

Research Article

Enhanced Photovoltaic Properties of the Solar Cells Based on Cosensitization of CdS and Hydrogenation

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The hydrogenated TiO₂ porous nanocrystalline film is modified with CdS quantum dots by successive ionic layer adsorption and reaction (SILAR) method to prepare the cosensitized TiO₂ solar cells by CdS quantum dots and hydrogenation. The structure and topography of the composite photoanode film were confirmed by X-ray diffraction and scanning electron microscopy. With deposited CdS nanoparticles, UV absorption spectra of H:TiO₂ photoanode film indicated a considerably enhanced absorption in the visible region. The cosensitized TiO₂ solar cell by CdS quantum dots and hydrogenation presents much better photovoltaic properties than either CdS sensitized TiO₂ solar cells or hydrogenated TiO₂ solar cells, which displays enhanced photovoltaic performance with power conversion efficiency (η) of 1.99% ($J_{sc} = 6.26 \text{ mA cm}^{-2}$, $V_{oc} = 0.65 \text{ V}$, and $FF = 0.49$) under full one-sun illumination. The reason for the enhanced photovoltaic performance of the novel cosensitized solar cell is primarily explained by studying the Nyquist spectrums, IPCE spectra, dark current, and photovoltaic performances.

1. Introduction

Photoelectrochemical solar cells based on TiO₂ nanocrystalline films sensitized with organic dyes have been studied intensely for the past 20 years as a potential promising low-cost alternative to traditional solid-state solar cells. Semiconductor nanocrystals with narrow band gap such as CdS, CdSe, PbS, and InP were also demonstrated as efficient sensitizers in the spectral range from the visible to mid infrared with advantages such as low-cost fabrication, good stability, multiple exciton generation, and the tunability of optical properties and electronic structure by changing the size of nanocrystals [1, 2]. The photoelectrochemical cells with semiconductor nanocrystals as sensitizers also were called quantum dot sensitized solar cells (QDSSCs) [1]. Among the common semiconductors, CdS is one of the most commonly used inorganic sensitizers [3] due to its optical properties and narrowed band gap adjusted by the particle size [4] and better UV stability [5] and it has reduced dark current [6]. However, the efficiencies of CdS sensitized TiO₂ solar cells have still stayed low up to now. It has been extensively studied how to enhance their photovoltaic efficiency. One common effective

method is cosensitization of more than one kind of quantum dots with different band gap. CdS and CdSe quantum dots cosensitized nanocrystalline TiO₂ is one of the most common cosensitization structures with a power conversion efficiency of more than 3% [7, 8].

The other alternative method is to modify TiO₂. In order to improve the photoelectric properties of TiO₂ under sunlight, some metal or nonmetal impurities [9, 10] were added to generate donor or acceptor states in the band gap and to modulate energy band structure [11]. Recently, the hydrogenated TiO₂ has attracted extensive attention. Chen et al. [12] demonstrated that the hydrogenated TiO₂ nanocrystal enhanced solar absorption by introducing disorder in the surface layers of nanophase TiO₂, and Wang et al. [13] reported hydrogen treatment was a simple and effective strategy to fundamentally improve the photoelectrochemical performance of TiO₂ nanowires for water splitting. Our previous work reported the self-sensitized effect of hydrogenated TiO₂ film which led to enhanced photovoltaic properties in the solar cell with hydrogenated TiO₂ as photoanode without adding any dye [14].

If a photoanode was prepared by depositing CdS quantum dots on the surface of hydrogenated TiO₂ film, the self-sensitized effect of hydrogenated TiO₂ film would work together with quantum dots sensitization, which should also be a kind of cosensitization to be a promising method to enhance the photovoltaic properties of quantum dot sensitized solar cells. In this work, hydrogenated TiO₂ films were fabricated on fluorine-doped tin oxide (FTO) glasses by screen printing and annealing under the specific temperature and time; then CdS quantum dots were attached to the surface of hydrogenated TiO₂ by successive ionic layer adsorption and reaction (SILAR) method. Structural characterization, photoelectrochemical properties, and photovoltaic performances were investigated and discussed. The cosensitized TiO₂ solar cells by CdS quantum dots and hydrogenation present much better photovoltaic properties than either CdS sensitized TiO₂ solar cells or hydrogenated TiO₂ solar cells. The reason for the enhanced photovoltaic performance of the novel cosensitized inorganic solar cell was also explained in detail.

