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Combination of Dragon Fruit, Hibiscus and Bitterleaf as Dye Sensitizer to Increase Efficiency of DSSC

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***Abstract*— Dye-sensitized Solar Cell (DSSC) from natural products was being prominently developed in recent years. The main advantage of DSSC was its fabrication which did not require high process technology, so the production cost was lower than solar cells from crystalline materials. There were important materials in Dye-sensitized Solar Cell technology, such as Zinc Oxide as an electrode, electrolyte solution and plant pigments as dye sensitizer to absorb light energy and convert it into a source of electrical energy. In this study, the combination of multiple dye mixture had been investigated to achieve high external quantum efficiency as sensitizers. Furthermore, each sample would be tested to determine the characteristics of natural dye and to calculate the power flow and efficiency of the electricity generated by each variable of the dye solution. There would be four variables in this study, which was the composition (% v/v) of the three main sources of natural dyes; hibiscus, bitter leaf and dragon fruit, respectively first (50% : 25% :25%), second (25% : 50% : 25%), third (25% : 25% : 50%) and fourth (33,33% : 33,33% : 33,33%). The result of the study showed that efficiency of variable 1 to 4 was 1.9076%, 2.8739%, 0.8566% and 0.8344%, respectively.**

***Keywords*— dye-sensitized solar cell; efficiency; natural dye**

I. INTRODUCTION

The energy crisis is a major problem for every country in the world. Fossil fuel is still used widely because of its relatively low price, ease of the usage and availability. However since the reserves are become decreasing, alternative energy resources especially which are eco-friendly and inexpensive are urged to be explored and utilized. One of the greens and inexpensive energy sources is the sun. Every hour the sunshine hits the earth giving energy more than enough to satisfy global energy needs for an entire year. Solar energy is the technology used to harness the sun's energy and make it useable. A solar cell (SC) is one of the several ways to convert solar energy into electrical energy. Silicon-based SCs have been mass-produced and applied in many applications with quite high efficiency of 15-25%. However, they have some drawbacks including high cost and requirement of a wide area to place the solar panel.

Dye-sensitized solar cell (DSSC) has been studied extensively on accounts of their attractive advantages, such as low cost, less toxic manufacturing, easy scale-up, light weight, and potential use of flexible panels compared to conventional silicon-based solar cells. Since the first time discovered by Professor Michael Grätzel in 1991, DSSC has become one of the research topics which is conducted by researchers around the world intensively. DSSC is also known as the first breakthrough in solar cell technology after silicon solar cells. In contrast to conventional solar cells, DSSC is a photoelectrochemical solar cell which uses an electrolyte as charge transport medium. DSSC consists of a sensitizing dye (photosensitizer) which acts as an electron donor raised when it absorbs light, transparent conducting oxide-coated glass as the substrate for the photoelectrode, nanocrystalline semiconductor film photoelectrode usually consists of porous Zinc Oxide (ZnO) or Titanium Dioxide (TiO_2) which acts as electron acceptor transferred from the oxidized dye, redox electrolyte contains I^-/I^{3-} (iodide/tri-iodide) redox ions which mediate electrons between photoelectrode and counter electrode thus producing cycle process in cell, and counter electrode with high electrocatalytic activity such as platinum or carbon as shown in Fig. 1 [1], [2].

The materials exist in nature are fruits, flowers, leaves, bacteria, and algae have various color and contain several pigments that can be easily extracted and used in DSSC. The advantage of using natural dyes as photosensitizers in DSSC is because they have a large absorption coefficient in the zone of visible light (visible region), ease of materials preparation and environmentally friendly. The most important point in the synthesis of natural dyes is cost-effective because it does not involve precious metals such as Ruthenium. These plant pigments show the electronic structure that interacts with sunlight and changes the wavelength which transmitted or reflected by both the plant tissue where each pigment has a maximum absorbance wavelength (λ_{max}). Natural pigment including chlorophyll, carotenoids, flavonoids, and anthocyanins could easily be extracted from natural products when compared to synthetic dyes.

