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Performance improvement of dye-sensitized solar cells by using natural chlorophyll and anthocyanin dyes

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ABSTRACT

Natural dye-sensitized solar cells (DSSC) have gained so much attention in recent years due to its low-cost fabrication process, ease of fabrication, and environmentally friendly. In order to improve the DSSC performance, the absorbance spectral of dyes must reach the maximum visible spectrum values. The combination of two dyes with different absorbance spectra can be utilized to expand the absorbance spectral. Here, we demonstrated the combination of natural chlorophyll and anthocyanin dyes from cassava leaves and black sticky rice, respectively, to enhance the DSSC performance. Our findings provide insights for increasing the DSSC performance by varying the combination of natural dyes. The highest efficiency was obtained from Chlorophyll:Anthocyanin 3:1.

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1. INTRODUCTION

Energy consumption in the world continues to increase every year, as well as in Indonesia. It is recorded that in 2000 to 2014, energy consumption in Indonesia increased by 65% and is expected to continuously increasing up to 80% by 2030 [1]. Being an equatorial region allows Indonesia to obtain optimal sunlight throughout the year. The sun capable to generate energy up to 532.6 GW [2]. Solar cells are leading innovation to renewable energy considering the recent advances in improving efficiencies [3]. However, the most commercial solar cells are considerably expensive regarding its initial cost [4].

Dye-sensitized solar cells (DSSC) is a third-generation solar cells that regarded as the most promising photovoltaic devices [5]. DSSC has been widely studied because of its characteristics that mimic photosynthesis which can convert light into electrical power, and produce robust solar conversion efficience [6]. Moreover, low cost, non-toxic, renewable, and abundant in nature become other factors that make DSSC increasingly developed [7], [8]. Basically, the component of DSSC consists of a working electrode, dye sensitizer, electrolyte, and counter electrode [9]. Among these components, a dye sensitizer plays a key role in determining the amount of light absorbed which then affects DSSC performances [10]. The overall performance of DSSC is reliant on the light absorption capability of the dye sensitizer, the diffusion of the ejected electron through the mesoporous TiO_2 film, relative rates of the dye regeneration and several deleterious interfacial charge recombination reactions [11], [12].

Traditionally, DSSC utilized inorganic materials such as Ruthenium complex dyes (N719), organic dyes such as porphyrin, as a photosensitizer in DSSC [1], [13]. Even though the performance of DSSC obtained from inorganic-organic materials are high (11-13%), due to high cost and quite difficulty to obtain this material, some researchers have switched to using natural dyes [13]–[17]. Nowadays, solar energy conversion relies on devices with natural materials due to its environmentally friendly aspect. Furthermore, by utilizing natural materials, it does not produce hazardous byproducts. Thus, many researchers investigate natural materials such as natural dye that can be utilized from plants, flowers, fruits, and protein crystals [18]–[23].

To improve the efficiency of DSSC, the optical properties of natural dye must be tuned to have maximum absorbance in the ultraviolet to visible light spectra. By extending the absorbance spectral, the electrons in the dye can absorb more energy to get excited. Thus, electrons in the ground state would be easier to get excited into higher energy levels and the reduction-oxidation reaction in DSSC happens. In the previous study, some researchers successfully demonstrated the combination of dye which can enhance the DSSC performance [20], [24].

In this work, we have successfully demonstrated the effect of chlorophyll and anthocyanin variation on DSSC performances. Chlorophyll and anthocyanin were obtained from cassava leaves and black sticky rice, respectively. Cassava leaves and black sticky rice were utilized because both plants are typical plants from Indonesia which means easy to find and extremely cheap. Cassava leaves and black sticky rice were used as a dye that has different optical properties. The variations of mixing these two pigments aim to maximize the absorbance of photon energy in the visible wavelength.

2. MATERIALS AND METHODS

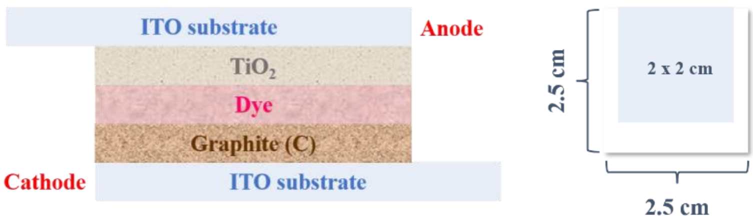
2.1. Materials

Titanium oxide (TiO_2), indium tin oxide (ITO) glass, and polyvinyl alcohol (PVA) were purchased from Sigma-Aldrich. Acetic Acid was purchased from Sari Kimia Raya. Ethanol Pro Analysis and Aquades were purchased from Makmur Sejati. Cassava leaves and black sticky rice were purchased from the traditional market.

