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CDS Quantum Dots Induced Surface Modification of ZnO Nanorods for Dye Sensitized Solar Cells

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Abstract

In this study, zinc oxide nanorods (ZnO NRs) were fabricated utilizing a straightforward sol-gel process at ambient temperature. A novel approach was reported to enhance the photovoltaic performance of dye-sensitized solar cells (DSSCs) by surface modifying ZnO NRs with CdS quantum dots (QDs). The CdS QDs-induced surface modification leads to improved light absorption, charge separation, and transfer efficiency. The ZnO-CdS nanostructures were characterized using XRD, TEM, and UV-Vis spectroscopy, revealing uniform decoration of CdS QDs on ZnO nanorods. An optical investigation showed that the sensitization of CdS QDs on to ZnO NRs led to a red-shift in the material. At the same time, the absorbed N719 dye molecule further boosts the visible light, which can significantly increase the overall efficiency of a solar device. The modified ZnO-CdS photoanode exhibited a significant enhancement in power conversion efficiency (PCE) of 26.57 % compared to bare ZnO nanorods. The improved performance is attributed to the enhanced electron injection and reduced recombination rate. The ZnO - N719 dye showed an efficiency of power conversion of 1.79 %, while the ZnO -CdS QDs showed an efficiency of 0.57 %. In addition to this, the ZnO: CdS: N719 co-sensitized device had a conversion efficiency of 2.07 %. This study demonstrates a promising strategy for designing high-efficiency DSSCs using semiconductor nanostructures.

Keywords: Quantum Dots, Semiconductor Metal Oxide, Modified Photoanode, Conversion Efficiency, Sensitized Solar Cells.

Introduction

Dye-sensitized solar cells (DSSCs) have emerged as a promising alternative to traditional silicon-based solar cells due to their low cost, ease of fabrication, and potential for high efficiency. However, the performance of DSSCs is limited by the photovoltaic properties of the photoanode material [1-3]. The limited light absorption and inefficient charge transport in sensitized solar cell (SSCs) make it difficult to attain maximum efficiency compared to DSSCs or other solar devices [4-6].So, the upcoming generation of solar cells were fabricated using a variety of alternative methods. Zinc oxide (ZnO) is proposed as an attractive substitute for titanium dioxide nanoparticles (TiO2 NPs) due to its similar physical and chemical features [7]. Additionally, it is considered as a feasible host material for solar device construction. Both ZnO and TiO2 have roughly same location for their conduction band edges, but electron mobility of ZnO is significantly higher and making it a promising candidate as an electron acceptor and transport material in

SSCs [8]. Also, ZnO nanoelectrodes are simple to synthesize and manufacture only adds to their usefulness [9]. However, ZnO-based DSSCs often face challenges with overall conversion efficiency due to the complexation of ZnO with dye molecules, which can impede the injection of electrons into the semiconductor [10-13].

To address this issue and enhance the efficiency of electron injection, various surface modification strategies for ZnO have been explored. According to the reported works, Ho et al. modified ZnO nanorods (NRs) by using Au NPs in an effort to boost the effectiveness of DSSCs, however the results showed a decrease in surface area as a result of the alteration [14]. To further manage the reduction reaction between ZnO and dyes for extremely effective DSSCs, Yahong Xie coated ZnO with thioacetamide. Importantly, it was found that the thioacetamide functioned as a surface modification rather than a sensitizer and that the efficiency seen was insufficient [9]. Because of this, the surface of ZnO must be altered with a variety of substances for application in a variety of situations. In order to improve the overall efficiency of the solar cell, CdS QDs were utilized in this study to change the ZnO surface, acting as both a surface modifying agent and a sensitizer. However, there are many factors that influence how well solar energy can be converted into electricity, including the function of the sensitizer, the design of the photo-anodic material, and the range of visible light [15-17].The QDSSCs were developed with a ternary hybrid structure that is both cost-effective and stable, and takes into account the properties of semiconductor nanocrystals [18-21]. In the present study, thin films of binder-free ZnO NRs were coated with CdS QDs. CdS QDs not only alter the surface area of ZnO but also lengthen UV to Visible region, improving solar cell efficiency [22]. After CdS QDs were made sensitive, the N719 dye molecule was anchored to film. In order to extend the optical absorption zone and increase the absolute loading of absorbing media on the semiconductor, the N719 dye molecules can be very helpful. Moreover, N719 dye has a high molar extinction coefficient, allowing efficient light absorption and conversion. N719 dye absorbs light across a broad spectrum, including visible and near-infrared regions, maximizing energy harvesting. The energy levels of N719 dye align well with the TiO2 conduction band and the iodide/triiodide redox couple, facilitating efficient electron injection and regeneration [2, 23]. The overall photovoltaic parameters are affected by the co-sensitized action of QDs and dye molecules, which boosts the efficiency of co-sensitized solar devices. When comparing co-sensitized solar devices to signally sensitized solar devices, we found that co-sensitized solar devices had nearly double the efficiency (2.07 %) under one solar irradiation (air mass 1.5 G).

