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Alloyed $CdTe_{0.6}S_{0.4}$ Quantum Dots Sensitized TiO_2 Electrodes for Photovoltaic Applications

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ABSTRACT

The photovoltaic performance of alloyed $CdTe_{0.6}S_{0.4}$ quantum dot sensitized solar cells (QDSSCs) is investigated. Fluorine doped Tin Oxide (FTO) substrates were coated with 20 nm-diameter TiO_2 nanoparticles (NPs). Pre-synthesized colloidal solution of alloyed $CdTe_{0.6}S_{0.4}$ quantum dots (QDs) of 4.2 nm was deposited onto TiO_2 NPs substrates using direct adsorption (DA) technique, by dipping for different times at ambient conditions. The FTO counter electrodes were coated with platinum, while the electrolyte containing I^-/I^{-3} redox species was sandwiched between the two electrodes. Compared to pure CdTe QDs and CdS QDs, $CdTe_{0.6}S_{0.4}$ QDs showed better photovoltaic performance. The J-V characteristic curves of the assembled QDSSCs were measured at AM 1.5 simulated sunlight. The short current density (J_{sc}) and efficiency (η) increase with dipping time. At 24 h dipping time, the open-circuit photovoltage V_{oc}, J_{sc}, fill factor (FF), and η were 0.46 volts, 1.54 mA/cm^2, 0.43% and 0.31%, respectively.

KEYWORDS

Alloyed $CdTe_{0.6}S_{0.4}$; Quantum Dot; Quantum Dots Sensitized Solar Cell; Photovoltaic Cell

1. Introduction

In the last two decades, semiconductor quantum dots (QDs) or nanoparticles (NPs) have been under intensive investigations, due to their unique size-dependent optical, electrical, thermal and magnetic properties [1-5], especially when their size is below the corresponding exciton radius. These attractive properties make QDs an important candidate in many applications such as, high efficiency thin film transistors, light-emitting diodes [6], electron-beam pumped lasers, electroluminescent devices and others [7,8]. Recently, QDs have become one of the most promising materials in solar cell fabrication [4,9, 10]. This third generation of solar cells is called quantum dots sensitized solar cells (QDSSCs). Tuning QDs properties by changing the particle size may cause problems in some applications, in particular, if unstable small particles (less than 2 nm) are used [11]. To overcome these problems, a new class of alloyed semiconductor nanoparticles (NCs) have been studied [11,12] to provide a way for continuous tuning of their energy band gap without changing the particle size. Alloyed AB_xC_{1-x} NCs are becoming increasingly important in many areas of nanoscale engineering through gradual variation of the composition variable x [13-17]. Alloyed $CdTe_xS_{1-x}$ QDs band gap can be adjusted to harvest the visible region of the solar spectrum by varying the tellurium molar ratio. This makes $CdTe_xS_{1-x}$ QDs a potentially favorable material for photovoltaic solar cell applications, where QDs of the same size but with varying optical properties might be preferable. QDs are deposited onto a mesoporous large band of semiconductors such as TiO_2 NPs [18,19], ZnO NPs [19], and SnO_2 NPs [20] to harvest the incident solar power. Mainly, two different strategies are used [4,21]: *in situ* growth of QDs by either a chemical bath deposition (CBD) containing both cationic and anionic precur-

sors [18] or the successive ionic layer adsorption and reaction deposition (SILAR) method [22]. These methods provide good surface coverage, but they limit the control of QD size and yield a broad size distribution [23]. These drawbacks can be avoided if the QDs are synthesized prior (*ex situ*) to being deposited [24] via electrophoretic deposition (EPD) [23], linker-assisted adsorption (LA) [25], or direct adsorption (DA) [16,26] using different dipping times. The last method was the one applied in this work. **Figure 1** shows an artistic depiction of the basic design of a QDSSC.

In this work, we synthesize alloyed $CdTe_{0.6}S_{0.4}$ QDs by organometallic pyrolysis method to be used as sensitizers in QDSSCs. These colloidal QDs were adsorbed onto TiO_2 NPs by DA technique for different dipping times under ambient conditions. The photovoltaic characteristic parameters, short circuit current density (J_{sc}), open circuit voltage (V_{oc}), fill factor (FF), and efficiency (η) for energy conversion) of alloyed $CdTe_{0.6}S_{0.4}$ QDSSCs were studied for four dipping times (1 h, 3 h, 6 h, and 24 h). To the best of our knowledge, this is the first time that DA technique has been used to deposit such kind of QDs onto TiO_2 NPs to assemble QDSSCs.

