

Employing 4-methoxybenzoic acid interfacial-modification to enhance the photovoltaic performance of CdS quantum dot-sensitized solar cells



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ABSTRACT

The effect of incorporating self-assembled monolayers of 4-methoxybenzoic acid (MBA) on CdS quantum dot-sensitized (QDSSC) solar cells based on ZnO nanorods was evaluated. The MBA was deposited on the ZnO photoanode, with the deposition time being a process variable. Photoluminescence spectra indicated that the MBA modification can suppress the photogenerated electron–hole recombination processes and presumably passivate the surface defects. The ultraviolet photoemission spectroscopy results prove that MBA forms an energy barrier at the interface of ZnO and CdS which may retard the back transfer of electrons. Overall the inclusion of MBA imparts a weak positive enhancement of the photovoltaic performance of these QDSSCs and this is an approach that may be developed to give stronger effects using alternative chemistries.

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1. Introduction

Quantum dot-sensitized solar cells (QDSSCs) have attracted considerable attention, since the sensitizers of semiconductor quantum dots exhibit amazing advantages such as higher exciton concentration, larger quantum yield and longer lifetime of hot electrons in contrast to the traditional dye [1–4]. However, so far, the photoelectric conversion efficiency of QDSSCs is far from 13% of DSSCs [5]. The strong photogenerated carriers' recombination and back electron transfer processes happened at the interface have been believed to be the key factors for such lower efficiency of QDSSCs. Thus, the interfacial modification is proposed as an effective way to solve these problems [6].

The self-assembled monolayers (SAMs), a two-dimensional molecular array that is spontaneously organized by adsorption of amphiphilic organic molecules on a solid inorganic surface, has been widely used to do the interface modification to improve the performance of photoelectric devices [7]. People have proved that it can significantly improve electrical coherence, adhesion and charge transfer properties of various oxides and metallic surfaces [8–14]. For instance, some works have reported that the SAMs deposition can effectively suppress the surface recombination of semiconductors, which will be beneficial for the charge separation

and transfer to improve the photovoltaic performance of solar cells [8–10]. Hau et al. reported that the utilization of a fullerene SAMs to modify the interface of TiO₂/P3HT solar cells can strongly enhance the photovoltaic performance. The main reason is that the SAMs can serve multiple functions to reduce the recombination of charges, passivate the inorganic surface trap states and improve the exciton dissociation efficiency at the polymer/ZnO interface, which finally leads to the better charge selectivity and improve solar cell performance [11]. Bruening et al. reported that the surface potential of inorganic semiconductors (i.e. CdTe and CdSe) can be decreased by the strong electron-donating substituent on benzoic acid [12–14]. Since the surface potential is directly proportional to the effective work function, the work function of the semiconductors decreases as well. Therefore, it is reasonable to deduce that the series of 4-substituted benzoic acid derivatives can strongly influence the surface potential and work functions of the inorganic semiconductors. It is well known that adjusting work function of semiconductor is an effective way to enhance the photovoltaic performance of solar cells. However, so far, little work has been performed to investigate how the SAMs of 4-substituted benzoic acid derivatives influence the photovoltaic performance of ZnO based QDSSCs.

Therefore, in this work, we employed 4-methoxybenzoic acid (MBA) derived from benzoic acid as SAMs to modify the surface of ZnO nanorods. The MBA/ZnO nanorods were used as photoanodes to fabricate CdS QDs synthesized solar cells. Fig. 1 illustrates the schematic structure of QDSSCs and the molecule formulas of MBA

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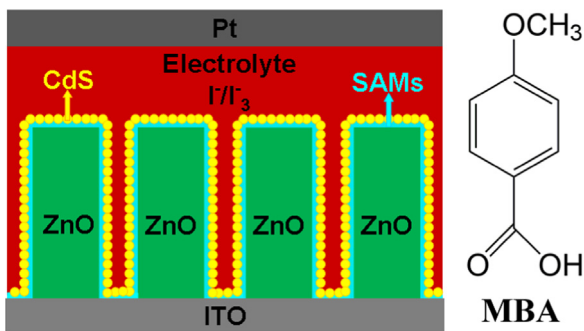


Fig. 1. Schematic structure of QDSSCs and the molecule formulas of MBA SAMs.

SAMs. We tried to deposit MBA on the surface of ZnO nanorods with different time. How they influence the surface structure, optical properties and photovoltaic performance of ZnO photoanodes were studied in detail. The related influence mechanism for photovoltaic performance was also revealed.

2. Experimental details

2.1. Preparation of MBA/ZnO photoanodes

The ZnO nanorods arrays were grown with chemical bath deposition method for 6 h under 95 °C with same procedure as our previous report [15]. Since the carboxylic anchoring group is acidic nature and can erode ZnO (potentially degrading the device) it was necessary to adjust the pH of the MBA/ethanol solution to ~ 7 with concentrated ammonia. We immersed the ITO substrates with ZnO nanorods into 10 mM MBA ethanol solutions for different time. After the MBA immersion step, the samples were rinsed into ethanol for 30 s to remove the excess MBA molecule. The obtained samples for immersing 1 min, 2 min, 3 min and 5 min were named as MBA(1 min)/ZnO, MBA(2 min)/ZnO, MBA(3 min)/ZnO and MBA(5 min)/ZnO, respectively.

