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by Noto Gultom

Submission date: 08-May-2021 09:43AM (UTC+0700)

Submission ID: 1581067600

File name: Motlan_Jurnan_EAST.pdf (1,004.66K)

Word count: 4714

Character count: 23789



The Effect of Post-Heating Time of ZnO Thin Film on the Efficiency of ZnO/*Hylocereus polyrhizus* DSSC

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Citation: Motlan and Siregar N. The effect of post-heating time of ZnO thin film on the efficiency of ZnO/*hylocereus polyrhizus* dssc (2019) Edelweiss Appli Sci Tech 3: 70-74.

Received: Dec 09, 2019

Accepted: Dec 27, 2019

Published: Dec 31, 2019

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Abstract

The efficiency of *hylocereus polyrhizus* based Dye-Sensitized Solar Cell (DSSC) has been improved by the ZnO thin-film that was used as a working electrode for DSSC. The ZnO thin-film was improved by varying the post-heating time during the annealing process which was synthesized by a sol-gel spin coating method. The preparation of dye solution was conducted by cutting the *hylocereus polyrhizus* into small pieces and put into a beaker glass. The *hylocereus polyrhizus* then was crushed with a mortar until it was soft. In order to obtain the extracted ethanol was added and kept for 24 hours in a dark room. The extract then was filtered by using filtered paper and put into a container that wrapped an aluminum foil and kept in place to avoid the extract from sun rays. The dye sample is then UV-Vis tested to find the highest absorbance value and wavelength of the sample. The extract solution was used to form the ZnO/dye solar cell where the ZnO thin film dipped into natural dyes solution with the ZnO thin film dipping up for 24 hours to let the dye adsorbed by the ZnO thin film. The ZnO thin film was dipped into extract *hylocereus polyrhizus* function as a working electrode and put together with a platinum counter electrode that separated by Surilyn. The pasting with Surilyn was conducted by pushing the working electrode and counter electrode and heated on a hot plate of the temperature of 70-80°C to perfectly put together. The working electrode which was put together to the platinum counter electrode was injected with liquid electrolytes through a small hole on the platinum counter electrode. Electrical testing is carried out after the DSSC has been assembled by making an electrical circuit between the DSSC with a measuring instrument. The series value of the *hylocereus polyrhizus* was 0.652 μA , at the wavelength of 538 nm. The maximum power of the DSSC is 0.10030 W/cm^2 and the efficiency of 0.0274%.

Keywords: Dye-sensitized solar cell, Post-heating time, ZnO thin-film, Sol-gel spin coating method, *Hylocereus polyrhizus*.

Abbreviations: DSSC-Dye-Sensitized Solar Cell, DEA-Diethanolamine, VOC-Open-Circuit Voltage, XRD-X-Ray Diffraction.

Introduction

Sunlight is one of the energy sources that have an abundant energy source; however, it has not been exploited well. The energy released by angry rays is actually only received by the surface of the earth at 69% of the total solar radiant energy [1]. The supply of solar energy from sunlight received by the earth's surface reaches 1024 J/year, this energy is equivalent to 1017 watts. The amount of energy is equivalent to Ten Thousand times the energy consumption throughout the world. In other words, by closing only 0.1% of the earth's surface with solar cell devices that 10% efficiency is able to meet the energy needs of the world. Solar cells are devices that convert solar energy into electrical energy, which is directly the current and voltage produced by solar cells depending on sunlight.

Solar cells based on current technological developments and manufacturing materials can be divided into three, namely: First, solar cells made of single silicon and multi-crystalline silicon. Second, thin-film solar cells and the third organic solar cells DSSC, the third generation of chip and reliable conversion of solar energy has gained an attraction to the development of thin-layer solar cells [2,3]. DSSC consists of several components such as oxide semiconductor, dyes, the counter electrode and electrolyte. There are several ways of improving the quality of the DSSC such as obtaining a growth window of the thin film semiconductor, the molecular structure of the dyes, electrolyte

pair of reduction-oxidation and electrode material. In the engineering of the semiconductor that is used as working electrode in the DSSC, the use of ZnO thin film has given high promise due to its higher bandgap compared to TiO_2 that previously used, therefore gives higher voltage, higher carrier mobility that can reduce carrier recombination, and has structure and morphology that can be controlled in its synthesis. In addition, ZnO also has a wide bandgap of 3.37 eV, has high optical transparency at room temperature and 60 meV electron binding ability [4,5]. ZnO thin film has been synthesized by several methods such as molecular beam epitaxy, RF magnetron sputtering, pulsed laser deposition, spray pyrolysis, chemical bath deposition, physical vapor deposition [6-14].

