American Scientific Research Journal for Engineering, Technology, and Sciences (ASRJETS)
ISSN (Print) 2313-4410, ISSN (Online) 2313-4402

http://asrjetsjournal.org/

The Photovoltaic Performance of CdS/Cu_2S (QD) Co-Sensitized Solar Cell

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Abstract

The performance of CdS/Cu_2S co-sensitized quantum dot solar cell (QDSSCs) is studied. CdS quantum dots (QDs) were adsorbed onto TiO_2 using Successive Ionic Layer Adsorption and Reaction (SILAR) method for different cycles (2, 4, 6, 8 and 10). The Cu_2S QDs were deposited onto TiO_2/CdS10 cycle photoanodes using the same method. The FTO counter electrodes were coated with platinum, while the electrolyte containing polysulfide redox species was sandwiched between the two electrodes. The current density- voltage (J-V) characteristic curves of the assembled QDSSCs were measured at different cycles and AM 1.5 simulated sunlight. The value of current density (J_{sc}) and conversion efficiency (η) of TiO_2/CdS/Cu_2S are 252% larger than the TiO_2/CdS values. The maximum values of J_{sc} and η are 3.56 mA/cm^2 and 1.21% respectively corresponding to CdS10/Cu_2S for 6 cycles of Cu_2S.

Keywords: Quantum dot sensitized solar cell (QDSSCs); CdS QD; SILAR; photovoltaic parameters.

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1. Introduction

In the last two decades, a great effort has been indicate to improve of the performance of the third generation solar cells, especially that of quantum dots sensitized solar cells (QDSSCs)[1]. In this kind of solar cells, quantum dots (QDs) are adsorbed onto a large band gap metal oxides such as TiO_2 nanoparticles (NPs)[2-7], ZnO NPs[7-11], and SnO_2 NPs[7] to act as light more harvesting sensitizers [12]. These QDs possess many attractive properties such as the ability to tunable band gaps, high absorption coefficients due to quantum confinement, large dipole moment leading to rapid charge separation and as well low manufacturing cost [13]. CdS has become a very attractive promising material for many specific applications in solar energy conversation and Cu_2S is highlighted because of its non-toxic, low cost and earth-abundant properties. The band-gap of bulk Cu_2S is 1.20 eV, which can make Cu_2S become a widely used photosensitizer for various wide band-gap semiconductor photoanodes [14].The adsorbing of Cu_2S QDs onto TiO_2 /CdS can make TiO_2 /CdS/ Cu_2S which would be more efficient for solar energy utilization than single semiconductor doped one. Their band gap matching for solar light absorption along wide band gap tenability and high absorption coefficient [15]. Synthesizing QDs techniques may involve different scientific processes, such as chemical, physical, biological, or some combination of these processes [16]. In some chemical methods, QDs are synthesized either by chemical bath deposition (CBD) technique or successive ionic layer adsorption and reaction (SILAR) method [16]. In our study we use SILAR, since it is simply adjusted by controlling the number of deposition cycles. A.Badawi and his colleagues employed CdSe QD sensitized TiO_2 using the direct adsorption technique (DA) to anchor CdSe QD onto TiO_2 substrate and the efficiency for energy conversion η has been increased from 0.11 % to 0.29 % [9]. N.Al-Hosiny and his colleagues studied $CdTe_xS_{1-x}$ QDs to deposit TiO_2 misporous, they tuned the efficiency via molar ratio control of $CdTe_xS_{1-x}$ QD and reported the maximum value of η was 0.31 % for x=0.6 of $CdTe_xS_{1-x}$ [10]. In our work CdS QDs and CdS /Cu_2S QDs were adsorbed onto the surface of the TiO_2 electrode by (SILAR) method. The effect of the CdS QDs and CdS /Cu_2S QDs number SILAR cycle on the QDSSCs photovoltaic parameter (short circuit current density J_{sc}, open circuit voltage V_{oc}, fill factor FF, efficiency for energy conversion η) has been investigated.

