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Investigation of Curcumin and Chlorophyll as Mixed Natural Dyes to Improve the Performance of Dye-Sensitized Solar Cells

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Abstract: This study investigates the natural dyes of curcumin and chlorophyll as sensitizers of dye-sensitized solar cells. Mixing two dyes can create a wider absorbance area, thereby increasing the performance of a solar cell. This study used variations in the volume ratio of the chlorophyll and curcumin mixture of 100:0, 75:25, 50:50, 25:75, and 0:100. The results were characterized by cyclic voltammetry, UV-Vis spectrophotometry, and solar simulator. The curcumin–chlorophyll dye mixture exhibited better light absorbance, bandgap energy, and efficiency than those of single dyes. The highest solar cell performance was achieved with a 75:25 mixture of chlorophyll and curcumin with electrical efficiency, short-circuit photocurrent density, open-circuit photovoltage, and fill factor values of 0.261%, 1.20 mA/cm^2, 0.535 V, and 0.405, respectively.

Keywords: curcumin; chlorophyll; natural dyes; dye-sensitized solar cells

1. Introduction

Solar energy from the sun is the earth's largest energy source, the usage of which can be classified into solar thermal and photovoltaic methods [1–3]. Solar thermal methods harness heat energy from the sun that can be used directly or converted into mechanical energy via approaches such as the drying of crops, solar stoves, Stirling engines, and solar chimneys. On the other hand, photovoltaic methods use photon energy converted into electrical energy via solar cell technology. The technology of solar cell can be classified into three generations: crystalline-silicon, thin film, and organic solar cell [1,4–6]. A dye-sensitized solar cell (DSSC) is one of the third-generation solar cells patented by Gratzel in 1991; it is easy to produce, inexpensive, and environmentally friendly compared to other types of solar cells [7–10].

DSSC is a photo-electrochemical solar cell with main components consisting of transparent conductive glass, a semiconductor, a dye sensitizer, electrolytes, and counter electrodes [11–13]. Dye that is applied to a DSSC is different from other color applications, such as those used in textiles and ink. In a DSSC application, dye is synthesized to produce electrons or active groups such as -OH, C=O, and -COOH. In addition, dyes for sensitizers must have good electron communication with semiconductors, for example TiO_2, ZnO, and NiO [12–17].

Generally, the dye sensitizers used in DSSC solar cells are synthetic ruthenium complexes, such as N621, N719, N3, Z-907, and black dye. The disadvantages of synthetic ruthenium dye, however, are expensive and require a complex manufacturing process [14,18]. On of promising alternative for synthetic dyes in DSSC are natural dyes. The natural dyes are generally in the form of chlorophyll, anthocyanin, xanthophyll, carotene, indigo, and flavonoids that can be produced from the flowers, fruits, leaves, and seeds of plants [19,20]. The advantages of natural dyes are that they are easy to obtain, easy to process, cheaper, and environmentally friendly [19–22].

Dyes have the ability to absorb light in certain wavelengths [6,8]. In all green plants, chlorophyll has two forms: chlorophyll-a and chlorophyll-b. Chlorophyll-a is green dominant with a chemical composition of $C_{55}H_{72}MgN_4O_5$, while chlorophyll-b is blue dominant with a chemical composition of $C_{55}H_{70}MgN_4O_6$. Chlorophyll is stored in the organelles, which are chloroplasts. Chlorophyll has a green color because it is not effective at absorbing green wavelengths and, thus, only reflects it. Chlorophyll achieves maximum absorption in blue and red wavelengths but absorbs very little yellow light [19,21]. Chlorophyll that is extracted from spinach leaves has high absorbance at the blue and red visible wavelengths: ±400–500 nm and ±600–700 nm, respectively. Chlorophyll has low absorption at ±500–600 nm green wavelengths [23,24].

Besides chlorophyll, curcumin dyes also have high potential as sensitizers of DSSC. Curcumin dye is extracted from turmeric (*Curcuma longa L.*) and has good absorbance in the visible wavelength of light, which is ±400–580 nm [25]. Kim et al. [26] extracted curcumin without purification to determine the performance of dyes using an ethanol solvent at concentrations of 1.5–2%. The research produced efficiency (η), open-circuit photovoltage (V_{OC}), short-circuit photocurrent (I_{SC}), and fill factor (FF) values of 0.36%, 0.5632 V, 1.0055 mA/cm^2, and 0.6399, respectively.

