Fabrication and characterization of porous CdS/dye sensitized solar cells

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Abstract:

Dye sensitized solar cells (DSSCs) were fabricated from porous cadmium sulfide (CdS) nanocrystalline thin films. The porous CdS nanostructured thin films were deposited onto FTO/glass substrates by the chemical bath deposition (CBD) method. The surface morphology, crystalline structure, and optical properties of the prepared nanocrystalline thin films were investigated. Rhodamine B, Malachite green, Eosine methylene blue, and Cresyl violet dyes were used to fabricate the DSSC devices. Comparing by the absorption spectrum of the prepared porous CdS nanocrystalline films, all dyes showed an absorption peak in the transparent range of CdS thin films indicating that they are suitable for the preparation of DSSCs with CdS. Current-voltage (I-V) characteristics showed that the fabricated solar cell using malachite green dye had the highest conversion efficiency of 0.83% while using Rhodamine B dye produces a solar cell with lowest efficiency of 0.38%. However, heat treatment to the fabricated solar cells caused significant enhancement in the output of all devices.

Keywords: DSSC, CdS nanostructure, solar cell.

تحضير خصائص خلايا الصبغات الشمسية المصنوعة من المركب CdS النانوي التركيب ودراستها

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الخلاصة:

ان تحضير خصائص المواد نانوية التركيب ودراستها يُعد غاية بالاهمية وذلك للخصائص الفريدة التي امتازت بها دون المواد الاخرى. المميزات الفريدة وخاصة الزيادة الكبيرة جداً في نسبة المساحة السطحية الى الحجم جعلت من هذه المواد تدخل في تصنيع كثير من النبائط الالكترونية وخصوصاً الخلايا الشمسية. في دراستنا هذه، حُضرت اغشية المركب CdS النانوية التركيب على قواعد موصلة وشفافة FTO باستخدام تقنية الترسيب الكيميائي (Chemical Bath Deposition (CBD) . بينت فحوصات المجهر الاكتروني FE-SEM ان الاغشية المحضرة بتقنية CBD كانت اغشية اسفنجية التركيب Porous . حُضرت خلايا شمسية باستخدام اربع صبغات هي Rhodamine B، Malachite green، Eosin-methylene blue، و Cresyl violet perchlorate. اظهرت الخلايا الشمسية المحضرة باستعمال عينة CdS الاسفنجي كفاءة تحويل اعلى من خلايا تركيب الجدران النانوية التركيب، اذ كانت اعلى كفاءة تحويل حُصل عليها 0.82%

لصبغة Malachite green. المعاملة الحرارية للخلايا المحضرة وادت الى تحسن كفاءة التحويل بشكل كبير لكلا النوعين اذ بلغت اقصى قيمة لخلية Cresyl violet باستعمال غشاء CdS الاسفنجي .1.25%.

الكلمات المفتاحية: الخلايا الشمسية الصبغية ، CdS نانوي التركيب ، الخلية الشمسية.

