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# Natural Dyes as a Photosensitizer from Dragon Fruit (*Hylocereus polyrhizus*) for Dye-Sensitized Solar Cells

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## Abstract

Natural dye is one of the important materials of DSSC which absorbs photons from the sun's rays and transmits the photoelectron. One of the natural dyes is dragon fruit dye. The characterization of the optical and electrical properties of a sensitizer DSSC used a UV-Vis spectrometer, FTIR spectrometer, and solar power meter. The absorption spectra of dragon fruit dye extracted were obtained by UV-Vis spectrophotometer, Fourier Transforms Infra-Red (FT-IR) was used to characterize the dye's components, and measure the radiated and transmitted sunlight by solar power meters. Result of UV-Vis absorbance, dragon fruit dye has the highest absorbance rate with a peak of 530 nm and bandgap of 2.09 eV, while the ZnO sensitized by dragon fruit dye has the highest absorbance rate with a peak of 535 nm and bandgap of 2.56 eV. From FT-IR spectrometer result shows that dye extracted from dragon fruit, and ZnO sensitized by dragon fruit dye contained C=O stretching vibrations at the peak of 1716 $cm^{-1}$, and the peak at 3423 $cm^{-1}$, and 3448 $cm^{-1}$ shows the O-H stretching vibration. The C-O-C which corresponds to the esters group for dragon fruit's dye is observed. It is a part of anthocyanin and chlorophyll. Zinc oxide (ZnO) paste is coated on the Indium Tin Oxide (ITO) as the thin film is dipped in the dragon fruit dye extracted for 30 hours. A thin film is prepared as the working electrode and counter electrode. The characteristic of current and voltage (I-V) as electrical properties are measured by the solar power meter. The best efficiency of dragon fruit dye extracted is 0.03%, and it can absorb the light from the sun. The performances of the dye absorption spectrum on the surface area are important aspects to determine the efficiency of the solar cell. The dragon fruit dye extracted coated ZnO thin film could be used as a sensitizer since it will increase the correlation between the ZnO thin film and dye. And the last, it will improve the quality of solar cells as well. The dragon fruit dye has the potential a natural sensitizer in DSSC.

**Keywords:**
Optical and electrical properties, zinc oxide, dragon fruit dye, sensitizer solar cells.

## 1 Introduction

Dye-Sensitized Solar Cell (DSSC) is a solar cell type that uses the color solution and is composed of a photo-anode, an oxide layer for electronic conduction, a monolayer of charge transfer dye, and a redox electrolyte, usually an organic solvent [1][2][3]. The function of the photo-anode is very important to convert light energy into electrical energy and is deposited in the conductive glass. Photo-anode is composed of a working electrode that had been coated with semiconductor materials [4][5]. The photo-anode has a large surface area for the dye attached and good electron transport capability to achieve high energy conversion efficiency performance. The components of a Dye-Sensitized Solar Cell (DSSC) are a working electrode, a counter electrode, and an electrolyte solution [6].

The electrodes are used as a separation between the light absorption function and the transport of the charge carrier. The DSSC functions as a medium for the absorption of solar radiation and widely-gap semiconductors such as $TiO_2$, ZnO, and $Nb_2O_3$ as charge carrier transport [7]. The dye-sensitized solar cell is a sandwich structure. It is made of transparent conductive glass, a porous nanometer membrane, electrolytes solution, and an electrode structure. The photoelectric conversion is complete in several interfaces dye and porous membrane, the dye molecules and electrolyte, and the electrolyte and the electrode. Dye sensitizers must have the absorption ability in the irradiation of sunlight. The dye molecules consist of hydroxide groups or carboxyl ones and can efficiently electron injection into the conduction band of the semiconductor. Dye sensitizers can be extracted from the natural dyes of plants [8][9].

In the previous research, there were types of research on natural dyes in DSSC. Natural dyes derived from dragon fruit [10] [11], rosella and blue pea flowers [12], red Sicilian orange, and purple eggplant fruits [13], dyes of *Prunus domestica*, *Mangifera indica*, and *Citrus limon* [14], *Aloe vera* and cladode of cactus extracts [15][16], etc. The natural dyes are suggested to the similar to the characteristic of the absorption coefficients highly. The synthesis of DSSC using natural dyes may offer a new possibility for the next application because the process is simple, low cost, green environment, has no pollution, and local wisdom [17][18].

