Semiconductor Quantum Dots for Performance Improvement in Solar Cells

Lekshmi Gangadhar, Akhila Rajan, P. K Praseetha

Abstract: ***The solar energy is the potential renewable green energy supply while considering the provision of sunlight in abundance and requirement for clean and renewable source of energy. The quantum dots sensitized solar cells (QDSC) provide an alternative perception to current photovoltaic devices. Quantum dots are colloidal nanocrystalline semiconductors possessing distinctive properties owing to quantum confinement effects. Quantum dots synthesis is based on an aqueous medium involving cadmium sulphate, zinc acetate, thiourea and ammonium hydroxide and assemble these quantum dots with nanoporous TiO_2 films for quantum dots-sensitized solar cell applications. Titanium dioxide nanowires (TNW) were fabricated by a wet chemical method. Physical and chemical properties of the formed nanoparticles were differentiated using field emission scanning electron microscope (FE-SEM), X-ray diffraction (XRD) and ultraviolet-visible light (UV-VIS) spectrometer. The establishment of TNWs allows the electrolyte to go easily within the film, escalating the surface contact among the nanowires, the quantum dots and the electrolyte, results in improvement in the efficiency of solar cell. Since quantum dots are solution processable, they are an attractive material for the recognition of low-cost, large-area, flexible and light weight photovoltaic devices. Various parameters of quantum dot sensitized solar cell were calculated. QDSC were fabricated by combining CdS and CdS/ZnS core-shell QDs with TiO_2 wire arrays. An efficiency of 7.02% was attained for the CdS/ZnS quantum dots-sensitized solar cells using the present method. Moreover these devices processed in ambient atmosphere have shown better performance and possess enhanced chemical, thermal and photochemical stability.***

Index Terms: Cadmium sulfide, sensitization, solar cells, quantum dots

I. INTRODUCTION

Nanoscale science and technology have emerged as the forefront of science and technologies. In recent years, properties of nano materials have generated a great interest because of the technological applications of these materials. Semiconductor quantum dots have attracted much attention as of their unique electric along with optical characteristics originating from quantum confinement effects and surface effects. Quantum dots are nanoscale semiconductor crystals with distinctive optical properties and surface chemistry which are beneficial for the development of biosensors and optoelectronic devices, in practical applications.

To advance the working of conventional solar cells, quantum dot sensitized solar cells implies nanotechnology and quantum mechanics. Quantum dots are semiconducting particles and by changing the quantum dot size, the energy levels are tunable which in turn tunes the bandgap. This bandgap tuning makes quantum dots attractive for solar cells. The dots may be synthesized over a wide range of sizes that facilitate to absorb a diversity of frequencies that are hard to absorb with traditional solar cells. A quantum dot solar cell makes infrared energy accessible as half of the solar energy reaching the earth is in the infrared region, mostly in the near infrared region.

Solar cells are attracted by fundamental consideration as of their potential applications in the energy generation devices. Photovoltaic (PV) effect was reported by Becquerel, who found that two silver electrodes in an electrolyte media upon light exposure can flow the current. According to O'Regan and Grätzel, dye-sensitized solar are examined widely throughout the world [1]. The quantum dot sensitized solar cell have established more considerations lately as they comprise certain advantages above dye sensitizers, like high absorption coefficients, tunable energy gaps and generation of multiple electron-hole pair with high energy excitation. The TiO_2 nanoparticle with photo electrode illustrates substantial power conversion efficiency over a large surface area [2]. The value of power conversion efficiency of photo electrodes exceedingly depends on the morphology and structure of TiO_2. So as to extend the photovoltaic performance with excellent electron transport and light scattering ability, nanostructures like nanowires, nanotubes or nanorods are considered as photoelectrode materials for sensitized solar cells.

The sensitization in quantum dot sensitized cell relies on the band gap variation because of the quantum confinement effect. Much research has been done on the fabrication of quantum dot sensitized cell within which quantum dots are embedded in mesoporous TiO_2 structure by chemical bath deposition technique [3,4]. The surface properties of quantum dots are often customized so as to improve the photo stability of the electrodes. CdS has shown much prospect as a sensitizer. The CdS quantum dots particles were coated with ZnS to ascertain a core/shell system, where the band gap of the core lies inside the band gap of the shell material and therefore the electrons and holes that are photo generated confined within the CdS [5]. An increase within the ZnS shell thickness will increase electron-hole pair generation resulting in massive free-carrier

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Lekshmi Gangadhar, Department of Nanotechnology, Noorul Islam Centre for Higher Education, Kumaracoil, Kanya kumari District. Tamilnadu, India.