When a dye molecule absorbs light, it leads to the excitation of electrons to an electronically excited state. The excited dye molecule injects an electron into the conducting band of the ZnO film. ZnO is commonly used because of inert, harmless, inexpensive price, have the nature of the good optical characteristics. ZnO works in visible light because it has a wide band gap, which is when the visible light on semiconductor material there will be electrons releasing. In the form of thin layers, this oxide material is transparent to the light because of its appropriate band gap. The crystal structure and grain size of the particles on the ZnO thin film greatly affect the optical properties and electric. Basically, the orientation of the nanocrystals which form a thin film is very dependent on the type of substrate. [3]. Then, the oxidized dye is restored by electron donation from the reducing ions in the electrolyte, usually an organic solvent containing a redox system. The donated electron is in turn regenerated by the reduction of conjugated ions in electrolytes. The circuit is competed by electron migration through an external load thus producing electricity as shown in Fig. 2 [4].

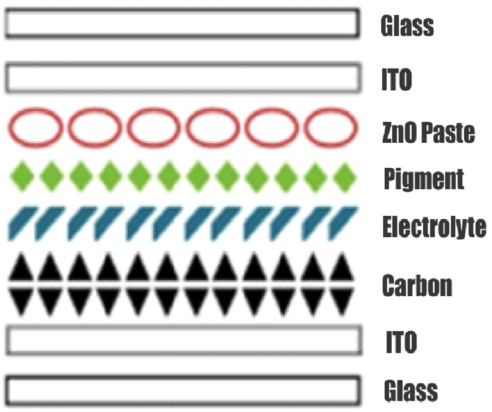


Fig. 1 Structure of DSSC

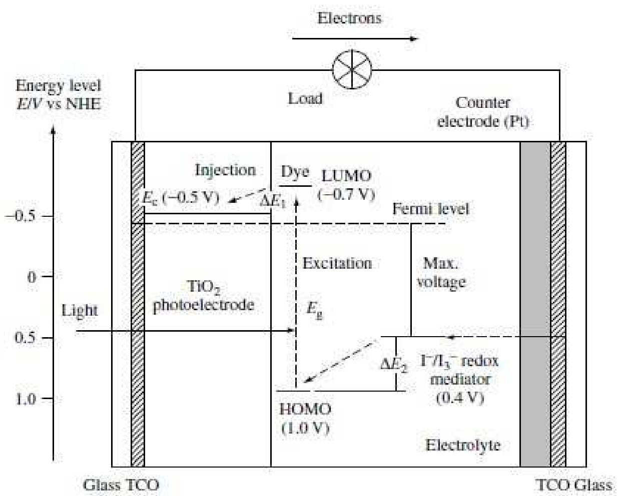


Fig. 2 Schematic energy diagram and operating principle of DSSC

Several previous studies have shown that the color (dye) has an important role in the absorption of light energy which also affects the power output of the solar cell. The efficiency of these type of solar cell which using constructed dye from organic material recently had reached only 2%. In the previous research, this method had been applied by utilizing pigment from various plants. For obtaining high efficiency in solar cells, dye elections need to consider several factors. Among them, the chemical structure of the dye must have small steric obstacle so that the maximum electron transfer process and dye extraction temperature also have a great impact on the photoelectric conversion efficiency of solar cells. Each plant body parts have its own pigment which can be used to construct a dye for DSSC. From the previous research, it has been found that the efficiency of DSSC can be improved by combining two or more multiple dyes [4], [5]. In this study, several plants which are hibiscus, bitter leaf and dragon fruit will be utilized as organic pigments in a combination of multiple dye mixture to increase the efficiency of DSSC. The material has been chosen because of its high efficiency in single dye and its plentiful resources in our hometown Balikpapan, Indonesia.

II. Materials and Methods

The plants (bitterleaf, hibiscus, and dragon fruit) as organic dyes were gathered in nearby places in Balikpapan. Indium Tin Oxide Glass acted as substrate. Zinc Oxide (ZnO) acted as an electrode (photoanode). Ethanol with 96% purity was used to extract the pigments from the plant. A mixture of Iodine 0.05 M and 0.5 M KI solution was made as an electrolyte solution. Carbon was used is graphite from candle soot as the counter electrode.