2. Experimental

2.1. Preparation of Hydrogenated TiO₂ Films and CdS Quantum Dots. Hydrogenated TiO₂ (H:TiO₂) films were prepared on fluorine-doped tin oxide (FTO) glasses with an area of 0.16 cm² via the same processing as reported previously [14]. Then CdS quantum dots were attached to the surface of hydrogenated TiO₂ by successive ionic layer adsorption and reaction (SILAR) method [15, 16]. In brief, 0.1 M Cd(NO₃)₂ in methanol was used as the cation source and 0.1 M Na₂S in 1:1 methanol and water as the anion source. The H:TiO₂ films with FTO substrates were successively dipped into the cation source and anion source for 5 min each. Following each dip, the films were rinsed for 1 min or more using pure ethanol to remove excess precursor, and the electrode was dried for 10 min before the next dipping. This dip cycle was repeated several times to obtain desirable CdS quantum dots on the surface of the H:TiO₂ films as photoanodes. According to the preliminary experimented research on the effect of SILAR cycles for CdS quantum dots, an optimized SILAR cycle was determined as 9 cycles. For comparison, the H:TiO₂ films without any quantum dots and TiO₂ films with CdS quantum dots were also fabricated as photoanodes.

2.2. Assembly of Solar Cells. To assemble solar cells, the mixed solution of water and methanol with volume ratio of 3:7 consisting of 0.2 M KCl, 0.5 M Na₂S, and 2.0 M S was used as polysulfide electrolyte. The photoanode, a platinized counter electrode, and the polysulfide electrolyte were sealed together with a hot-melt polymer film (Surlyn 1702-25, DuPont) to constitute a sandwich-like solar cell to measure photoelectrochemical properties. The active area of the cell is 0.16 cm².

2.3. Testing Device and Characterization Method. The crystal structure of TiO₂ films was characterized by an X-ray

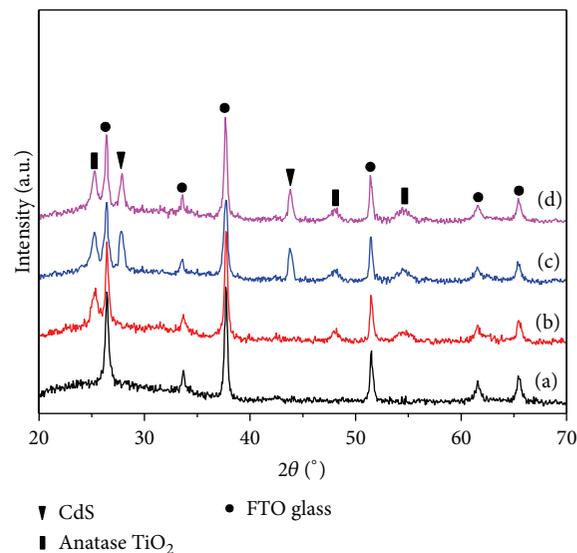


FIGURE 1: X-ray diffraction patterns of (a) FTO glass substrates; (b) H:TiO₂ films; (c) CdS sensitized TiO₂ films; (d) CdS sensitized H:TiO₂ films.

diffraction (XRD; X' Pert Pro MPD) with Cu K α 1 radiation ($\lambda = 0.154$ nm) and the film surface morphology was observed by SEM (INSPECT F, FEI Company). Ultraviolet-visible absorption spectra were measured by Shimadzu UV2100 Spectrophotometer. In order to measure photovoltaic performance of the solar cells, simulated sunlight (AM 1.5 G) was irradiated using a solar simulator with a Xe lamp (calibrated with a standard Si-based solar cell), and the current-voltage (I - V) curve was recorded by a CHI-660 electrochemical workstation. The photocurrent density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF), and corresponding photoenergy conversion efficiency (η) were calculated from the obtained I - V curves. The incident photon to current conversion efficiencies (IPCE) spectra were obtained by Newport QE-PV-SI. Electrochemical impedance spectroscopy (EIS) was also measured at the CHI-660 electrochemical workstation with sinusoidal perturbations of 10 mV at frequencies from 0.01 Hz to 10 kHz with zero bias potential.