2. Experimental

2.1. Deposition of the CdS QDs onto TiO_2 NPs photoanode

The nano TiO_2 paste was prepared by the following procedure of G.Syrrokostas et.al [17].The CdS QDs were sensitized ontoTiO_2 nanoparticles using SILAR method [18].The photoanode is immersed for one min in 0.1 M ($Cd(NO_3)_2.H_2O$) in methanol as a cation source (Cd^{+2}) and to be adsorbed on the TiO_2 electrode. The TiO_2 electrode was then rinsed with methanol to remove the excess Cd^{+2} cations. Then the photoanode is immersed in 0.1 M Na_2S methanolic solution as the anion source (S^{-2}) for one min in order to react with Cd^{+2} cations followed by methanol rinsing. This procedure was referred to as one SILAR cycle. Five samples obtained at increasing number of cycles labeled a, b, c, d and e for 2, 4, 6, 8 and 10 cycles, respectively. The counter electrodes were prepared by coating another Florien-doped Tin Oxide (FTO) substrate of resistance of 7 Ω/sq

with platinum (Pt).

2.2. Deposition of the Cu_2S QDs onto TiO_2/CdS10 QDs photoanode

In this part, the Cu_2S QDs were deposited onto TiO_2/CdS10 (sample e which has high efficiency) by the same method (SILAR technique). The photoanode is immersed for 1 min in 0.1 M ($CuNO_3$)$2.4H_2O$) in methanol as a cation source (Cu^{+2}) and to be adsorbed on the TiO_2 electrode. The TiO2 electrode was then rinsed with methanol to remove the excess Cu^{+2} cations. Then the photoanode is immersed in 0.1 M Na_2S methanolic solution as the anion source (S^{-2}) for 1 min in order to react with Cu^{+2} cations followed by methanol rinsing. We get also five samples at different cycles labeled a, b, c, d and e for 2, 4, 6, 8 and 10 cycles respectively.

2.3. Assembly of QDSSCs

QDSSCs were built by assembling the TiO_2 photoanodes sensitized with (CdS and CdS/Cu_2S) , aqueous polysulfide electrolyte and the counter electrodes. Parafilm was used as spacer between the two electrodes and also as a sealing to prevent the evaporation of the electrolyte. The space between the two electrodes was filled by polysulfide electrolyte which was prepared by dissolving 1.1M Na_2S, 1.2 M S, and 0.12 M KCl in deionized water [19].

2.4. Characterizations and measurements

The UV-Vis absorption spectra were recorded using (Jasco 670 with 1cm quartz cuvettes) spectrophotometer. The size and morphology was determined using high resolution transmission electron microscopy (HRTEM, JEOL JEM-2100 operated at 200KV with high resolution Gatan CCD bottom camera, Orius SC200). Photovoltaic solar cell measurements were performed using a solar simulator device (San-Ei Electric XES-40S1) at AM 1.5 with 1 sun illumination intensity (100 mW/cm^2), and current density–voltage (J–V) data were recorded using a source meter unit (Keithley SMU 2600).

3. Results and Discussion

3.1. Optical measurements of CdS QD

For TEM measurement, TiO_2/CdS layer in ethanol was scratched from FTO coated substrate without disturbing FTO layer and placed on Cu-coated carbon grid .Figure 1 shows the TEM image of TiO_2/CdS for sample (e).The average particle size for CdS QD were estimated to be approximately 5.4 nm and average particle size for TiO_2 is 22 nm.

Figure 2 show UV-visible absorption spectra for (TiO_2 and TiO_2 / CdS layer with increasing number of cycles by SILAR method). It can be seen that the TiO_2 layer shows an absorbance edge at 400 nm corresponding to 3.1 eV band gap. Also, there is a blue shift of absorbance edge with decreasing number of cycle. These are 401 nm, 414 nm, 431 nm, 451nm and 465 nm for samples (a, b, c, d and e) respectively. This blue shift is due to quantum confinement effect [20].

