimental

TiO_2 with a thickness of $7.5\ \mu\text{m}$, CdS/CdSe QDs, and a ZnS passivation layer were synthesized according to literature methods [16]. Other experimental details are described below.

2.1. Materials and preparation of FeS counter electrode

Iron nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$), sulfur (S), triethanolamine, ethanol, isopropanol, and *t*-butanol were used to

prepare FeS thin film and purchased from Sigma Aldrich. CE thin films were deposited on a fluorine-doped tin oxide (FTO) substrate using the facile CBD method. Prior to deposition, FTO substrates were cleaned ultrasonically with acetone, ethanol, and deionized water for 10 min each.

In a 20-ml mixture of triethanol amine and ethanol (1:1), 0.5 mmol of iron nitrate nonanhydrate was added and stirred vigorously to obtain a transparent solution. Subsequently, 0.6 mmol of sulfur was added into the reaction mixture and stirred for 30 minutes at room temperature. The cleaned FTO glass substrates were immersed into the growth solution horizontally and placed in a hot air oven. Mixing the iron source directly with triethanol amine should be avoided and may cause phase impurity and irregular deposition. Considerable amounts of energy along with prolonged reaction durations are needed, so the deposition was carried out at a constant temperature of 90°C for 180 min. A similar experiment was done using isopropanol and *t*-butanol and replacing ethanol in the experimental procedure. The obtained FeS thin films were washed with ethanol solution and dried under nitrogen atmosphere. Depending on the solvents used, the obtained FeS thin films were named as E-FeS, P-FeS, and B-FeS for ethanol, isopropanol, and *t*-butanol, respectively.

2.2. Assembly of the QDSSCs and symmetric cells

The prepared CdS/CdSe/ZnS photoanode and CEs (FeS) were sealed using a $25\text{-}\mu\text{m}$ sealant (SX 1170–60, Solaronix) at 100°C . The internal space between the electrodes was filled with polysulfide electrolyte consisting of sodium sulfide (1 M), sulfur (2 M), and sodium hydroxide (0.2 M) in a solution with 7:3 ratio of methanol and water. Symmetric electrodes of FeS films were sandwiched between a $25\text{-}\mu\text{m}$ sealant (SX 1170–60, Solaronix) at 100°C and filled with a redox liquid electrolyte containing 1 M Na_2S , 2 M S, and 0.2 M KCl in a mixture containing 3 mL of DI water and 7 mL of methanol.

2.3. Characterization techniques

The morphology and thickness were measured by Schottky emission high resolution scanning electron microscope (SEM; SU-70, Hitachi) in the Busan KBSI. The thickness and elemental compositions of the electrodes were investigated using a field emission scanning electron microscope (FE-SEM, S-2400, Hitachi) equipped with energy-dispersive X-ray spectroscopy (EDX) operated at 15 kV. High resolution X-ray diffraction (XRD) analysis was performed on a D8 ADVANCE with a DAVINCI (Bruker AXS) diffractometer using $\text{Cu K}\alpha$ radiation and operated at 40 kV and 40 mA. UV–vis spectroscopic analysis was carried out using an OPTIZEN 3220 UV spectrophotometer. The surface roughness of

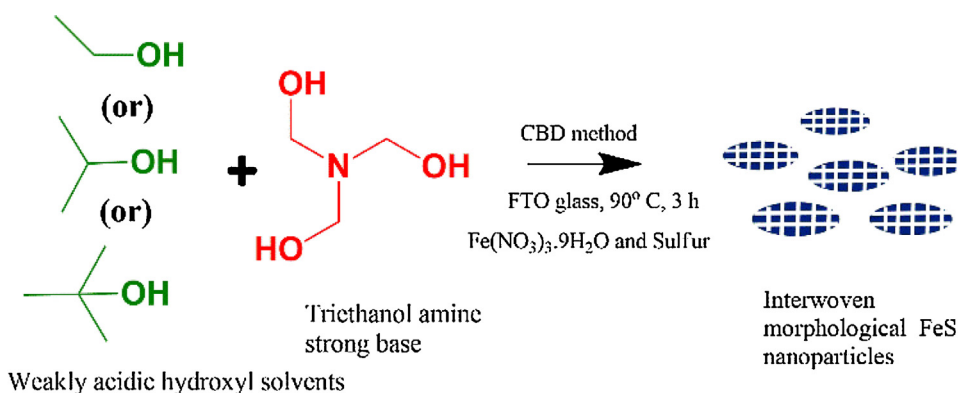


Fig. 1. Deposition of interwoven Iron sulfide nanoparticles on FTO substrate.

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