2. Experiment

2.1. Preparation of Alloyed $CdTe_{0.6}S_{0.4}$ QDs

Alloyed $CdTe_{0.6}S_{0.4}$ QDs sample was synthesized as the method of Talapin *et al*. [27] by varying the amount of the second precursor. Cadmium solution was prepared by 0.3 g of CdO added to 3.0 g of stearic acid, and heated up to 170˚C till the red color of CdO disappears to ensure that the reaction between CdO and stearic acid is complete and CdO completely transform to Cd sterate. 2.0 g of tri-*n*-octylphosphine oxide (TOPO) and 1.0 g of hexadecyleamine (HDA) are added to the reaction mixture and heated at 200˚C. Tellurium solution was prepared by mixing 0.1 g of tellurium in 0.1 mL trioctylphosphine (TOP). Sulfur solution was also prepared by dissolving 0.2 g of sulphur 2.0 mL (TOP). Appropriate amounts of sulfur and tellurium solutions were mixed together to give the desired ratio (Te/S: 0.6/0.4). The mixture was then injected into the cadmium solution at a temperature 200˚C. A QDs sample with molar ratios (x = 0.6) was obtained from the reaction mixture at time interval of 5 minutes. The sample was separated using centrifuge. It was then washed several times with methanol to remove excess ligands. The QDs were precipitated with toluene at room temperature.

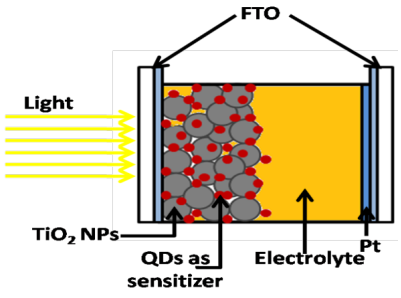


Figure 1. A depiction of the basic design of a QDSSC.

2.2. Preparation of Solar Cell Electrodes

A colloidal paste of TiO_2 NPs was prepared by the method of G. Syrrokostas *et al*. [28]. Three grams of commercial TiO_2 nanopowder (20 nm) (Degussa P-25) was ground in a porcelain mortar and mixed with a small amount of distilled water (1 ml) containing acetyl acetone (10% v/v) to create the paste. Acetyl acetone was used as a dispersing agent, since it prevents coagulation of TiO_2 nanoparticles and affects the porosity of the film. The paste was diluted further by slow addition of distilled water (4 ml) under continued grinding. The addition of water controls the viscosity and the final concentration of the paste. Finally, a few drops of a detergent (Triton X-100) were added to facilitate the spreading of the paste on the substrate, since this substance has the ability to reduce surface tension, resulting in even spreading and reducing the formation of cracks. The TiO_2 paste was deposited on a conducting glass substrate of SnO_2:F with sheet resistance of 7 Ω/sq and >80% transmittance in the visible region, using a simple doctor blade technique. This was followed by annealing at 450˚C for 30 min. and the final thickness was 8µm after the solvent evaporation. Then the TiO_2 films were dipped into a colloidal solution of pre-synthesized alloyed $CdTe_{0.6}S_{0.4}$ QDs to form working electrodes for different times (1 h, 3 h, 6 h, and 24 h). The counter electrodes were prepared by coating another FTO substrate sheet with a resistance of 7 Ω/sq with Pt.

2.3. Assembly of QDSSC

The Pt counter electrode and alloyed $CdTe_{0.6}S_{0.4}$ QDs sensitized TiO_2 electrode were assembled as a sandwich type cell using clamps. Both electrodes were sealed using a hot-melt polymer sheet (solaronix, SX1170-25PF) of 25 µm thickness in order to avoid evaporation of the electrolyte. Finally, Iodide electrolyte solution was prepared by dissolving 0.127 g of 0.05 M Iodine (I_2) in 10 mL of water-free ethylene glycol, then adding 0.83 g of 0.5 M potassium iodide (KI). The electrolyte was inserted in the cell with a syringe, filling the space between the two electrodes.

