RESEARCH ARTICLE

Improving the photovoltaic parameters in CdS quantum dot sensitized SnO_2 based solar cells through incorporation of chemically deposited compact SnO_2 layer

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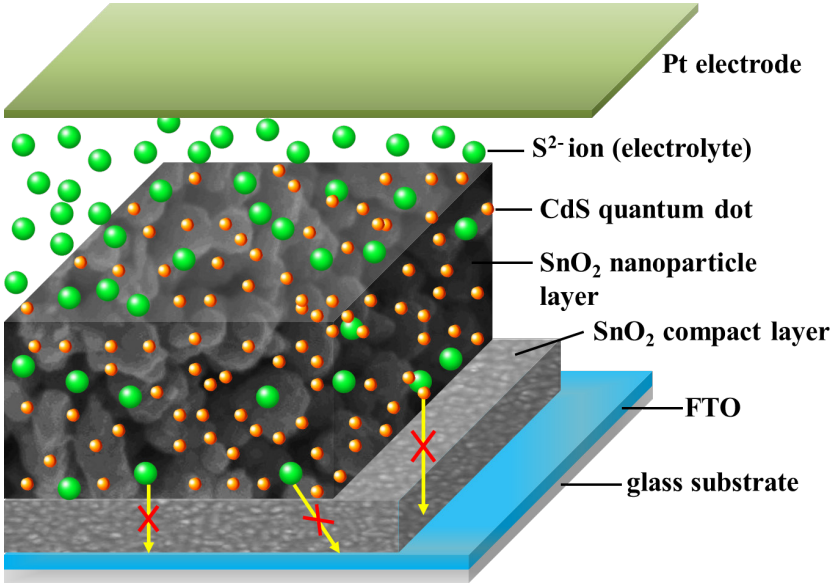

Highlights

- Compact layer incorporation has improved the solar cell performance by 250%.
- The compact layer facilitates charge extraction and reduces carrier recombination.
- The photoanode with compact layer displays a longer lifetime (τ).

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Improving the photovoltaic parameters in CdS quantum dot sensitized SnO_2 based solar cells through incorporation of chemically deposited compact SnO_2 layer

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Abstract: Mitigation of the recombination of electrons within the quantum dot sensitized solar cells (QDSSCs) and hence the improvement in the performances of the devices can be achieved by incorporation of a compact layer in between the transparent conducting substrate and the semiconducting materials used in these devices. In this work, a facile and cost-effective method of incorporation of a compact layer of SnO_2 over the Fluorine doped Tin oxide (FTO) substrate and its effect on the efficiency enhancement of the CdS sensitized SnO_2 QDSSCs have been studied by means of current-voltage characteristics, Electrochemical Impedance Spectroscopy, and electron lifetime estimation. The incorporation of SnO_2 compact layer improved the overall efficiency of the device by 250% as compared with the devices fabricated with no compact layer under the illumination of 100 mW cm^{-2}. The improvement in the open-circuit voltage and the significant enhancement in the short circuit current density (~200%) together with the increase in the electron lifetime in the QDSSC with compact layer suggested that the compact layer has acted as a weak energy barrier, which increased the electron density in the mesoporous SnO_2 film. The enhancement in the short circuit current density and the efficiency mainly stems due to the decrease in the series resistance and the increase in the recombination resistance of the device fabricated with the compact layer.

Keywords: SnO_2 compact layer; CdS sensitization; SILAR; SnO_2 photoanode.