Conductive glass for the electrode is made from soda limestone glass which has been coated with a transparent conductive oxide material of fluorine-doped tin (II) oxide. The ITO glass has a resistivity of 18-24 ohms per cm square with a coating thickness of 8 Angstroms. The conductive oxide is coated on one side of the glass using chemical vapor coating techniques or chemical vapor deposition (CVD). The conductive glass substrate acts as a current collector as well as the supporting structure of the cell and a barrier layer between the cell with the open-air [6].

A. *Extraction of Hibiscus, Dragon Fruit, and BitterLeaf Dye*

Extraction hibiscus, dragon fruit, and bitterleaf were carried out by maceration method. The advantage of maceration technique is a simple process although it requires time tends to be much longer [7]. Hibiscus and dragon fruit were stripped from its skin, while the bitterleaf is picked from its plant. Then, the materials were mashed with mortar and pestle. By using Ethanol with 96% purity, the materials then immersed for 24 hours in the ratio of 1 : 5 volume to extract its pigments.

The solution then filtered and kept in sample bottles as shown in Fig. 3. Selection of the ratio was due to the greater amount of solvent. Contact between the solvent with the solid material to be perfect, so that the pigment is dissolved more. Ethanol used as a solvent because it has an ethyl group which is non-polar and polar hydroxy to dissolving pigments.

Fig. 3 The filtered solution in sample bottles

B. *Fabrication of Photoanode ZnO Paste*

ZnO paste was made by mixing 1 gram of ZnO powder with 2 ml of dye solutions which were extracted earlier. Then it is stirred until it forms a suspension with a soft texture and perfectly coated by coloring dye as shown in Fig. 4.

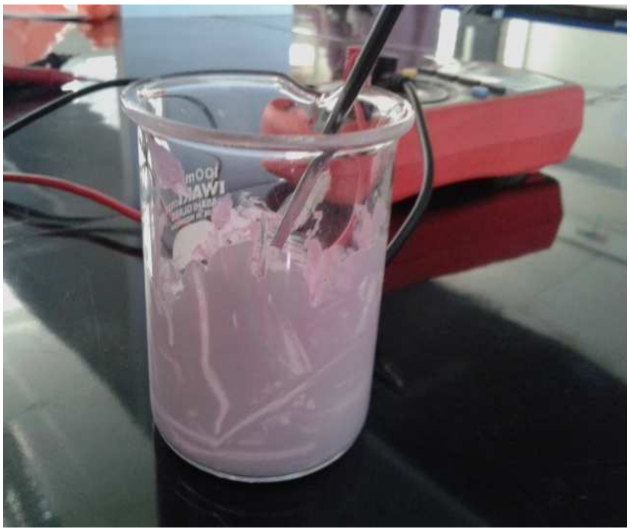
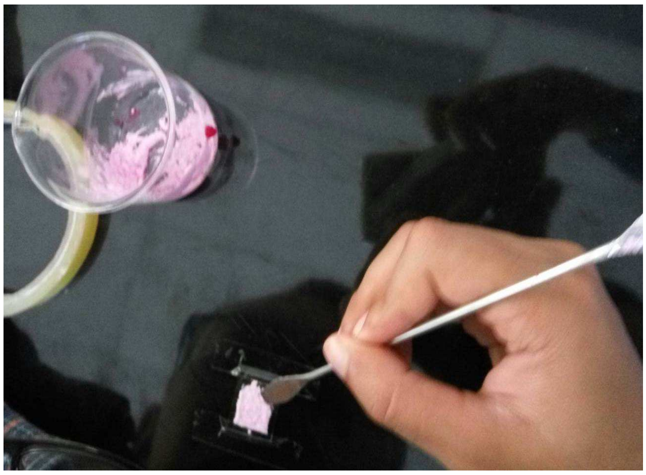