Experimental Details

Materials

The fluorine doped tin oxide (FTO, sheet resistance: 8-10 Ω) glass was used for depositing bare ZnO NRs and considered as optically transparent electrode (OTE). The various AR grade chemicals such as, mercaptopropionic acid (MPA), 98.00%), cadmium chloride monohydrate (CdCl2.H2O, 98.00%), sodium sulfide (Na2S), zinc acetate (Zn (CH3CO2)2), TritonX-100, ethylene glycol ((CH2OH)2), etc were purchased from Sigma-Aldrich.

Synthesis of Zno Nanorods

Initially, 100 mL of zinc acetate was stirred at room temperature by slowly adding 2M of ammonia solution to get complete clear solution. Then,5 mL of ethylene glycol and 5 mL TritonX-100 added in this solution and heated this solution at 70 °C to 80 °C with constant stirring till to the formation of white precipitate. The mixture is cooled at room temperature, centrifuged and washed several times with ethanol. The powder is dried and calcinated in at 450 °C for 4 h. Then, the synthesized powder was used for further characterizations.

Surface Modification of Zno by CDS QDS and Device Fabrication Within719 Dye

In ultrasonic bath, FTO substrates were cleaned using detergent solution for about 30 min. and then rinsed with double distilled water. Again, it is rinsed with acetone and dried at 110 °C for 30 min. in an electric oven. These substrates were used for the deposition of ZnO NRs by using low-cost doctor-blade technique. In this protocol, ZnO paste was prepared by sonication of the ZnO NRs in ethanol solution and continuously stirred to form a gel. The obtained paste is the deposited-on surface of FTO. Furthermore, the prepared ZnO films were sintered at 450 °C for 30 min. and cooled naturally to the room temperature. Subsequently, capped CdS QDs were deposited on ZnO films by using spin coater. Then, these modified ZnO-CdS thin films were rinsed with the water and dried in oven at 110°C for 15 min. After sensitization of CdS QDs, the surface of ZnO NRs becomes rough (as displayed in the scanning electron microscope (SEM) images). Finally, the N719 dye were anchored on the surface of modified ZnO:CdS QDs film by dipping films into N719 dye solution for 24 h. Then the electrodes were washed with equimolar mixture of tetra-butyl alcohol and acetonitrile (1:1) for removing the free dye molecules and considered as a photoanode. To design sandwich solar device of ZnO: CdS: N719 dye is used as a working electrode and platinum coated ITO as a counter electrode are pressed through Surlyn sheet and put in an electric oven at 110 °C. After proper binding of device electrolyte is inserted through counter electrode and whole device is placed in vacuum for the electrolyte filling on surface of the ZnO: CdS: N719 electrode.

Results and Discussion

The as-synthesized nanomaterials with their composites and fabricated devices are characterized by various characterization tools. In order to determine the optical properties of the nanomaterials, ultra-violet visible diffuse reflectance spectroscopy (UV-Vis DRS) is used with 200 to 800 nm wavelength range. X-ray diffractometer (XRD) is used to analyse crystallite size and phase purity. Morphology of the ZnO NRs is determined by using SEM images. After fabrication of sandwich solar cell devices, the photovoltaic measurement is carried out under solar simulator.