2.2. Preparation of substrate

The structure of DSSC and the design of ITO glass were shown in Figure 1. The structure of DSSC was constructed using a sandwich structure which consists of substrates, TiO_2 paste, dye, electrolyte, and counter electrode, as shown in Figure 1(a). For a single DSSC cell, two substrates are required, the working electrode (anode) and the counter electrode (cathode).

The substrate used in this research was ITO glass which had a resistivity value of 15-25 ohm/sq. The ITO glass size was 2.5×2.5 cm^2, and the work area was 2×2 cm^2. To create the work area on ITO glass, the scotch tape was attached to cover the non-coated conductive parts. As shown in Figure 1(b), the blue part is the work area and the white part is the area covered by scotch tape.



(a) (b)

Figure 1. Preparation of DSSC substrate (a) The structure of DSSC and (b) the design of ITO glass

2.3. Preparation of dye solution

Preparation of dye solution is shown in Figure 2. Boneless cassava leaves that have been cleaned were weighed 20 g and mashed until soft using mortar. Then, the mashed cassava leaves were soaked in 50 mL ethanol solvent and stored in the dark bottle. Afterward, the solution was stirred using a magnetic stirrer for 30 minutes, as shown in Figure 2(a). Black sticky rice that has been cleaned was weighed 20 g and mashed using mortar. Then, 42 mL of ethanol, 5.6 mL of acetic acid, and 22.4 mL of aquades were mixed with the mashed black sticky rice until homogenous and stored in the dark bottle. Afterward, the solution was stirred using a magnetic stirrer at 60 °C for 60 minutes, as shown in Figure 2(b).

After the mixing solution was stirred, chlorophyll and anthocyanin extracts were stored for 24 h to maximize the extraction process. Afterward, chlorophyll and anthocyanin extract were filtered with filter paper, then keep in the dark bottle. In order to get 5 samples with different variations of dye, the extraction of the chlorophyll and anthocyanin dye is combined with the ratio of 1:3, 1:1, and 3:1, as shown in Figure 2(c).