Akhila Rajan, Department of Nanotechnology, Noorul Islam Centre for Higher Education, Kumaracoil, Kanya kumari District. Tamilnadu, India.

P.K Praseetha, Department of Nanotechnology, Noorul Islam Centre for Higher Education, Kumaracoil, Kanya kumari District. Tamilnadu, India.

concentration that ends up in improvement of nonlinear optical absorption. The optical limiting threshold decreases as the ZnS shell thickness increases. High luminescence efficiency and stability have achieved for core-shell nanoparticles [6]. The organometallic precursors bring into being high quality quantum dots but the precursors are not air-stable and highly toxic.

The working electrode for QDSC is TiO_2 coated FTO, sensitized with CdS and CdS/ZnS quantum dots and the counter electrode is graphite FTO glass [7]. The energy-conversion efficiency of solar cell is likely to be reliant on the morphology and structure of the quantum dots-adsorbed TiO_2 film [8, 9]. The electrical properties of quantum dot sensitized solar cells (QDSC) under illumination and non-illumination conditions are investigated [10]. To improve the effective surface area and to absorb more quantum dots, TiO_2 films are used as photoanodes and thus more light absorption and greater efficiency can be achieved [11,12]. The high alteration efficiency achieved by quantum dot sensitized solar cell may attribute to porous titania film, quantum dots and the electrolyte [13]. To enhance electron transport, we have to afford a huge surface area to adsorb the sensitized quantum dots and enhance incident light harvest. The conversion efficiency can be further enhanced by optimizing the thickness of the TiO_2 electrode.

In the present work, the quantum dots were synthesised via chemical as well as green route. Synthesized quantum dots were successfully used as in QDSC. Effect of titanium dioxide nanowire on structural and optical properties of QDs was examined. Different parameters of quantum dot sensitized solar cell were determined. We focused on quantum dot solar cells fabricated by combining CdS and CdS/ZnS coreshell QDs with TiO_2 wire arrays. Moreover these devices processed in ambient atmosphere have shown better performance and possess enhanced chemical, thermal and photochemical stability.

II. MATERIALS AND METHODS

A. Preparation of CdS quantum dots

The starting materials are Cadmium sulphate as a Cd^{++} ion source and thiourea as an S^{--} ion source. The pH of the reaction mixture was adjusted by using ammonia. Ammonium hydroxide is added dropwise to cadmium sulphate solution at 50°C to adjust pH to 11. Thiourea is added dropwise to these results in yellow colour precipitate of CdS quantum dots. Filter the precipitate and wash certain times along with double distilled water and kept for drying in the oven at 70°C to collect yellow powders of CdS quantum dots.

B. Preparation of CdS/ZnS quantum dots

The precursors for core CdS are Cadmium sulphate and thiourea. The pH of the reaction mixture was adjusted by using ammonia. Ammonium hydroxide is added dropwise to cadmium sulphate solution at 50°C to adjust pH to 11. Thiourea is added dropwise to this which results in yellow colour precipitate of CdS quantum dots. Zinc acetate solution is added to this followed by addition of Ammonium hydroxide. Thiourea is added dropwise to these results in precipitate of CdS/ZnS quantum dots. Filter the precipitate and wash certain times along with double distilled water and kept for drying to the oven at 70°C to collect nanopowders. This results for the formation of CdS/ZnS quantum dots.

C. Preparation of TiO_2 nanowires

Take 0.1gram of anatase TiO_2 and 40 ml of 10 M NaOH, heated under stirring at 100° C for 5 hours. The mixture is kept in oven at 200° C for 24 hours. The resultant sample is filtered. The filtrate was taken and 1M HCl was added dropwise under stirring until the pH became 7. The precipitate formed is filtered, then it washed along with double distilled water and also ethanol was used.The sample is dried in hot air oven at 70° C for 8 hours. A fibrous white product was obtained.

D. Fabrication of Solarcell

The QDSC is composed of four main components: wide bandgap semiconductor film (TiO_2) crystalline nanoparticles on FTO glass, QDs adsorbed onto these nanoparticles, hole conductor electrolyte penetrating the nanocrystalline semiconductor network and a graphite counter electrode. The coating of TiO_2 nanowires paste in FTO electrode is done by doctor blading. Chemical Bath Deposition (CBD) is used for sensitization of QDs on nanostructured wide bandgap semiconductors which provide high surface coverages for QDs. The direct connection between the QD and the wide bandgap semiconductor leads to efficient charge injection from the QD into the wide bandgap material. For the application of quantum dot sensitized solar cell, potassium iodide electrolyte was developed. A solvent consisting of potassium iodide and ethylene glycol is prepared and stored in a dark container.