INTRODUCTION

Within the third-generation photovoltaic cells, quantum dots (QDs) based solar cells (QDSSCs) have recently attracted much attention due to the distinct properties of the quantum dots. High molar extinction coefficients, ability of multiple exciton generation, high stability to heat, tunable energy gap due to the quantum confinement effect, and high theoretical efficiency are some of the major advantages of quantum dots based solar cells (Tian and Cao, 2013; Pan *et al*., 2018; Zahu *et al*., 2020). Quantum dots such as CdS (Chang and Lee, 2007; Chen *et al*., 2009; Sudhagar *et al*., 2009 ; Tak *et al*., 2009; Zhu *et al.*, 2011), CdSe (Robel *et al*., 2007; Lee *et al*., 2008; Chen *et al*., 2010) and PbS (Hoyer and Konenkamp, 1995; Plass *et al*., 2002; Leventis *et al*., 2010) are widely used in these QDSSCs. Among these QDs, CdS has attracted a wide interest due to its suitable optical properties for the solar cells, such as efficient light photon absorption in the visible region of the solar spectrum (Nozik, 2002; Chang and Lee, 2007; Chen *et al*., 2009; Zhu *et al*., 2011). Typically, in the construction of QDSSCs, suitable quantum dots have been used to sensitize a semiconducting material such as TiO_2. Improvement in the power conversion efficiencies (PCE) of QDSSCs from less than 5% to 12% over have been reported with the usage of complex quantum structures and modifications of photoanode over the past decade (Du *et al*., 2016; Zhang *et al*., 2019; Pan *et al*., 2019; He *et al*., 2020). However, the efficiencies of CdS sensitized solar cells fabricated without any post treatments, combinations or modifications on QDs are still lying in the range of 0.7-1.1% (Kim *et al*., 2011; Zhou *et al*., 2013). Since these QDs do not interconnect via chemical bonding with the semiconducting material, the structure and the morphology of the photoanode are a crucial factor determining the power converting efficiency (PCE) of the device especially in the charge transfer processes (Tian and Cao, 2013; Pan *et al*., 2018; Sahu *et al*., 2020;). Photoanodes fabricated with TiO_2 based substrates are currently the most attractive structures in fabrication of QDSSCs. This is due to the higher efficiencies achieved compared to other semiconductors such as SnO_2 or ZnO (Ju *et al*., 2010; Tian and Cao, 2013). However, further improvements in the device performances have been limited due to the low electron mobility and transport properties of the TiO_2 (Hendry *et al*., 2006). In this scenario, SnO_2 is a promising candidate having a wide band gap (3.6 eV) and higher electronic mobility. Moreover, its low sensitivity to UV degradation and better long term stability, make the SnO_2 as a very promising material for these devices (Park *et al*., 2004). However, application of SnO_2 as a photo anode is confined due to the poor adhesion to the substrate materials like Fluorine doped Tin oxide glasses (FTO). Moreover, comparatively little work has been carried out on SnO_2 to achieve the results suggested by theoretical studies. Most

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of the devices showing comparable efficiencies with the TiO_2 photoanodes are fabricated with a combination of CdS with CdSe (Liu *et al*., 2020). However, to the best of our knowledge, only very few studies have been carried out with CdS sensitized solar cells with SnO_2. In this context, Zhou *et al*. (2013) have reported CdS sensitized SnO_2 solar cells with 0.22% PCE and very recently Liu *et al*. (2019) have reported CdS sensitized sol-gel derived thin films of self-patterned micro-blocks of closely-packed SnO_2 nanoparticles as high-performance photoanodes in alkaline solution of methanol with higher photo current density. However, overall device performances with PCE were not reported. In this study, a novel photoanode structure comprising with compact layer of SnO_2 was introduced in the first time in the fabrication of CdS sensitized SnO_2 solar cells towards the efficiency enhancement. The device performances have been compared with and without the compact layer in the photoanode.

EXPERIMENTAL

Fabrication of SnO_2 electrode

$SnCl_2$ (2.0 g) was ground well with 4.0 mL of 1 M H_2SO_4 for 15 minutes. $SnCl_2$ reacted with H_2SO_4 to form $SnSO_4$. This paste was spin coated on the pre cleaned FTO at 3000 rpm for 60 seconds. Then, the $SnSO_4$ coated FTO glass was sintered at 500 ºC for 2 hours. At 500 ºC, $SnSO_4$ oxidized and SnO_2 was formed. For future reference, this layer is referred as the SnO_2 compact layer (CL). Then, 3.0 mL of SnO_2 colloidal suspension, 10 drops of glacial acetic acid, and 0.02 g of MgO were ground well. Then, 5 drops of Triton X-100 and 40.0 mL of absolute ethanol were added to the mixture. The resulting nanoparticle (NP) colloidal sample was sonicated for 20 minutes. FTO/SnO_2 CL was placed on a hot plate at 150 ºC. Then the colloidal mixture was sprayed over for several times. Then, the SnO_2 CL/ SnO_2 NP electrode films were sintered at 500 ºC for 30 minutes.