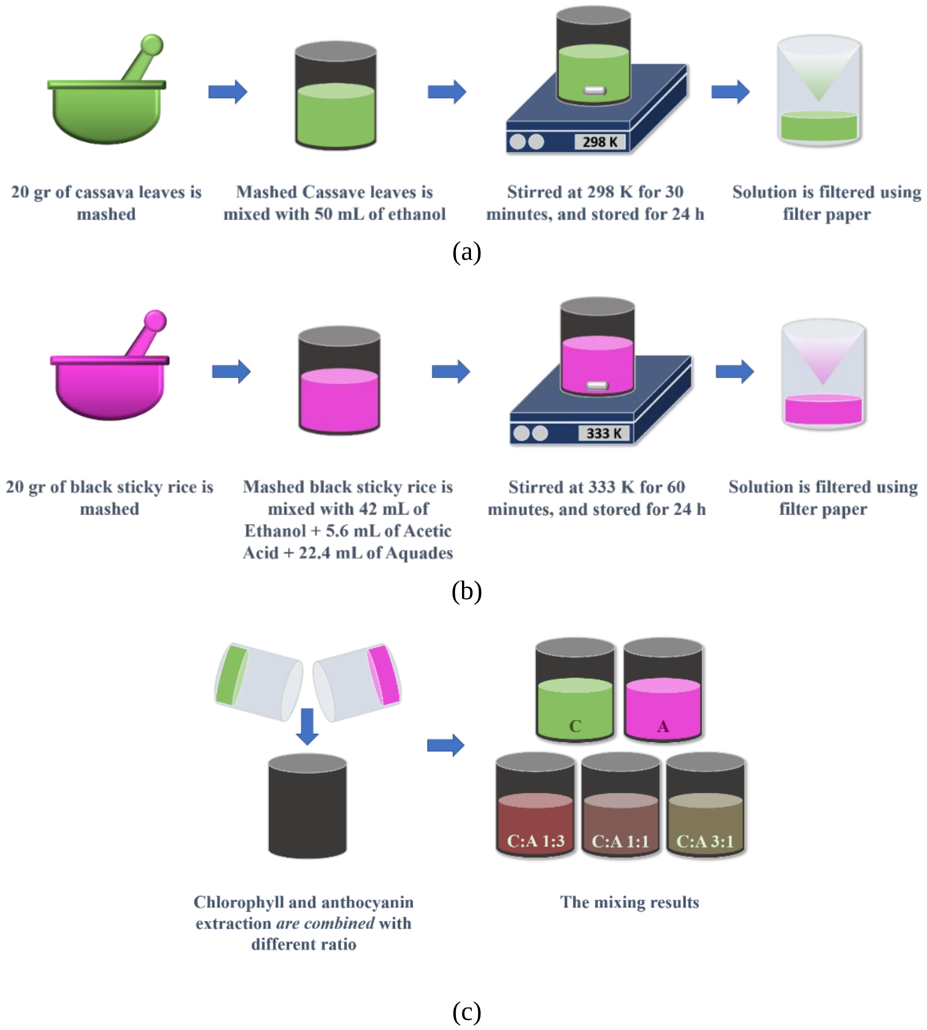


Figure 2. The extraction process of dye (a) chlorophyll, (b) anthocyanin, and (c) the mixing process of dye variations

2.4. Preparation and deposition of TiO_2 and dye

Preparation of TiO_2 paste was done by mixing 1.5 g PVA and 13.5 mL Aquades. In order to mix those two materials, the suspension was stirred using a magnetic stirrer and stirrer bar at 45 °C for 30 minutes. Then, 7.5 mL of the stirred solution was mixed with 0.5 g TiO_2 slowly and stirred until the solution becomes homogeneous.

Afterward, 0.25 mL TiO_2 paste was deposited onto an ITO substrate (2×2 cm^2) via the spin-coating method. The spin coater was set at 975 rpm, and the paste deposition was carried out for 10×10 seconds. After the TiO_2 paste was almost dry for about 5 minutes, the scotch tape attached to the ITO glass was removed. Then, the substrates were heated at 250 °C for 15 minutes. After the temperature of ITO glass dropped to room temperature, ITO glass was soaked in dye solution for 30 minutes, as shown in Figure 3.

2.5. Preparation and deposition of electrolyte solution

Preparation and deposition of electrolyte solution are shown in Figure 4. The electrolyte was prepared by mixing 0.8 g KI into 9 mL acetonitrile and 1 mL aquades. Afterward, 0.127 g I_2 powder was added to the solvent and stirred with a magnetic stirrer for 30 minutes. In order to prevent the photo-decomposition process, the solution was stored in a dark bottle. ITO glass that was soaked into dye was dropped by 0.25 mL electrolyte, as shown in Figure 4(a).

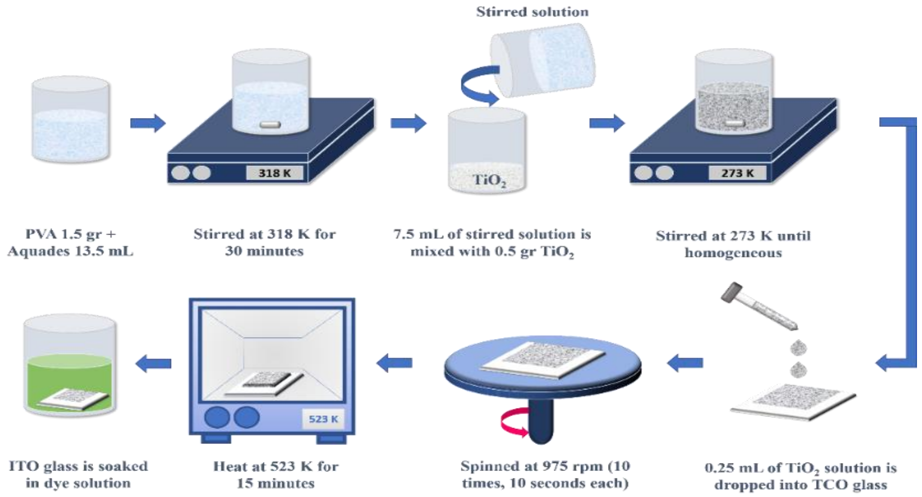


Figure 3. The preparation and deposition process of TiO_2 paste

2.6. Preparation counter electrode

In order to prepare the counter electrode, the conductive side of ITO glass was burned above the candle for 1 minute until the conductive side was covered by carbon, as shown in Figure 4(b). Afterward, the area outside the work area was cleaned using tissue. The area of counter electrode was 4 cm^2.

