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CdS quantum dots sensitized ZnO spheres via ZnS overlayer to improve efficiency for quantum dots sensitized solar cells

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Abstract

This paper reports the preparation of three-dimensional ZnO spheres by using a hydrothermal method and their application to quantum dots sensitized solar cells (QDSSCs). After achieving the desired thickness of sensitized CdS quantum dots (QDs) for ZnO spheres, ZnS overlayer was deposited on the surface of CdS/ZnO photo-anodes to further improve the photoelectric properties. CdS QDs and ZnS overlayer were deposited by successive ionic layer adsorption and reaction (SILAR) method. The surface morphology and crystal structure of the samples were verified by field-emission scanning electron microscopy (FE-SEM), Transmission electron microscopy (TEM) and X-ray diffraction (XRD). The CdS QDs sensitized solar cells were ameliorated via using ZnS as a protection-layer between quantum dots and electrolyte. As a result, the power conversion efficiency (η) has been increased from 0.60 to 1.43% after being treated by ZnS overlayer for CdS/ZnO photo-anodes.
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Keywords: ZnO spheres; CdS QDs; ZnS overlayer; Solar cells

1. Introduction

Since the publication of O′Regan and Grätzel′s paper for dye-sensitized solar cells (DSSCs) [1], the use of DSSCs as power conversion devices has attracted much interest. However, the dyes in traditional DSSCs are costly, high toxicity and instability, so new substances are detected to supersede the organic dyestuff [2–5]. A new type of DSSCs, known as ultra-thin film sensitized solar cells or QDSSCs, has aroused the attention of scientists [6–8]. The major sensitizers contain some narrow-gap semiconductors QDs such as CdS [9,10], CdSe [11], PbS [12], PbSe [13] and Sb_2S_3 [14] that have plenty of excellent characteristics like photostability, tunable band gap, high molar extinction coefficients and size-dependent optical properties in QDSSCs [15–17]. Compared to DSSCs, QDSSCs offer the advantageous position of its optical properties that can be regulated by controlling the size of quantum dots rationally. In addition, quantum dots can make quantum yield greater than 1 by the impact ionization due to the effect of multiple-exciton generation [18–21]. So theoretically QDSSCs will have higher conversion efficiency than DSSCs.

The base materials of photo-anode also influence the performance of solar cells. Over the last few years, the effect of TiO_2 photo-anodes has been researched to achieve the respectable η in QDSSCs [22–24]. The best performance up to 5.4% has been obtained by depositing Mn-d-CdS/CdSe multi-layers on TiO_2 electrodes through SILAR method [24]. Simultaneously as the potential substitute for TiO_2, other suitable semiconductor oxides are being searched [25–28]. With the energy-band structure and physical properties similar to those of TiO_2, a wide variety of ZnO nanostructures are developed including nanoparticles, nanowires, nanorods, nano-tubes, nanosheets and nanobelts during the past several decades. ZnO is so easier to be synthesized for anisotropic growth. Moreover, ZnO has a higher electron mobility that reduces the odds of electron recombination at the electrolyte–electrode interface.

In this paper, CdS QDs are deposited on ZnO films to combine with the advantages of QDs and ZnO in the light absorption and electron transport. CdS QDs were used to sensitize the photo-anodes of ZnO spheres for QDSSCs with improved efficiency by using ZnS overlayer in the experiment.

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CdS QDs and ZnS overlayer were prepared via SILAR method with different cycles and sedimentation time. The QDSSCs have greater stability and efficiency after adding ZnS overlayer, because ZnS has a wide bandgap minimum higher than CdS and irradiation stably.

2. Experimental section

2.1. Synthesis of ZnO spheres

All reagents and chemicals were of analytical grade without any other purification. The synthesis method of ZnO spheres was similar to that reported by Shuang Xu [29]. To prepare ZnO spheres, 1.0 g $Zn(NO_3)_2$ was dissolved by 24 ml deionized water and stirred at room temperature until all the reagents dispersed well. Then, 4–8 ml triethanolamine (TEA) was dripped into the solution until it was clear. The suitable solvent system for reaction was transferred into an autoclave and kept at 160 °C for 2 h. After the reaction, the autoclave was placed at room temperature and cooled naturally. The final product was washed with deionized water to remove the organic residual. After being dried over night at 80 °C, the whole products were annealed at 450 °C for 30 min.

2.2. Fabrication of ZnS/CdS/ZnO photo-anodes

ZnO paste was prepared by mixing with 2 ml terpineol, 0.2 g ethyl cellulose and 5 ml absolute ethanol. Then, 1.0 g ZnO spheres were added to the mixture under stirring and keeping at 100 °C for 3 h. ZnO photo-anodes for QDSSCs were prepared by coating the ZnO paste on the FTO with doctor-blade technique [30] and annealing at 450 °C for 30 min to remove the organics.