2.4. Measurements

The sizes of the QDs were measured by high resolution transmission electron microscope (HRTEM) (JEOL JEM-2100 operated at 200 KV and equipped with Gatan CCD

higher resolution camera). X-ray diffraction (XRD) patterns were carried out with an automated powder diffractometer (Bruker D8-advace diffractometer) with Cu X-ray tube (Wavelength: kα1 = 1.540598), the tube potential is 40 KV and the current is 40 mA. The alloyed $CdTe_{0.6}S_{0.4}$ QDs absorption spectra QDs (before and after adsorption on TiO_2 electrodes) were recorded using a UV-Visible spectrophotometer (JASCO V-670). In addition, The current density-voltage (J-V) characteristics were recorded (with a Keithley 2400 voltage source/ammeter using GreenMountain IV-Sat 3.1 software), with the alloyed $CdTe_{0.6}S_{0.4}$ QDSSCs subjected to the irradiation of a solar simulator (ABET technologies, Sun 2000 Solar Simulators, USA) operating at 100 mW/cm^2 (AM1.5G). A Leybold certified silicon reference solar cell (Model: [57863]) were used to calibrate the incident solar illumination. The J-V characteristic curves of alloyed $CdTe_{0.6}S_{0.4}$ QDSSC was also studied at various illumination intensities using attenuators and calibrated by the previous Si reference solar cell. All experiments were carried out under ambient conditions.

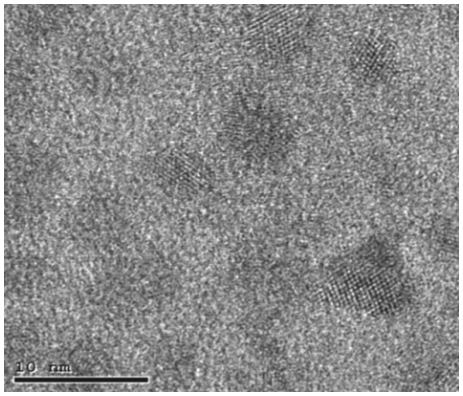
3. Results and Discussion

3.1. Characterization of Alloyed $CdTe_{0.6}S_{0.4}$ QDs

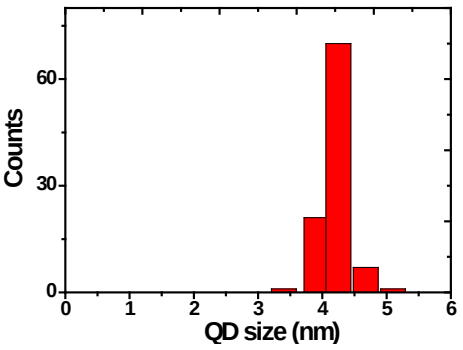
The average particle size distribution of the synthesized $CdTe_{0.6}S_{0.4}$ QDs was measured using HRTEM (JEOL 311UHR operated at 300 KV). Specimens were prepared by depositing a drop of hexane solution onto a Formvar-coated copper grid and letting it to dry in air. **Figures 2(a)** and **(b)** show the HRTEM micrograph and a histogram of particle size distribution for $CdTe_{0.6}S_{0.4}$ QDs respectively. It is observed that the particles size of the sample was about 4.2 ± 0.3 nm.

The alloyed $CdTe_{0.6}S_{0.4}$ QDs were characterized using X-ray diffractometer. The diffraction patterns seemed to be single phase with cubic sphalerite structure only. However, trials to apply MAUD program for Rietveld analysis yield bad pattern fitting as a single phase. So, we applied X'pert HighScore Plus program to identify the phases present in the samples. Two phases were identified, CdS wurtzite hexagonal structure and CdTe cubic structure. The phase composition and crystal structure of one of the samples $CdTe_{0.6}S_{0.4}$ is further investigated applying Rietveld method. **Figure 3** illustrates the pattern fitting resulting from the Rieveld analysis. The analysis yields a phase percentage of 58.2% for CdTe and 41.2% for CdS, which indicates that ternary alloys are formed.

Figure 4 shows the UV-Vis. absorption spectra of $CdTe_{0.6}S_{0.4}$ QDs in a colloidal solution. The excitonic absorption edge is easily observed at 610 nm which correspond to 2.03 eV energy band gap. The energy gap of the bulk (E_g (bulk) $CdTe_{0.6}S_{0.4}$ alloy) can be calculated using Vegard's law as follows [14,29]:



(a)



(b)

Figure 2. (a) HRTEM micrograph for $CdTe_{0.6}S_{0.4}$ QDs and (b) the histogram of particle size distribution.