3. Results and Discussion

3.1. Characterization of TiO₂ Films Cosensitized by CdS and Hydrogenation. X-ray diffraction patterns of the H:TiO₂ film, CdS/TiO₂ film, and CdS/H:TiO₂ film are shown in Figure 1. TiO₂ in all the films is in the anatase phase, the same as reported in our previous work [14]. With the successive ionic layer adsorption in 9 cycles, the peaks at 27.9° and 43.8° corresponding to (100) and (110) planes of CdS quantum dots, respectively, can be observed in the patterns of CdS sensitized TiO₂ and H:TiO₂ films as shown in Figures 1(c) and 1(d), respectively. No obvious difference is observed between the XRD patterns of the CdS sensitized TiO₂ films with or without hydrogenation. Figure 2 shows SEM surface morphology of pure H:TiO₂ film and CdS/H:TiO₂ film. The

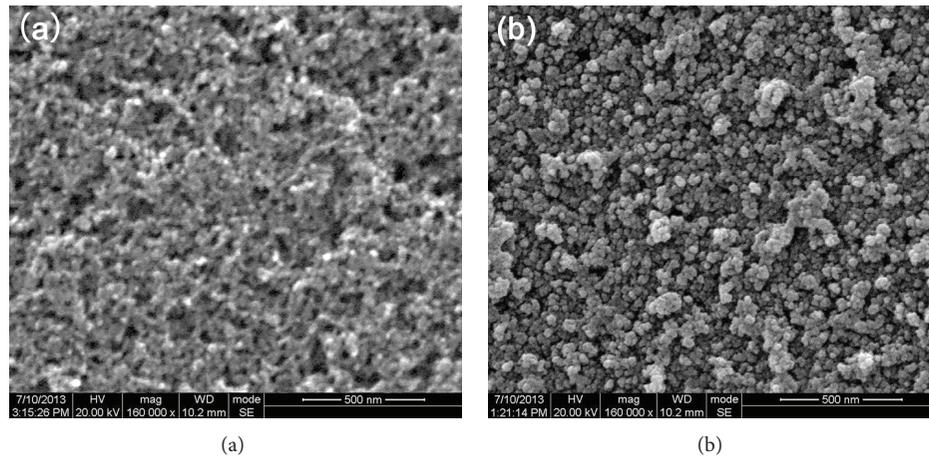


FIGURE 2: SEM surface morphology of (a) pure H:TiO₂ film and (b) CdS/H:TiO₂ film.

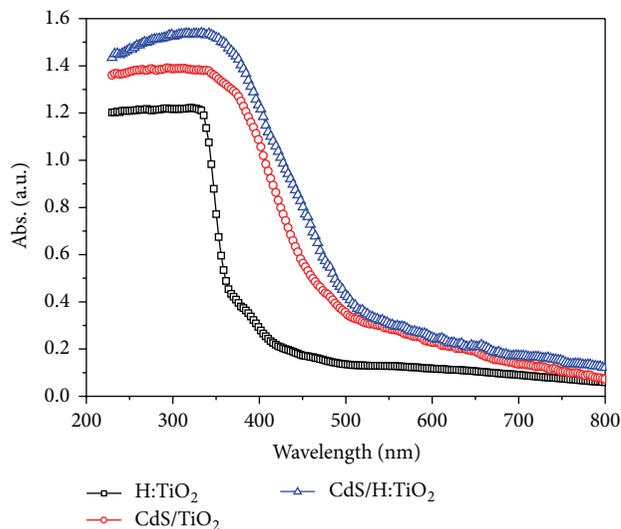


FIGURE 3: UV-visible absorption spectra of the H:TiO₂ film, the CdS sensitized TiO₂ film, and the CdS sensitized H:TiO₂ film.

