



Research paper

Quantum dot sensitized solar cells: Light harvesting versus charge recombination, a film thickness consideration



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ABSTRACT

Sensitizer loading level is one of the key factors determined the performance of sensitized solar cells. In this work, we systemically studied the influence of photo-anode thicknesses on the performance of the quantum-dot sensitized solar cells. It is found that the photo-to-current conversion efficiency enhances with increased film thickness and peaks at around 20 μm . The optimal value is about twice as large as the dye counterparts. Here, we also uncover the underlying mechanism about the influence of film thickness over the photovoltaic performance of QDSSCs from the light harvesting and charge recombination viewpoint.

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1. Introduction

Quantum dot sensitized solar cells (QDSSCs) is one of the most promising candidates for the third generation solar cells. It has a similar device structure with traditional dye-sensitized solar cells except that replacing organic molecular dyes with inorganic semiconductor quantum dots [1–3]. At present, the certified PCE for liquid junction devices reached 11.6% based on Zn–Cu–In–Se QDs [4]. However, the efficiency is still lag behind that of dye-sensitized devices and far below the theoretical expectation (44%) [5,6]. The lower QDs loading amount is one of the reasons leading to the moderate performance of QDSSCs [7,8]. Because of the quantum dots have much larger sizes than dye, which make the photo-anode (mainly TiO_2) covered at much lower loading levels when the film thicknesses remains constant [7]. Consequently, fewer electrons are generated. Furthermore, the structure of photo-anodes and the larger QDs also prevent even QD coverage on the TiO_2 surfaces. The lesser and inhomogeneous QDs coverage can produce bare surfaces of photo-anodes. Then, these bared photo-anode surfaces would become recombination centers in the interfaces of photo-anodes/electrolyte [8], which lead to the lower photo-to-current conversion efficiency (PCE).

So, it has attracted increasing research attention on the optimization of TiO_2 mesoporous film to increase QD loadings. Cao et al. demonstrated the best PCE $\sim 4.62\%$ on 11 μm thick photo-

anode films with optimized porosity [9]. Lin et al. obtained an optimal thickness of 22 μm employing CdS/N719 co-sensitized photo-electrodes [10]. Zhong et al. revealed that the optimal TiO_2 transparent layer thickness is around 9 μm on 6 μm light-scattering layer for colloidal CdSe QDs [11]. But, there is still not a universal standard for the film thickness and lack of specifically investigation to uncover the underlying mechanism. Moreover, there are limited literatures about the optimization of the film thickness by the light harvesting versus charge recombination.

In order to deeply understand the influence of film thickness on PCE, we prepared the different thickness photo-anodes films and sensitized them with CdS CQDs/CdSe. All cells were tested using polysulfide as electrolyte and PbSe as counter electrode. The materials, photo-anodes and solar cells were characterized with scanning electron microscopy (SEM), current-voltage traces, absorption spectrum, electrochemical impedance spectroscopy (EIS) and Mott-Schottky (M-S) measurements. We found that PCE increases with a growing film thickness and peaks at around 20 μm . The best PCE on 20 μm -thickness photo-anode cell is attributed to the highest QD loading level with optimized light harvesting capacity, charge carrier density and longer electron lifetime in the present work.

2. Experimental

2.1. Materials

Cadmium oxide (CdO, 99.99%), sulfur powder (99.99%), oleylamine (OAm), 1-octadecene (ODE, 90%) and 3-Mercaptopropionic

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acid (MPA, 99%) were purchased from Aladdin. Oleic acid (OA, 90%) is obtained from Alfa. Terpeneol and ethyl cellulose (EC, Fluka) were purchased from Aldrich. All these reagents were used without further purification. P₂₅ purchased from Germany Degussa Co. Fluorine doped tin oxide (FTO) glass (7Ω/square) was purchased from WuHan GEAO company.

2.2. The preparation of CdS QDs and phase transfer

Oleylamine capped CdS QDs with the size about 4 nm were synthesized according to the literature of Zhong's group with the minor modification [12]. Briefly, 10 mL of OAm and 2 mmol S powder were mixed in three-necked bottle and heated to 160 °C under nitrogen atmosphere with stirring. 6 mL of Cd stock solution (0.4 M) were prepared by dissolving CdO in ODE/OA (1:1) at 250 °C. Then, injected S stock solution into Cd reaction flask and grew 5 min at 250 °C. The obtained CdS nanocrystals were purified by precipitation and centrifugation processes with the actone. The purified oil-soluble QDs were transferred into water phase by ligand exchange process with MPA [13,14]. Finally, the water-soluble QDs were redispersed into deionized water for use in the next step. The absorption spectra and TEM images of QDs were shown in Fig. 1.