Optical Absorption Study

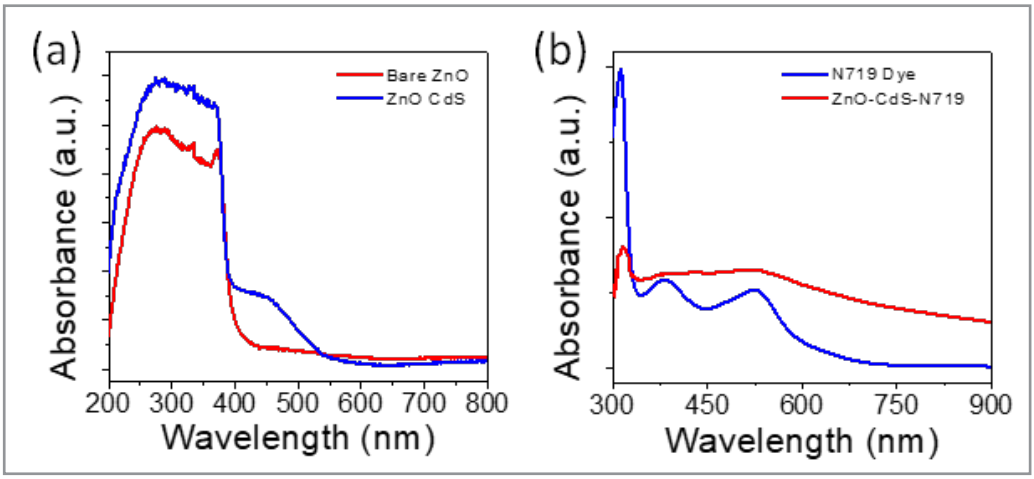

Figure 1: (a) UV-Vis DRS of bare ZnO, CdS sensitized ZnO film and (b) N719 dye and of ZnO: CdS: N719 thin films.

The optical properties of the bare ZnO and CdS sensitized ZnO NRs are analysed by UV–Vis DRS as shown in Fig. 1. From DRS spectra, it's clear that ZnO NRs shows absorption spectra at 390 nm and obtained band-gap value is 3.17 eV. For bare ZnO case, the spectral curve does not display any significant deviation. However, ZnO:CdS thin film shows the absorption edge is higher than that of bare ZnO indicating the influence of CdS QDs as displayed in Fig. 1a of the binary composite [24]. The DRS spectra of ternary nanocomposites shows broad visible region, due to N719 dye (Fig.1b) [25]. Therefore, the existence of N719 affects optical properties of the ternary nanocomposites that is responsible for the red shift in absorption spectrum. This improved absorption in the visible wavelength region results in the generation of abundant electron–hole pairs in the ZnO:CdS and ZnO:N719 based device under UV-visible illumination, which can lead to greater photo-current activity.

FTIR Analysis

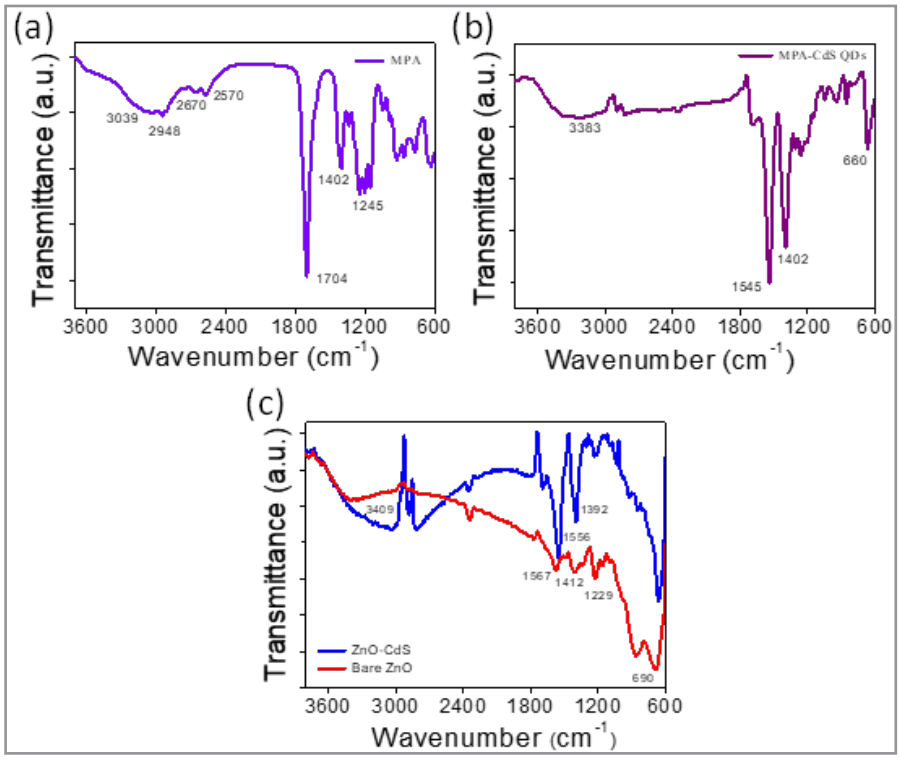

Figure 2: FTIR spectra of (a) bare MPA, (b) MPA capped CdS QDs and (c) bare ZnO and CdS sensitized ZnO thin films.