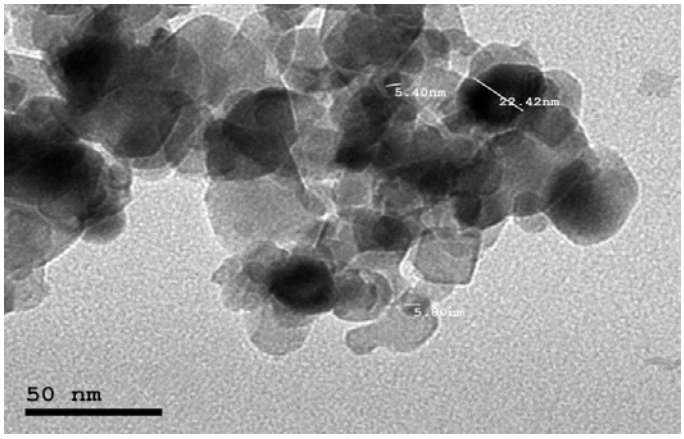

Figure 1: TEM image of TiO_2 / CdS QD for sample (e)

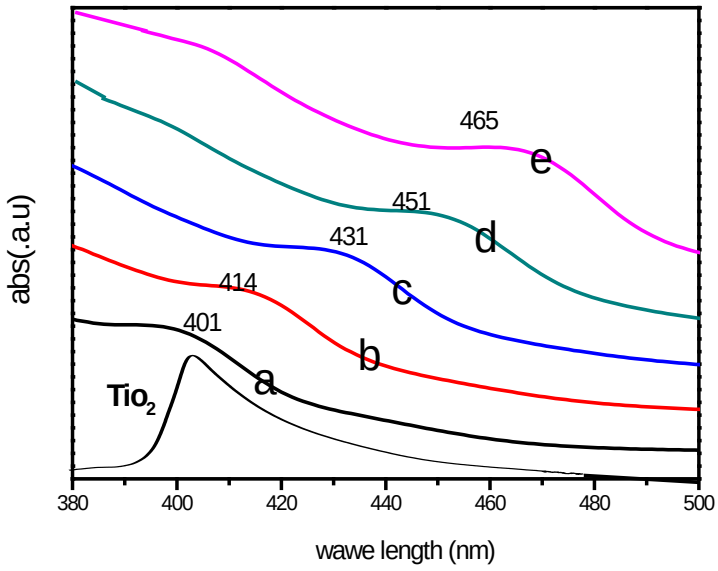

Figure 2: show UV-vis. absorption of TO_2/CdS QD electrode with different number of cycles by SILAR method

The particle size of CdS QD is calculated using the effective mass approximation (EMA) model [21-23].

$$E_{gn}(R) = E_{gb} + \frac{h^2}{8m^* R^2} - \frac{1.8e^2}{4\pi\varepsilon\varepsilon_0 R} \quad (1)$$

where E_{gb} (2.4 ev) is the bulk band gap [21] , E_{gn} is the nano band gap , R is the radius of the CdS quantum dots , m^* (7.62 x 10^{-32} kg) is the reduced mass [21], and ε (7.1) is the dielectric constant for CdS [21]. The average particle sizes of CdS QDs are obtained from UV.spectra and applied equation 1. The obtained values of CdS particle sizes ranged from 3.2 nm for sample (a) to 4.4 nm for sample (e) in close agreement with that

obtained by TEM measurements.The elemental analyses of the CdS QDs sensitized TiO_2 working electrode was measured using (EDX) (model JEOL JED-2300). Figure 3 shows the EDX spectra of TiO_2/CdS QDs sensitized working electrode where Cd, S, Ti and O atoms are indicated. This result ensures the adsorption of CdS QDs onto the TiO_2 electrode.