However, this method involves a rather complicated procedure carried out because it requires sophisticated equipment, instead of using the sol-gel spin coating method because the equipment is simple and low cost, does not use space with high vacuum and very good microstructure. Research on DSSC has been carried out using various types of semiconductors with variations in heating temperature, heating holding time, spin-coating speed and various types of dyes using thin-film TiO_2 with calcination temperature of 450°C and holding time 120 minutes and using dye extract from spinach leaves, DSSC efficiency was obtained 0.13% [15].

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Thin-film TiO₂ with calcination temperature of 450°C and dye extract from Male Flower Leaves (*Luffa Cylindra-L*) obtained efficiency DSSC 1.3% [16]. Thin-film TiO₂ and dye extracts from various fruits and leaves, the result is that the magnitude of DSSC efficiency depends on the fruit or leaf extract used [17,18]. TiO₂ thin films and the variation of solvents in dye extracts from *Melastoma Malabatricum* Leaves fruit results that the magnitude of DSSC efficiency was influenced by solvents [19]. Varying the thickness of TiO₂ thin films the result is that the magnitude of DSSC efficiency is influenced by the thin film thickness [20]. Variations in the temperature of ZnO thin films (300 °C, 350 °C, 400 °C and 500°C), the result is the highest DSSC efficiency of 3.92% at 400°C [21].

It cause ZnO has demonstrated multifunctional properties with high energy binding strength, low resistivity, great light catching characteristics, high optical transparency at room temperature, 3.37 eV wide bandgap, and the ability to bind free electrons at 60 meV. The quality of the thin film is a crucial point in the improvement of the efficiency of the DSSC. In this work, we tried to improve the quality of the ZnO thin film by varying the Post-heating time of the ZnO thin film during the sol-gel spin coating. The aim of this research is seeing the effect of Post-Heating time of ZnO thin film on ZnO/dye-based DSSC efficiency.

Experiment Method

ZnO Thin Film Synthesis

Materials used in this research were Zinc Acetate Dehydrate, Isopropanol and Diethanolamine which successively used as basic materials, solutions, and stabilizers. The ZnO thin film was synthesized by using sol-gel spin coating method. Zinc Acetate dehydrates (Zn(CH₃COOH)₂·2H₂O) was diluted in isopropanol, stirred with a magnetic stirrer for 10 minutes, then little by little 1.72 ml Diethanolamine (DEA) was dropped into the solution. The solution which was in the form of the gel was dropped on an FTO glass substrate and spin with a speed of 5000 rpm. The sample then was heated with pre-heating 250°C and Post-Heating 500°C with heating with a variation of 30, 60, 90, 120 and 150 minutes. The ZnO thin film samples were then characterized by (X-Ray Diffraction) XRD, SEM, and UV-Vis.

The Preparation of Dye Solution (Sensitizer)

The preparation of dye solution was conducted by cutting the *hylocereus polyrhizus* into small pieces and put into a beaker glass. The *hylocereus polyrhizus* then was crushed with a mortar until it was soft. In order to obtain the extracted ethanol was added and leaf for 24 hours in a dark room. The extract then was filtered by using filtered paper and put into a container that wrapped an aluminum foil and kept in place to avoid the extract from sun rays. The dye sample is then UV-Vis tested to find the highest absorbance value and wavelength of the sample.

DSSC Synthesis

The extract solution was used to form the ZnO/dye solar cell where the ZnO thin film dipped into natural dyes solution with the ZnO thin films facing up for 24 hours to let the dye adsorbed by the ZnO thin film. The thin film was dipped into extract *hylocereus polyrhizus* function as a working electrode and put together with a platinum counter electrode that separated by Surilyn. The pasting with Surilyn was conducted by pushing the working electrode and counter electrode and heated on a hot plate of the temperature of 70-80°C to perfectly put together. The working electrode which was put together to the platinum counter electrode was injected with liquid electrolytes through a small hole on the platinum counter electrode.

Efficiency Measurement

Electrical testing is carried out after the DSSC has been assembled by making an electrical circuit between the DSSC with a measuring

instrument, namely a digital multimeter as shown in Figure 1. This test is based on the lighting method of the light beam to determine the performance and efficiency of the cells obtained when the solar cell object is exposed to light with certain intensity at the top of the electrode (anode). DSSC outputs are an Open-Circuit Volt (Voc) and Short Circuit Current (Isc) DSSC. Then the amount of fill factor (FF) and DSSC efficiency are calculated (η).



Figure 1: DSSC Efficiency Measurement.

Result and Discussion

The crystal structure was determined by using XRD. Figure 2 shows the XRD pattern of the ZnO thin film of a sample with pre-heating 250°C and Post-Heating 500°C with the variation of 30, 60, 90, 120 and 150 minutes.