Deposition of CdS quantum dots

CdS quantum dots were deposited on the SnO_2 electrode using successive ionic layer adsorption and reaction (SILAR) method. 0.4 M cadmium chloride ($CdCl_2$) and 0.1 M sodium sulfide (Na_2S) aqueous solutions were prepared for cationic (Cd^{2+}) and anionic (S^{2-}) precursors respectively. For the SILAR method, the SnO_2 film was dipped in the cationic precursor for 1 minute and thoroughly washed in ethanol : deionized water (1:1) mixture. The same procedure was carried out with the anionic precursor with the same dipping time. 10 SILAR cycles were carried out to fabricate the best CdS quantum – dot sensitized photoanode.

Preparation of the polysulfide electrolyte

Liquid polysulfide electrolyte was prepared by the following method. 2 M sulfur and 2 M Na_2S were dissolved in a mixture of deionized water and methanol in the ratio of 3:7 (v/v). The mixture was stirred for 30 minutes.

Solar Cell assembly

An appropriate amount of the polysulfide electrolyte was applied on to the CdS sensitized photoanode and then sandwiched it with the platinized counter electrode as schematically shown in Figure 1. Platinum coated FTO glass substrate was used as the counter electrode. Then the solar cell was clipped together using couple of steel clips.

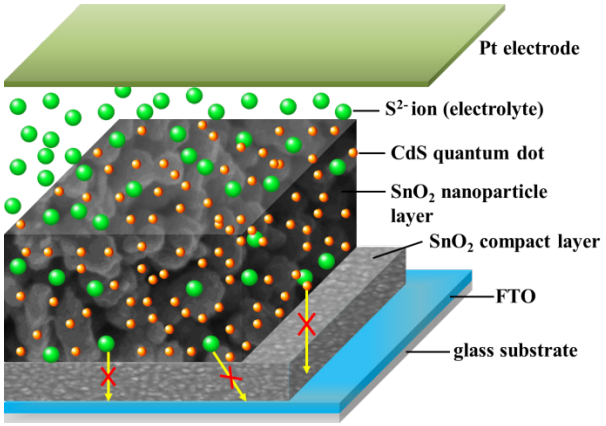

Figure 1: Schematic diagram showing the CdS sensitized solar cell with SnO_2 compact layer and porous nanoparticle (NP) SnO_2 layer.

Current -Voltage characterization

In order to compare the photovoltaic performance of the QDSSCs, with and without the compact layer of SnO_2, current density-voltage (*J-V*) measurements of solar cells were measured under the illumination of 100 mW cm^{-2} with AM 1.5 spectral filter using a computer controlled multi-meter (Keithley 2000 model) coupled with potentiostat/ galvanostat unit (HA-301). The active area of the QDSSC was 0.25 cm^2.

Electrochemical Impedance Spectra (EIS) measurements

In order to determine the interfacial resistance values of the solar cells, electrochemical impedance spectra of the QDSSC was obtained by using PGSTAT128 N with an FRA 32M Frequency Response Analyzer (Metrohm) under the illumination of 100 mW cm^{-2} (Solar simulator with AM 1.5 spectral filter) in the frequency interval from 0.01 Hz to 1 MHz. Carrier transport resistance, recombination resistance and series resistance of the interfaces of the QDSSCs were calculated by fitting the electrochemical impedance spectroscopy (EIS) data with an appropriate equivalent circuit of the QDSSC. Electron lifetimes in the solar cells were estimated with Bode phase plots.