Fig. 1 FTO glass coated with titanium dioxide nanowires

E. Assembly of the QDSCs

The working electrode used was TiO_2 coated FTO, sensitized with CdS and CdS/ZnS quantum dots. The counter electrode was graphite FTO glass. The cell was fabricated with great care in sandwich form by clamping working electrode and counter electrode, so as to take leads from both the electrodes. An effective working area of 0.25 cm^2 was exposed to light source, while all other areas were masked. Prior to clamping, one or two drops of electrolyte freshly prepared were placed above the working electrode. It should be ensured that the whole aperture was covered by the electrolyte and the electrodes were firmly held with the help of crocodile clip. The I-V characteristics of QDSCs (FTO/TiO_2/QDs/Graphite/FTO) were measured with a solar simulator, the sun simulator Oriel Sol 3A which includes a Xenon lamp at an intensity of 100mW/cm^2 (AM1.5). An

active area around 0.25 cm^2 (0.5 cm × 0.5 cm) of the resulting cell is exposed to light. The efficiency of solar cell under illumination and non-illumination conditions (with light (wl) and without light (wol)) is also calculated. The structure of QDSC is shown in figure 2.

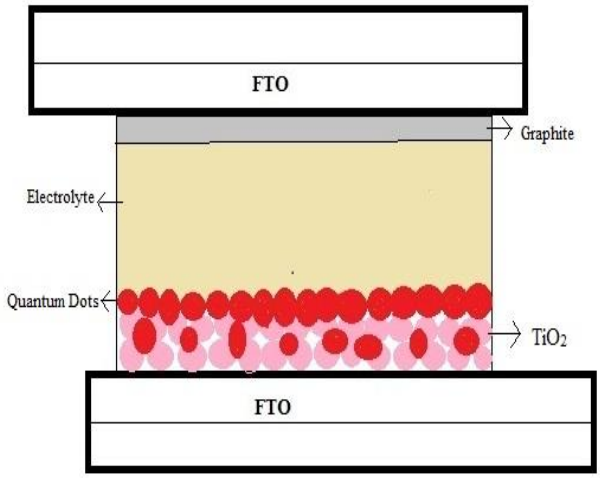

Fig. 2 Structure of quantum dot solar cell

III. RESULTS AND DISCUSSION

A. Transmission Electron Microscopy (TEM)

TEM is performed to confirm the particle size and to verify the effective formation of core-shell structure. fig. 3, indicates the clear morphology of CdS/ZnS QDs NPs among the a... size range as of 6 nm to 8 nm, illustrates that CdS Q... ...e effectively coated with ZnS shell by chemical method. Fig.3 clearly shows that have no agglomeration of QDs also we are able to attain the fine solution or powder of equivalently dispersed nanoparticles for different applications. The crystal constant change from CdS to ZnS established the formation of CdS/ZnS core/shell structures after the overgrowth of ZnS shells. Lattice fringes are clearly showed in HRTEM images throughout the entire particles is due to their high crystallinity.

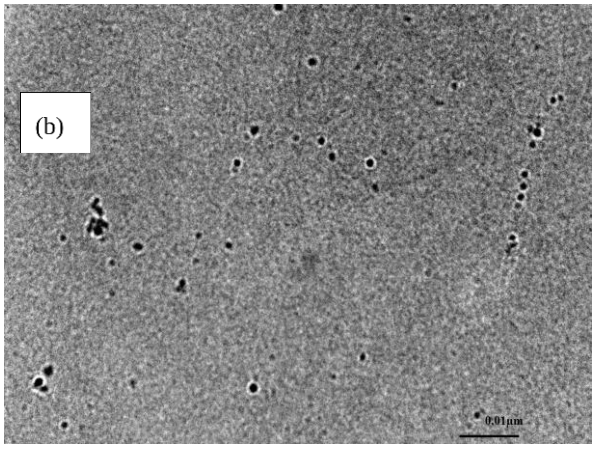

Fig. 3 TEM image of CdS/ZnS quantum dots

B. Scanning Electron Microscopy (SEM) observations of TiO_2 nanowires and quantum dots

In Fig.4, SEM pictures revealed that quantum dots and TiO_2 nanowires successfully prepared. SEM pictures shows needle like self-assembled one-dimensional nanostructures which may be regarded as TiO_2 nanowires. The average length of the wires was about 2-4 μm and the average diameter was 40-50 nm. From Fig.5 Spherical like structures were observed in QDs prepared in chemical bath deposition (CBD) method. The observed particles are in good crystalline nature. The average diameter of the CdS quantum dots was about 6-8 nm from Fig.5 (a) and the average diameter of the CdS/ZnS quantum dots was about 7-9 nm from Fig.5 (b).