H:TiO₂ film is typical porous nanocrystals as in Figure 2(a), while the CdS/H:TiO₂ film displays dense-packing CdS quantum dots with the grain size of about 25 nm according to Figure 2(b).

3.2. Absorption of TiO₂ Films Cosensitized by CdS and Hydrogenation. The UV-visible absorption spectra of H:TiO₂ film, CdS/TiO₂ film, and CdS/H:TiO₂ film are showed in Figure 3. It has been demonstrated that hydrogenation treatment can improve light absorption of TiO₂ due to the generated dangling bonds and disordered surface layers on the surface of TiO₂ nanophase [12]. Compared with the H:TiO₂ film with a band gap of about 3.10 eV, the CdS/TiO₂ film has a much stronger absorption in the range of 300~800 nm in that CdS has a much smaller band gap (2.25 eV in bulk) than H:TiO₂ [17]. When CdS quantum dots are deposited on the surface of the H:TiO₂ films, the obtained CdS/H:TiO₂ film combines

the advantages of both hydrogenation treatment and CdS quantum dots and thus reveals the strongest absorption as shown in Figure 3.

3.3. Impedance Spectra of the Solar Cells Based on Cosenitization of CdS and Hydrogenation. In order to analyze the internal electron transport process of solar cells, the electrical impedance spectra (EIS) for the sensitized solar cells based on CdS/H:TiO₂, CdS/TiO₂, and H:TiO₂ are shown in Figure 4(a), and Figure 4(b) shows the relevant equivalent-circuit model. Similar to a typical DSSC system [18, 19], R_{ct1} and CPE1 represent the electron transfer resistance and interfacial capacitance at the interface between counter electrode and electrolyte, respectively. R_{ct2} and CPE2 are electron transport resistance and interfacial capacitance at the interface between electrolyte and photoanode, respectively. R_s is ohmic series connection resistance of the whole cell. Z_N is the electrolyte Nernst diffusion impedance. Theoretically, the solar cell impedance spectroscopy has three semicircles representing high-, middle-, and low-frequency features, associated with R_{ct1} and CPE1, R_{ct2} and CPE2, and Z_N , respectively [20]. In Figure 4(a), the largest semicircle at middle-frequency almost covers the other two, which is attributed to carrier transportation and recombination at photoanode/electrolyte interfaces. The fitted values of R_{ct2} are 18 Ω , 32 Ω , and 56 Ω for the solar cells with photoanode of CdS/H:TiO₂, CdS/TiO₂, and H:TiO₂, respectively. According to our previous work, hydrogenation treatment of TiO₂ can reduce R_{ct2} because the band gap is narrowed down and meanwhile the oxygen vacancy density increases. The loading of CdS nanoparticles on pure TiO₂ also decreases R_{ct2} of the CdS/TiO₂/electrolyte interfaces significantly. For the CdS/H:TiO₂ photoanode, both of hydrogenation self-sensitization and CdS sensitization work together and result in the lowest R_{ct2} . The lower R_{ct2} reflects quicker electrons transport at the interface between electrolyte and photoanode, which implies that smaller probability of interface recombination occurs at the photoanode/electrolyte interface.