1. Introduction

The world urgently needs alternative sources of environmentally sustainable energy supply for industrial development and electricity supply. Solar power is one of the important clean sources of power, Thus, conversion this power to useful energy have attracted a great deal of attention Dye-Sensitized Solar Cells (DSSCs) are one of the solar cells that show promising properties, use of environmentally friendly materials, ease of preparation, and low-cost of production [1-3]. Nanomaterials are widely used to fabricate solar cells, especially DSSC types, due to their unique physical properties. Cadmium sulfide (CdS) is an n-type semiconductor with a wide band gap of 2.42eV (in bulk) that makes it a very suitable material to prepare optoelectronic devices, especially as a window layer in solar cells [4, 5]. Nanocrystalline CdS thin films can be prepared by a variety physical and chemical methods like vacuum evaporation [6], pulsed-laser deposition [7], sol–gel [8], successive ionic layer adsorption and reaction (SILAR) [9], and chemical bath deposition (CBD) [10]. The CBD method produces nanocrystalline, uniform, large area, as well as inexpensive thin films [4, 11]. CdS nanocrystalline thin films, which are grown by CBD, show important interesting properties. Thus, it is used for electro-optical applications [12]. CdS nanoparticles are used to fabricate DSSCs with different nanomaterials such as TiO_2 and ZnO [13-15]. M.F. Hossain et al. [16] deposited CdS nanoparticles by the CBD method onto nanoporous TiO_2 to fabricate DSSCs and they obtained a conversion efficiency of about 1.13%. S.S. Mal and co-workers [17] found that the efficiency for TiO_2 microspheres covered by CdS nanoparticles reached 2.34%, while H. Choi et al. [18] obtained 3.14% from the device that they synthesized based on TiO_2/CdS quantum dots. Moreover, Y. Meng et al. [19] prepared devices with ZnO spheres/CdS quantum dots and obtained an efficiency of about 1.39%.The use of porous electrodes in DSSCs is much different to thin films because the inherent conductivity of the film is very low as well as the small size of the nanocrystalline particles does not support a built-in electrical field. Additionally, porous thin films can help in bringing about ionic conduction when it is in contact with an electrolyte and that led to efficient electrical conductivity in the device [20]. Therefore, using porous electrodes in DSSCs led to efficient transport of photogenerated electrons and this enhances the conversion efficiency. Moreover, the electrolyte penetrates the porous film all the way to the back-contact making the semiconductor/electrolyte interface essentially three-dimensional [21]. In the present work, we used porous CdS nanocrstalline thin film as a photoelectrode, instead of ZnO or TiO_2 , to fabricate DSSCs using Rhodamine B, Malachite green, Eosin methylene blue and Cresyl violet perchlorate dyes. The morphology, crystalline structure, optical and electrical characteristics of the fabricated solar cells were investigated.

2. Experimental details

2.1 Synthesis of CdS nanocrystalline thin films

Fluorine-doped tin oxide (FTO) coated **glass**, with a resistivity of 10 $\Omega.cm^{-1}$ used as substrates were washed in hot distilled water and then cleaned ultrasonically for 2 minutes using diluted HCl, acetone, ethanol, and isopropanol solutions, respectively. Finally, the substrates were cleaned ultrasonically with distilled water for 5 min and left to dry naturally. Porous CdS nanocrystalline thin films were deposited on the FTO/glass substrates using the chemical bath deposition (CBD) method in an alkaline aqueous solution containing 0.05M of cadmium nitrate [$Cd(NO_3)_2$] and 0.1M thiourea [$CS(NH_2)_2$]. These were used as Cd^{2+} and S^{2-} ion source, respectively. Ammonium acetate (NH_4CH_3COO) with a concentration of 1M was added to the solution as a buffer to control the reaction rate. Some drops of ammonia solution (28%) were added to adjust the pH of the reaction to 10 and the total volume was 100ml. After stirring the solution for 5 min, the FTO/glass substrates were immersed vertically in the beaker and then placed on the hot plate under the preparation temperature of 75 ^oC for 45 min. Finally, the samples were taken out of the solution and washed by distilled water to remove any contaminants. The preparation process was repeated to increase the thickness of the CdS nanocrystalline thin film. The thickness of the prepared CdS nanocrystalline thin film was 725nm by using the Swanepoel method [22]. The surface morphology of the obtained CdS nanocrystalline thin films was analyzed using scanning electron microscopy (Zeiss Supra 55VP). X-ray diffraction (XRD;PANalytical X'Pert PRO-MRD with CuKα (1.5406˚A) was employed to determine the crystalline structure of the CdS nanocrystalline thin films. To fabricate a CdS/dyes solar cell, the dye is spread onto the deposited CdS nanocrystalline thin film on FTO/glass. Then the back contact (black carbon spreadelectrolyte) is injected between the dye and carbon electrode via a small hole. The current–voltage (I–V) was investigated pre and post-hot of the device under various temperatures for 30min depending on the type of the dye used. The current–voltage (I–V) measurements were performed using a Keithley2400 source meter in the dark and under the illumination of one sun (AM 1.5, $1000W/m^2$).

2.2 Preparation of electrolyte

A suitable polysulfide liquid electrolyte was prepared for the application based on CdS thin film dye-sensitized solar cells (DSSCs). A solvent consisting of methanol and water in the volume ratio of 7:3 has been found to be the optimum solvent for preparing the liquid electrolytes [23]. This solvent ratio gave a higher conversion efficiency of the solar cell compared with pure methanol or water as a solvent. Furthermore, Na_2S and S gave rise to a good redox couple in the electrolyte for DSSCs operation, and the optimum concentrations required were 0.5M and 0.1M, respectively. Addition of potassium chloride (KCl) to the electrolyte further enhanced the performance. Then, the solution was stirred for 2 hours. Finally, the obtained solution was kept in the dark to use in DSSC device fabrication.