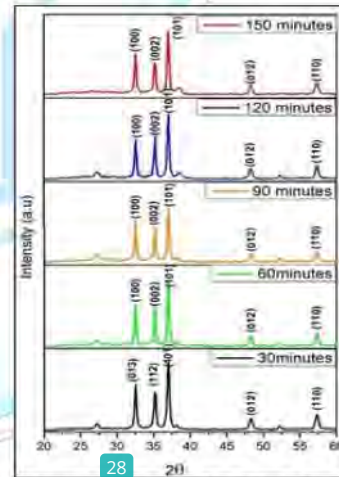


Figure 2: X-ray diffraction spectra of ZnO.

The X-ray diffraction pattern of the sample is analyzed using search march by showing the sample has the same crystal planes namely fields (100), (002), (101) and has same growth peak and oriented to the plane (101). Analysis of the XRD pattern shows that samples are hexagonal wurtzite and growth orientation toward the C axis perpendicular to the substrate surface along with standard data of ZnO of JCPDS 80-0075 card. The ZnO crystal size measured using Scherrer equation [22].

$$D = \frac{0.9 \lambda}{\beta \cos \theta} \quad (1)$$

Whereas D=crystal size, λ=wavelength, β=FWHM (Full Width Half Maximum), θ=diffraction angle.

Crystal sizes of the ZnO thin films calculated using equation 1 are shown in Table 1.

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Post-Heating Time (minutes)	Phase	Peak		Crystal size (nm)
		2θ (degree)	FWHM (degree)	
30	ZnO	36.9775	0.375	22
60	ZnO	36.9513	0.3049	27
90	ZnO	36.9582	0.3264	27
120	ZnO	36.9545	0.3376	25
150	ZnO	36.9428	0.339	25

Table 1: Crystal size of ZnO.

The size of the crystal is influenced by the value of FWHM, if the FWHM value decreases the size of the crystal is large and vice versa. The size of the crystals obtained for all samples is almost the same in the range of 22-27 nm. This shows that the duration of heating does not affect the size of the crystal, because the difference in the duration of heating is very small. The increase in Post-Heating temperature is in line with the decreasing FWHM value and increasing crystal size, this is due to the higher heating temperature in the growth of ZnO thin film crystals, the energy obtained by ZnO atoms to form the higher the crystal field as well, so the better the Crystal formed [19-23].

14 The Optical Property of the ZnO Thin Film

Transmittance and absorption spectra for all samples were taken in the wavelength range of 300-800 nm which is in the range of its application for solar cells. Figure 3a shows there is a sharp increase in transmission of the ZnO thin film in the wavelength range of 350-430 nm which is in the ultraviolet range. The highest and lowest transmission value is when the Post-Heating of 30 and 120 minutes. According to the heating mechanism the higher, the heating temperature and the longer the Post-Heating time the better is the compacting of the powder and the stronger the bond among granules and porosity of the material.

The high transmission value of ZnO thin film, make it a good application for solar cell [24]. Absorbance spectrum on the ZnO thin film is shown in Figure 3b. There is a sharp decrease in the adsorption for all samples in the wavelength range of 350-430 nm and the highest and lowest adsorption consecutively when Post-Heating 120 and 30 minutes. Therefore, the ZnO thin film has high transmittance at visible light and can be applied in optoelectronic. The ZnO thin film has a direct bandgap, therefore the bandgap can be calculated by using the following equation 2 [25,26].

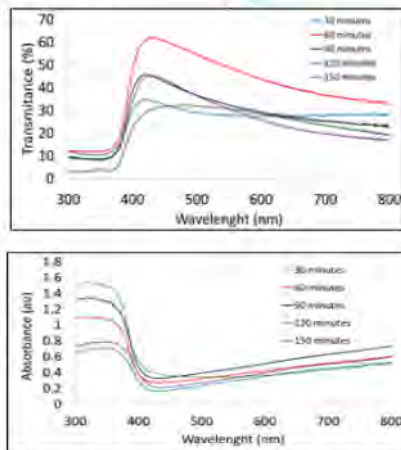


Figure 3: Optical properties of ZnO thin film: (a) transmittance, (b) absorbance. $[(\alpha h\nu)]^2 = C_D (h\nu - E_{opt})$ (2)

The bandgap with variations in the duration of heating to Post-Heating of various Post-Heating time of ZnO thin film which was derived using the Tauc Plot method is shown in Figure 4.

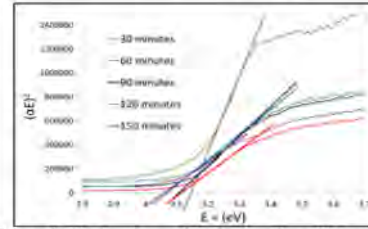


Figure 4: The bandgap of ZnO using Tauc Plot method.