According to SILAR method, CdS and ZnS were deposited onto the ZnO photo-anodes. The ZnO photo-anodes synthesized by the above described process were immersed in 0.1 M $Cd(NO_3)_2$ alcohol solution for 10–30 min. Then the photo-anodes were rinsed with alcohol. Subsequently, they were put into 0.1 M Na_2S methanol solution for another 10–30 min followed by another rinsing with alcohol. All these steps were one SILAR cycle. After five cycles, the intermediate production denoted by CdS(5)/ZnO was ready and the 5 was defined as the SILAR cycles number. Finally, in order to enhance the efficiency, ZnS overlayer was deposited on the CdS(5)/ZnO films through the same SILAR method. The CdS(5)/ZnO films were immersed alternately into 0.1 M $Zn(NO_3)_2$ alcohol solution and 0.1 M Na_2S methanol solution each for 10 min and rinsed with alcohol. The experiments were executed for 2, 4, and 6 cycles which were defined as ZnS(X)/CdS(5)/ZnO to investigate the different photovoltaic performance while the X was the SILAR cycle number.

2.3. Assembly of QDSSCs

After being sensitized, the composite electrodes both with and without ZnS overlayer were used as the photo-anodes. The composite photo-anodes and the platinum electrodes were sandwiched together, and then the electrolyte was dropped into the aperture between the two electrodes. The liquid electrolyte was an acetonitrile solution containing 0.6 M tetrabutyl ammonium iodide, 0.5 M 4-*tert*-butylpyridine, 0.1 M iodine and 0.1 M lithium iodide.

2.4. Characterization

The surface morphologies of all samples was characterized by field-emission scanning electron microscopy (FE-SEM, Hitachi S-4800, Japan) and Transmission electron microscopy (TEM, JEOL JEM-2010). The crystal structures were analyzed by X-ray diffraction (XRD, Cu *K*α radiation, SmartLab 3 kW, Rigaku, Japan). The optical performances of the samples were studied by a UV–vis spectrophotometer (UV-2550, Shimadzu, Japan). Photoelectrochemical tests were carried out by measuring the current density–voltage (*J*–*V*) characteristic curves under simulated AM 1.5 solar illumination at 100 mW cm^{-2} from a xenon arc lamp (CHFXM500, Trusttech Co. Ltd., China) in an ambient atmosphere. Electrochemical impedance spectroscopy (EIS) measurements were analyzed in the dark. All measurements were performed at a CHI660C electrochemical workstation (CH Instrument Inc., China).

3. Results and discussion

3.1. Microstructure and optical performance of ZnS/CdS/ZnO spheres

ZnO spheres were synthesized by the chemical reaction of $Zn(NO_3)_2$ and TEA in the distilled water. This mechanism was similar to the "oriented aggregation" as reported [29,31]. In the hydrothermal system, as the temperature raised, the solution became gel at 50 °C, because Zn^{2+} could react with H_2O and came into the gel of $[Zn(OH)_2]_n$. Then TEA worked with $[Zn(OH)_2]_n$ and became a stable complex of $[Zn(OH)_2TEA]_n$. When the temperature reached 160 °C, $[Zn(OH)_2TEA]_n$ set loose of H_2O and formed ZnO spheres of TEA package. Due to the hydroxyl groups in peripheral TEA molecules attached the adjoining hydroxyl groups of the other ZnO nanoparticles, as the result that ZnO nanoparticles with hydroxyl groups began clustering together to a ball shape of aggregation. Fig. 1A showed that ZnO spheres dispersed well and the diameter of the spheres was about 1–2 μm. From Fig. 1B, it could be observed that ZnO spheres were assembled by ZnO nanoparticles and the surface of ZnO spheres was rough with plenty of holes. We could conclude that ZnO spheres had a strong ability to absorb the QDs. The typical image of the synthesized ZnS/CdS/ZnO nanostructure was shown in Fig. 1C. Fig. 1D–F showed the TEM images of ZnO/CdS/ZnS nanostructure. The obvious coverage on the surface after decoration of ZnO/CdS spheres with ZnS could be observed. It could be noticed that the thickness of the CdS QDs and ZnS coverage was estimated to be 10–20 nm. The energy-dispersive X-ray spectroscopy (EDS) in Fig. 1G illuminated that the CdS was well deposited on ZnO spheres and the existence of ZnS would be discussed later.

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