2.2. SILAR technique to deposit CdS QDs

We utilized a successive ionic layer adsorption and reaction (SILAR) technique to in-situ deposit the CdS QDs on the surfaces of MBA/ZnO and pure ZnO nanorods [16]. The detailed procedure is similar to the description in the literature [17]. Firstly, the CdS/MBA/ZnO and CdS/ZnO nanostructures were immersed into a 0.1 M $\text{Cd}(\text{NO}_3)_2$ solution for 5 min and then washed with deionized water. Subsequently, they were immersed into a 0.1 M Na_2S solution for another 5 min followed by another washing with deionized water. These two dipping procedures are termed as one SILAR cycle. In our case, the SILAR cycles were repeated about 20 times to realize full deposition of CdS QDs on the surface of MBA/ZnO and pure ZnO nanorods. After 20 cycles, the CdS/MBA/ZnO samples were thoroughly washed with ethanol and deionized water for several times and then dried at room temperature for fabricating solar cells.

2.3. Cell fabrication

The device structures consisted of a CdS/MBA/ZnO photoelectrode and an ITO substrate sputter coated with a 20 nm platinum layer. These were positioned face-to-face to assemble sandwich-type cells. The iodide-based electrolyte of 1 M LiI and 0.05 M I_2 alcohol solution was injected into the interelectrode space by capillary action.

2.4. Characterization and measurements

To evaluate the microstructure of samples, the JEM-2100 transmission electron microscope was used to record the transmission electron micrographs (TEM) and high-resolution transmission electron microscopy (HRTEM) images. The VG ESCALAB Mark II X-ray photoelectron spectroscopy (XPS) were utilized to obtain the quantitative compositional information of samples, which was carried out in an ultra-high vacuum chamber (a pressure lower than 1.333×10^{-7} Pa) with Mg $\text{K}\alpha$ radiation ($h\nu = 1253.6$ eV). The photoluminescence (PL) spectra were taken from the Renishaw *inVia* spectrometer with 325 nm excitation from a continuous He–Cd laser at a power of 2 mW. The room temperature ultraviolet-visible (UV–vis) absorption spectra were recorded from a UV–vis spectrophotometer (UV-5800PC, Shanghai Metash Instruments Co., Ltd.). The current–voltage curves of solar cells were measured using a Keithley 2400 source-measure unit. The cells were illuminated by Oriel 91,160 solar simulator equipped with an AM1.5G filter at a calibrated intensity of 100 mW/cm^2 . Measurements with the ultraviolet photoemission spectroscopy (UPS) were conducted in ultra-high vacuum (UHV) at $\sim 10^{-10}$ mbar, by irradiating with 21.22 eV photons (He I line) with a sample bias of -8 V.

3. Results and discussions

To observe the morphology variation after depositing MBA SAMs and CdS QDs, the detailed microscopic structures were characterized by TEM technique. To avoid the effect from the ultrasonic preparation process, the ZnO and MBA/ZnO nanorods were peeled off from the ITO substrates with blade and collected on the copper grid. Subsequently, a few drops of ethanol were dropped on the copper grid to remove the contaminations on the nanorods. Fig. 2(a), (c), (e) and (g) shows the TEM images of ZnO nanorod, MBA(2 min)/ZnO, MBA(5 min)/ZnO and CdS/ZnO, respectively, which clearly shows that the average diameter of these nanorods is 150 nm. From Fig. 2(c) and (e), we can see that the surface of ZnO nanorods have some amorphous materials after depositing MBA for 2 min and 5 min. Fig. 2(b), (d) and (f) show the HRTEM images of ZnO nanorods, MBA(2 min)/ZnO nanorods and MBA(5 min)/ZnO nanorods, respectively. From Fig. 2(b), we can distinguish that the lattice fringe space is ~ 0.26 nm, which matches well with the d-spacing of the (002) planes of wurtzite ZnO [12]. Moreover, from Fig. 2(d), we can observe a ~ 1.8 nm thickness amorphous layer attaches on the surface of ZnO nanorods. From Fig. 2(f), we can see that the thickness of thin amorphous layer on the surface of ZnO nanorods increase to ~ 3.1 nm. According to the report by Bang et al. [18], we know that the thickness of SAMs is typically 1–2 nm. Since our samples are rod-shape, the evaluation for the thickness of SAMs from HRTEM images might not be accurate, which only presents some tendency for the variation of SAMs. Meanwhile, according to the report in the dissertation of Michael Salinas [19], we know that the thickness of the SAM (d_{SAM}) is depending on the length of molecule (l_{molecule}) and the tilt angle (θ), which is following the $d_{\text{SAM}} = l_{\text{molecule}} \cos \theta$ relationship. The tilt angle should be related to the packing density of molecule. Therefore, in our case, the prolonging immersion time will increase the packing density of molecule, which decrease the tilt angle so that the thickness of MBA SAMs increases step by step. From this aspect, we might believe that such kind of amorphous layer appeared in HRTEM images should be originated from MBA SAMs, which will be further testified by the XPS and contact angle measurement in the following parts.

In addition, as shown in Fig. 2(a), the surface of ZnO is smooth. While, from Fig. 2(g), we can observe that a layer of ultrafine

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