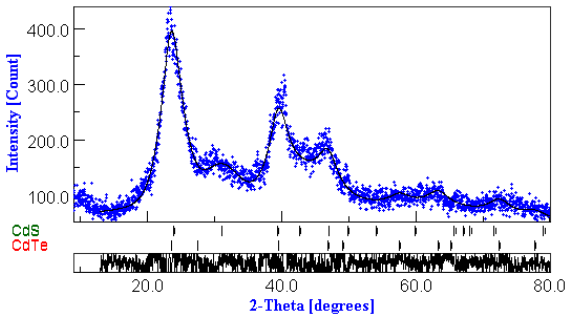


Figure 3. XRD pattern of alloyed $CdTe_{0.6}S_{0.4}$ QDs.

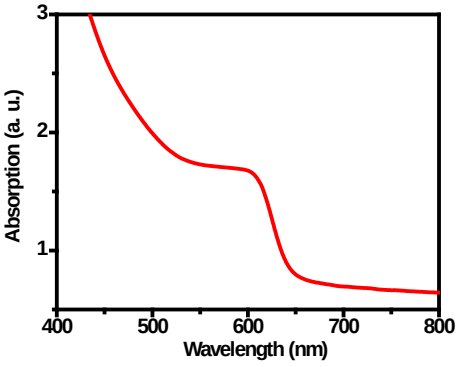


Figure 4. UV-Visible absorption spectrum for $CdTe_{0.6}S_{0.4}$ QDs.

$$\chi(CdTe_xS_{1-x}) = x\chi(CdTe) + (1-x)\chi(CdS) - x(1-x)b$$

Where χ stands for E_g (bulk) for CdTe (= 1.47 eV [30]) and for CdS = 2.42 eV [30], x (= 0.6) is the Te mole fraction, and *b* is the bowing parameter = 3.17 [13] for $CdTe_xS_{1-x}$ alloy. The calculated value of E_g (bulk) for $CdTe_{0.6}S_{0.4}$ alloy is 1.106 eV. The blue shift to shorter wavelength with respect to bulk $CdTe_{0.6}S_{0.4}$ alloy is due to the quantum confinement effect.

3.2. Characterization of $CdTe_{0.6}S_{0.4}$ QDs Sensitized TiO_2 Electrodes (The Working Electrode)

The UV-Vis. absorption spectra of alloyed $CdTe_{0.6}S_{0.4}$ QDs sensitized TiO_2 NPs electrode for different dipping times (0, 1 h, 3 h, 6 h, and 24 h) are shown in **Figure 5**. It is clearly seen that the absorption increases as the dipping time increases up to 24 hours, due to the increase of the adsorbed amount of alloyed $CdTe_{0.6}S_{0.4}$ QDs, while the onset of the absorption edge of $CdTe_{0.6}S_{0.4}$ QDs is clear at 24 hours dipping time as shown in **Figure 5**.

3.3. Characterization of Alloyed $CdTe_{0.6}S_{0.4}$ QDSSCs

The J-V characteristics curves of the assembled $CdTe_{0.6}S_{0.4}$ QDSSCs are shown in **Figure 6** for four dipped times (1 h, 3 h, 6 h, 24 h) using TiO_2 photoelectrodes and 100 mW/cm^2 from a solar simulator. The values of V_{oc}, J_{sc}, FF, and η of the assembled QDSSCs are given in **Table 1**. It is observed that both J_{sc} and η increase as the dipping time increases up to 24 hours. These results could be explained as follows; as the dipping time increases, the adsorbed amount of alloyed $CdTe_{0.6}S_{0.4}$ QDs increases, which leads to increase in the absorption of the incident light. Our earlier measurements of J_{sc} and η for CdTe QDSSCs [4] and CdS QDSSCs [16] having the same

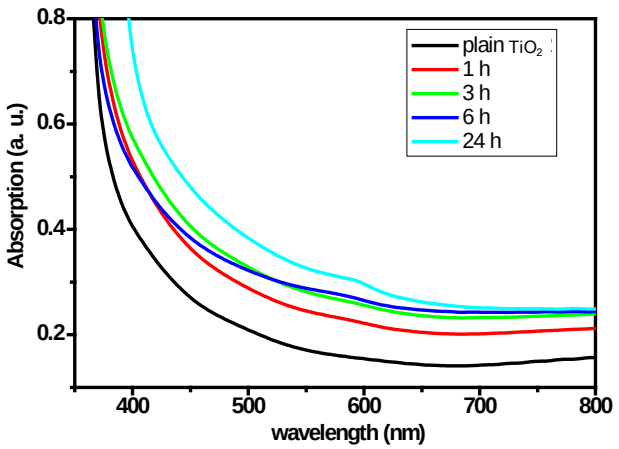


Figure 5. UV-Vis. absorption spectra of alloyed $CdTe_{0.6}S_{0.4}$ QDs/TiO_2 electrode for different dipping times.

