Research Article



New Approaches to the Fabrication and Design of Dye and Quantum Dot Sensitized Solar Cells

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Abstract

In the present study, we have been used three new and innovative methods to fabricate sensitized solar cells (SSCS). In the first method, the photoanode of the solar cell is sensitizing by CdS quantum dots (QDs) and dye extracted from Ligustrum Texanum (LT) fruit. At the second approach, the dye solution obtained from LT fruit made the preparation of cation and anion precursors for CdS QDs nucleation was using in the (SILAR) technic instead of distilled water and we sensitized the cell's photoanode with these quantum dots. Finally, according to the last procedure, we used two photoanodes to trap more of the landing photons, with the first photoanode sensitized by CdS QDs and the second photoanode become sensitized by the dye. The efficiency of solar cells made by the first approach was 2.38%; the second approach was 3.16% and the third approach was 5.91%. According to the results, a significant increase in the performance and efficiency of solar cells observed (M5 Cell).

Keywords: Quantum Dot-Sensitized Solar Cells; Natural Dye; Cds Quantum Dots; Efficiency

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Introduction

Sensitized solar cells (SSCS investigated due to their low-cost production and reasonable conversion efficiency [1-3]. Many researchers try to increase efficiency for practical applications porpose. There are two distinguish ways to improve efficiency, one is the improvement of photovoltage and the other is the enhancement of the photocurrent of solar cells. For improving the photovoltage, some metal oxides used with more negative conduction band potential compare to TiO₂ such as NbO₅ [4] or SrTiO₃ [5] used as a TiO₂ substitution material. To enhance photocurrent, the improvement of absorbance in the longer wavelengths region is important because the solar spectrum has large photon flux in a wavelength region of 500-1000 nm [6]. The dye extracted from Ligustrum Texanum (LT) fruit can absorb solar light from 400 nm to 600 nm, but at the longer wavelength region (>600 nm) have not enough efficiency. In order to overcome this limitation, some light scattering materials such as metal oxide particles with diameters of several hundred nanometers added [7, 8].

In this work, we present our new and innovative approach to increasing the amount of photocurrent and thus for increasing the efficiency we used quantum dots (QDs) and dye to sensitize photoanode of solar cells. Nanoscale light absorbers can quickly separate light-producing carriers in two different environments. One challenge in sensitized solar cells is to develop the range of light absorption in whole areas of the solar spectrum to produce maximum photo-current. The absorption wavelength range of molecular dyes in dye-sensitized solar cells is usually narrow. Hence, in recent years, semiconductor nanomaterials have been using as light-absorbing materials for this type of cell. The small size QDs create a larger bandgap due to the effects of quantum confinement; this can be adjusting the absorption of light by solar cells. Hence, we expect photosensitization with two layers consisting of QDS and dye is a good way to increase light absorption and improve cell performance [9-14].

Experimental Procedure

Preparation of ${\rm TiO}_{_2}$ nanoparticles by Hydrothermal Hydrolysis method

The synthesis of $\mathrm{TiO}_{_{2}}$ nanoparticles involved the following steps.

Step 1: Preparation of Amorphous TiO₂

Initially, 0.1 mol titanium tetrachloride (TiCl_4) was mixed with 100 ml distilled water for 30 minutes in an ice bath. At this stage the Ti⁺⁴ ion is formed but the acidic solution (PH <2) [15-16]. Then the aqueous solution of NH4OH was added dropwise to a solution of PH=7 to give a white precipitate [17-18]. At this stage, the solution was continuously stirred in the ice bath for one hour. Finally, a centrifuge was used to remove the precipitate and was washed twice with distilled water four times to remove impurities. The precipitate was dried at 40 °C for 30 h [19-21].

Step 2: Preparation of TiO₂ crystalline nanoparticles

At this stage, 0.4 gr of amorphous TiO_2 prepared in the first step was immersed in 20 ml hydrogen peroxide for 30 minutes in an ice bath, under stirring. Then, 80 mL of hydrogen peroxide was dropped and added slowly [22-23]. The resulting solution was vigorously stirred in an ice bath for 11 hours until a clear yellow solution was got. This solution was transferred to an autoclave and heated in the furnace for 14 hours at 180 °C [24-25]. The precipitate was centrifuged to collect the precipitate, and the precipitate turned white [26, 27]. Sedimentation in three stages, 4 hours at 40 °C, 4 hours at 100 °C and 4 hours at 250 °C dried. Finally, the prepared sample was annealed at 130 °C for 16 hours to obtain the appropriate crystallinity in the prepared nanoparticles [28-29].