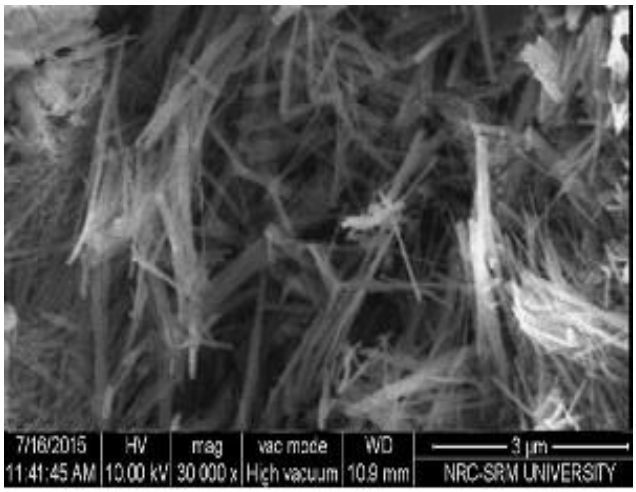

Fig.4 SEM images of TiO_2 nanowires

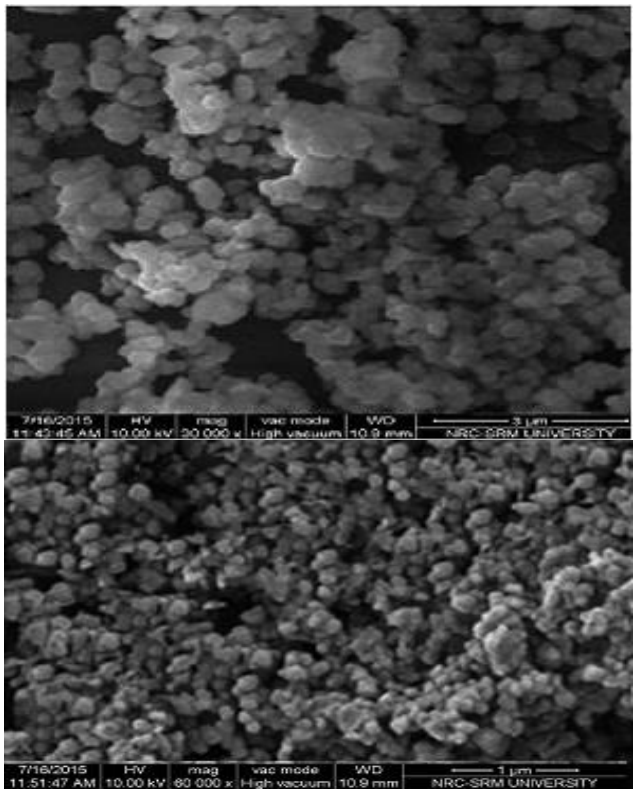

Fig.5 SEM image of (a) CdS quantum dots and (b) CdS/ZnS quantum dots

C. **Powder X-Ray Diffractometry (PXRD)**

Fig.6 shows the X-ray diffraction (XRD) pattern of TiO_2 Nanowire. In case of TiO_2 nanowires, the prominent peaks of d=0.325 and d=0.1628 indicates good crystallinity of the TiO_2 anatase phase. Fig.7(a) shows the XRD pattern of the CdS QDs deposited by the chemical bath deposition (CBD) method. The pattern of the diffraction peaks corresponds to the hexagonal phase of CdS. The lattice parameters were determined and found to be = 4.14 Å and c = 6.715 Å. All these diffraction peaks can be completely indexed which is in accordance with that of the standard spectrum (JCPDS, No. 89-2944). High intensity peaks present (2θ = 26.168°) shows that the particle size of CdS which is small and found to be around 3.6 nm corresponding to the (002) plane for CdS quantum dots.