RESULTS AND DISCUSSION

Photovoltaic performance

Figure 2 shows the performances of the QDSSCs with and without the compact layer of SnO_2. Curve (1) shows the current– voltage (*J–V*) characteristics of the devices

with no compact layer in the photoanode, while curve (2) shows the same characteristics with the photoanode with SnO_2 compact layer under the illumination of 100 mW cm^{-2}. Device parameters namely, short circuit current density (J_{SC}), open circuit voltage (V_{OC}), fill factor (FF) and the power conversion efficiency (η) determined from those curves and are tabulated in Table 1. The highest efficiency of 0.77% is achieved from the QDDSC fabricated with the photoanode, comprised with a compact SnO_2 layer which is ~250% higher than that of the device assembled with no compact layer in the photoanode with the same thickness of mesoporous SnO_2 layer. As it is evident from these two curves and also from the values tabulated in the Table 1, both the V_{OC} and J_{SC} of the CdS QDSSCs have been greatly enhanced upon the addition of the SnO_2 compact layer. However, the most noticeable is the enhancement in the J_{SC}, which has increased from 2.28 mA cm^{-2} to 6.82 mA cm^{-2}. The fill factor has not been drastically affected by the novel SnO_2 layer. This improvement in photovoltaic performance of compact layer based DSSC could be attributed to the decrees in the electron recombination within the device. The efficiency of the reference cell with no compact layer was recorded to be 0.21%. This is in well agreement with the efficiency value of 0.22% reported by Zhou *et al.* (2013) in CdS sensitized SnO_2 solar cells.

Electrochemical impedance spectroscopy (EIS)

In order to get an idea about the electronic and ionic processes and recombination resistances of the QDSSCs, Electrochemical impedance spectroscopy (EIS) measurements were performed on the devices fabricated with and without the compact layer of SnO_2. Figure 3 shows the Nyquist plots of the CdS sensitized QDSSCs and the equivalent circuit model used. As it is depicted in the figure, two characteristic semicircles including a smaller one at higher frequency and a larger one at intermediate frequency can be seen. In equivalent circuit, R_S is the series resistance which represents the FTO/SnO_2 interface, while R_{1CT} is the resistance at the counter electrode/electrolyte interface and R_{2CT} is the resistance at the photoanode/electrolyte interface which is known as the recombination resistance. CPE_1 and CPE_2 are the constant phase elements and W is the finite Warburg impedance element which is related with diffusion process. Low recombination resistance favors the electron recombination, which negatively affects the cell performance. Hence, a higher recombination resistance is favorable to higher cell performance. EIS parameters are listed in the Table 2 (Dissanayake *et al.*, 2019). Table 2 shows the impedance parameters of the devices estimated from the above equivalent circuit. As it is evident from the Table 2, the series resistance R_S of the QDSSC fabricated with SnO_2 compact layer in the photoanode is lower than those of the device fabricated without such a compact layer in the photoanode. On the other hand, significant increase in both charge transfer resistances R_{1CT} and the R_{2CT} can be observed. Even though there is no evidence to correlates the increase in the R_{1CT} with the enhancement of cell performances, the increase in the R_{2CT}, which represents the recombination resistance can be correlated with the increase in the J_{SC} as well as in the V_{OC} as tabulated in the Table 1. Therefore, EIS data are in correlation with the increase in the efficiency due to the enhancement in both J_{SC} and V_{OC} of the QDSSC with SnO_2 compact layer in the photoanode (Dissanayake *et al.*, 2020).

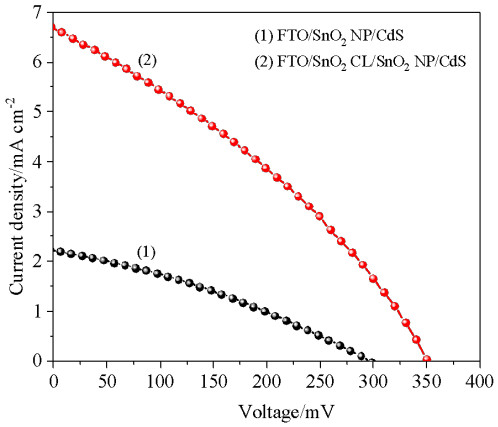

Figure 2: Current – voltage characterization of CdS QDSSCs (1) without and (2) with SnO_2 compact layer in the photoanode under the illumination of 100 mW cm^{-2}.