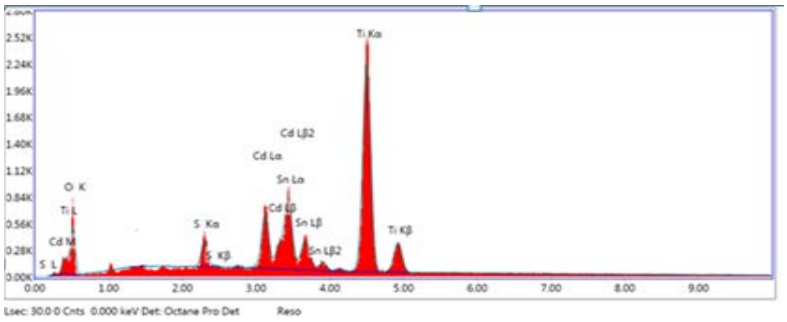

Figure 3: EDX of TiO_2/ CdS QD working electrode for sample e

3.2. Photovoltaic performance of CdS QDSSCs

Figure 4.a shows the J-V characteristics curves of CdS QDSSCs measured under a simulated sunlight with an intensity of 100 mW/cm^2 (AM 1.5G). The value of J_{sc} vs number of SILAR cycle of CdS QD are shown in Figure 4.b. The photovoltaic parameters V_{oc},J_{sc}, FF and η for produced CdS QDSSCs are given in Table 1. It is clearly seen that as CdS QDs size increases, the values of J_{sc} and η increase. The maximum values of J_{sc} and η are 2.24 mA/cm^2 and 0.48% respectively for 10 cycles (sample e) of CdS QD. S.Abdallah et. al [24,25] has made measurement on CdS QD sensitized TiO_2 using the direct adsorption technique (DA) to anchor CdS QD onto TiO_2 substrate . They reported that, there are an increase of J_{sc} (from 0.31 mA/cm^2 to 0.67 mA/cm^2) and η (from 0.05% to 0.18%) as the CdS particle size increase. Our results shows that an increase of J_{sc} (from 1.18 mA/cm^2 to 2.24 mA/cm^2) and η increase (from 0.15 to 0.48) with increasing particle size of CdS QDs in agreement with their work .They attributed the increase in J_{sc} with increase particle size in a red shift and thus causes relativity high absorption of the incident photon from solar spectrum .Therefor, particle size of CdS QD 4.4 nm (correspond to 465 nm) harvest more visible photon than the other particle. However , our results show an improvement of the J_{sc} and (η) than their work by 266 % which may be ,due to the SILAR technique is being better method than DA technique to give high-quality surface spread, lower aggregation ,higher photocurrents, longer lifetimes of electron and solve problem of surface chemistry which lead to improve QDSSCs [26].

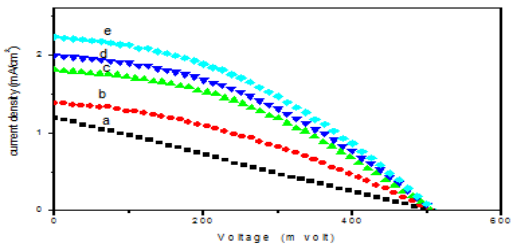

Figure 4.a: J-V characteristic curve of QDSSCs of CdS QDs with different size (different cycle)

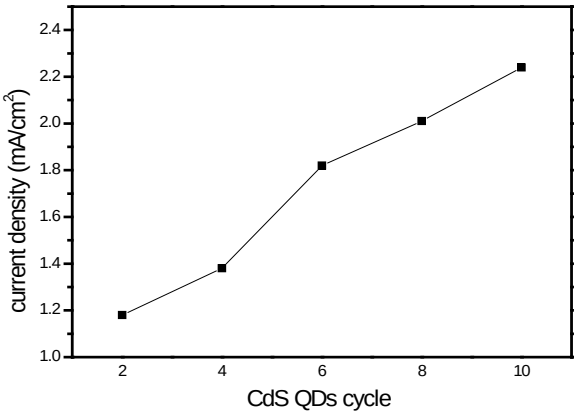

Figure 4.b: Variation of the current density as a function of SILAR cycles of CdS QD

Table 1: J -V characteristics parameters of a CdS QDSSCs for different QDs sizes, under 1 sun illumination.