2.7. DSSC assembly

The DSSC was assembled using TiO_2 coated dye sensitizer as the working electrode and carbon as the counter electrode. The working electrode and counter electrode were assembled as shown in Figure 4(c). The conductive side of both substrates was attached tightly using the clip.

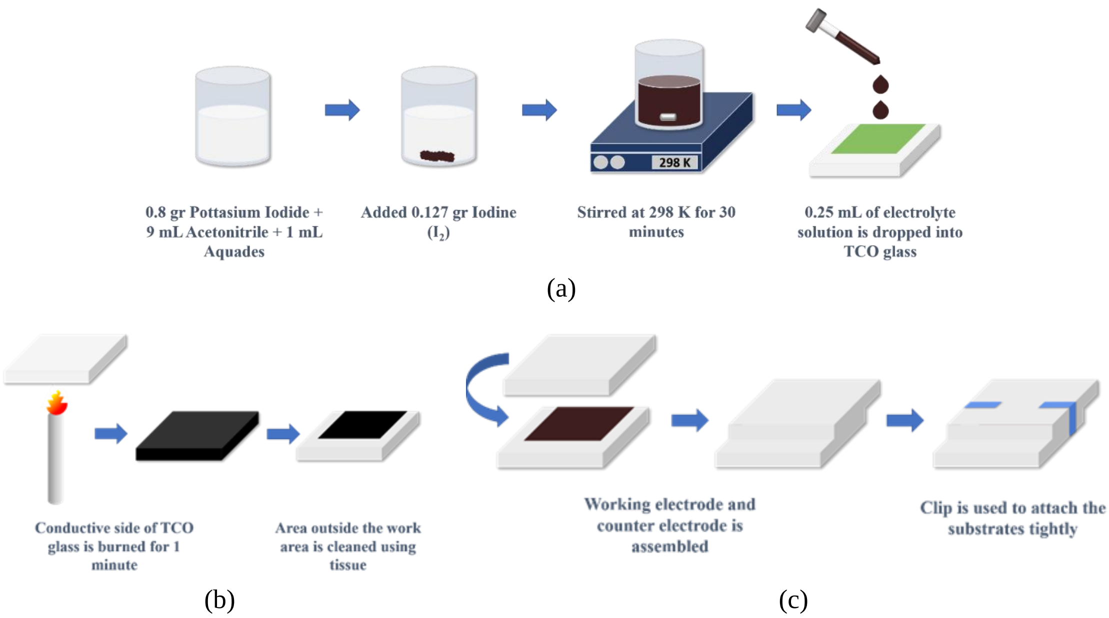


Figure 4. Preparation and deposition of electrolyte solution (a) the preparation and deposition process of electrolyte, (b) the preparation of counter electrode, and (c) the assembly process of working electrode and counter electrode