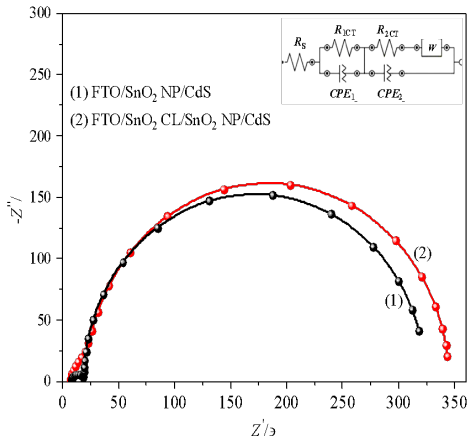

Figure 3: Nyquist plots of CdS QDSSCs (1) without and (2) with the SnO_2 compact layer in the photoanode, under the illumination of 100 mW cm^{-2} with AM 1.5 spectral filter.

Table 1: Photovoltaic parameters of CdS QDSSCs under the illumination of 100 mW cm^{-2}.

Photoanode	V_{OC} (mV)	J_{SC} (mA cm^{-2})	FF (%)	Efficiency (%)
FTO/SnO_2 CL/SnO_2 NP/CdS	350.95	6.82	32.21	0.77
FTO/ SnO_2 NP/CdS	297.85	2.28	31.01	0.21

Figure 4 shows the Bode phase plots obtained based on the EIS measurements of CdS QDSSCs with and without SnO_2 compact layer. Life-time of the electron τ can be calculated from the frequency corresponding to the maximum phase angle f_{max} using the following equation (Kumari *et al*., 2019) and estimated values are tabulated in Table 3.

$$\tau = \frac{1}{2\pi f_{max}} \quad \text{..... (1)}$$

The electron lifetime is directly proportional to the recombination resistance (R_{2CT}), which is consistent with the data. Further, the higher electron lifetime positively affects the photocurrent as well (Kumari *et al*., 2019). As shown in the Table estimated electron life-time of the device fabricated with the compact layer is higher than that of the device without a compact layer in the photoanode. This means that in this novel photoanode configuration electrons diffuse and transfer more easily than that of the photoanode with no compact layer. Higher electron lifetime also implies reduced recombination of photoelectrons between SnO_2 photoanode and the electrolyte, which contributes to the enhancement in J_{SC} leading to overall efficiency enhancement (Kumari *et al*., 2019).

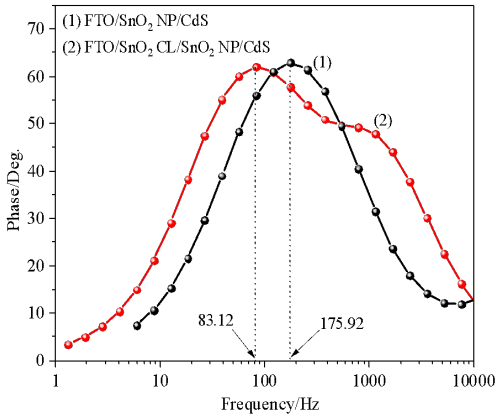

Figure 4: Bode phase plots of CdS QDSSCs (1) without and (2) with the SnO_2 compact layer in the photoanode, under the illumination of 100 mW cm^{-2} with AM 1.5 spectral filter.

Table 2: : EIS parameters of CdS QDSSCs under the illumination of 100 mW cm^{-2} with AM 1.5 spectral filter.

Photoanode	R_S (Ω)	R_{1CT} (Ω)	R_{2CT} (Ω)
FTO/SnO_2 CL/SnO_2 NP/CdS	7.42	322	15.3
FTO/ SnO_2 NP/CdS	7.88	305	11.0

Table 3: Comparison of electron lifetime values with photovoltaic parameters.