Sample TiO_2/CdS	V_{oc} (m Volt) ± 0.01	J_{sc} (mA/cm^2)	FF	η(%) ± 0.01
a	508	1.18± 0.01	0.24	0.15
b	505	1.38± 0.02	0.35	0.24
c	504	1.82± 0.01	0.39	0.37
d	509	2.01± 0.01	0.38	0.42
e	510	2.24± 0.01	0.40	0.48

3.3. Optical measurements of CdS10/Cu_2S QD

In this part, the Cu_2S QDs were deposited onto TiO_2/CdS(10) photoanodes by SILAR method (which has higher J_{sc} and η). Figure 5 shows the UV spectra of TiO_2/CdS10/Cu_2S photoanode for different number of cycle. The absorption edges varies between 487 nm for sample a with a corresponding band gap of 2.55 eV to 553 nm for sample e with a corresponding band gap of 2.24 eV which should be ascribed to the contribution of Cu_2S, since its band gap was reported to be 1.2 eV [15].It was also noted that, the absorption intensity of this band gradually increased with the increasing amount of Cu_2S in CdS.

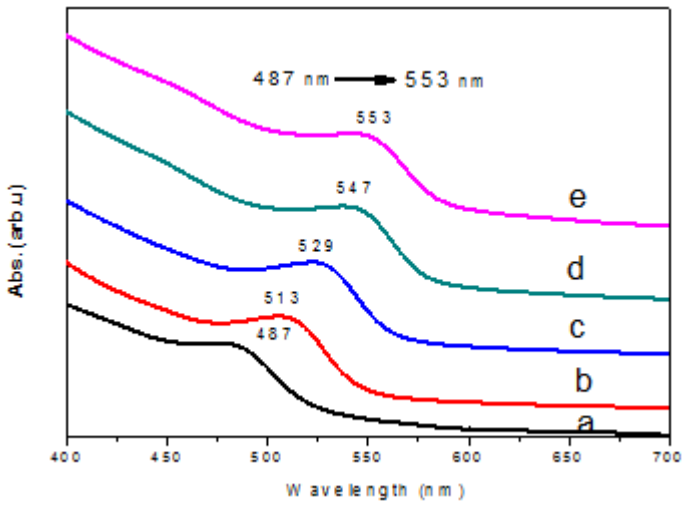

Figure 5: show UV-vis. absorption of TO_2/CdS10/Cu_2S QD electrode with different number of cycles by SILAR method

3.4. Photovoltaic performance of Cu_2S/CdS QDSSCs

$_2$S Cu Figure 6 shows the J-V characteristics curves of (CdS10/Cu_2S) QDSSCs measured under a simulated sunlight with an intensity of 100 mW/cm^2 (AM 1.5G). The photovoltaic parameters (V_{oc},I_{sc}, FF and η) for all the sample are obtained and given in Table 2.

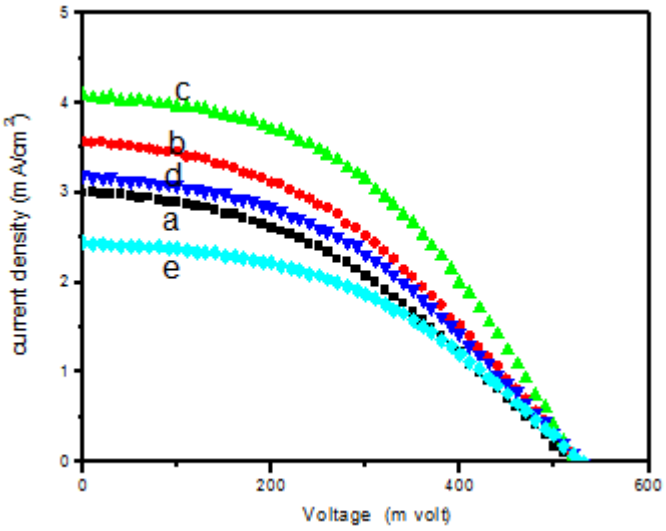

Figure 6: J-V characteristic curve of QDSSCs of CdS10/Cu_2S QDs with different cycle