Fig. 2represents the FTIR patterns for bare mercaptopropionic acid (MPA), MPA capped CdS QDs and bare ZnO NRs as well as CdS QDs sensitized ZnO NRs. For MPA (Fig.2a) the bands observed at ~3400-3000 cm-1,~2948 cm-1,~2570 cm-1, ~1704 cm-1 and ~1402 cm-1 are due to stretching vibrations of the functional moieties such as -OH, -CH2, S-H, C=O and C-O, re-

spectively. In addition, a small band value at ~2670 cm-1 is due to bending vibration of -OH. However, in FTIR pattern of MPA capped CdS QDs (Fig. 2b), the stretching band of carboxylic O-H group is disappeared due to its deprotonation; while the peak position of C=O group is shifted from 1704 to 1545 cm-1 [26]. In addition, the stretching band of S-H is also not observed because of its high pKa value; which reveals that the interaction of sulfhydryl moieties of MPA with Cd2+ ions and hence, the co-ordination between the different moieties avoids the deprotonation of the sulfhydryl group of MPA [27].

Similarly, in FTIR spectra of bare ZnO NRs, the broad absorption band at ~3400-3000 cm-1and ~1400-1500 cm-1 are due to O–H stretching and deformation of C–OH groups of water molecules. Zn–O bond stretching vibrations appears at 690 cm-1. However, ZnO:CdS composites also shows O–H stretching vibrations in the range of 3000-3500 cm-1 range, while Zn–O bond ranges between 500 and 600 cm-1. The peak at 1392 cm−1 in the binary composite is assigned to C–O bending vibration. From the FTIR spectra of ZnO:CdS composite, the composites have the characteristic peak of oxygen containing functional groups particularly at 1704 cm-1 are weakened and the O–H stretching peak decreases. This is mainly attributed to the loss of oxygen containing functional groups. Overall, present study reveals that, surface of ZnO NRs are covered through CdS QDs.

XRD Analysis of ZnO: CdS Thin Film

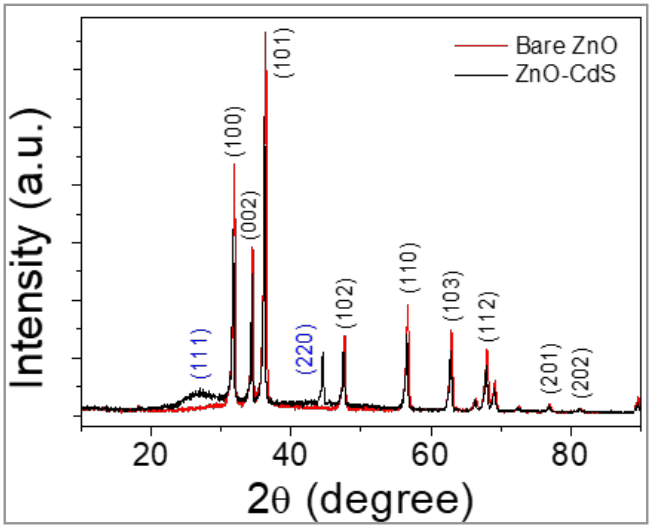

Figure 3: XRD patterns of bare ZnO NRs and CdS QDs sensitized ZnO thin films.

X-ray diffraction (XRD) technique is used to determine crystallinity, phase of the ZnO NRs, CdS QDs and their composites. Typical XRD pattern of the synthesized ZnO NRs, ZnO:CdS composites are shown in Fig. 3. The various reflections are appeared at 2 values of 31.86, 34.58, 36.32, 47.63, 56.71, 62.86, 68.08, 69.26 and 76.88, with corresponding reflection of (100), (002), (101), (102), (110), (103), (112), (201) and (202), respectively. All these peaks are indexed and these are well matches with Wurtzite (hexagonal) structure of ZnO (JCPDS card no. 36-1451) [28]. Furthermore, absence of any characteristic impurity peaks indicates formation of high quality ZnO NRs.