This type of DSSC of natural dyes as a photosensitizer has been widely used in research on the fabrication and characterization of DSSC by dipping in titanium dioxide ($TiO_2$) and zinc oxide (ZnO). Zinc oxide (ZnO) has a similar bandgap to $TiO_2$ [19][20][21][22]. It has higher electron mobility which helps in the process of electron transportation. The natural dye as a photosensitizer is dragon fruit dye. In the previous research, the $TiO_2$ layer added with dragon fruit dye obtained a visible spectrometer absorption of around 535 nm at the annealing temperature of 450 $^0$C and resulted in an electrical efficiency of 0.22% [23]. The $TiO_2$ layer coupled with the dragon fruit dye showed absorption spectra between 450 nm to 700 nm with a peak value of 535 nm. It has shown types and pigments of invisible red spectra [24]. The black olive extracted has a higher photosensitization, broader absorption ability of sunlight to more energy in DSSC, and better charge transfer between the black olive extracted and $TiO_2$ surface [25][26]. A study on the fabrication of the prototype of Dye-Sensitized Solar Cells (DSSC) by dragon fruit and $TiO_2$ was conducted [27][28].

In this work, the preparation of DSSC used zinc oxide (ZnO) and dragon fruit extracted. The Dragon fruit dye is doped on a ZnO as a sensitizer DSSC. Dragon fruit is one of the tropic fruits that 30-35% part peels which contains anthocyanin and is red. ZnO has electronic properties such as an energy gap (Eg) and a conduction band which is widely used in DSSC [29]. ZnO has interesting properties including higher electron mobility than $TiO_2$. DSSC constructed with ZnO as a photoanode can absorb more dye, improve the photon utilization rate and provide a rapid collection for the photo excited carriers [30]. The performance of the DSSC depends on the sensitizer dye and wide bandgaps. Dye absorption is doped on the surface of thin films ($TiO_2$, ZnO, etc.), and is very important to determine the performance of the solar cells [31][32]. Dragon fruit dye contains high enough natural anthocyanin dyes. Anthocyanins are dyes that have a role to give red color and have the potential to become natural dyes for food and can be used as an alternative to synthetic dyes that are safer for solar cells. The effect of dragon fruit dye can be got environment-friendly, easy to obtain, and low in cost.

## 2 Research Methodology

### 2.1 Materials

The materials in this experiment are the conductive glass substrates, ITO (25 mm × 25 mm, 15 Ω/sq), zinc oxide (ZnO) powder (Sigma Aldrich), nitric acid solution, Triton X-100, dragon fruit dye extracted, iodine (I3, Sigma Aldrich) and carbon from the candle.

### 2.2 Characterizations

The bandgap of the dragon fruit dye and ZnO paste on ITO glass substrate as a sensitizer were tested using a UV spectrophotometer and Fourier Transform Infrared (FTIR) spectrophotometer. The absorption of UV spectra was recorded by UV spectrophotometer Shimadzu type UV 1800 and with a wavelength of 400 to 700 nm. Meanwhile, FTIR spectra were recorded on Shimadzu 8201 PC with 1 $cm^{-1}$ resolution with the wavenumber 400 to 4000 $cm^{-1}$. The sample was blended as a KBr pellet and scanned into a blank KBr pellet before measurement. The electrical properties of the dragon fruit dye and ZnO paste on ITO glass substrate as a sensitizer were tested using sunlight as the light source. The radiated sunlight was measured by the solar power meter.

### 2.3 Research Design

1. Preparation of the dragon fruit dye as a sensitizer.

The Dragon fruit was mixed into 500 ml of distillate water at room temperature and milled for 10 minutes until homogenous. The mixture is put into a glass beaker and covered with aluminum foil to prevent evaporation for 24 hours to get the dragon fruit dye. The dragon fruit dye extracted is evaporated for 48 hours. The result of the evaporation is purple color. Then, the dye was diluted using distilled water to obtain a pure Dragon fruit dye solution as in Fig. 1.