It is clearly seen that as Cu_2S QDs SILAR cycles increases, the values of J_{sc} and η increases peaking at 4.01 mA/cm^2 and 1.21% respectively for 6 cycles (sample c) which about 252% larger than TiO_2/CdS. These values decrease for sample d (3.56 mA/cm^2 and 0.96%) and sample e (2.60 mA/cm^2 and 0.73%). The increase of J_{sc} and η can be explained by two effect .First effect , due to the relative narrow band gap of Cu_2S (1.2 ev) [15] , Cu_2S can increase the utilization efficiency of the incident light , and result in the more efficient charge

separation in CdS QD , which can improve the transportation of the photosensitized electron and reduce the recombination of the photosensitized electron and hole , so the performance of the whole efficiency will be promoted in turn [27].

Table 2: J -V characteristics parameters of CdS10/ Cu_2S QDSSCs for different QDs cycles, under 1 sun illumination.

sample	V_{oc} (mVolt) ± 0.01	J_{sc} (mA/cm^2)	FF	η(%) ± 0.01
a	520	3.01± 0.01	0.41	0.52
b	531	3.72± 0.02	0.40	1.06
c	**527**	**4.01± 0.03**	**0.49**	**1.21**
d	525	3.56± 0.01	0.48	0.96
e	527	2.60± 0.01	0.41	0.73
CdS10	510	2.24	0.42	0.48

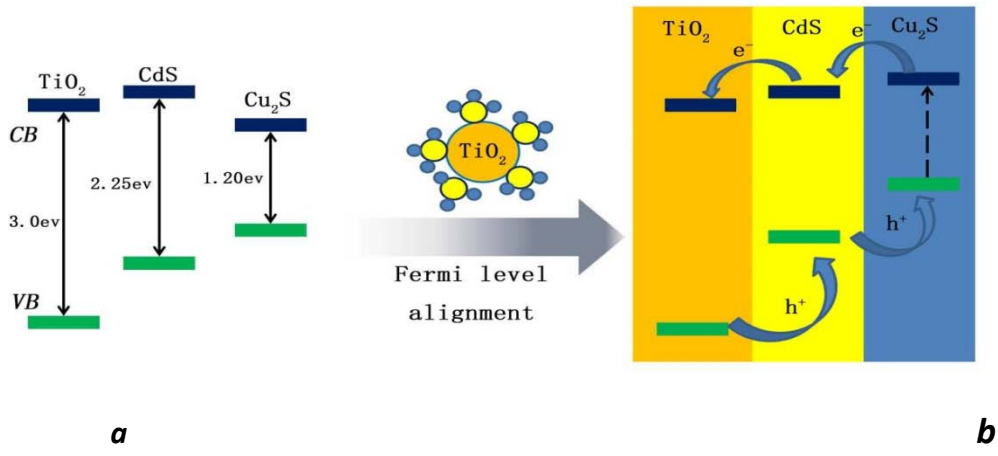

Figure 7.a: Relative band positions of TiO_2, CdS and Cu_2S in bulk and ideal stepwise structure

Figure 7.b: band edges for the efficient transport of the excited electrons and holes in TiO_2/CdS/Cu_2S photoelectrode