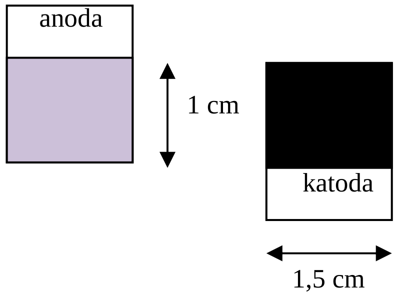
Fig. 4 ZnO paste

C. *Deposition of ZnO Paste*

ITO glass was prepared and washed with 70% alcohol. ZnO paste was deposed in the conductive site of the glass by using doctor blade's method with the help of stirring bar to flatten pasta. On the ITO glass with an area of 1.5 x 1.5 cm square is molded areas for ZnO deposition with the area of 1 x 1.5 cm square above the conductive surface as shown in Fig. 5(a) and 5(b).



(a)



(b)

Fig. 5 (a) ZnO paste deposition, (b) ITO glass construction as photoanode and cathode

The formed substrate layer then dried with a hair dryer within 15 minutes. If the dye color intensity was reduced during the drying, then 2 drops of dye are added to the surface of the active area of the solar cell, and then ZnO paste is dried again for 5 minutes.

D. *Fabrication of Carbon Counter Electrode*

The function of the carbon is a catalyst to accelerate the reaction in the DSSC. Carbon coated ITO glass were used as comparison electrode which was sintered from the wax candle as shown in Fig. 6.

Fig. 6 Carbon electrode from wax candle sintering

E. Fabrication of DSSC Sandwich

DSSC's sandwich consists of substrate's glass which covered with ZnO sensitized dye as shown in Fig. 7. The electrode was etched by two drops of electrolyte solution then covered with carbon coated glass face to face and finally clamped on its both side.

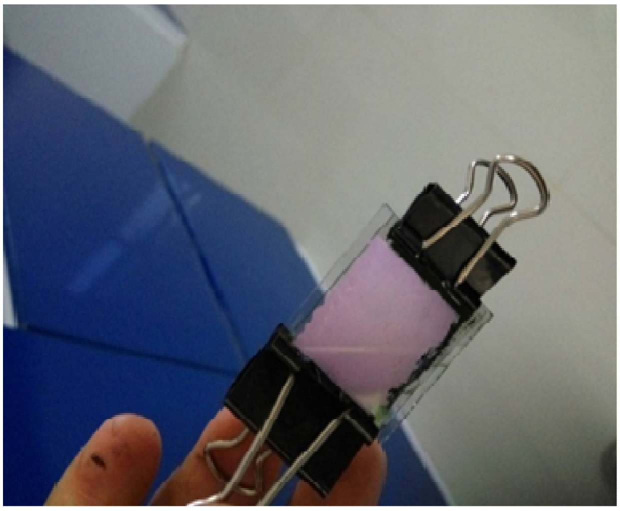


Fig. 7 Prototype of DSSC

F. Dye Characteristic Measurement

Absorption profiles of the pigments (anthocyanin and chlorophyll) were analyzed using UV-VIS spectrophotometer as shown in Fig. 8. The dye solution will absorb the sunlight spectrum at a wavelength of 400-700 nm.

Fig. 8 Dye characteristic measurement using UV-VIS Spectrophotometer

G. DSSC's Performance Measurement

Dye Sensitized Solar Cell's performance was carried out based on direct exposure to a source of light. The measurement test was carried out when the sun reaches the highest intensity. It has been reported that the intensity of the sun highest in Indonesia occurred at 11:00 to 13:00 thus the data collection was done in the time range [8]. DSSC was placed parallel at an angle of 180° since the sun will reach the highest position at that time. In this work, sunlight was used as a light source and measured its intensity with a light meter as shown in Fig. 9.

Fig. 9 DSSC's performance measurement

III. RESULTS AND DISCUSSION

A. Dye Characteristics Analysis

Radiant energy consists of a large number of electromagnetic waves with a different wavelength. The visible light spectrum is only a small part of the entire electromagnetic radiation consists of color components where each color has a different wavelength as shown in Fig. 10.

Pigments are substances which change the color of visible light as a result of the selective absorption process the wavelength in a certain range. Pigment molecule absorbs energy at a wavelength thus reflect certain other visible wavelengths.