Interestingly, ZnO:CdS composites shows an additional peak at 27.42°, 44.86° which is due to presence of (111) and (220) crystal plane of CdS QDs on ZnO NRs; which clearly indicate that the proper depositions of CdS QDs take place on surface of ZnO NRs. The crystallite size of bare ZnO NRs and ZnO:CdS composites determined from the Debye Scherrer's equation and is shown in Table 1. Moreover, after sensitization; the crystallite size of ZnO:CdS is decreases.

Table 1: Structural parameter of ZnO:CdS thin film.

Sample	(hkl)	d value	Parameters			Crystal size (nm)	Dominance crystal structure
			a (Å)	c (Å)	V(Å3)		
Bare ZnO	101	2.471	3.23	5.18	54.04	9.34	Wurtzite hexagonal
	002	2.590					
	110	1.621					
Bare CdS	111	2.38	5.85	-	200.20	1.89	Cubic
	220	2.07					
	311	1.76					
ZnO:CdS	101	2.482	3.21	5.19	54.57	9.39	Wurtzite hexagonal
	002	2.585					
	110	1.602					

Scanning Electron Microscopy

The surface morphology of all pristine materials as well as the binary (ZnO-CdS) nanocomposites was carried out using SEM technique, as depicted in Fig. 4. ZnO shows NRs shaped morphology without sensitization of CdS QDs as shown in Fig. 4a. It is observed that the surfaces of the bare ZnO NRs appear clear and soft. Furthermore, SEM images of CdS QDs deposited on ZnO surfaces are shown in Fig. 4b, that appears rough and tarnished surfaces, which indicates that deposition of the CdS QDs on the surface of ZnO NRs. Randomly scattered spherical particles with uniform size-distribution of CdS QDs are good resolved on the ZnO surface helps for the maximum anchoring of N719 dye molecule.

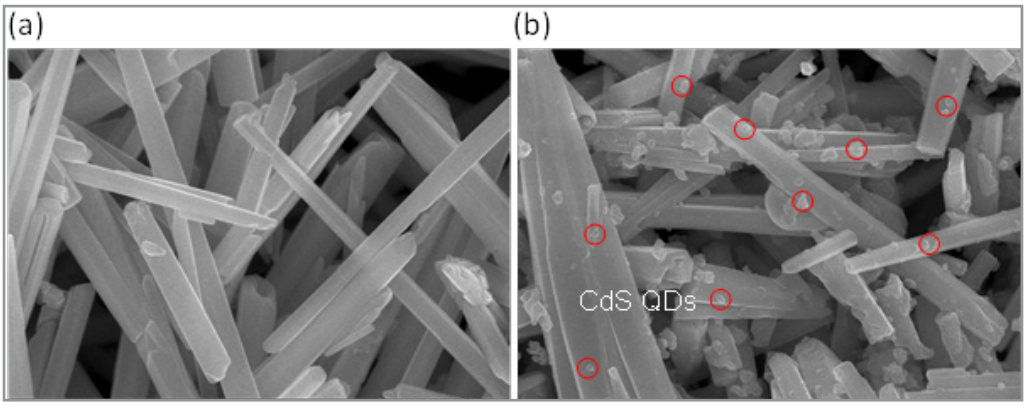

Figure 4: SEM images of (a) bare ZnO NRs and (b) CdS QDs sensitized ZnO NRs.

Photovoltaic Studies: Photocurrent Density-Voltage (J-V) Measurement

Wei et al. designed CdS QDs-sensitized Zn2SnO4 solar cells by surface treatment with Al3+ ions[29] and obtained results are displayed in Table 2.

Table 2: Cell parameters of CdS QD SSCs composed of Zn2SnO4 treated with Al^{3+}.

Electrode	Jsc (mA/cm^2)	Voc (V)	Fill factor (%)	Efficiency (%)
Zn2SnO4	0.49	0.487	0.455	0.109
Zn2SnO4/Al-1	0.64	0.468	0.521	0.157
Zn2SnO4/Al-2	0.53	0.444	0.530	0.126

Similarly, Yin et al. fabricated CdSe-CdS QD co-sensitized ZnO hierarchical hybrids for solar cells to improve the efficiency of solar device [30] and obtained results are shown in Table 3.

Table 3: Photovoltaic parameters of the assembled devices.