3. Principle of DSSC operation:

Porous n-CdS nanostructured thin film was used as the photoelectrode to fabricate DSSC devices while conductive carbon was used as a counter electrode. Polysulfide electrolyte is injected to fill the

space between. the porous CdS photoelectrode and that covered by dye and the counter electrode. When the solar cell is illuminated by light, charge carriers will be generated Figure 1 shows a scheme of the charge separation process at the Donor:Acceptor interface of a DSSC. At first, light induces the dye to produce photo-excitation into excitons (D^*) as described by the following equation:

$$D/CdS + h\upsilon \xrightarrow{Photo-excitation} D^*/CdS \quad ..1$$

Then, the excitons will migrate and separate at the CdS/dye interface and inject electrons into the conduction band of the CdS semiconductor, leaving holes (D^+) in the dye as shown by Eq 2:

$$D^*/CdS \xrightarrow{Electron-excitation} D^+/CdS + e^-/CdS \quad ..2$$

The injection process of electrons from the excited dye molecules to the conduction band of the CdS should be very fast to obtain a efficient DSSC device.

The dye will regenerate by accepting electrons from the reduced state of the redox couple (RE), producing the oxidized state of the redox couple (OX) in the electrolyte:

$$D^+/CdS + RE \xrightarrow{Dye-regeneration} D/CdS + OX \quad ..3$$

After that, electrolyte regeneration occurs via OX accepting electrons from the counter electrode and producing RE:

$$OX + {}^-e/CE \xrightarrow{Electrolyte-regeneration} RE \quad ..4$$

The injecting electron will recombine by donating electrons to D^+:

$$e^-/CdS + D^+/CdS \xrightarrow{Recombination-by-D^+,Dark-reaction} D/CdS \quad ..5$$

Finally, the injected electrons recombine by donating electrons to OX:

$$e^-/CdS + OX \xrightarrow{Recombination-by-OX,Dark-reaction} RE \quad ..6$$

Fast photo-generation of electrons and translation from the conduction band of CdS to the FTO electrode is very important to obtaining an efficient DSSC device.

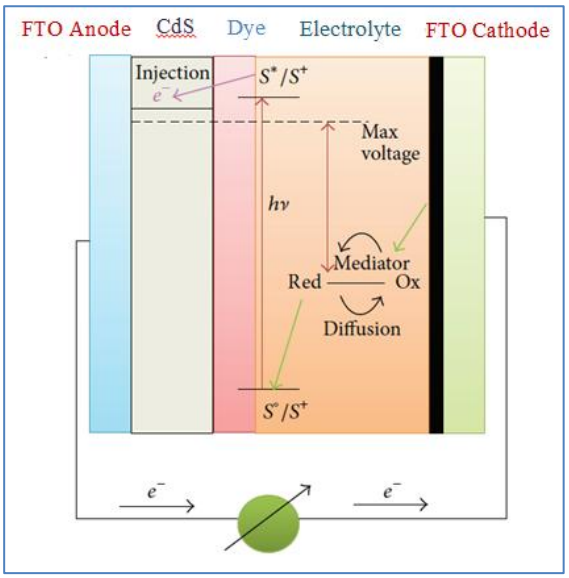

Figure 1: Structure and operating principle of dye-sensitized solar cells [24]

4. Results and discussion

4.1 Surface morphology

Figure 2A shows the FE-SEM images of a prepared CdS nanocrystalline thin film. The CdS is grown on the surface of FTO-glass substrate as a porous structure. The pores with different diameters are distributed over the surface of the CdS thin film that covers the entire area of the substrate. The type of nanostructure prepared by the CBD method can be controlled by adjusting the synthesis parameters such as molar concentration, temperature, rate of deposition, as well as the type of substrate [11]. Two ideas have been proposed for describing the mechanism of synthesis of thin films by the CBD method they are ion-by-ion and cluster.However, according to the ion-by-ion mechanism, the Cd^{2+} and S^{2-} ions are absorbed on the surface of the substrate and then combined to form CdS nanoparticles that act as seeds to grow the nanostructure. S.R. Gosavi et al. [20] suggested that the CdS nanoparticles will aggregate together to form a uniform layer which in turn led to the formation of a porous nanostructure. Figure2 inset shown

in the FE-SEM micrograph of the prepared sample at higher magnification shows clearly that the CdS nano-layers are organized and surrounded the pores as the porous nanostructure is formed. Porous structure of CdS is also prepared using the simple chemical route nucleates and found that the synthesized thin films is suitable to use as a photosensor [20]. However, other methods such as sol-gel can be used to prepare porous materials such as ZnO [25].