Preparation of precursors for the synthesis of CdS quantum dots (QDS)

 $Cd(NO_3)_2$ and Na_2S were using as the precursors for the synthesis of CdS QDS [30-32]. CdS QDs were deposited on the TiO₂ films by the successive ionic adsorption and reaction (SILAR) method. Briefly, the TiO₂ films were immersed in 0.5 M Cd(NO₃)₂ distilled water solution for 1min, washing with distilled water and drying in the air [33]. Then, the as-prepared films were submerged in 0.5 M Na₂S distilled water solution for 1min, washing and drying again. This process is counted as one cycle and the number of repetitions is 12 [34-35].

Preparation Of Dye Solution

The dye of the LT fruit is obtained by the following method. Wash the fruits with distilled water [36-38], then pour and mash them in a container [39-41], then mix half of them in distilled water solvent and the other half with ethanol solvent to extract the color [42-44]. After 24 hours, pass the extracted dye

through filter paper to remove the solid residue [45-47]. Finally, the dye solution was protecting from direct light exposure and stored in a refrigerator at 5 $^{\circ}$ C [48].

Preparation Of Tio, Electrode (Photoanode)

Fluorine doped tin oxide on glass (FTO glass, 7Ω /sq. Sharifsolar) was used as a substrate. The FTO glass was treated with an aqueous solution of TiCl₄ (40 mM) at 70 °C for 30 min [3]. The semiconductor paste was prepared by blending 2gr of synthesized TiO₂ nanopowder of anatase structure, 8 ml of ethanol, and 0.8 ml of Acetylacetone, and 0.1 ml Triton X-100. The resulting suspension was stirred for 2 days to make it uniform [4,6]. Transparent and scattered TiO₂ films, each with an area of 0.25 cm2, were coated on the TiCl₄ layer by a rod–doctor Blade technique using TiO₂ pastes [29]. TiO₂ films were annealed at 500 °C for 15 min. After cooling down to 80 °C, the sintered TiO₂ electrode was soaked into ethanol solutions of the natural dye extracts [33,34].

Preparation Of Electrolyte Solution

Liquid I-/I3- electrolyte were preparing from a mixture of 2 gr KI and 25 ml ethylene glycol and add 0.2 gr I2. The solution was stirring for 2 h at 50 °C and then stored in a sealed bottle [17].

Preparation Of Counter Electrode

Transparent Pt counter electrode was prepared by the following method: coating a few drops of 5mM H2PtCl6 hydrogen ethanol solution on FTO conductive glass, and then heating it at 450 °C for 15 minutes [15].

Assembly of DSSC

The dye-sensitized TiO_2 electrode and Pt counter-electrode is assembled into a sandwich type cell [19-21]. The electrolyte is injecting into the gap between the photoanode and counter electrode. Thus, the typical sandwich type of DSCs was making (Figure. 2) [18, 22].



Figure 1 : A schematic of DSSCs

Fabrication of DSSCS

In this work, five types of DSSCS containing natural dye and QDs sensitizers were fabricating and put into practice, individually, co-sensitized and additional layers. For individual solar cells, a nanocrystalline TiO_2 film on a FTO glass that was treating with an aqueous solution of TiCl_4 (0.04 M) at 70 °C for 30 min was coating. The dye extracts were adsorbing by dipping the coated glass for overnight in natural dyes (M1 cell) and CdS QDs were depositing on the TiO₂ films by the successive ion-

ic adsorption and reaction (SILAR) method for 12 cycles (M2 cell). Then, the film was washed with an ethanol solvent. Oxidation-reduction ions of iodide/ triiodide (I-/I3-) were employing as the electrolyte. Next, the sensitized TiO_2 electrode with dye and CdS QDS, the Pt counter electrode, and the electrolyte solution are assembled into a sealed sandwich- type solar cell.

For the configuration of co-sensitized solar cells, the photoelectrode was preparing by creating two layers of sensitizers. Small QDs result a larger bandgap due to the effects of quantum confinement, thus adjusting their light absorption. Hence, it is expecting that photosensitization with two layers consisting of QDs and dye is a good way to increase light absorption and improve cell performance. In order to fabricate such a cell, CdS QDs was first sensitized to the photoanode for 12 cycles by the SILAR method. The QDs-sensitized photoanode is then immersing overnight in the dark in a dye solution from the LT fruit with an ethanol solvent to apply a second layer of sensitizer (M3 cell). In fabricating (M4) cell for preparation of cationic and anionic precursors in the deposition stage of CdS QDs by the SILAR method instead of the solvent of distilled water in a solution of precursors from LT fruit pigment solution with ethanol solvent in preparation of precursors of CdS QDs we used.