Device	Counter	Jsc (mA cm−2)	Voc (V)	Fill factor (%)	Efficiency (%)
12 h ZnO	Cu2S	1.12	0.15	33.16	0.04
4CdS/24 h ZnO	Cu2S	3.07	0.49	30.53	0.46
4CdS/12 h ZnO	Cu2S	2.29	0.49	52.86	0.59
8CdS/12 h ZnO	Cu2S	4.01	0.57	37.98	0.87
12CdS/12 h ZnO	Cu2S	3.63	0.53	39.85	0.77
3CdSe-8CdS/12h ZnO	Cu2S	4.32	0.62	52.98	1.39

The solar devices are fabricated using the composition of FTO/ ZnO:CdS:N719/ (I-/I3-) /Pt-ITO and tested under1 sun (100 mW cm-2AM 1.5G) solar illumination conditions for photovoltaic studies. Similarly, DSSCs-based on ZnO:N719 dye and ZnO:CdS QDs also designed and their photovoltaic performances compared with surface modified CdS QDs DSSCs. Fig.5 shows the current density-voltage (J-V) characteristics of the hybrid cells i.e., ZnO:CdS, ZnO:N719 and surface modified ZnO:CdS:N719 devices and corresponding photovoltaic parameters are listed in Table 4.

Table 4: Photovoltaic properties of ZnO: N719, ZnO:CdS and ZnO:CdS:N719 devices.

Compositions	Jsc (mA/cm2)	VOC (mV)	FF (%)	η (%)
ZnO-CdS QDs	3.97	310	47.08	0.57
ZnO-N719 dye	6.41	550	50.84	1.79
ZnO-CdS QDs-N719 (dye)	6.73	580	53.24	2.07

From the photovoltaic parameters, it's observed that without surface modification of ZnO-N719 device shows poor performance, which may be due to complex formation of dye molecule with ZnO NRs. However, same ZnO NRs sensitized with CdS QDs shows 0.57 % power conversion efficiency that confirms CdS QDs acts as a sensitizer. Therefore, CdS QDs are used as surface modifying agent as well as sensitizer in ZnO based DSSCs. After loading CdS QDs on the surface of ZnO N719, dye anchored on it and resulting efficiency of device is huge increment of solar device. Specially, Voc of the modified device is increased from 310 mV to 550 mV and corresponding efficiency is also increased from 1.79 % to 2.07 %. This is attributed to the near about double efficiency is increased for ZnO: CdS: N719 based device.

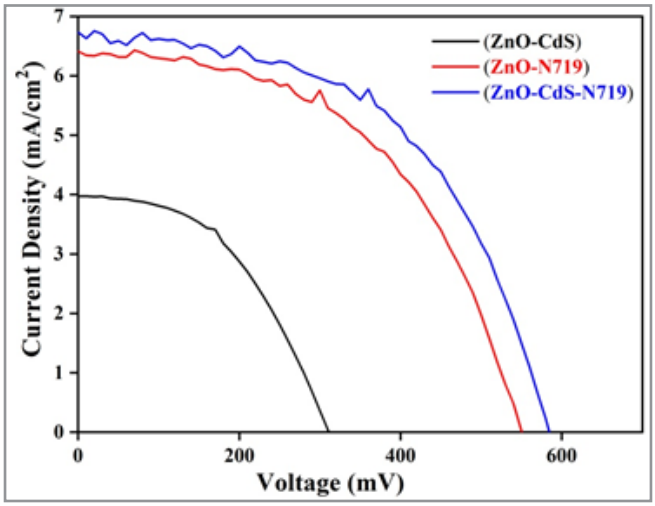

Figure 5: J-V measurement of the ZnO-CdS, ZnO-N719, ZnO-CdS-N719 based devices.

Conclusion

In conclusion, the surface modification of ZnO nanorods with CdS quantum dots has been successfully demonstrated to enhance the photovoltaic performance of dye-sensitized solar cells. The CdS QDs-induced surface modification leads to improved light absorption, charge separation, and transfer efficiency, resulting in a significant enhancement in power conversion efficiency. The ZnO NRs sensitized with the N719 dye showed an efficiency of power conversion of 1.79 %, while the ZnO NRs that were sensitized with CdS QDs showed an efficiency of 0.57 %. In addition to this, the ZnO:CdS:N719 co-sensitized device resulting conversion efficiency of up to 2.07 %. The findings of this study suggest that the ZnO-CdS nanostructures have great potential for application in high-efficiency DSSCs.

Conflicts of Interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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