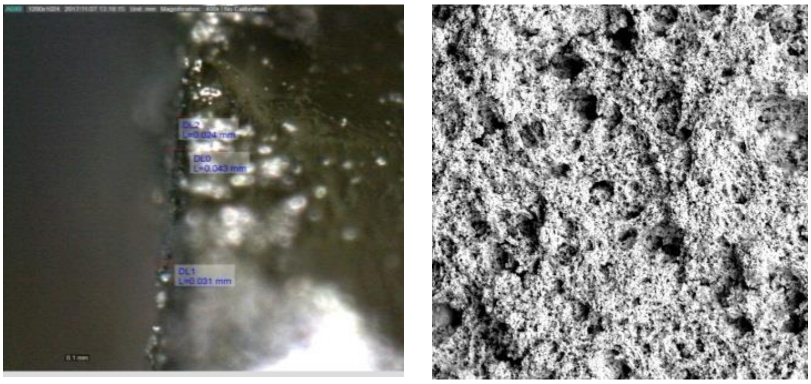
2.8. Characterization tools

TiO_2 thickness was determined using a digital microscope (Dino-Lite AM4115 Series) while TiO_2 structure was determined using scanning electron microscope (SEM, Phenom G2 Pro). The optical properties were performed using a UV-visible spectrophotometer (UV-1800 Shimadzu). Open circuit voltage (V_{oc}) and short circuit current (I_{sc}) measurement was carried out under AM 1.5 G and LED lamp cool daylight 7 Watt using multimeter (Sanwa CD771 and Hyelec MS8229). A Lux meter (Krisbow KW06-288) was used to control the light source.

3. RESULTS AND DISCUSSION

3.1. Characterization of TiO_2 structure

Characterization of TiO_2 structure are shown in Figure 5. A digital microscope was used to further investigate the thickness of TiO_2. Based on the result using 400 times magnification, the thickness value of TiO_2 film on ITO glass ranges from 0.024 to 0.043 mm, with the average value of 0.033 mm, as shown in Figure 5(a). Afterward, SEM measurement was carried out to identify the structure of TiO_2. As shown in Figure 5(b), the SEM images of TiO_2 reveal a porous morphology.

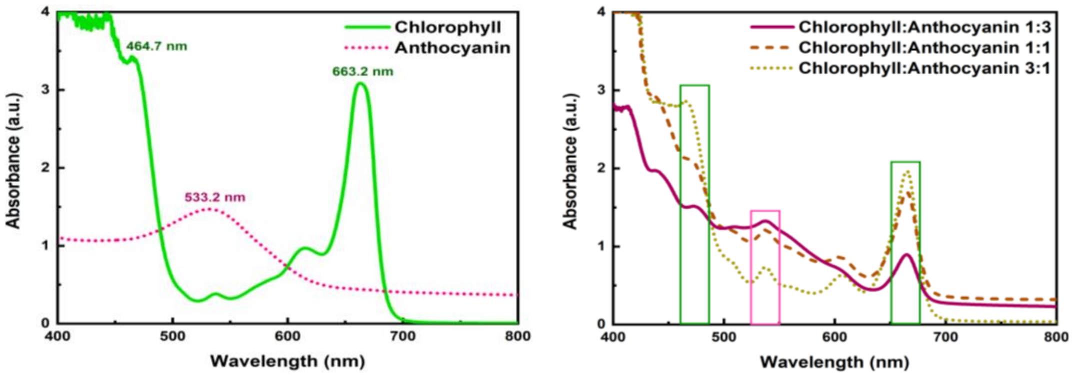


(a) (b)

Figure 5. Characterization of TiO_2 structure (a) the thickness of TiO_2 and (b) SEM images of TiO_2

3.2. Optical properties of chlorophyll and anthocyanin

The absorbance measurement of dye extraction was conducted to determine the ability of dye to absorb visible light with a wavelength of 400 to 800 nm, as shown in Figure 6. According to Figure 6(a), chlorophyll dye exhibits optical absorption in the visible regions with an absorbance peak of 400 and 663 nm. Meanwhile, the absorbance peak of anthocyanin dye is at a wavelength of 533 nm. As the chlorophyll and anthocyanin dyes are combined with different ratios, all of the combinations show the strongest absorption in both chlorophyll and anthocyanin peak characteristics at 464, 533, and 663 nm. As shown in Figure 6(b), the highest peak of chlorophyll is obtained from Chlorophyll:Anthocyanin with the ratio of 3:1, 1:1, and 1:3, respectively, while the highest peak of anthocyanin is obtained in the ratio of 1:3, 1:1, and 3:1, respectively, shows the success in the combination process.



(a) (b)

Figure 6. UV−visible absorption spectra of (a) chlorophyll and anthocyanin and (b) the variation of chlorophyll and anthocyanin

Based on Tauc plot (αhν) 0.5 data, as shown in Figure 7(a), (b), the estimated band gaps of chlorophyll and anthocyanin are 1.79 and 1.97 eV, respectively. However, when chlorophyll and anthocyanin are combined, the estimated bandgap of the combination with the different ratios are slightly the same, which are 0.811, 0.810, and 1.799 eV, respectively, for Chlorophyll:Anthocyanin 1:3, 1:1 and 3:1, as shown in Figure 7(c)-(e).