Fig. 10 Absorbance and complimentary color

In Dye-sensitized Solar Cell (DSSC), an organic pigment is an important component in the conversion of sunlight energy into electricity, so it is necessary to determine its characteristics for an optimal pigment which will be used in the fabrication of DSSC. Before utilized as a sensitizer, dye solution was first characterized using UV-VIS spectrophotometry. This characterization was conducted to determine the absorption of light by each pigment contained in the raw materials. Normally sunlight scatters 7% of ultraviolet ray, 47% of visible ray, and 46% of infrared ray [9].

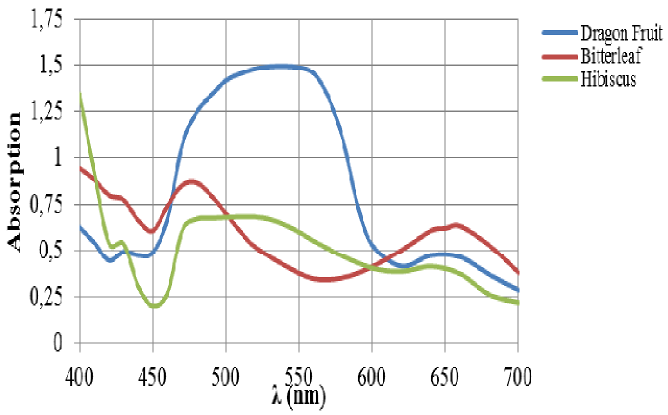


Fig. 11 Absorption of single dye with UV-Vis Spectrophotometer

Dye absorbance analysis was conducted to determine the characteristics of the dye in the form of single dye and multiple dyes (mixture) based on the selected variables on their ability to capture visible light. The absorbance spectrum of a colored sample can be measured in the wavelength range of 400-700 nm. Results of the dye absorption rate graphs are shown in Fig. 11. Based on these results, each single dye contained pigments which have the various capability to absorb energy based on a certain range of wavelength. In dragon fruit's dye, it has been obtained that a fairly wide of absorption spectrum occurred at 480-570 nm wavelength with its maximum wavelength (λ max) was 540 nm. The same results of the experiment had been conducted by obtaining maximum absorption at λ = 533 nm. While in hibiscus's dye has been obtained that maximum wavelength (λ_{max}) occurred at 510 nm. It showed that dragon fruit and hibiscus have anthocyanin pigment to absorb light energy. Anthocyanin pigments that contained in the dragon fruit have a complementary color red to reddish purple absorbs green to yellowish green [10], [11].

In bitterleaf's dye, it has been obtained two dominant ranges of light absorption spectrum which are 460-490 nm with λmax = 480 nm and at λ = 640-670 nm range with λmax = 660 nm. Bitterleaf itself contained chlorophyll and carotenoid pigments. In higher plants, there are two kinds of chlorophyll, which are dark green (chlorophyll-a) and light green (chlorophyll-b). These types of chlorophyll have two regions of wavelength absorption, which are at 400 nm - 490 nm and at 620 nm - 680 nm [12]. At λmax = 480 nm indicates the presence of carotenoid mixtures which contained the bitter leaf. This energy absorption mechanism was also utilized in the process of transduction energy in solar cells [13].

Next, analysis of multiple dyes mixture (anthocyanin and chlorophyll) was carried out furthermore with several composition variables. Results of the dye absorption rate graphs are shown in Fig. 12. Based on these results, a combination of three pigments will expand the range of light absorption in the visible light range, and its range is wider than the mixture of two pigments or a single dye. This is caused by the ability of each pigment which contained the substance is capable of absorbing light in accordance its own wavelength region. There is three maximum absorbance peak [9]. The influence of dye mixture will be determined furthermore in the measurement of its performance as DSSC.