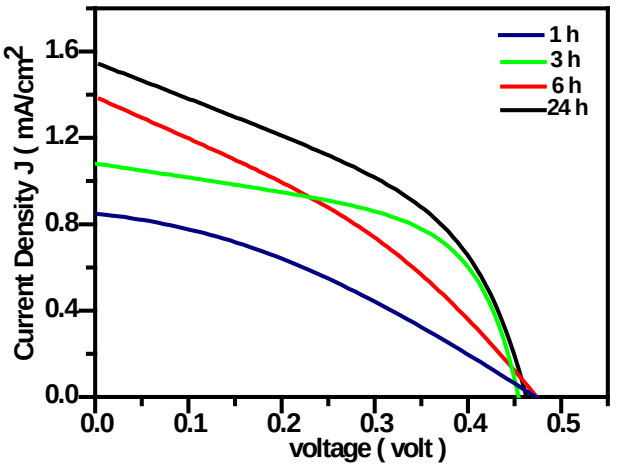


Figure 6. J-V characteristic curves of alloyed $CdTe_{0.6}S_{0.4}$ QDs QDSSCs for different dipping times.

Table 1. J-V characteristics parameters of alloyed $CdTe_{0.6}S_{0.4}$ QDSSCs for different dipping times.

Dipping time (hour)	V_{oc} (Volt) ±0.01	J_{sc} (mA/cm^2) ±0.01	FF	η (±0.01) %
1	0.47	0.84	0.37	0.15
3	0.45	1.07	0.55	0.24
6	0.47	1.38	0.35	0.26
24	0.46	1.54	0.43	0.31

dipping time are 1.037 mA/cm^2 and 0.18% for CdTe and 0.41 mA/cm^2 and 0.11% for CdS. Therefore, Compared to pure CdTe QDs and CdS QDs, $CdTe_{0.6}S_{0.4}$ QDs show better photovoltaic performance. This enhancement of J_{sc} and η may result from improved photo-absorption efficiency, and charges separation of $CdTe_{0.6}S_{0.4}$ QDs, when Te/S approaches to 0.6/0.4, more interfaces between CdTe and CdS were formed, improving charges separation. Thus, more electrons can be injected to TiO_2 electrode, leading to up-rise of TiO_2 quasi-Fermi level and further increasing J_{sc} and η values. Additionally, the DA technique, which was used to deposit the alloyed $CdTe_{0.6}S_{0.4}$ QDs onto the TiO_2 NPs, leads to the direct pinning of the alloyed $CdTe_{0.6}S_{0.4}$ QDs bands onto those of the TiO_2 NPs without any energy barriers. This causes a direct electronic interaction between the two semiconductor materials (alloyed $CdTe_{0.6}S_{0.4}$ QDs QDs and TiO_2 NPs). The DA technique minimises the electron injection time from the CBM of alloyed $CdTe_{0.6}S_{0.4}$ QDs to that of the TiO_2 NPs.

Furthermore, Voc is approximately constant (= 0.46 ± 0.01 V) for all dipping times. Since the electrons inject quickly from the conduction band minimum (CBM) of alloyed $CdTe_{0.6}S_{0.4}$ QDs to the lowest conduction band (CB) energy of the TiO_2 NPs, indicating that the CB level of TiO_2 NPs and the valance band (VB) of the electrolyte dictate the Voc of the assembled QDSSCs.

4. Conclusion

Alloyed $CdTe_{0.6}S_{0.4}$ quantum dots (QDs) of 4.2 nm were adsorbed onto TiO_2 nanoparticles (NPs) using the direct adsorption (DA) method, for different dipping times (0 to 24 h), as a sensitizer for solar cells. Our results show that both short current density (J_{sc}) and energy conversion efficiency (η) increase as the dipping time increases. The maximum values of Jsc and η are 1.54 mA/cm^2 and 0.31% respectively for 24 h dipping time. The enhancement of J_{sc} and η may result from improving the photo-absorption efficiency, and charges separation of alloyed QDs. The open circuit voltage (V_{oc}) is approximately constant (= 0.46 ± 0.1 volt) for all dipping times, since it is only dictated by the conduction band (CB) of TiO_2 NPs and valance band (VB) of the electrolyte. The performance of such quantum dots sensitized solar cells (QDSSCs) can further be enhanced by tuning the QDs energy band gap to harvest more solar spectra.

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