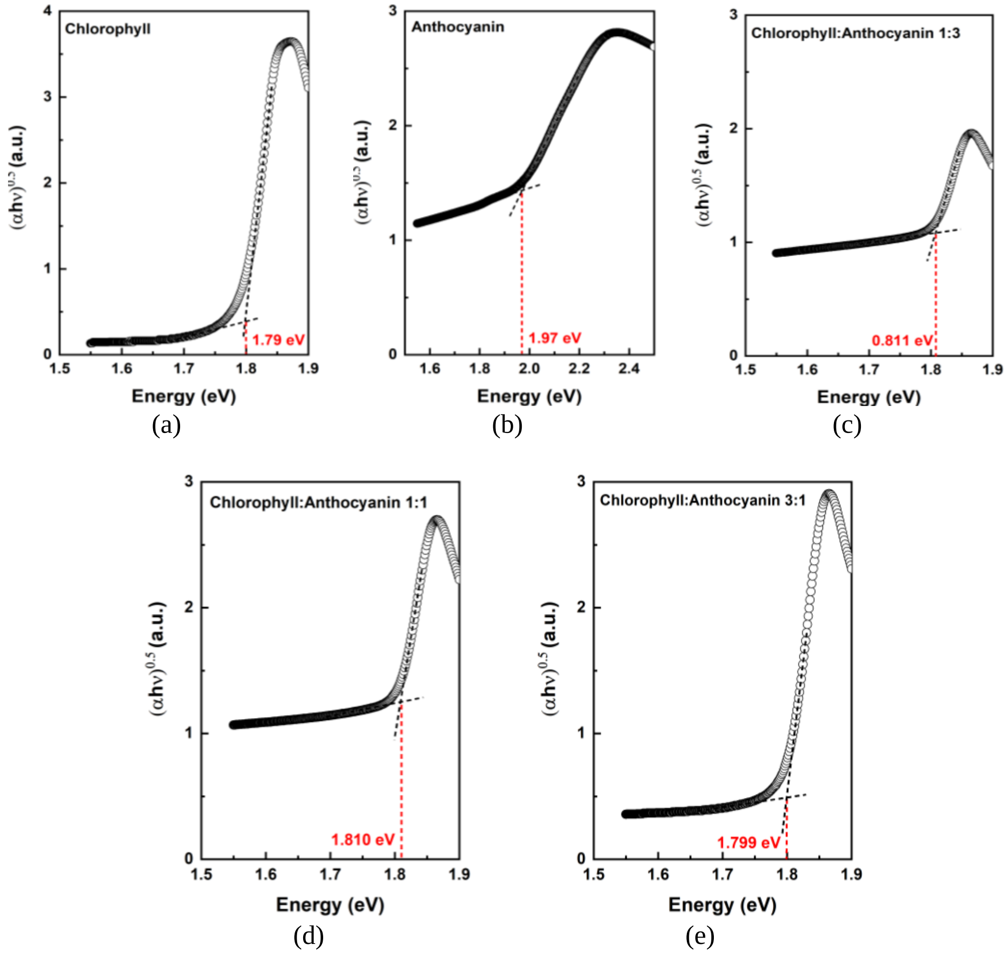


Figure 7. Tauc plot of (a) chlorophyll (C), (b) anthocyanin (A), (c) C:A 1:3, (d) C:A 1:1, and (e) C:A 3:1

3.3. DSSC performance of chlorophyll and anthocyanin

The voltage and current measurements were conducted to study the different dye combination performances. In this research, the voltage and current measurements were performed under AM 1.5 G and 7 Watt cool daylight LED irradiation. Measurement under two different irradiation was conducted to identifiy the effect of illumination strength on DSSC performances.

3.3.1. DSSC performance under AM 1.5 G irradiation

DSSC performance under AM 1.5 G irradiation was conducted by measuring the length of the object shadow from 07.30 to 10:15 AM. The air mass value was obtained according to (1).

$$AM = \sqrt{1 + \left(\frac{s}{h}\right)^2} \quad (1)$$

AM is *Air Mass*, s is the shadow length, and h is the object length. As shown in Table 1, AM 1.5 occurs from 08:15 to 08:30 AM. Thus, the open-circuit voltage (V_{oc}) and short circuit current (Isc) measurements were carried out from 08:15 to 08:30 AM.

