

Research Article

The Synthesis and Characterization of ZnO/Natural Dyes for Dye-Sensitized Solar Cells

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Abstract

The ZnO/Natural Dyes had been successfully synthesized as Dye-Sensitized Solar Cells (DSSC). The natural dyes were extracted from tamarillo, red dragon fruit, red spinach, boat lily flower, and purple hibiscus flower. Preparation of ZnO thin films were carried out on an indium thin oxide (ITO) glass substrate using sol-gel method. The ZnO thin films were characterized by XRD and UV-Vis. The ZnO/natural dyes thin films were formed by dipping the ZnO thin film into natural dye solutions with the ZnO thin films facing up for 24 hours to let the dyes adsorbed by the film. DSSC efficiencies were measured by a simple circuit that consists of DSSC, multimeter both analog and digital. The results show that the ZnO crystal structure was of wurtzite hexagonal shape with size of 27.3 nm, and the band gap was 3.25 eV. The maximum efficiency of DSSC was of purple hibiscus flower which was 0.10017 %.

Keywords

- DSSC
- Crystal structure
- ZnO thin film
- Natural dyes
- Efficiency

INTRODUCTION

Dye-sensitized solar cell (DSSC) is a potential alternative for solar cells. DSSC does not require high-purity material and therefore lower cost [1-3], and it has a wide band gap [4,5]. Nanoparticles TiO₂ and ZnO solar cell has been achieved [6]. ZnO is an n-type semiconductor material, II-VI group with wide energy band gap of 3.37 eV and a binding energy of 60 meV [7-9]. The structure and optical properties of ZnO thin films are influenced by synthesis conditions such as solvent concentration [10], heating temperature [11], and coating spinning speed [10]. Dyes which were used as a dye sensitizer could be either synthetic or natural dye [12]. Natural dyes can be extracted from various fruits and plants, while synthetic dyes are expensive and generally use organic metal based ruthenium complex that contains heavy metals that can contaminate environment [13].

Previous researches had used natural dyes from spinach and ipomoea [14], rosella and blue pea flowers [15]. Resent research also had used ZnO layer and dye extracted from tamarillo, however its efficiency was still low [1] size and characterize the structural and optical properties of ZnO thin film and to measure the efficiencies [6]. The aims of this research are to synthesize and characterize the DSSC ZnO/natural dyes. The natural dyes were tamarillo, red dragon fruit, red spinach, boat lily flower, and purple hibiscus flower.

MATERIALS AND METHODS

Synthesis of ZnO thin films

ZnO thin film were Synthesized by sol-gel method. Zincacetate

dehydrate of 4,0 grams was added into isopropanol solvent of 35.72 ml then stirred with a magnetic stirrer and DEA of 1.92 ml was added into the solution to form sol-gel. The sol-gel was dropped on to ITO glass substrate and with spin coating of 1800 rpm. To improve the ZnO thin films, it was annealed to 250°C (preheating) for 30 minutes and to 500°C (post-heating) for 30 minutes.

Synthesis of the ZnO/natural dyes

Tamarillo, red dragon fruit, red spinach, boat lily flower, and purple hibiscus flower were each milled and then the milling was soaked with distilled water for 24 hours. The extracted dyes were heated for 5 hours and diluted with distilled water to obtain dye solutions. The solution then, filtered with filter paper and put into an aluminium foil to be tested by a UV-Vis to obtain the highest absorption. The best solution then was used to form the ZnO/natural dyes. The ZnO thin film was dipped into natural dye solution where the ZnO thin films facing up for 24 hours to let the dyes adsorbed by the film.

Electricity measurement

In order to measure the DSSC efficiency a simple circuits that consists of DSSC. Each sample was stacked with a layer of carbon electrodes then clamped with a paper clip and connected to a series circuit to determine the value of current and voltage.

RESULTS AND DISCUSSION

X-ray diffraction (XRD) diffraction pattern of the ZnO thin film is shown in Figure 1. It shows that the ZnO thin film structure is *hexagonal*.

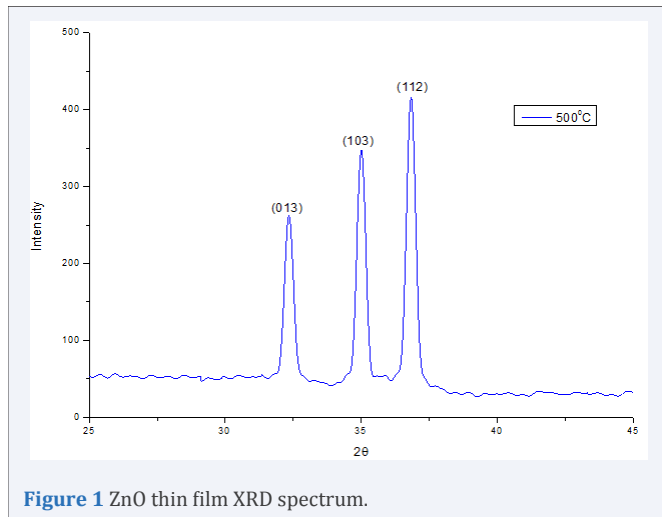


Figure 1 ZnO thin film XRD spectrum.