It is worth mentioning that porous materials are very interesting nanostructures that are used to fabricate optoelectronic devices, especially solar cells. Energy dispersive X-ray spectroscopy (EDX) analysis showed the ratio of cadmium to sulfur (Cd/S) in porous CdS nanocrystalline thin films was 1.42 (Fig.2B).

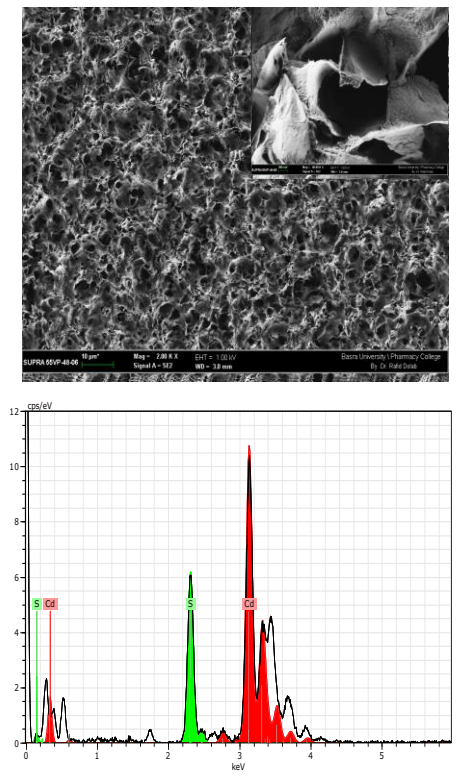

Figure2:(A) FE-SEM micrograph of porous CdS nanocrystalline thin film, inset high magnification of thin film, (B)

4.2 Crystalline structure

The crystalline structure of the prepared porous CdS nanocrystalline thin film was investigated by the XRD as shown in Figure 3. The XRD pattern contains diffraction peaks corresponding to (101), (200) and (110) planes of the FTO-glass substrate and other peaks are related to the CdS structure. However, the diffraction peaks located at 26.64^{o} could be related to the (111) plane of CdS cubic (zincblend) structure or to the hexagonal (wurtzite) phase of CdS according to the standard data base PDF-4 (00-001-0647, and 00-001-0780). In addition, the diffraction peak at 44.08^{o} could correspond to the c(220) or h(110) plane. The close location of the diffraction peaks of the cubic and hexagonal phases of CdS makes it difficult to recognize the structure type, but the absence of (101) and (100) hexagonal planes may indicate that the crystalline structure of the CdS porous thin film is zincblend [4].

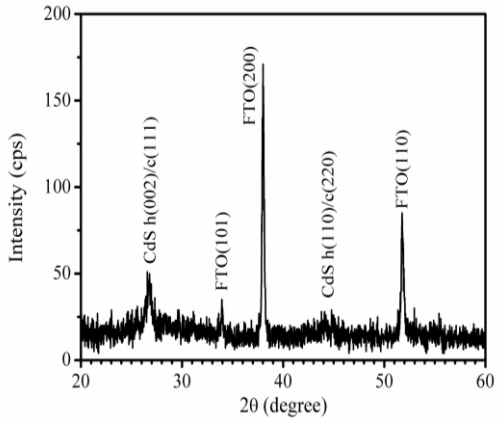

Figure3: XRD pattern of porous CdS nanocrystalline thin film prepared onto FTO/glass substrate.