Fig. 1. Dragon fruit dye extracted.

The type of sample used is sample 1 which is dragon fruit dye purely and sample 2 is a mixture of dragon fruit dye with ZnO. Sample 2 was made with a ratio of 1:1 by dissolving 1 ml of ZnO into 1 ml of dragon fruit dye purely. It was tested using a UV-vis and FTIR spectrometer to determine the absorbance spectra and the functional groups of each sample. The solar power meter is used to measure the radiated and transmitted sunlight.

2. Preparation of the ZnO paste.

Powder of 0.2 g ZnO was blended with 0.4 ml nitric acid solution in a glass beaker. The solution blended was stirred using a magnetic stirrer for 15 minutes at a rate of rotation of 700 rpm and 80 °C until homogeneous. Then, it was added the Triton X-100 and covered with aluminum foil. The solution of ZnO and nitric acid can be coated on the side of the ITO conductive glass substrate using doctor blade technique as a thin film.

3. Preparation of the ITO conductive glass.

The ITO conductive glass as a substrate was cleaned using ethanol and dried using a hot plate for 5 minutes at a temperature of 100 °C to remove moisture.

4. Preparation of counter electrode.

Preparation of counter electrode use doctor blade technique as seen in Fig. 2. shows the ITO glass was coated as a carbon layer on the counter electrode by using a candle flame. Then the slide was heated on a hot plate at 150 °C for 15 minutes. For the carbon paste on the counter electrode, the carbon paste was prepared 0.1 g of carbon was added and mixed with ZnO pasta for 5 minutes.

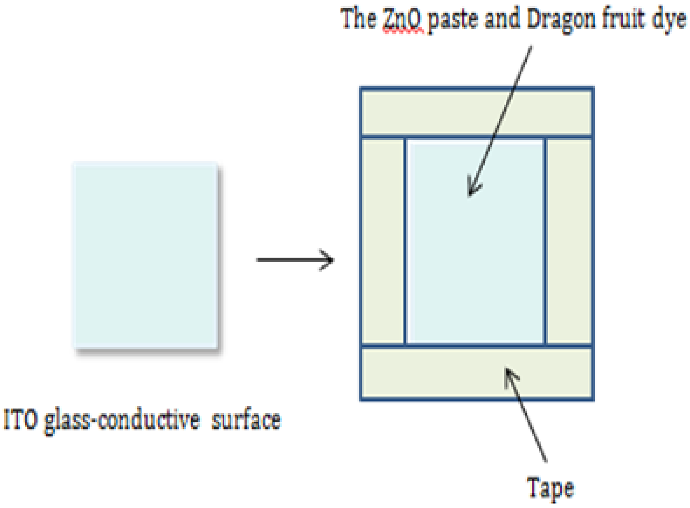

Fig. 2. ZnO paste and dragon fruit dye on ITO glass substrate using doctor blade technique.

The carbon paste was heated on a hot plate at the sintering temperature of 300 °C for 1 hour. Then, the deposition of the carbon paste doped on ITO glass was done by the doctor blade method. After the paste becomes dry, the scotch tape was removed from the ITO glass.

5. Preparation of the dragon fruit dye and ZnO paste on the ITO glass (thin film) as a working electrode.

ZnO paste on the ITO conductive glass substrate (thin film) is dropped by dragon fruit dye extracted as much as four times. Then, it is rotated using a spin coater until it the evenly dispersed. The samples are SL-1, SL-2, and SL-3, respectively. SL-1, SL-2, and SL-3 are one, two, and three drops of dragon fruit dye extracted. The pre-annealing thin film is being done using a hotplate at a temperature of 100 °C for 15 minutes. Pre-annealing is used to remove water vapor in a thin layer. Then, the heating (annealing) thin film is done using a furnace at a temperature of 450 °C for 30 minutes. The annealing of the thin film process serves to improve the crystal structure.