Photoanode	f_{max} (Hz)	τ (ms)	J_{SC} (mA cm^{-2})	Efficiency (%)
FTO/SnO_2 CL/SnO_2 NP/CdS	83.12	1.91	6.82	0.77
FTO/ SnO_2 NP/CdS	175.92	0.90	2.28	0.21

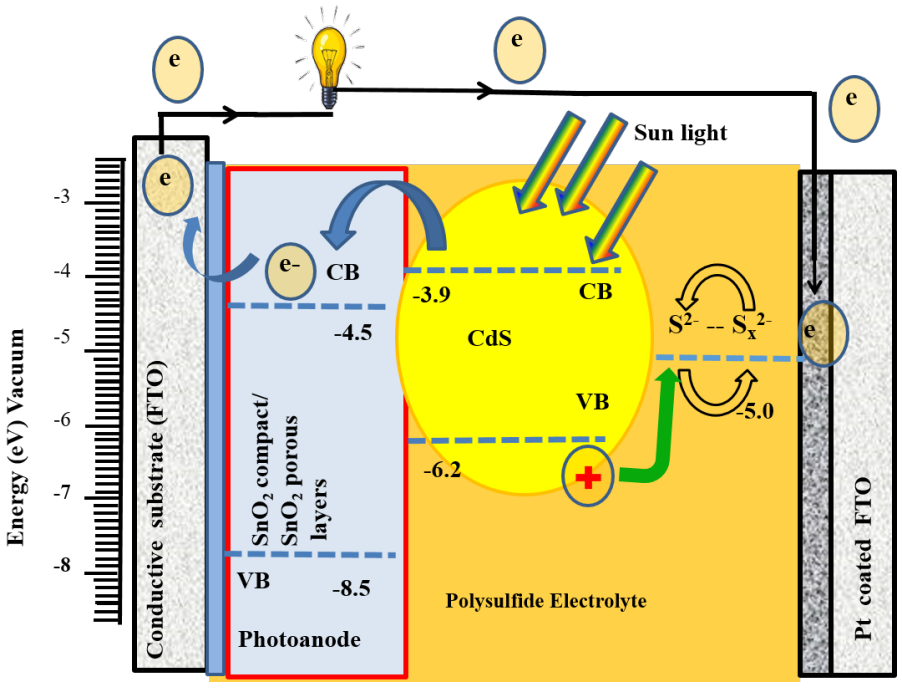

Figure 5: Energy band diagram of SnO_2 and working mechanism of CdS sensitized SnO_2 photoanode with compact SnO_2 layer.

In order to show the above charge transfer process within the device, band positions and possible charge transfer mechanism are schematically shown in Figure 5 (Lee and Chang, 2008; Kim *et al*., 2011; Zhou *et al*., 2013). As it is well known when the QDSSCs are exposed to the sunlight, the QDs harvest some photon energy in the light corresponding to their band gap values. Accordingly in this case CdS QDs with 2.3 eV band gap absorb the corresponding photons and generate the photo-excited electrons. These excited electrons are then injected from its conduction band (CB) to the CB of the SnO_2 semiconductor and then subsequently migrated to the external circuit through the conductive substrate where SnO_2 is deposited (FTO). These electrons then migrate towards the counter electrode. Meantime holes produced by the photo-excitation process synchronously in the valence band (VB) of the QDs, are immediately transferred to the redox electrolyte to oxidize it. The oxidized electrolyte obtained the electrons in the counted electrode from external circuit in the device. One of the crucial factors determining the overall efficiency is the electron separation and transport before recombining within the device. Therefore, in the present case due the existence of the compact SnO_2 layer in between the FTO and the porous SnO_2 the back electron transfer from the FTO substrate must be minimized as confirmed by the EIS measurements.

CONCLUSION

In summary, here we report the introduction of the compact SnO_2 layer by a simple chemical solution deposition method, which is instrumental in improving the overall photovoltaic performance of the SnO_2 based CdS quantum dot sensitized solar cells. Incorporation of this compact layer resulted in 250% efficiency enhancement in CdS sensitized SnO_2 based solar cells favoring the forward direction electron transfer and prevents the electron recombination either or both with the redox mediator and the holes produced in the CdS quantum dots.

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STATEMENT OF CONFLICT OF INTEREST

The authors declare no conflict of interest.

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