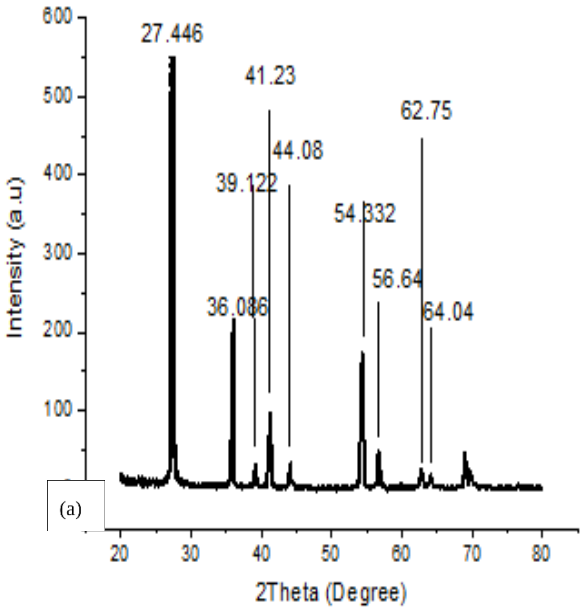

Fig.6 XRD image of TiO_2 nanowires

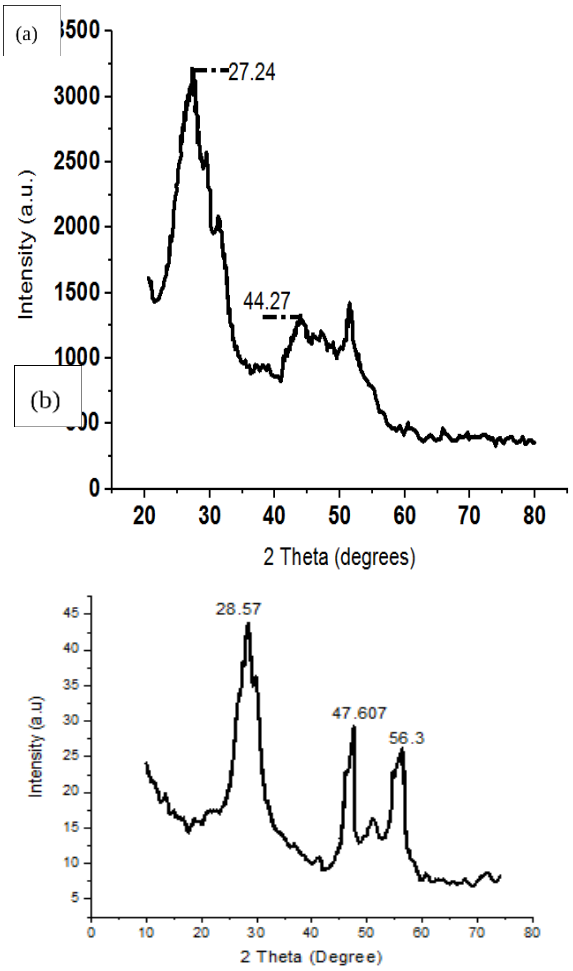

Fig.7 XRD images of (a) CdS quantum dots and (b) CdS/ZnS quantum dots

In Fig. 7(b) The most prominent peaks are similar as ZnS QDs i.e (002) direction along with the other reflections at (200) and (201) planes. Here the peaks indicates that the ZnS also present at the outlying surface in the form of a shell. Debye Scherrer formula was used to calculate the average particle size of CdS/ZnS core shell and was found around 4.2 nm. According to XRD both CdS and ZnS peaks were present in the core shell structure. The presence of high intensity peaks (2θ = 27.58°) clearly shows that the particle size of CdS/ZnS is very small corresponding to the (002) plane for CdS/ZnS quantum dots.

D. Fourier Transform Infra-Red (FTIR) Spectroscopy

The relative IR absorbance significantly depends on short range environment of oxygen coordination around cations in the lattice, crystal geometry and oxidation state of its cations. A typical IR spectrum of the sample was recorded on a NaCl crystal which is shown in Fig.8. TiO_2 - anatase form nanowires having characteristic peaks at 638, 513 and 397 cm^{-1} were observed in the FTIR spectrum. An initiation of the prominent peak from 404 cm^{-1} is a specifically characteristic of TiO_2 - anatase. A broad absorption band around 3400 cm^{-1} is assigned to O-H vibration of absorbed H_2O. The absorption band present at 644 cm^{-1} corresponds to Cd-S stretching. The band around 1500 cm^{-1} is due to NH_2 bending. The band near 2353 cm^{-1} can be attributed to C=O residue probably due to atmospheric CO_2.

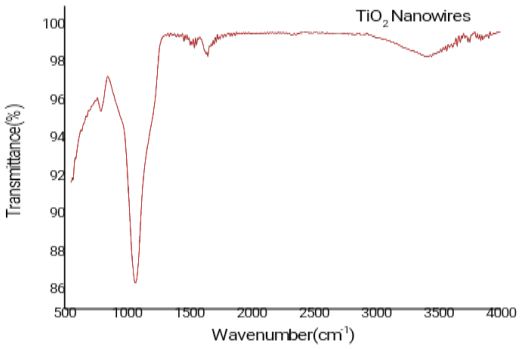

Fig.8 FTIR spectra of TiO_2 nanowires

In Fig.9 (a), the absorption band present at 644 cm^{-1} corresponds to Cd-S stretching. The band around 1500 cm^{-1} is due to NH_2 bending. The band near 2353 cm^{-1} can be attributed to C=O residue probably due to atmospheric CO_2. The broad peak at 2120 cm^{-1} and 1383 cm^{-1} were assigned to C=C , C-H characteristic vibrations in the CdS sample.