Table 1. Measurement results of shadow length's object

Time	Shadow Length	AM
8:15	17.7 cm	1.57
8:30	15.8 cm	1.48

According to Table 2, the highest voltage was generated by the variation of Chlorophyll:Anthocyanin with a ratio of 3:1. However, the highest current was generated by the variation of the Chlorophyll:Anthocyanin with a ratio of 1:1. Afterward, maximum power point voltage (V_{MPP}) and maximum power point current (I_{MPP}) were calculated. Then, based on the V_{MPP} and I_{MPP} calculations, maximum power (P_{MAX}), and efficiency can be calculated according (2) to (4).

$$FF = \frac{V_{MPP}\ x\ I_{MPP}}{V_{oc}\ x\ I_{sc}} \quad (2)$$

$$P_{MAX} = V_{oc}\ x\ I_{sc}\ x\ FF \quad (3)$$

$$\eta = \frac{P_{MAX}}{P_{IN}}\ x\ 100\% \quad (4)$$

Based on Table 3, all DSSC with the combination of dyes has higher cell performance than both single individual dyes. The highest maximum power and efficiency were produced by the variation of the Chlorophyll:Anthocyanin with the ratio of 3:1 with the value of 6.720×10-7 W and 1.805×10-4 %, respectively. The smallest maximum power and efficiency were produced by chlorophyll dye with the value of 2.750×10-8 W and 7.388×10-5 %, respectively.

Table 2. Open circuit voltage (V_{oc}) and short circuit current (I_{sc}) measurement of DSSC under AM 1.5 G irradiation

Sample	V_{oc} (mV)	I_{sc} (μA)
Chlorophyll (C)	440	2.5
Anthocyanin (A)	460	2.4
C:A 1:3	461	5.1
C:A 1:1	507	5.3
C:A 3:1	517	5.2

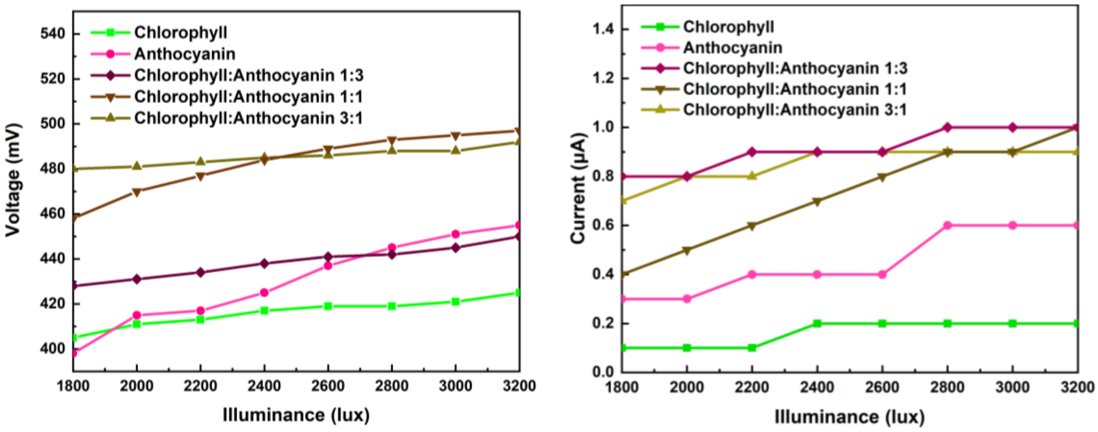
Table 3. Calculation results of DSSC characteristic

Sample	Pmax (W)	η (%)
Chlorophyll	2.750×10^{-8}	7.388×10^{-5}
Anthocyanin	2.760×10^{-8}	7.414×10^{-5}
C:A 1:3	15.880×10^{-7}	1.579×10^{-4}
C:A 1:1	6.718×10^{-7}	1.804×10^{-4}
C:A 3:1	6.720×10^{-7}	1.805×10^{-4}

3.3.2. DSSC performance under 7 watt cool daylight LED irradiation

For further investigation, DSSC performance under 7 watt cool daylight LED irradiation was conducted. The measurement was done with the illuminance ranging from 1800 to 3200 lux, as shown in Figure 8. According to Figure 8(a), all DSSC show the same pattern, the greater the strength of the illumination, the greater the open-circuit voltage generated. The highest mean value of open-circuit voltage (V_{oc}) that was generated by the variation of the Chlorophyll:Anthocyanin with the ratio of 3:1. As shown in Figure 8(b), the highest mean value of short circuit current (I_{sc}) can be generated from the variation of the Chlorophyll:Anthocyanin with the ratio of 1:3.