The XRD results, was analyzed using origin, shows orientation peak of (112) that shows wurtzite hexagonal structure in accordance with JCPDS 80-0075 card.

The size of the ZnO crystal was calculated using Scherrer equation:

$$D = \frac{0,9\lambda}{\beta \cos \theta} \quad (1)$$

Where as D = crystal size, λ = wavelength, β = *FWHM* (full width half maximum) and θ = diffraction angle. The average size of the ZnO thin film was 27.3 nm.

Transmission spectrum is shown in Figure 2. A sharp increase in transmission is shown in the region of 338 to 410 nm wavelength and stable in the region of the wavelength of >410. The lowest and highest intensities are consecutively 23.1% and 96.6%. The adsorption spectrum is shown in Figure 3. The sharp decrease in the adsorption spectrum is in the region of 338 nm to 410 nm wavelength and then stable at the wavelength of > 410 nm wavelength. The lowest and the highest adsorptions are 0.05 a.u and 0.49 a.u. Therefore the ZnO thin film has a high transmission in the visible region that can be used as transparence window in optoelectronic devices [17].

ZnO thin film is a compound semiconductor which has a direct band, and optical band gap can be obtained from the following equation [18]:

$$(\alpha hv)^2 = C_D(hv - E_{opt}) \quad (2)$$

where α = absorbance coefficient, h = Planck constant, ν = photon frequency, C_D = constant, E_{opt} = gap energy. The band gap of the ZnO thin film is 3.25 eV.

Figure 2 and 3 shows a sharp increase in the transmittance value in the wavelength range of wavelength of 338 nm to 410 nm and stable in the range over 410 nm wavelength. The lowest and highest transmittance value are consecutively 23.1% and 96.6%. The absorbance spectrum shows a sharp decrease in the wavelength range of 338 to 409 nm. The lowest and highest absorbance values are consecutively 0,4% and 49%. The band gap energy is 3.25 eV.

The UV-Vis spectra of tamarillo, red dragon fruit, red spinach, boat lily flower, and purple hibiscus flower dyes were taken in the range of 200 nm to 800 nm, shown in Figure 4. The absorbance intensities and their position for each Tamarillo, red dragon fruit, red spinach, boat lily flower, and purple hibiscus flower consecutively: 2.78 a.u and 330nm, 2.79 a.u and 328 nm, 3.30 a.u and 242 nm, 2.50 a.u and 415 nm and 2.90 a.u and 246. These results show each color has its anthocyanin pigment and different abilities in absorbing energies [19], where the bigger its anthocyanin content the bigger its energy absorption ability [20]. The mechanism of transduction energy was also used in solar cell [21] Photoelectric effect [22].

DSSC efficiency

Efficiency is the percentage of power ratio produced by *DSSC* compared to the power produced by the sun to illuminate the earth.

$$\eta_{ij} = \frac{(J_{sc}) \times (V_{oc}) \times FF \times 100}{P_m} \quad (3)$$

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

The electrical properties as well as their efficiencies of each

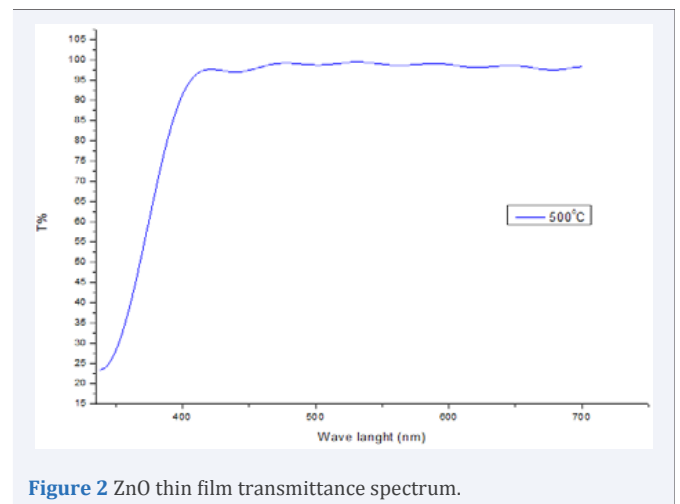


Figure 2 ZnO thin film transmittance spectrum.