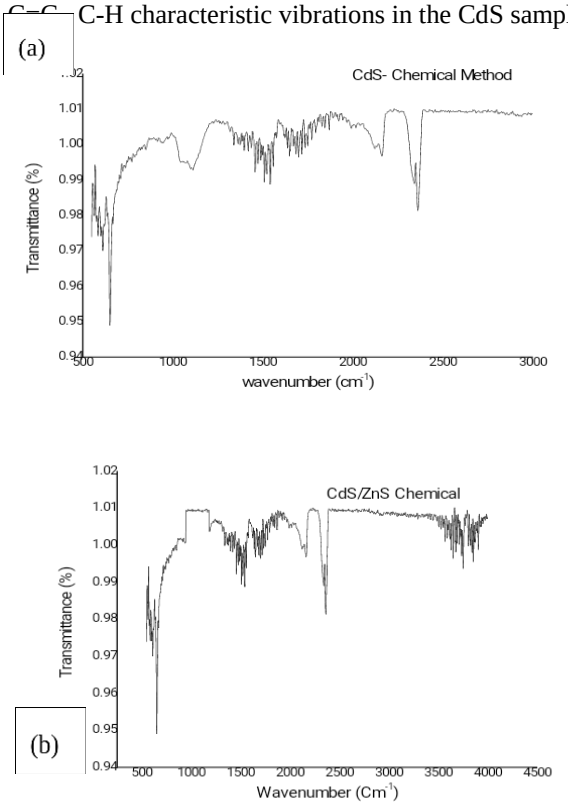

Fig.9 FTIR spectra of (a) CdS quantum dots and (b) CdS/ZnS quantum dots

In Fig. 9(b), the absorption band present at 644 cm^{-1} corresponds to Cd-S stretching. The band around 1500 cm^{-1} is due to NH_2 bending. The band near 2353 cm^{-1} can be attributed to C=O residue probably due to atmospheric CO_2. The broad peak at 2120 cm^{-1} and 1383 cm^{-1} were assigned to C=C , C-H characteristic vibrations. The peak at 1119 cm^{-1} shows (C-N) amines with two small peaks at 1626 cm^{-1} for (N-H) amines and 1401 cm^{-1} for O-H occured in CdS/ZnS quantum dots. These results show that how effectively ZnS covers the CdS core and minimize the toxicity of CdS materials as ZnS is nearly non toxic.

E. U-V Visible Spectroscopy

The optical absorption spectrum of TiO_2 nanowires and CdS quantum dots are shows in the Fig.10 (a). In absorption spectrum, the peak at 252 nm indicates the absorption due to anatase TiO_2. From the spectrum the optical absorption excitonic peak of CdS quantum dots is shifted to the lower wavelength side relative to that of bulk CdS (~530 nm) crystals. The absorption edge of the core/shell structure is lies between CdS and ZnS quantum dots because surface to volume ratio in core/shell is more than CdS but less than ZnS. This continuous blue shift shows that quantum detention occurred in the prepared quantum dots. In Fig. 10(b) CdS and ZnS quantum dots have absorption at 298 nm respectively. The UV-visible absorbance spectra for the quantum dots have a long tail extending to the far end of the visible region. CdS has a higher conduction band edge than that of TiO_2 and spontaneous electron transfer is possible from the CdS QDs to TiO_2. This electron injection will result in electron trapping on the surface of TiO_2. This electron trapping causes spectral changes in the red region of the visible spectrum during application.

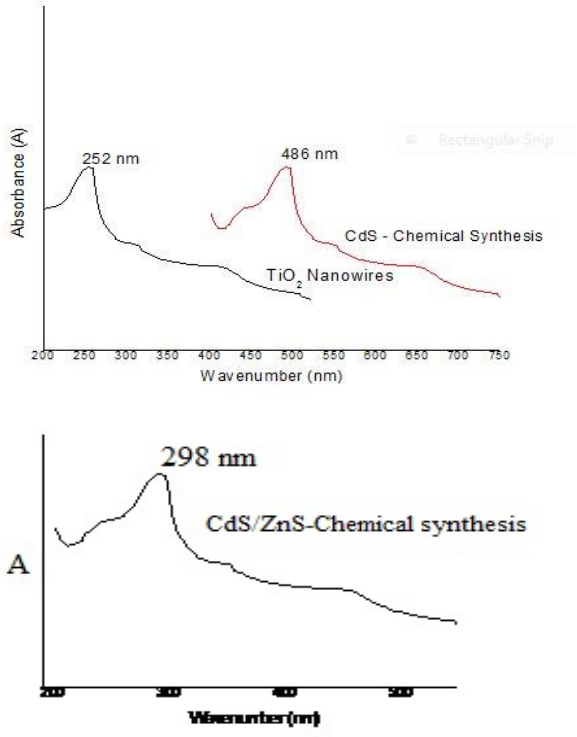

Fig.10 UV-Visible images of (a) TiO_2 nanowires and CdS quantum dots and (b) CdS/ZnS quantum dots