Based on Table 4, the highest maximum power and efficiency are produced by the variation of the Chlorophyll:Anthocyanin with the ratio of 3:1, while the smallest maximum power and efficiency are produced by chlorophyll dye. Based on these data, it was evident that by mixing the variations of chlorophyll and anthocyanin dye can produce greater maximum power and greater efficiency than single chlorophyll or anthocyanin dye.



(a) (b)

Figure 8. DSSC performance of chlorophyll and anthocyanin dyes (a) voltage-illuminance curves of DSSC and (b) current-illuminance curves of DSSC under 7 watt cool daylight LED irradiation

Table 4. Calculation results of DSSC characteristic

Sample	FF	P_{MAX} (W)	η (%)
Chlorophyll	0.25	1.740×10^{-8}	2.49×10^{-7}
Anthocyanin	0.25	4.820×10^{-8}	6.89×10^{-7}
C:A 1:3	0.25	9.980×10^{-8}	1.43×10^{-6}
C:A 1:1	0.25	8.780×10^{-8}	1.26×10^{-6}
C:A 3:1	0.25	1.040×10^{-7}	1.48×10^{-6}

According to the band diagram shown in Figure 9, double-excited electrons would be generated from both TiO_2 and dye. However, the excited electrons from the dye have a shorter time to transport into the TiO_2 conduction band. Thus, the excited electron from dye plays a key role in determining the DSSC performance. The smaller the bandgap of dye, the easiest the electron to get excited from ground state to excited state, in this state, energy is produced. Therefore, Chlorophyll:Anthocyanin 3:1 has the best performance due to its lowest bandgap energy. Besides that, the performance of DSSC was also affected by the pH of the dye. Anthocyanins are stable at low pH [25], while chlorophyll works optimally at pH 7-5. When anthocyanin and chlorophyll are combined, they need a proper ratio to get a stable condition.

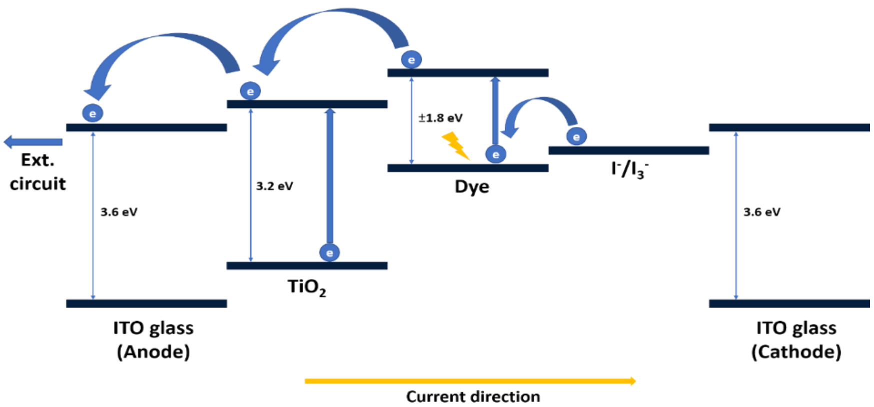


Figure 9. Band diagram of DSSC

4. CONCLUSIONS

This work reports the preparation and photovoltaic performance of a natural organic-based generate DSSC obtained from cassava leaves and black sticky rice. The combination of chlorophyll and anthocyanin was capable to expand the wavelength absorption range which significantly affect the DSSC performance. The best DSSC performance under AM 1.5 G and 7 Watt cool daylight LED irradiation were both obtained from the variation of Chlorophyll:Anthocyanin with the ratio of 3:1, with the efficiency of 1.81×10^{-4}% and 1.48×10^{-6}%, respectively. We successfully demonstrated that the integration of a natural dye with a combination of chlorophyll and anthocyanin can pave the way to generate DSSC with higher performance.

ACKNOWLEDGMENTS

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