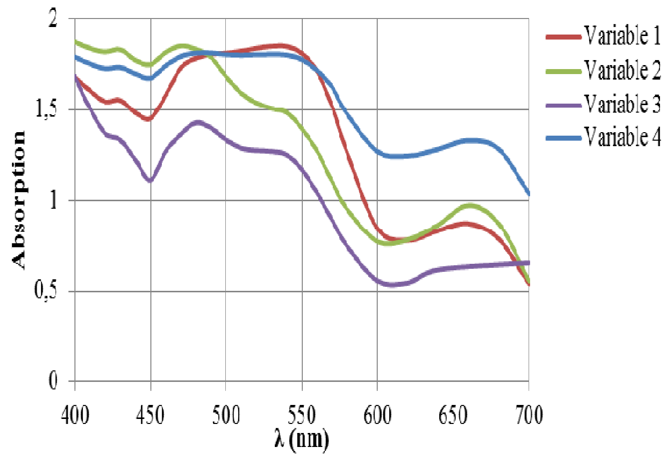
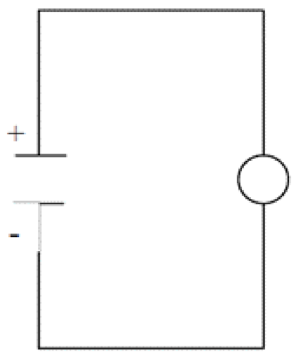


Fig. 12 Absorption of multiple dyes with UV-Vis Spectrophotometer

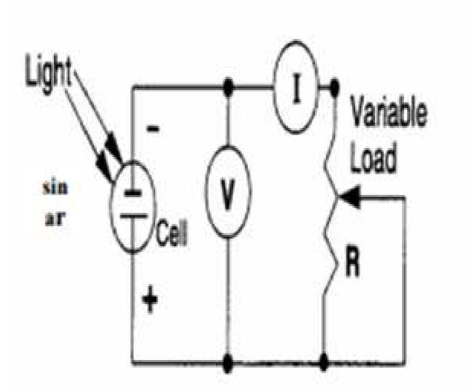
B. Measurement of Current, Voltage, and Efficiency of DSSC

This work was carried out to determine the dye mixture influences in DSSC. Current and voltage were measured under the illumination of direct sunlight (outdoor). DSSC output parameters include of open circuit voltage (open circuit), V_{oc} and the short circuit current (short circuit), I_{sc}. Measurement was performed with a multimeter with a circuit as shown in Fig. 12(a) and Fig. 12(b).

While Fill Factor (FF) is the ratio between the maximum power DSSC, when given resistance variation, with the multiplication of V_{OC} and I_{SC}. The maximum power is the result of multiplying the current and voltage of DSSC voltage when the resistance variation changed (forming I-V curve).



(a)



(b)

Fig. 12 (a) Open circuit, (b) Closed circuit

Current and voltage obtained are symbolized by I_m and V_m.

Fill Factor equation is given by [11] :

$$FF = \frac{\mathrm{Im}\, xVm}{IscxVoc} \quad (1)$$

While efficiency (η) of light conversion to electrical energy can be determined by the following equation :

$$\eta = \frac{IscxVocxFF}{IGxA} \quad (2)$$

In the measurement performance test, voltage arose caused by the difference of energy conduction in ZnO as semiconductor electrode and the potential of the electrolyte solution. The current density raised was influenced by the intensity of the light source which will determine the number of photons absorbed by the dye in its conversion process [15]. The measurement results are shown in the DSSC performance in Table 1.

According to Table 1. it can be seen that the efficiency of the single dye-sensitized solar cell of dragon fruit, bitterleaf and hibiscus each by 0.8112%, 0.7443%, and 0.4765%. Dragon fruit's dye has the highest efficiency compared bitter leaf and hibiscus. This is due to the voltage generated from DSSC dragon fruit has great value. While bitterleaf DSSC has strong currents greater than the dragon fruit and hibiscus because the level of excitation energy from chlorophyll is better than anthocyanin pigments [1]. The difference in the efficiency of solar cells occurred because of the differences of dye molecules in energy transfer mechanism.