F. Photocurrent- voltage (I-V) characteristics QDSCs

The I-V characteristics of quantum dot sensitized solar cell were calculated using a solar simulator and the results are shown in Fig.11, Fig.12 and Fig.13. An active area of approximately 0.25 cm^2 (0.5 cm × 0.5 cm) of the resulting cell is exposed to light. The efficiency of solarcell under illumination and non-illumination conditions (with light (wl) and without light (wol)) is also calculated. Under illumination, a large leakage current causes low open-circuit voltage V_{oc}. When light illuminates over the cell, the I_{sc} always increases slightly and V_{oc} decreased.

TiO_2 nanowires has a great potential for the absorption of the photon energy from the solar energy source due to large surface area of nano structure as high internal surface

area are most enviable for high efficiency. Fig.11 shows I-V characteristics of solar cell with working electrode TiO_2 nanowires coated FTO and the counter electrode as graphite FTO glass under illumination and non-illumination conditions. The efficiency of solar cell under non-illumination conditions without light (wol) is calculated as η=1.42%, and with light (wl) illumination is calculated as η=3.5%.

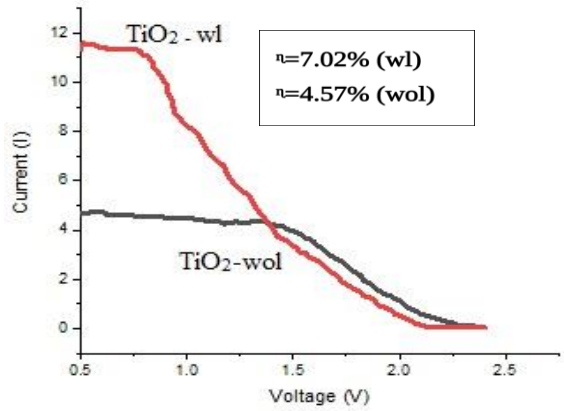

Fig.11 I-V characteristics of TiO_2 nanowires with light (wl) and without light (wol)

In order to improve the photo response, the TiO_2 electrode is sensitized with CdS quantum dots and the corresponding I-V characteristics are also obtained. Since the size of quantum dot is in the range of an atom, it can effectively utilize whole of the solar spectrum producing more photo electrons. Fig. 12 shows I-V characteristics of CdS quantum dot solar cell with light (wl) and without light (wol). The working electrode for QDSC is TiO_2 coated FTO sensitized with CdS quantum dots synthesized by chemical method and the counter electrode is graphite coated FTO glass. The efficiency of solar cell under non-illumination conditions without light (wol) is calculated as η=3.25%, and with light (wl) illumination is calculated as η=4.3%.

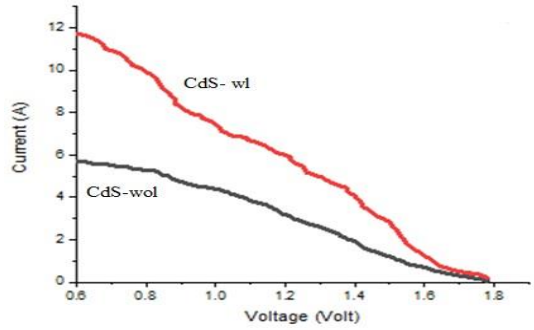

Fig. 12 I-V characteristics of CdS quantum dot solar cell with light (wl) and without light (wol)

Fig. 13 shows I-V characteristics of CdS/ZnS quantum dot solar cell with light (wl) and without light (wol). The working electrode for QDSC is TiO_2 coated FTO sensitized with CdS/ZnS quantum dots synthesized by chemical method and the counter electrode is graphite coated FTO glass. The efficiency of solar cell under non-illumination conditions without light (wol) is calculated as η=4.57%, and with light (wl) illumination is calculated as η=7.02%.