4.3 Optical properties

Figure 4 shows the UV-VIS absorbance and reflectance spectra of the prepared porous CdS nanocrystalline thin films on the FTO/glass substrate. The absorption edge was observed around the wavelength of 500nm (2.48eV), and the

prepared thin films appeared to have a low reflection value (~0.5%) for the wavelength range of 500-700nm. The optical absorption of the dyes is shown in Fig. 5. Comparing with the absorption spectrum of CdS nanocrystalline films, all dyes show absorption peaks in the transparent range of CdS. The absorption peaks of 548, 580, 620, and 650nm can be absorbed by the dyes , Rhodamine B, Cresyl violet, Malachite green, and Eosin, respectively. The optical band gap (E_g) of porous CdS nanocrystalline thin film is calculated from the reflection spectrum using the relationship [26]:

$$2\alpha d = \ln[\frac{R_{max} - R_{min}}{R - R_{min}}].....7$$

where α is absorption coefficient, d is the thickness of thin film, R is the reflectance at any wavelength, and R_{max} and R_{min} are the maximum and minimum values of the reflectance. Thus, the optical band gap can be estimated from Fig. 6 in which the square of $h\upsilon\{\ln[(R_{max}-R_{min})/(R-R_{min})]\}$ is plotted against $h\upsilon$. The absorption coefficient (α) increases sharply with photon energy beyond the absorption edge. The calculated optical band gap of the prepared CdS nanocrystalline thin film is 2.51eV and is higher than that for bulk CdS of 2.42eV [27]. The increase in the optical band gap of the prepared porous CdS thin film can be related to the quantum size effect that appears in nanocrystalline structures [28]. The optical properties of the CdS nanocrystalline thin films indicate that they are suitable for use in fabricating DSSCs.

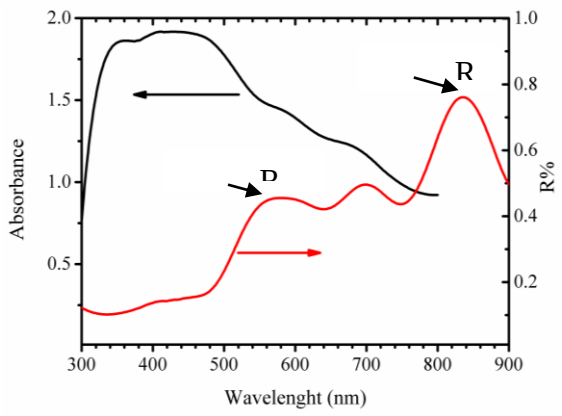

Figure 4: Optical absorbance and reflection of porous CdS nanocrystalline thin film prepared. onto FTO/glass substrate

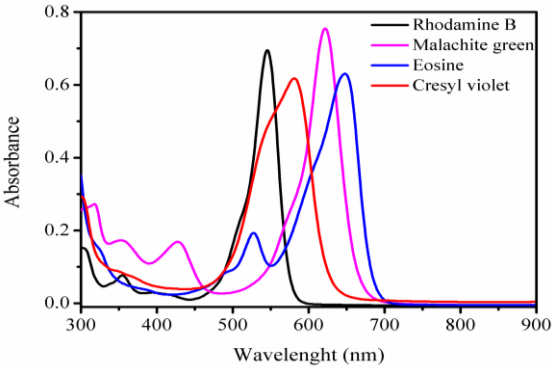

Figure 5: Optical absorbance of dyes.

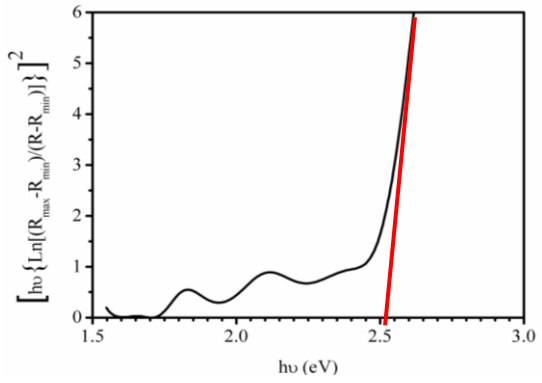

Figure 6: Optical band gap determination of porous CdS film from reflection spectra.