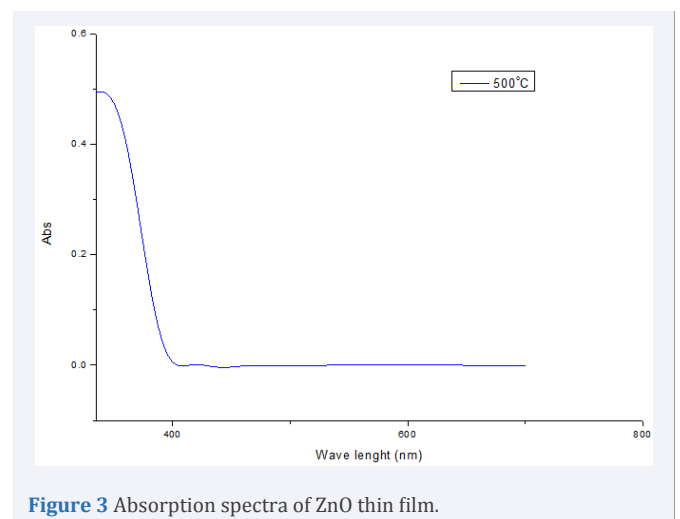


Figure 3 Absorption spectra of ZnO thin film.

tamarillo, red dragon fruit, red spinach, boat lily flower, and purple hibiscus flower were obtained by using equation 3 and 4. The results are shown in Table 1.

The different in DSSC efficiencies are due to the different in the molecules of color substances in the mechanism of energy transfer [19]. The extract of purple hibiscus flower has a higher photosensitivity performance compare to others. This is due to charge transfer between the color substance molecules of purple hibiscus flower and the ZnO thin film are better than those of tamarillo, red dragon fruit, red spinach, boat lily flower. This related to the compatibility of ZnO thin film surface and the structure of the molecules of the color substances [23]. The highest efficiency of the DSSC of the purple hibiscus flower and the lowest efficiency of the red spinach are corresponding previous works [24,25]. The DSSC efficiency depends on the degree of anthocyanin of the extract color substance molecules.

CONCLUSION

Based on the results of the study and discussion, it can be concluded that the crystal structure of ZnO thin film is *hexagonal wurtzite* and ZnO crystal size is 27.3 nm. The spectrum of transmittance and absorbance curve shows an increase in the transmittance value which is quite sharp and a fairly sharp decrease in the wavelength range 380 nm to 410 nm and is stable at a wavelength > 410 nm. Value of ZnO thin film band gap is

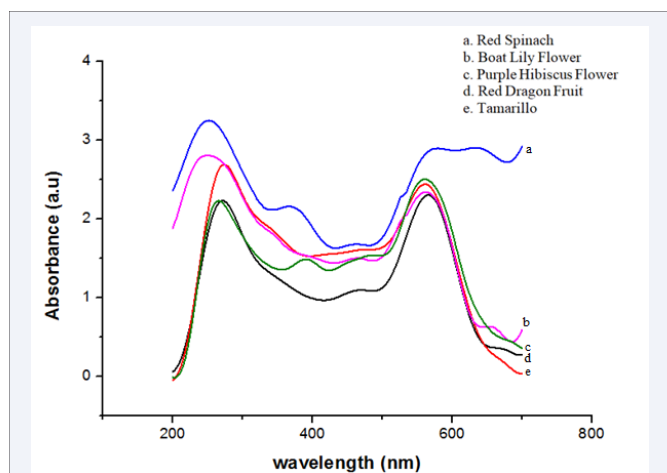


Figure 4 Wavelength vs. Absorption Graph of Natural dyes solution.

Table 1: Electrical voltage values, current density and DSSC Dye efficiency.

| Variety of Natural Dye | V_{oc} (V) | J_{sc} (mA/cm ²) | P_{max} (W/cm ²) | P_{in} (W/cm ²) | FF (%) | η (%) |
|------------------------|--------------|--------------------------------|--------------------------------|-------------------------------|--------|------------|
| Tamarillo | 0.45 | 0.400 | 0.035 | 36.5 | 19.794 | 0.097 |
| Red Dragon Fruit | 0.50 | 0.281 | 0.030 | 36.5 | 48.214 | 0.082 |
| Red Spinach | 0.35 | 0.187 | 0.015 | 36.5 | 17.778 | 0.041 |
| Boat Lily Flower | 0.45 | 0.712 | 0.101 | 36.5 | 31.676 | 0.278 |
| Purple Hibiscus Flower | 0.45 | 0.487 | 0.036 | 36.5 | 29.250 | 0.100 |

3.25 eV. DSSC efficiency maximum with dye extract from hibiscus flowers that is 0.10017 %.

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