The third kind of configuration was the case where two sensitized photoanodes, one with QDs and the other with pigment, were integrating into an additional layer in a cell. Its configuration is fluorine-doped oxide (FTO) glass/TiO₂/QD/TiO₂/ dye/electrolyte/Pt-FTO glass (M5 cell).

Characterization

The structure and morphology of photoanode films coated with and without CdS QDs were analyzed by X-ray diffraction (XRD) and Field Emission Scanning Electron Microscopy (FESEM) [23-25]. Energy-Dispersive X-ray Spectroscopy (EDX) carried out for elemental identifications. UV–Vis Spectrophotometer used to study the light absorption properties of the sensitized photoanodes [27, 29]. The photocurrent-voltage (J-V) test of SSCs was recorded under one sun illumination using a solar simulator [26, 28].

Result And Discussion

Figure. 2 (a, b) illustrates the X-ray diffraction patterns of the photoanode decorated with and without CdS QDS. The diffraction peak at 28.2° indicates that CdS QDS has been successfully deposited on the photoanode. FESEM was performing to characterize the morphology and structure of TiO_2 films deposited with and without CdS QDS. Figure. 3 (a) shows the FE-SEM image of the bare TiO_2 photoanode that has a polyporous surface that is convenient for the deposition of QDs.



Figure 2: X-ray diffraction patterns of (a) TiO, nanoparticles and (b) the TiO, photoanode decorated with CdS QDS

The FESEM image of TiO_2 photoanode sensitized with CdS QDs in Figure. 2 (b) did not show any significant difference compared to Figure. 2(a), and there is only a little increase in particles size that is in agreement with XRD results and a slight change in the porosity. The energy-dispersive X-ray spectroscopy (EDX) used to indicate the existing elements of Ti, O, Cd, and S in samples and their concentrations are presenting in Table 1. These results confirmed the CdS QDs were deposited on the TiO₂ nanostructure layer.



Figure 3: Surface FESEM images of TiO_2 nanoparticles before (a) and after (b) deposition of CdS QDS and (c) EDX spectra of TiO_2 photoanode decorated with CdS QDS

The EDX analysis shows the purity of the sample because no other elements were detected in Figure. 3(c), and the quantitative histograms of these elements are shown in the inset of Figure.3(c). Also, the atomic composition of elements in TiO_2 photoanode decorated with CdS QDs indicated in Table 1.

Element	Line Type	Apparent Con-	k Ratio	Wt%	Wt% Sigma	Atomic %
		centration				
0	K series	10.11	0.03401	30.63	0.37	66.18
S	K series	7.76	0.06687	14.83	0.18	15.99
Ti	K series	1.01	0.01007	2.55	0.15	1.84
Cd	L series	19.67	0.19672	52.00	0.37	15.99
Total:	-	-	-	100.00		100.00

Table 1: The atomic composition of elements in TiO₂ photoanode decorated with CdS QDs

Figure. 4 show the UV–Vis absorption spectra of the natural dye extracts of LT in ethanol solvent (Figure.4.a) and TiO_2 photoanode sensitized with CdS QDs (Figure. 4.b). It can be seen by adding LT to FTO/TiO₂/12CdS and using a new layer arrangement according to the M5 cell, the absorption of most of the solar spectrum can be enhanced (Figure.4.c).



Figure 4: UV-Visible absorbance spectrum of natural dye extracted from LT fruit (a) FTO/TiO₂/12CdS (b) and M5 Cell (c)

Using these natural dyes as photosensitizers, a photovoltaic test was performed on the produced DSSC by measuring the J-V characteristics of each cell under the condition of 100mW/cm2 under the irradiation of a tungsten halogen lamp. The performance of the natural dyes as sensitizers in the DSSCs evaluated based on the short circuit current density (JSC), open-circuit voltage (VOC), fill factor (FF), and energy conversion efficiency (η). By using the JV and PV results, we can calculate the fill factor (FF) and energy conversion efficiency (η)

$$FF = \frac{J_m \times V_m}{J_{SC} \times V_{OC}} \quad (2)$$

where Jm is the photocurrent density and Vm is photovoltage for maximum power output Pm.

Figure. 5 presents the typical J–V curves of the as prepared SSCs using the sensitizers extracted from LT in ethanol solvent and CdS QDS for 12 cycles (M1 and M2 cells).



Figure 5: J-V characteristic curves of M1 and M2 cells

The performance parameters obtained from the J-V curves are listed in Table 2, including open circuit voltage (VOC), short-circuit current density (JSC), fill factor (FF), and energy conversion efficiency (η).