In brief, Fig. 3 depicts design of DSSC. The counter electrode of the ITO conductive glass substrate that has been layered with carbon from the candle. The working electrode is arranged on the counter electrode. Between the electrodes, some drops of electrolyte solution (iodine) are given. Then, both sides are clamped together using the binder clips. Sunlight enters the ITO glass and attacks dye molecules and is absorbed on the ZnO surface. Sunlight was a primary source of the energy and was used to produce electricity.

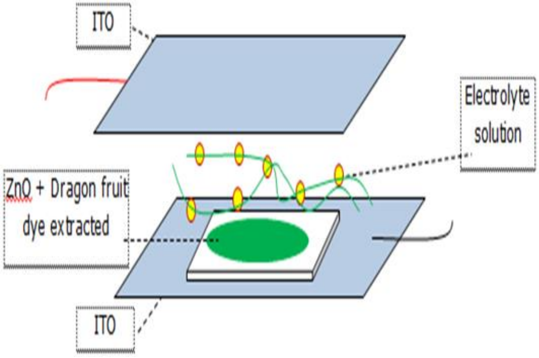

Fig. 3. Design of DSSC.

The photoanode of DSSC has two functions the support for sensitizer loading and the transporter of the photo-excited electron from the sensitizer to the external circuit. A large surface area is important to ensure high dye loading. The sample can be applied to the testing by an I-V meter as seen in Fig. 4.

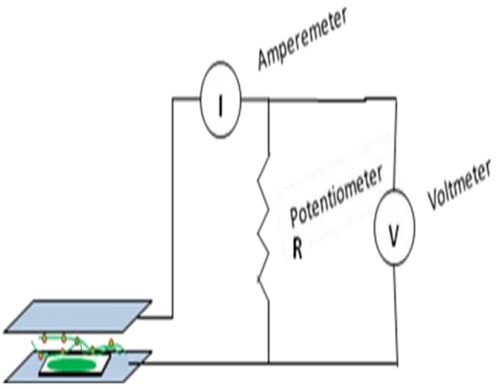

Fig. 4. Design the testing of DSSC using a solar power meter.

## 3 Results and Discussion

The absorption spectra of the dragon fruit dye extracted were obtained by UV-Vis spectrophotometer as shown in Fig. 3. Dragon fruit dye extracted can be absorbed in a wide range of visible light. The wavelength range is 400 nm to 700 nm. The dragon fruit dye has the highest peak absorption at 530 nm. Dragon fruit dye extracted can absorb light from 465 nm to 575 nm wavelength as shown in Fig. 5.

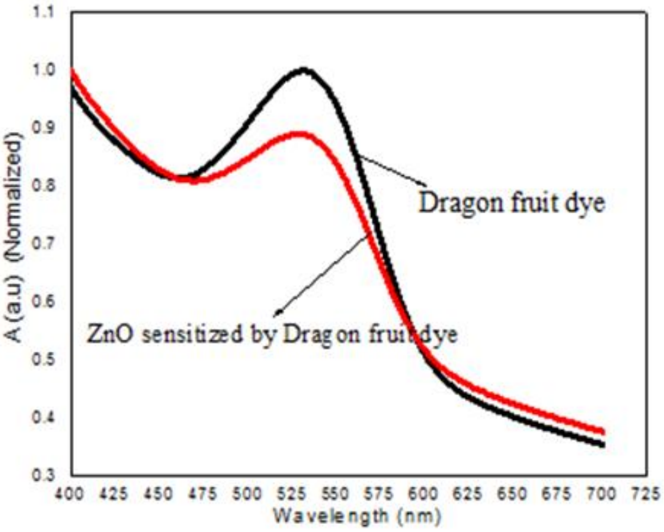

Fig. 5. Absorption spectra of dragon fruit dye and ZnO sensitized by dragon fruit dye.

The spectra of dragon fruit dye have an absorbance of 0.74 and transmittance of 9.01%, respectively. The dragon fruit shows a good absorption spectrum between 450 nm to 600 nm with a peak value of 535 nm. The ZnO thin film was sensitized by dragon fruit dye and has good absorption at a wavelength of 535 nm. It is a higher absorbance of dragon fruit dye purely. The decrease in absorption ability is due to the very short soaking time of 1 day. The longtime of immersion affects the number of dye molecules that fill the pores in the ZnO thin layer so that absorption capacity is high. A mixture of ZnO and dragon fruit dye can absorb the light because there are anthocyanin molecules on dragon fruit dye adsorbed on the surface of ZnO particles.