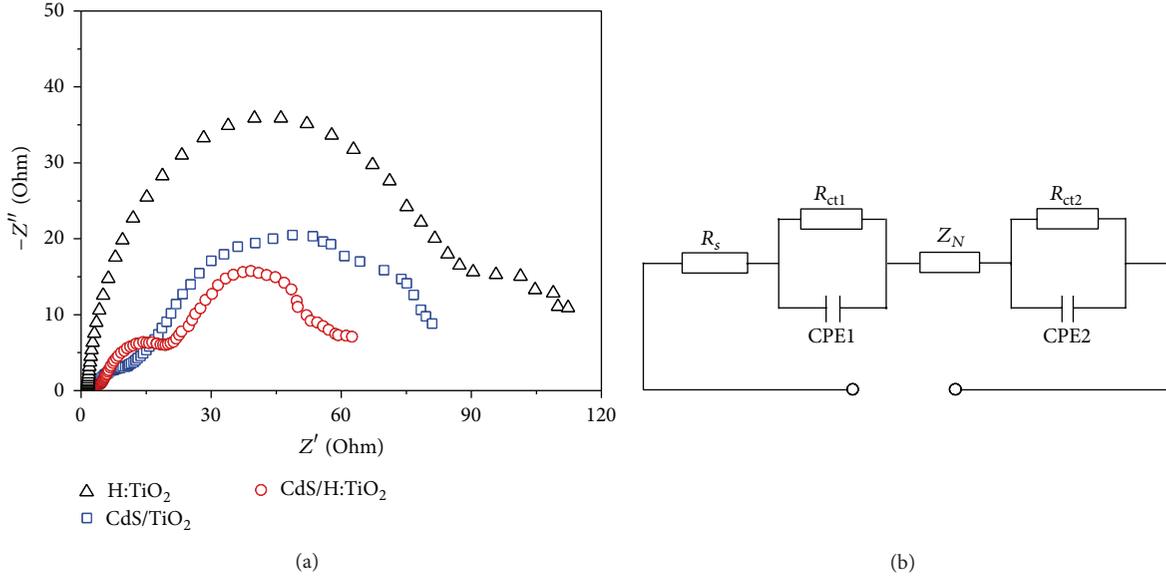


FIGURE 4: (a) Electrochemical impedance spectra of the solar cells with different photoanodes; (b) the corresponding equivalent circuit.

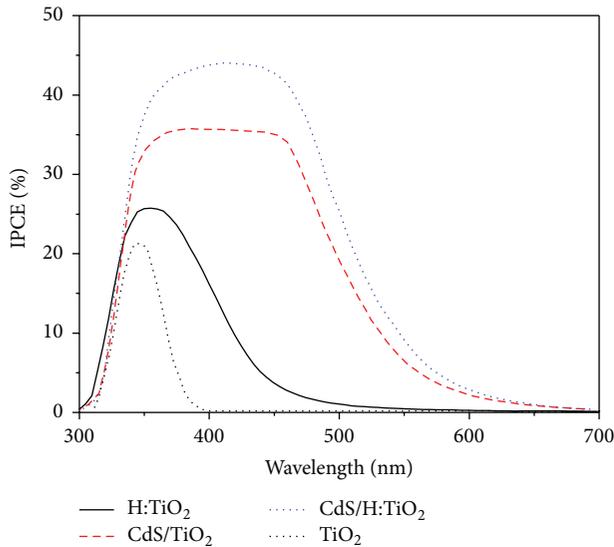


FIGURE 5: IPCE spectra of the solar cells with different photoanodes.

3.4. Photovoltaic Performances of the Solar Cells Based on Cosensitization of CdS and Hydrogenation. The incident photon to current conversion efficiencies (IPCE) spectra of the solar cells with different photoanodes are shown in Figure 5. The IPCE can be expressed as [21]

$$\text{IPCE}(\lambda) = \eta_{\text{LHE}} \eta_{\text{inj}} \eta_{\text{cc}}, \quad (1)$$

where η_{LHE} is the light harvesting efficiency, η_{inj} is the electron injection yield, and η_{cc} is the charge collection efficiency. It can be seen that the profile of IPCE plot corresponds well with the UV-vis absorption spectra in Figure 3. The IPCE of TiO_2 nanocrystal film without any sensitizer is low in the UV region and negligible in the visible region. The $\text{H}:\text{TiO}_2$

TABLE 1: The photovoltaic performances of the solar cells with different photoanodes.