TABLE I
EXPERIMENT RESULT FOR CURRENT DENSITY, VOLTAGE AND EFFICIENCY OF DSSC

Variables	1st Circuit		2nd Circuit
	Voc (mV)	Isc (μA)	η (%)
Dragon Fruit (N)	437,4	75,3	0,8112
Bitterleaf (S)	397,7	67,7	0,7443
Hibiscus (H)	441,2	65,3	0,4765
Variable 1 (50%H: 25%S: 25% N)	516	154,4	1,9076
Variable 2 (25%H: 50%S: 25% N)	531	216,3	2,8739
Variable 3 (25%H: 25%S: 50% N)	441	239,4	0,8566
Variable 4 (33%H: 33%S: 33% N)	466	84,5	0,8344

*)**Notes** : N is Dragon fruit's dye ; S is bitterleaf's dye and H is Hibiscus's dye

Based on this test, the highest efficiency was obtained from Variable 2 (25%H: 50%S: 25%N). It has been obtained that chlorophyll composition from bitterleaf has a strong influence in improving the efficiency. Multiple dyes mixture effect on increasing the electron transfer mechanism of each dye to the semiconductor electrode and dissociation process mechanism photon absorbed by the dye so that the resulting output current density becomes larger [4].

In general, there are two methods of dye molecules adsorption to the surface of the ZnO paste. The first method is deposited ZnO paste to conductive glass area. Next, the glass which has been coated will be sintered in a furnace at a temperature of 350 °C for 30 minutes. Afterward, the glass is immersed in a dye solution for 24 hours. The second method is by mixing the dye directly into the ZnO paste. The previous researchers have reported that the efficiency of DSSC with the technique of mixing ZnO with red ginger extract dye having an efficiency of 0.78%, while the ZnO is soaked into dye red ginger extract has only 0.002% efficiency [16]. It means that the efficiency of DSSC with the second method yield the higher efficiency. This is what underlies the selection of both methods in this study to obtain the optimum value of efficiency. The advantage of this technique is the dye molecules can come into contact with the ZnO structure thereby the suspension paste has a lighter color and shorter time spent and does not require a long time immersion [11]. Related research of DSSC fabrication of dragon fruit, bitterleaf, and hibiscus using a blending technique has not been done before.

DSSC efficiency is affected by five main factors, namely dye, electrodes, cell thickness, surface area, the contact surface between ZnO and dye's electrolyte, and type of electrolyte. ZnO photoanode thickness will affect the capacity of storing dye, the thicker the layer means more dye can be stored. However, the layer thickness will increase the drop of transfer speed; it will affect the strength of the current density (Isc) and voltage (Voc) produced. The value of Isc and Voc of all variables fluctuate due to differences in photoanode thickness which were not included in these work variables. The Wider contact surface between the dye-electrolyte with ZnO will increase the intensity of excited electron and whole transfer from dye to the electrolyte. Nanopore particles are the key factor to obtain effective wide contact area for DSSC. Transfer rate and recombination must be balanced because various factors can influence i.e. different potential between two electrodes, the morphology of ZnO, and the conductivity of the electrolyte [17].

IV. CONCLUSION

Organic pigments from dragon fruit, bitter leaf and hibiscus can be used as a dye sensitizer in DSSC fabrication. Anthocyanin pigments from the dragon fruit reach maximum absorption wavelength (λmax) at 540 nm, the pigment chlorophyll from the bitter leaf at 480 nm and 660 nm, while the pigment hibiscus at 510 nm. The combination of the dyes will expand the range of absorption wavelengths thus increase the excitation electron energy for photoelectric conversion. Multiple dyes DSSC has a higher efficiency than single dye. The combination of pigments from the third substrate capable of increasing current density generated by photon dissociation of dye molecules, so it has been increased DSSC's efficiency. The highest efficiency of DSSC was achieved in Variable 2 (25%H: 50%S: 25% N) with chlorophyll pigment was dominant than other.

NOMENCLATURE

A	active surface area of DSSC	cm^2
FF	fill Factor	
I_G	sunlight global intensity in certain air mass	mW/cm^2
I_m	maximum current of DSSC	mA/cm^2
I_{SC}	short current of DSSC	mA/cm^2
V_m	maximum voltage of DSSC	V
V_{OC}	open voltage of DSSC	V
η	efficiency of DSSC	%

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