Two types of optical properties such as direct and indirect are observed in the optical bandgap and are determined by optical absorption spectra. The optical absorption coefficient, α(λ) for photon energy is measured by [33], where A is the absorbance spectra and d is the thickness of the thin film (m).

$$\alpha(\lambda) = \frac{2.303\ A}{d} \tag{1}$$

The optical absorption coefficient α(λ) as a function of photon energy hν can be used by the Davis and Mott equation which is the optical energy bandgap that determines the type of optical properties [eq.2], where B is the energy-independent constant, $E_{opt}$ is the optical energy bandgap, hν is photon energy and m is a constant.

$$\alpha(h\nu) = \frac{B\left(h\nu - E_{opt}\right)^m}{h\nu} \tag{2}$$

The result of the energy bandgap of dragon fruit dye and ZnO sensitized by dragon fruit dye can be seen in Fig. 6. It also depicts the energy band gap of dragon fruit dye and ZnO sensitized by dragon fruit dye is 2.09 eV and 2.56 eV. The effect of the dragon fruit's dye-doped on ZnO thin can increase the energy bandgap by around 18.4%. The results of FTIR spectra can be shown in Fig. 7.

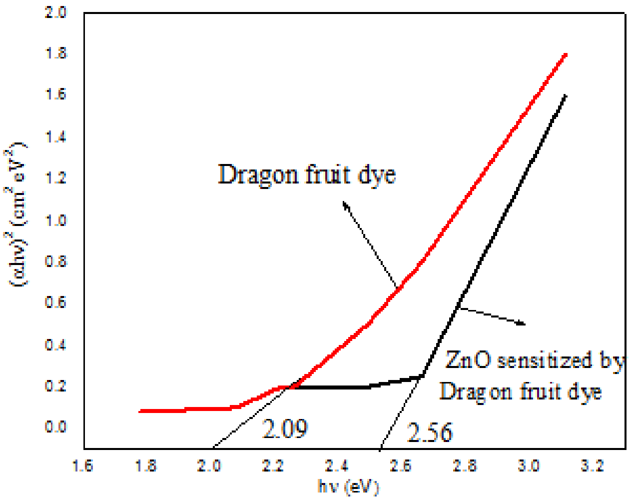

Fig. 6. The energy gap band of dragon fruit dye and ZnO sensitized by dragon fruit dye.

Fig. 7 shows that the result of the FTIR spectra. The board and sharp absorption wavenumbers at 3200-3400 cm$^{-1}$ and 1600–1700 cm$^{-1}$. Two broad bands at 3423 and 3448 cm$^{-1}$ are for an H-bonded, and vibration of the free hydroxyl group. The C=O stretching vibration is at 1716 cm$^{-1}$ for the dragon fruit dye compound. The C-O stretching is at 1407 cm$^{-1}$. The sharp peak shows the wavenumber range at 1030–1060 cm$^{-1}$ is C-O-C stretching vibration. It is esters acetates. The effect of anthocyanin on DSSC can be used in a photoelectric conversion. In the future, the research about the dragon fruit dye extracted will be fulfilled about the C-H bond which is derived from organic compound impurities trapped in the cavities of the ZnO thin layer.

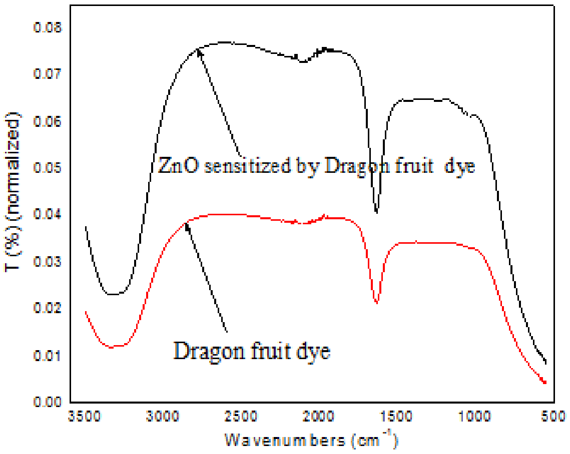

Fig. 7. FTIR spectra of the dragon fruit dye and ZnO sensitized by dragon fruit dye.