Second effect, when TiO_2, CdS and Cu_2S are in bulk. Their band gap are 3.2 eV, 2.25 eV and 1.20 eV, respectively can be seen in Figure 7.a.Therefor, their band gap cannot match well to provide a good path for the transportation of the electrons due to that the conductor band of Cu_2S is lower than that of CdS, which inhibit the photosensitized electrons transport from the Cu_2S to CdS [27].When Cu_2S nanoparticles were coated on the CdS QDs a great number of nanoscale heterostructure between the two semiconductors could be produced. The band gap alightment of TiO_2 CdS/ Cu_2S has been reported by Y.Chen and his colleagues [15], as shown in Figure 7.b. It is easily observed that the conduction band (CB) of CdS is lower than (CB) of Cu_2S lead to the electron transfer occurred from Cu_2S to CdS, and eventually to TiO_2 [27]. While the hole transfer occurred from TiO_2 to CdS and then from CdS to Cu_2S due to the different Fermi levels alignment when the system reached equilibration state. And when the Cu_2S SILAR reached 6 cycles, the interaction between Cu_2S and CdS achieved the best effect. Our result show the increase of J_{sc} from 2 cycle (Sample a) 3.01 mA cm^2 to 6 cycle (sample c) 4.01 mA cm^2 support this argument. The decrease of J_{sc} for sample d and e are attributed to beyond 6 cycle CdS QD nearly covered by Cu_2S nanoparticles, which would reduce the formation of more heterostructure and affect the efficient utilization of light by CdS nanoparticles. Additionally, the increase of Cu_2S cycles led to an increase in the thickness which takes a longer time for the electron to reach the TiO_2. Therefore, the probability of recombination or trapping of the generated holes would be higher, causing the photocurrent to decrease [28].

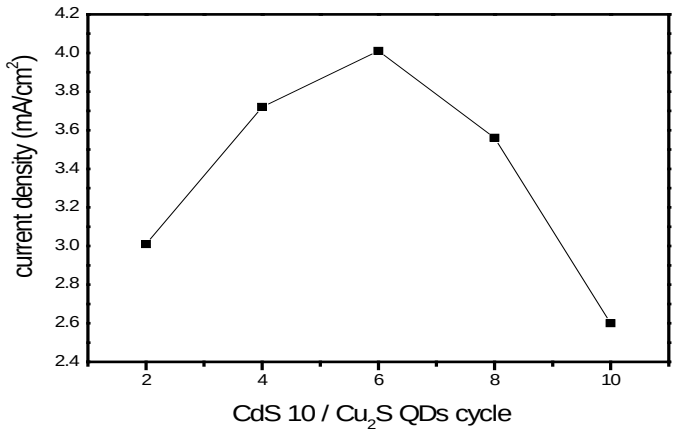

Figure 8: Variation of the current density as function of SILAR cycles of Cu_2S QD

The value of J_{sc} vs number of SILAR cycle of Cu_2S QD is shown in Figure 8.

4. Conclusions

CdS QDs were adsorbed onto TiO_2 nanoparticle electrode using SILAR method to act as sensitizer for QDSSCs. The absorption edges of TiO_2/CdS photoanode are red shift as the number of cycle increases from 2 to 10 .EDX measurements ensures the success of adsorption of CdS QDs onto the TiO_2 electrode. The values of J_{sc} and η for CdS QDs solar cell increase as the number of cycle increase. Such an increase may be result from the

improving of phtoto-absorpation efficiency and charge separation of TiO_2/CdS solar cell. Furthermore, Cu_2S were adsorbed onto TiO_2 / CdS (10) cycles (which has higher efficiency) with different cycles. The absorption edges of TiO_2/CdS10/Cu_2S photoanode are red shift from 487 nm to 553 nm as the number of cycle increases from 2 to 10 which should be ascribed to the contribution of Cu_2S. The maximum values of J_{sc} (3.56 mA/cm^2) and η (1.21%) are obtained for sample CdS10/Cu_2S for 6 cycles of Cu_2S which about 252% larger than TiO_2/CdS. The improvement of J_{sc} and η are due to the formation of hetrostructure between CdS and Cu_2S which causes the conduction band (CB) of CdS is lower than (CB) of Cu_2S . Thus more electrons can be transfer from Cu_2S to CdS and injected to TiO_2 nanoparticle electrode**.**

Acknowledgements

The authors would like to express their sincere gratitude to Ain Shams University and the Physics Department at Ain Shams University for their financial and technical support in performing and finishing this work.

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