Samples	V _{oc} (mV)	J _{sc} (mA/cm ²)	FF	η%
M1	721	12.35	0.5	4.45
M2	515	5.6	0.33	0.95

$$\eta = \frac{FF \times J_{SC} \times V_{OC}}{P_{in}} \times 100\% \quad (1)$$

Figure. 6 shows the J-V curve of CdS QDs and dye-sensitized solar cell (M3 cell), and Table 3 lists the photovoltaic parameters of the cell. The lower performance and efficiency of this cell compared to the solar cell sensitized using LT pigment with ethanol solvent can attribute to the reduction of binding sites for the bonding of pigment molecules on titanium dioxide nanoparticles due to the filling of these sites with quantum dots that had almost filled these positions before the pigment molecules. Also, the reason for the high performance of this cell compared to the solar cell sensitized with the CdS QDs can be attributing to the presence of pigment and the use of two types of sensitizers to expand the absorption range in the whole of the solar spectrum.



Figure 6: J-V characteristic curves of M3 cell

Table 3: The photoelectrochemical parameters of the M3 cell

Samples	V_{OC} (mV)	J _{sc} (mA/cm ²)	FF	η%
M3	660	7.7	0.47	2.38

In order to solve the problem of binding sites and the balance between pigment molecules and the binding between quantum dots and titanium dioxide nanoparticles, we considered another method in the construction of solar cells. In this new approach in order to make cationic and anionic precursors in the deposition stage of CdS QDs, we used from SILAR technic and instead of distilled water solvent in precursor solution from LT pigment, we used ethanol solvent as a cationic and anionic precursor. Figure. 7 shows the curves related to the J-V analysis, and Table 4 shows the photovoltaic characteristic parameters of the M4 cells.



Figure 7: J–V characteristic curves of M4 cell

Table 4: The photoelectrochemical parameters of M4 cell

Samples	V _{oc} (mV)	J_{SC} (mA/cm ²)	FF	η%
M4	645	9.8	0.5	3.16

The most-reported articles and research used titanium dioxide nanoparticles with a size of several hundred nanometers to trap and scatter more light entering the solar cell, while, in

this paper, two thin layers of titanium dioxide film are used with different sensitivity in each. Figure. 8 shows a schematic diagram of this cell (M5).



Figure 8: A schematic diagram of the make-up and the operational principle of M5 cell

With attention to this point that titanium dioxide film is annealing on a transparent conductive glass at a temperature of 500 °C after the coating layer, so the first layer in this cell cannot be sensitizing with pigment, because pigment molecules are degrading at temperatures above 120 °C. Figure. 9 shows the photovoltaic performance of the manufactured solar cells. Table 5 summarizes some relevant parameters.



Figure 9: J-V characteristic curves of M5 cell

The efficiency of solar cells made by the first (M3) approach was 2.38%; the second approach (M4) was 3.16% and the third approach (M5) was 5.91%, which according to the results, a significant increase in the performance and efficiency of solar cells observed.

Table 5: The photoelectrochemica	l parameters of M5 cell
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Samples	V _{oc} (mV)	J _{sc} (mA/cm ²)	FF	η%
M5	700	14.24	0.59	5.91

Conclusion

In this paper, we present an investigation on the use of two types of sensitizers in the fabrication of SSCS. We tried to improve the energy conversion efficiency of clean SSCS by substituting the sensitizer TiO₂ nanoparticles as photoanode with other nanomaterials. The photoanode is made by extracting natural dye from ligustrum texanum fruit and CdS QDs as a photosensitizer. The fabricated SSCs showed enhancement in the solar-to-electrical energy conversion. Also, according to experimental results; this new method in the fabrication leads to a significant increasing in power conversion efficiency. The photovoltaic performance of SSCs evaluated under simulated solar light irradiation. The SSCS has produced short-circuit photocurrent densities (JSC) ranging from 5.6 to 14.24 mA/cm2, open-circuit voltages (VOC) varied from 515 to 700 mV, and the fill factors varied from 0.33 to 0.59. Excellent photovoltaic performance and highest energy conversion efficiency (η =5.91%) are provided by sensitive solar cells with two types of sensitizers and two layers of titanium dioxide film (M5). The reason for this can due to the scattering and trapping of more incoming light by producing two layers of LT fruit in the photoanode, as well as more absorption of the solar spectrum due to the substitution of two types of sensitizers.

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Conflict Of Interest

The authors declare that they have no conflict of interest.

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