The electric current in the DSSC circuit is the bonding of the anthocyanin dye molecule with ZnO. Anthocyanin dye molecules that bind to ZnO in the presence of light cause the excitation of electrons to the ZnO semiconductor conduction band through the glass conducting. These electrons cause an electric current. The bond that occurs in the anthocyanin dye with ZnO can occur through a chelate bond. This bond can occur between the hydroxyl groups of anthocyanin.

A solar power meter is a device that can measure the radiated and transmitted sunlight. The measurement of the I-V was obtained to determine the energy performance of solar cells (DDSC) with the resulting output parameters, namely maximum voltage ($V_{max}$), maximum current ($I_{max}$), maximum power ($P_{max}$) and solar cell efficiency (ɳ) can be seen in Fig. 4. The conversion efficiency of solar cells is calculated by eq. 3.

$$\eta = \frac{P_{maksimum}}{P_{cahaya}} \times 100\% \quad (3)$$

The results of the solar power meter can be seen in Table 1. I-V measurement with the intensity of the sun and the ambient temperature at the time of measurement is 129 lux and 30 °C. It shows that the best efficiency is obtained from the SL-3 layer, namely 0.03%.

Table 1. I-V Measurement of solar power meter.

| Model | $V_{oc}$ (mV) | $I_{sc}$ (μA) | $P_{max}$ (mwatt) | ɳ (%) |
|---|---|---|---|---|
| SL-1 | 0,4 | 4,9 | 1,96x10$^{-9}$ | 0,002 |
| SL-2 | 2,5 | 6,5 | 16,3x10$^{-9}$ | 0,02 |
| SL-3 | 2,44 | 13 | 31,7x10$^{-9}$ | 0,03 |

It can be seen the resulting current is higher than the SL-1, and SL- 2 layers. The thickness of the SL-3 layer can absorb the light from the sun. The performances of the dye absorption spectrum on the surface area are important aspects to determine the efficiency of the solar cell. The efficiency value obtained in SL-1, SL-2, and SL-3 is still below the efficiency value of solar cells with silicon-based materials [34][2]. The result of the current and voltage DSSC from dragon fruit dye extracted coated ZnO obtained, indicated that the energy bandgap could be optimized to make a wide bandgap. ZnO thin film needs the dragon fruit dye to decrease the surface and move electrons in the bandgap.

The ZnO paste and dragon fruit dye on ITO conductive glass substrate as a thin film show that the thickness of the thin film will affect the electrical properties. The thickness of the thin film will increase, so the current value will increase continuously. It is because of the electrical properties of the thin film that have changed from conductor to semiconductor and the crystallinity of grain boundaries.

## 4 Conclusion

The ZnO paste and dragon fruit dye on ITO conductive glass substrate as a thin film have been fabricated and tested in the DCCS as a sensitizer was done. Result of UV-Vis absorbance, dragon fruit dye has the highest absorbance rate with a peak of 530 nm and bandgap of 2.09 eV, while the ZnO sensitized by dragon fruit dye has the highest absorbance rate with a peak of 535 nm and bandgap of 2.56 eV. The energy bandgap can be optimized to make a wide bandgap. The FTIR spectra show the presence of the dragon fruit dye molecules attached to the surface of the ZnO layer. The best efficiency of the result solar power meters from the third sample (SL-3) showed the more dye that is coated on a thin film of ZnO, so higher the value of the current generated. Dragon fruit extracted dye can absorb the light so that it can be used to make DSSC as a working electrode. ZnO paste is suggested to improve the surface and move electrons in the band gap between the thin film and dye. It can be concluded that the natural dye of the dragon fruit dye extract can be applied as a sensitizer.

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## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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