The bandgap of Figure 4 of ZnO thin film as a function of Post-Heating. Figure 4 shows that the highest bandgap is 3.140 eV and the lowest is 3.073 eV of Post-Heating time of each 120 and 150 minutes. Generally, the bandgap increases as the Post-Heating time increase up to 120 minutes, beyond that temperature the band gap decreases. The increase in bandgap along with the increase in Post-Heating time is due to the compacting of the granules, and the decrease of porosity size and number [27-29]. The increase in density of the smaller granules will increase the surface energy as well as the bandgap. However, by further increasing the Post-Heating time will start forming bigger granules and reduce the energy gap [30].

The Absorbance of *Hylocereus polyrhizus* Dye Solution

The absorption of the sensitizer extracted red dragon dye solution which was determined by UV-Vis is shown in Figure 5.

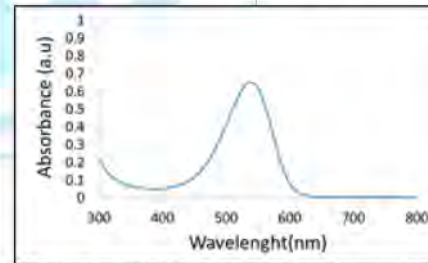


Figure 5: Graph of wavelength Vs absorption of extracted *hylocereus polyrhizus* dye solution.

The absorption intensity of the *hylocereus polyrhizus* dye solution as shown in Figure 5 is 0.652 a.u at the wavelength of 538 nm. Previous work showed that the peak absorption of the *hylocereus polyrhizus* was in the range of 420-580 and absorption intensity was affected by solution concentration, the higher the solution concentration the higher is the solution absorption value [31,32]. Absorption peaks in the range of the wavelength of 500-600 nm show that there is the content of anthocyanin in the dye solution [33-35]. The peak absorption of *hylocereus polyrhizus* at a wavelength of 535 nm which is in the range of 450-600 nm [36]. The photon absorption by the solar cell is very crucial in solar cell technology, the higher the absorption the bigger the photon excitation the bigger is the amount of solar energy converted into electricity.

The efficiency of DSSC

The percentage of efficiency is obtained by comparing the power produced by the DSSC (P_{max}) prototype with the power which is given by solar input (P_{in}) shown by the following equation 3 and the output power can be obtained from the following equation 4.



$$\eta (\%) = \frac{(J_{SC}) \times (V_{OC}) \times FF \times 100}{P_{in}} \quad (3)$$

$$P_{max} = V_{max} \times J_{max} \quad (4)$$

Table 2 Shows the efficiency ZnO/DSSC of the *hylocereus polyrhizus* based on a variation of Post-Heating time during the synthesis of ZnO thin film is shown in Table 2.

Post-Heating Time Minutes	Vmax (V)	Jmax (mA/cm ²)	Pmax (W/cm ²)	Pin (W/cm ²)	FF (%)	η (%)
30	0.4	0.00258	0.00205	36.5	198.54	0.0056
60	0.4	0.0352	0.0015	36.5	10.638	0.0041
90	0.4	0.0075	0.00275	36.5	91.667	0.0075
120	0.4	0.013	0.1003	36.5	192.78	0.0274
150	0.4	0.0025	0.00533	36.5	532.5	0.015

Table 2: The values of voltage, current density, and DSSC efficiency with Post-Heating time variation and *hylocereus polyrhizus* dye.

Table 2 shows that the highest efficiency of the DSSC is when the Post-Heating time of the ZnO is 120 minutes which 0.0274%. The increase in the bandgap, as well as the increase in energy efficiency, is due to the improvement in crystal growth during the Post-Heating time. The increase in the formation of small granules, and at the same time decrease the porosity has improved the quality of the DSSC. The increase in the number of granules will facilitate electrons flow from conduction band into valence band, that facilitates photocatalyst reaction and bigger absorption by the dye, and therefore bigger spectrum. Semiconductors with a wide bandgap will multiply electrons flowing from the conduction band to the valence band, which makes the photocatalyst reaction chamber and absorption by the dye will become more so that the spectrum becomes wide [37].

Conclusion

Dye-Sensitized Solar Cell can be fabricated by using ZnO thin film with the variation of Post-Heating and extracted dye from *hylocereus polyrhizus*. The smallest size of the ZnO thin-film was 22 nm for Post-Heating of 30 minutes and the bandgap 3.140 eV for the Post-Heating time of 120 minutes. The intensity of extracted dye from the *hylocereus polyrhizus* Sensitizer is 0.652 a.u and peak position at 433 nm. The ZnO thin-film and *hylocereus polyrhizus* based DSSC can convert solar energy into electrical energy. The maximum power obtained in this research was 0.10030 W/cm² and the maximum efficiency of 0.0274% for the Post-Heating time during synthesis ZnO thin-film of 120 minutes.

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