Increased light absorbance by dyes can be achieved by mixing and combining more than one type of natural dye [27–29]. In this study, two types of natural dyes were mixed as photosensitizers in DSSC: chlorophyll and curcumin. Chlorophyll extracted from spinach leaves can absorb light in the blue spectrums (±400–450 nm wavelengths) and red spectrums (±625–700 nm wavelengths) but can absorb very little green light at ±500–600 nm. Furthermore, curcumin dye extracted from turmeric can absorb light at wavelengths of ±420–580 nm. The mixing of these two natural dyes is expected to increase the absorbance area at visible wavelengths. The novelty of this paper is comprised by the proposed mixed-natural dyes for the improvement of the absorbance area. Increasing the absorbance area will thereby increase the intensity of photons injected from the photoanode of a DSSC. As a result, the electron injection process in a photoanode will be improved, and the performance of the DSSC will also increase.

2. Materials and Methods

2.1 Extraction of natural dyes

Curcumin was extracted from turmeric and chlorophyll was extracted from spinach leaves that were cleaned using water. The spinach leaves and turmeric leaves were cut into small pieces, and the pieces were dried under the sun to evaporate the water content; the dried pieces were crushed into powders. The spinach and turmeric leaf powders were dissolved into ethanol with a ratio of 1 g/10 ml. The extraction process using a maceration method was carried out for ±2–3 hours with a heating temperature of 70 °C [19]. Next, the liquid from the extraction process was filtered by using a Whatman filter paper. The liquid from the filtering process was processed into the rotary evaporator machine for 1 hour at temperature of 80 °C. Then, both the chlorophyll and curcumin were dissolved into ethanol with concentrations of 8 g/100 ml, and both dyes were mixed with the specified volume ratio. This study mixed chlorophyll and curcumin with volume ratio variations of 100:0, 75:25, 50:50, 25:75, and 0:100.

The natural dye was examined by using a UV-Vis spectrophotometer to determine the absorbance values at visible wavelengths. In addition, a cyclic voltammetry test was used to determine the value of the bandgap energy for the natural dyes.

2.2 Fabrication of DSSC

Semiconductor for DSSC in this research used the TiO_2 anatase nanoparticle (~21 nm particle size) from Sigma Aldrich. The semiconductor was deposited on the transparent conductive oxide (TCO) substrate by using the doctor blade method. Fluorine-doped tin oxide (FTO), one of TCO glass was used as the DSSC-assembling substrate.

Counter electrode in DSSC structure was fabricated by using sputtering method. In sputtering method, a catalytic platinum was deposited on the FTO glass in a vacuum tube with initial pressure of 9.5 × 10^{-5} Torr (0.012665625 Pa). Argon gas was injected at 4 mTorr or 0.53328947 Pa into vacuum chamber with conditions of high electrical potential difference between platinum and FTO glass (404 V, 125 mA).

I_3^-/I^- electrolyte solution in DSSC was synthesized by mixing 3.3 g sodium iodide (99.95%), 523.875 mg pure iodine (99.95%), 5.481 g heteropolyacid (HPA), and 30 mL acetonitrile.

After DSSC elements were assembled with interval space of 30-μm thermoplastic, the electrolyte solution was injected through the hole in the counter electrode, and sealed by glass glue. The solar cell characterization was tested by using a solar simulator under irradiance of 100 mW/cm^2. As a result, the current–voltage (I–V) curve of solar cell can be measured by a digital multimeter (Keithley 2401) to obtain V_{OC}, I_{SC}, FF, and η of DSSC.

3. Results and Discussion

3.1 Light absorbance

Figure 1 explains that the absorbance of the extraction of turmeric (curcumin) and spinach leaves (chlorophyll) in the visible light region is 400 to 750 nm. In this work, the maximum absorbance of natural dyes is around 3.5 at 400-550 nm. Even though it is more than 1.5, it is still acceptable to justify its performance as sensitizer in the DSSC.

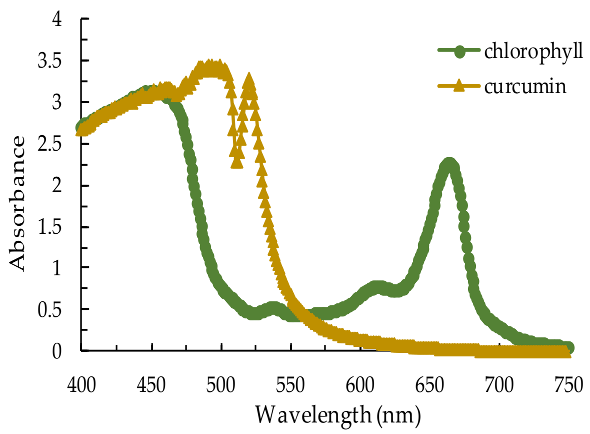

Fig. 1: Light absorbance of the chlorophyll and curcumin dyes.

Curcumin has two absorption peaks at ±480–500 nm and at 520 nm. Then, at the 521 nm wavelength, the absorbance starts to decrease slowly until 750 nm. This is in accordance with previous studies, where curcumin was found to only be able to absorb visible blue, green, and yellow light at ±400–580 nm [26].