4.4 J-V Characteristics

Figure 7 shows the current density-voltage characteristics of as-fabricated solar cells under illumination. The porous CdS/cresyl violet device shows the highest open circuit voltage (V_{oc}) of 500.3mV

while the CdS/Rhodamine B was the lowest V_{oc} of 245.5mV. In addition, the DSSC prepared on porous CdS/Malachite green gave the highest value of J_{sc} of 5.7 mA/cm^2 in contrast to the CdS/Cresyl violet solar cell showed the lowest value of 3.1 mA/cm^2 (Table 1). We observed strong enhancement of the output of the fabricated DSSCs after heat treatment (see Fig.8). The short circuit photocurrent density J_{SC} increased for all prepared solar cells and that could be attributed to enhancement of the contact between the porous CdS thin films and the dyes. In addition the heat treatment could have caused an increase in the crystallinity of dyes that in turn led to an increase in the J_{sc} of DSSCs. The power conversion efficiencies of the solar cells were calculated using the relationship [29]:

$$\eta = (\frac{J_m V_m}{P_{in}}) \times 100\% \text{.....8}$$

Where P_{in} is the power of the incident light, J_m is the current density at the maximum power point, V_m is the voltage at the maximum power point. The fill factor (FF) is calculated using the relationship [29]:

$$FF = \frac{J_m V_m}{J_{sc} V_{oc}} \text{,.....9}$$

The fabricated solar cell using Malachite green dye shows the highest conversion efficiency before annealing of 0.83% while that using Rhodamine B dye had the lowest conversion efficiency of 0.38%. All fabricated solar cells show a low value of the fill factor in Table1. The main reason for the low conversion efficiencies could be the poor fill factors of the as prepared DSSC devices. However, the electrolyte type is another important parameter that could affect the value of the FF and η of the DSSC. The low value of fill factor may be ascribed to the lower hole recovery rate of the polysulfide electrolyte, which leads to a higher probability for charge recombination [30]

Furthermore, heat treatment of the fabricated solar cells caused significant enhancement in the output of all devices as shown in Table 2. The η of the solar cells fabricated using Rhodamine B, Malachite green, Eosin methylene blue and Cresyl violet dyes is increased by the ratio of 231%, 140%, 230%, and 219% respectively, after the heating treatment process.

The series resistance (Rs) is another parameter that can affect cell performance and high Rs value reduces the output voltage under load, thereby decreasing the FF. Thus Rs should be low because increasing the Rs of a solar cell increases the voltage drop within the solar cell [29]. The value of Rs for all prepared solar cells decreased after heat treatment as shown in Table 2. At the same time, the shunt resistance (R_{sh}) of DSSCs increased after heat treatment for two devices (except for the Creysl and Eosin solar cells). This contributed to the increase in the J_{sc} value.

Decreasing the Rs and increasing R_{sh} could be a main factor which lead to increase in the FF and efficiency of the solar cells. In addition, surface recombination is another parameter that has a significant effect on the J_{sc} and V_{oc} values of DSSC devices. The V_{oc} of all prepared DSSCs (except for the Cresyl violet device) increased after heat treatment indicating the enhancement of the CdS/dye coupling that led to fast charge injection (electrons) from the dye molecules to the porous CdS nanostructure thin films and then transfer to the FTO substrate with simultaneous transfer of the

holes from the dye to the electrolyte. B. Sankapal et al. [31] fabricated DSSCs based on the structure FTO/CdS-flat/CdS nanowires/dye using Rhodamine B and Rose Bengal dyes. They obtained η of the solar cell prepared using Rhodamine B of 0.12% while the other one showed η of 0.125%.

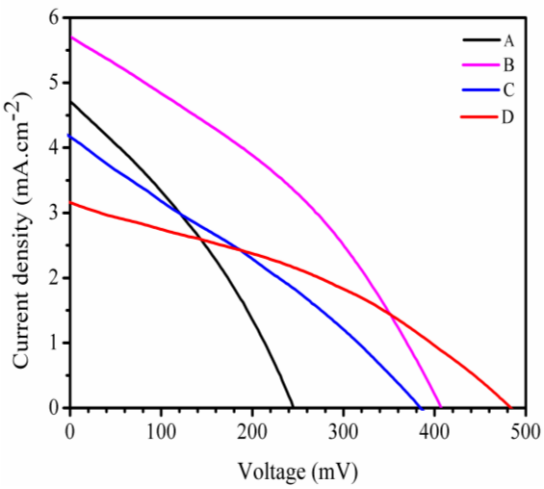

Figure 7: J-V characteristics of the prepared DSSCs (A) porous CdS/Rhodamine B dye , (B) porous CdS/Malachite green dye, (c) porous CdS/Eosin-methylene blue dye and (D) porous CdS/Cresyl violet dye.