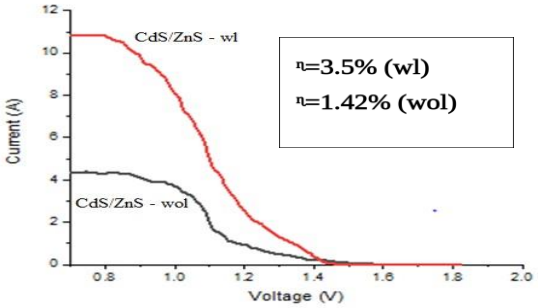

Fig.13 I-V characteristics of CdS/ZnS quantum dot solarcell with light (wl) and without light (wol)

To enhance the performance of CdS quantum dots sensitized TiO_2 solar cells and to improve the electron diffusion length, the surface of the CdS QDs are coated with ZnS which inturn results in increasing the electron lifetime, electron mobility and electron diffusion length. The CdS quantum dots particles were covered with ZnS (3.5eV) to establish a core/shell system, where the band gap of the core lies energetically within the band gap of the shell material and the electrons and holes which are photogenerated confined inside the CdS (2.42eV). An increase in the ZnS shell thickness increases electron-hole pair generation leading to large free-carrier concentration which results in enhancement of nonlinear optical absorption. Also the optical limiting threshold decreases as the ZnS shell thickness increases. The photo-degradation of CdS core was improved after ZnS shell growth significantly. The ZnS shell layer can improve the optical properties of quantum dots and so high luminescence efficiency and stability have been achieved for core-shell nanoparticles. Precise control of the size, shape and composition of both the core and the shell enable the emission wavelength to be tuned over a wider range of wavelengths than with individual semiconductor. As the bandgap of the core is smaller than that of the shell, both the conduction and valence band edges of the core lie within the bandgap of the shell, which confines both electrons and holes in the core. The emission wavelength due to radiative electron-hole recombination within the core is slightly red shifted compared to uncoated CdS. Depositing a surface passivation layer of ZnS protects the QDs from chemical corrosion in addition to suppressing the back-reaction of the photo-injected electrons. The CdS core with ZnS shell shows increment in conversion efficiency of QDSC. The iodide/triiodide couple has a suitable redox potential and have slow recombination kinetics thus making them a favorable candidate for solar cells. The calculated P_{max} and efficiency of solar cells from I-V curve with light (wl) and without light (wol) in an active area of 0.25 cm^2 is shown in table 1.

Table 1. P_{max} and efficiency of solar cells from I-V curve

	Solar cell	P_{max} without light	P_{max} with light	Experimental efficiency without light (%)	Experimental efficiency with light (%)
1	TiO_2 nanowires	0.56	1.40	1.42	3.5
2	CdS (Chemical)	1.30	1.72	3.25	4.3
3	CdS/ZnS (Chemical)	1.82	2.806	4.57	7.02

IV. CONCLUSION

The quantum dots are promising sensitizers for photo catalytic applications and it is evident from the results that CdS and CdS/ZnS quantum dots sensitized TiO_2 shows a better photo response than bare TiO_2. Electron transfer across the QD-TiO_2 junction, one of the first steps necessary to generate usable photocurrent from QDSC was studied. The present study was undertaken with standard TiO_2 layer sensitized with QDs and standard electrolytes to explore the best counter electrode materials resulted in higher efficiencies. The photo conversion efficiency of FTO/TiO_2/CdS/ZnS QDs/Graphite/FTO solar cell is higher than the efficiency of FTO/TiO_2/CdS QDs/Graphite/FTO solar cell. Core/shell semiconductor nanocrystals CdS/ZnS that prepared are safe, low-cost and environmental friendly. Absorbing sunlight from appropriate portion of the solar spectrum and tuning the bandgap of the semiconductor by tuning the size of the quantum dots allow optimizing the performance of the devices. It is expected that the conversion efficiency can be increase further by optimizing the thickness of the TiO_2 electrode which is well suited for harvesting more light energy in solar cell applications. If the performance of the devices is optimized, these low-cost, high-throughput devices could make a huge impact in the energy conversion industry across the globe.

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AUTHORS PROFILE

Lekshmi Gangadhar, received the Bachelor of Technology in Electronics and Communication Engineering from Mahatma Gandhi University, Kerala, India and Master of Technology in Nanotechnology from Noorul Islam University. She is currently working towards the Ph.D degree in Department of Nanotechnology, Noorul Islam Centre for Higher Education, Tamilnadu, India. Her current research interests include photovoltaics systems based on quantum dots, experimental study of nanostructured materials, the design of high-performance QDSC and large-area devices characterization.

Akhila Rajan, Research scholar, Department of Nanotechnology, Noorul Islam Centre for Higher Education, Kumaracoil, Kanya kumari District. Tamilnadu, India.

P.K Praseetha, Associate Professor and Head, Department of Nanotechnology, Noorul Islam Centre for Higher Education, Tamilnadu, India. Her current research interests include nanobiotechnology, nanomedicine, biomaterials, green synthesis of nanostructured compounds and targeted drug delivery.