2.3. The preparation of photoanodes and the assembly of QDSCs

TiO₂ thin films were prepared by spin-coating method. The home-made P₂₅ pastes were spin-coating 5, 10, 15, 20 times on FTO glass substrate, followed by sintering at 500 °C for 30 min to obtain the different thickness photo-anodes. The as-prepared films were sensitized with water-soluble CdS suspension. Then, TiO₂/CdS films were sensitized with CdSe QDs by the successive ionic layer adsorption and reaction (SILAR) method. ZnS barrier layer coated the sensitized photo-anodes by alternately dipping electrode into Zn(OAc)₂·2H₂O (0.1 M) and Na₂S·9H₂O (0.1 M) aqueous

solutions for 1 min. This procedure was repeated four times. Finally, cells were assembled by sandwiching the TiO₂/CdS (colloidal)/7CdSe photo-anodes, PbSe counter electrode prepared by electrode position according to the our previous work [15] and the polysulfide electrolyte (1 M Na₂S, 1 M S and 0.1 M NaOH aqueous solution) with a binder clip and separated by double sticky tapes.

2.4. Characterization

The phase and purity of CdS QDs were characterized by X-ray diffraction (XRD) with Cu Kα radiation (λ=1.5418Å). The morphology and size of the as-prepared QDs were determined by transmission electron microscope (TEM, JEOL JEM-2100). The thickness and compositions of the photo-anodes were evaluated by scanning electron microscope (SEM, Philips-FEI Quanta200). The UV–Vis absorption spectra of films were obtained by the UV-Lambda 950 spectrophotometer. The IR spectra were recorded by Bruker Tensor 27 spectrometer. The photovoltaic measurements of cells were completed by Keithley 2400 source meter under irradiation of simulated solar light (AM1.5G, 100 mW/cm²). The active area of cells is 0.25 cm². Electrochemical impedance spectroscopy (EIS) data were collected at –0.7 V bias potential, 5 mV of amplitude and the frequency range from 0.1 Hz to 100 kHz under dark condition. Mott-Schottky plots (MS) were tested based on three electrodes system at the 1 kHz of frequency, 10 mV of amplitude and the bias voltage from –1 to –0.3 V in dark condition.

3. Results and discussion

The powder X-ray diffraction (XRD) patterns for CdS QDs before and after exchange with MPA are shown in Fig. 1a. The diffraction patterns are well matched with cubic phase of CdS (JCPDS PDF#65-2887). The major peaks at 2θ values 26.46°, 43.93° and 51.95° can be assigned to the (111), (220), (311) facets. In Fig. 1b, a distinct

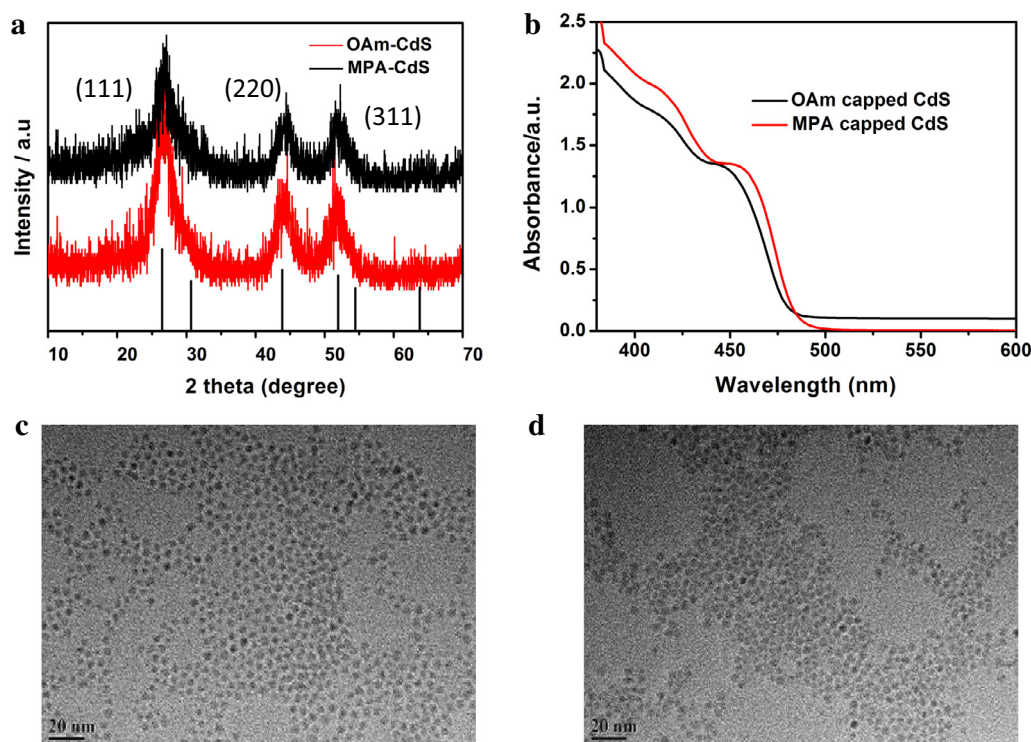


Fig. 1. XRD pattern (PDF#65-2887) (a), UV absorption spectra (b), TEM images (c–d).

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