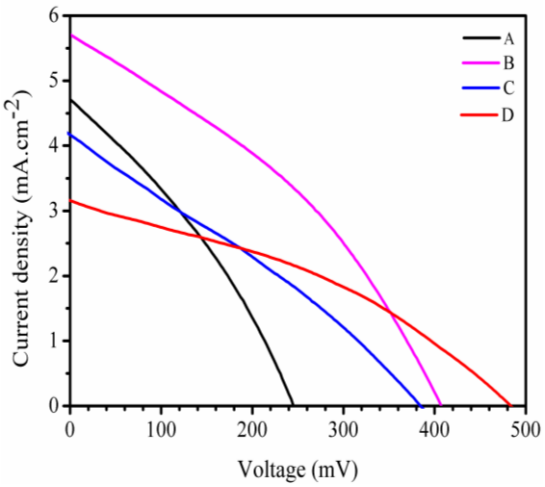

Figure 7: J-V characteristics of the prepared DSSCs (A) porous CdS/Rhodamine B dye , (B) porous CdS/Malachite green dye, (c) porous CdS/Eosin-methylene blue dye and (D) porous CdS/Cresyl violet dye.

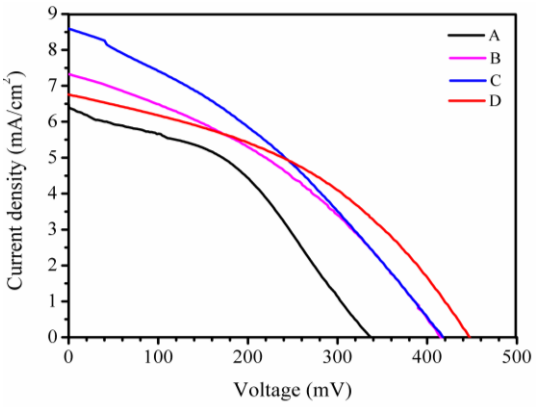

Figure 8: J-V characteristics of prepared DSSCs after annealing process (A) porous CdS/Rhodamine B dye , (B) porous CdS/Malachite green dye, (c) porous CdS/Eosin-methylene blue dye and (D) porous CdS/Cresyl violet dye.

Table1: Output of as prepared porous CdS nanocrystalline thin films/dyes DSSCs

Dye	J_{SC}(mA/cm^2)	V_{oc}(mV)	R_{sh} (Ω)	R_s (Ω)	FF%	η %
Rhodamine B	4.7	245.5	1257	507	33	0.38
Malachite green	5.7	405	1930	594	35	0.82
Eosin methylene blue	4.2	385.9	1561	1017	33	0.53
Cresyl Violet	3.1	500.3	3495	1235	38	0.57

Table2: Output of prepared porous CdS nanocrystalline thin films/dyes DSSCs after heat treatment process

Dye	Ann. temperature	J_{SC} mA/cm^2	V_{oc} (mV)	R_{sh} (Ω)	FF%	η %
Rhodamine B	185 ^oC	6.39	338.0	1617	412	41
Malachite green	125 ^oC	7.35	414.5	2281	477	37.5
Eosin methylene blue	90 ^oC	8.67	416	1312	513	34
Cresyl Violet	130 ^oC	6.77	447	3178	435	41.5

5. Conclusions

Porous CdS nanocrystalline thin films can be prepared by CBD onto a FTO/glass substrate. The porous CdS thin films could be structured with cubic phase as concluded from the XRD pattern. Porous CdS/dye solar cells were fabricated using Rhodamine B, Malachite green, Eosine methylene blue, and Cresyl violet dyes. . The DSSCs that prepared using Malachite green dye showed a high conversion efficiency of 0.88% compared with other dyes while the CdS/Rhodamine B solar cell showed the lowest efficiency of 0.38%. Heat treatment of the fabricated DSSCs lead to enhancement of the performance of the output of all devices. The conversion efficiency was increased for CdS/Rhodamine B from 0.38% to 0.88% when the device was annealed at 185 ^oC for 30min. However, the maximum efficiency of 1.25% was obtained from a Cresyl violet solar cell after heat treatment at 130 ^oC. The enhancement in the output of fabricated

DSSCs after heat treatment could be attributed to decreasing the Rs and increasing Rsh compared with as-fabricated solar cells.

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