Photoanodes	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF	η (%)
$\text{H}:\text{TiO}_2$	0.14	0.28	0.38	0.015
CdS/TiO_2	4.68	0.56	0.42	1.10
$\text{CdS}/\text{H}:\text{TiO}_2$	6.26	0.65	0.49	1.99

solar cell exhibits stronger IPCE. The hydrogenated TiO_2 increases oxygen vacancy density and has much more trap states near the conduction band, leading to the enhancement of the absorption as reported in our previous work [14]. The absorption enhancement implies the increase of the light harvesting efficiency, which increases the IPCE value according to the above formula. Significantly enhanced IPCE can be observed in the CdS sensitized solar cells, which can be attributed to the fact that CdS quantum dots have much higher absorption coefficients of 10^5 to $10^6 \text{ M}^{-1} \text{ cm}^{-1}$ above the band gap energy [22]. The $\text{CdS}/\text{H}:\text{TiO}_2$ solar cell exhibits the strongest IPCE due to the cosensitization of CdS and self-sensitization of hydrogenated TiO_2 . The maximum IPCE of the $\text{CdS}/\text{H}:\text{TiO}_2$ solar cell can reach 41.7% using a polysulfide electrolyte, suggesting that the cosensitization of CdS and hydrogenation has the potential to enhance the photoelectric properties.

Figure 6 shows the illuminated and dark J - V characteristics of the solar cells with different photoanodes, and the photovoltaic performances of them are listed in Table 1. The self-sensitization effect of $\text{H}:\text{TiO}_2$ photoanode with polysulfide electrolyte is very weak, exactly as our previous results of $\text{H}:\text{TiO}_2$ photoanode with the I^-/I_3^- electrolyte. The CdS/TiO_2 QDSSC also displays the ordinary photovoltaic performance as others' work with an energy conversion efficiency of 1.1%. Compared with the other two solar cells, the $\text{CdS}/\text{H}:\text{TiO}_2$ solar cell, which is cosensitized by CdS and

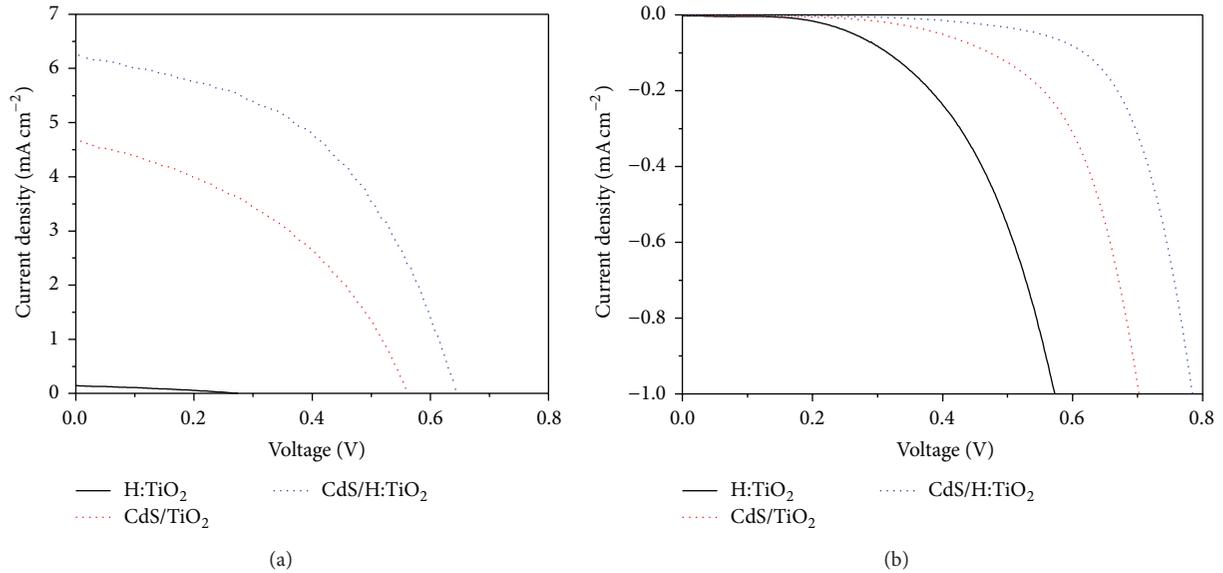


FIGURE 6: The J - V characteristics of the solar cells with different photoanodes (a) under simulated AM 1.5 solar spectrum irradiation at 100 mW cm^{-2} and (b) in the dark.

hydrogenation, exhibits enhanced photovoltaic characteristics with short circuit current density (J_{sc}) of 6.26 mA/cm^2 , open-circuit voltage (V_{oc}) of 0.65 V , fill factor (FF) of 0.49 , and power conversion efficiency (η) of 1.99% .

The enhancement of J_{sc} can be explained by the equation: $J_{sc} = J_{inj} - J_{rec}$, where J_{inj} and J_{rec} are the electron injection current density and the recombination current density [23]. The stronger absorption at the UV-vis (as shown in Figure 3) and the higher IPCE (as shown in Figure 5) of CdS/H:TiO₂ hold out the potential of increasing J_{inj} and consequently enhancing the photoelectric effect. On the other hand, the recombination current is derived from the interface recombination mostly caused by the reduction reaction with the electron in the conduction band of TiO₂ and S_n^{2-} in the electrolyte, which would form the dark currents and reduce photovoltaic performance of solar cells. As shown in Figure 6(b), the dark current density of the solar cell with CdS/H:TiO₂ photoanode is obviously lower than that of the CdS/TiO₂ or H:TiO₂ solar cell, which should be attributed to lower R_{ct2} of the CdS/H:TiO₂ solar cell than that of the other two solar cells. The enhanced J_{inj} and the suppressed dark current density of the CdS/H:TiO₂ solar cell lead to the enhancement of the J_{sc} .

Under constant illumination, the solar cell reaches a photostationary situation, and V_{oc} corresponds to the increase of the quasi-Fermi level of the semiconductor (E_{Fn}) with respect to the dark value (E_{F0}), which equals the electrolyte redox energy ($E_{F0} = E_{redox}$). V_{oc} can be determined by the following equation [24]:

$$V_{oc} = \frac{E_{Fn} - E_{redox}}{e} = \frac{k_B T}{e} \ln \left(\frac{n}{n_0} \right), \quad (2)$$

where e is the positive elementary charge; $k_B T$ is the thermal energy; n is the electron concentration in conduction band of the semiconductor photoanode under illumination; n_0 is

the electron concentration in the dark condition. Here, n and n_0 can be characterized by IPCE and dark current density, respectively. The stronger IPCE as shown in Figure 5 implies the higher n of the CdS/H:TiO₂ solar cell than the H:TiO₂ solar cell and the CdS/TiO₂ solar cell, while the lower dark current density as shown in Figure 6(b) manifests the lower n_0 of the CdS/H:TiO₂ solar cell than the other two solar cells. As a result, the higher n and the lower n_0 lead to the improvement of V_{oc} for the CdS/H:TiO₂ solar cell as shown in Figure 6(a).

4. Conclusions

In summary, a cosensitized TiO₂ photoanode by CdS quantum dots sensitization and self-sensitization of hydrogenated TiO₂ film was achieved by depositing CdS quantum dots on the surface of hydrogenated TiO₂ film. By comparing solar cells with different photoanodes of H:TiO₂, CdS/TiO₂, and CdS/H:TiO₂, the cosensitization effect by CdS and hydrogenation in the CdS/H:TiO₂ solar cell enhanced photovoltaic performance with power conversion efficiency (η) of 1.99% , which was increased by more than 80% compared with CdS/TiO₂ solar cells. The cosensitization effect combined the quantum dots sensitization and self-sensitization of hydrogenated TiO₂ films and caused larger extension absorption in the visible light range, quicker electrons transport, smaller probability of interface recombination, and consequently better photovoltaic performance. This study will give some useful enlightenment to the development of novel inorganic low-cost solar cells.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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