The extraction of spinach leaves (chlorophyll) has strong absorbance at wavelengths of 400 to 450 nm because the spinach leaves are dominated by chlorophyll-a, which absorbs at a maximum of ±400–450 nm wavelengths, while chlorophyll-b absorbs only at ±400–500 nm wavelengths [23]. Then, at wavelengths of 450–600 nm, the absorbance value is low. These results were in accordance with the chlorophyll properties, which has low absorbance in the green wavelength areas of ±500–600 nm. This indicates that, when polychromatic light via UV-Vis spectroscopy passes through the chlorophyll, it will reflect a green color. After that, the absorbance value rises from 600 nm and reaches its maximum peak at the 664 nm wavelength.

Figure 2 shows the effect of mixing variations of chlorophyll and curcumin. The mixed dye with composition ratios of 75:25, 50:50, and 25:75 had higher absorbance and a wider light absorption area than did the single dyes made of 100% chlorophyll and 100% curcumin. This phenomenon was caused by the two dyes (chlorophyll–curcumin) having strong absorption at wavelengths of 400–500 nm, so that, when mixed, they can improve absorption in those same wavelengths. The addition of curcumin dye affected the absorbance values at wavelengths of 400–550 nm. The greater the composition of curcumin in the mixture of chlorophyll–curcumin, the greater the absorbance value. The addition of chlorophyll, however, causes the area of light absorption to be wider at 550–750 nm, as shown in Figure 2.

The comparison of absorbance ability was analyzed by calculating the absorbance area for each of the dyes. The absorbance areas of the 100:0, 75:25, 50:50, 25:75, and 0:100 chlorophyll–curcumin mixed natural dyes were 426.54, 502.53, 494.63, 466.27, and 442.66 (arbitrary unit), respectively.

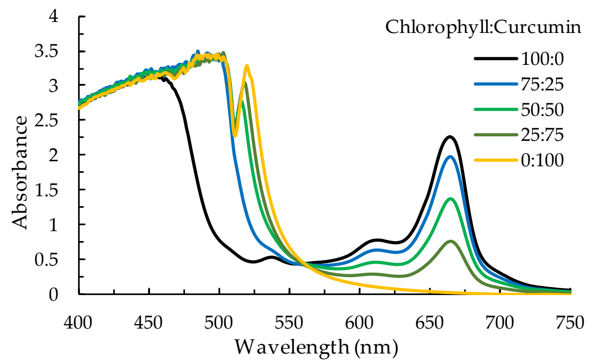

Fig. 2: Light absorbance of chlorophyll–curcumin mixed natural dyes.

Moreover, Figure 1 and 2 show that the absorbance peaks are more than one. Absorbance scale normally runs from zero to one, but could go higher than that in extreme cases (up to more than 90% of a wavelength of light is absorbed) based on Beer-Lambert law.

3.2 Band gap energy

The energy level of natural dyes was analyzed by using cyclic voltammetry. The analyzed energy level in this study were the reduced the energy (E_{red}) level and the oxidation energy (E_{ox}) level. Oxidation energy is the energy needed by molecules to release electrons. The reduction energy is the energy that molecules need to add electrons. Those energy levels can be converted to the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO). HOMO is an orbital that acts as an electron donor, because it is the outermost orbit (highest energy) that contains at least one electron. LUMO is an orbital that acts as an electron receiver, because it is the innermost orbit (lowest energy) that has space to receive electrons. The HOMO energy (E_{HOMO}) and LUMO energy (E_{LUMO}) can be obtained by using equations 1 and 2. Then, the bandgap energy can be derived from the difference between E_{LUMO} and E_{HOMO}.

$$E_{HOMO} = -e[E_{ox} + 4.4] \quad (1)$$

$$E_{LUMO} = -e[E_{red} + 4.4] \quad (2)$$

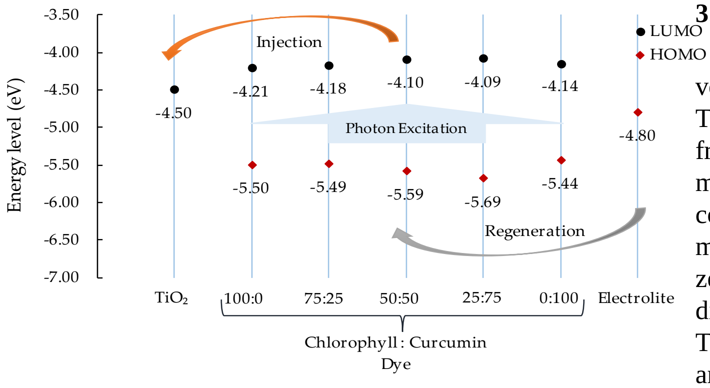

Fig. 3: HOMO–LUMO energy levels for chlorophyll–curcumin mixed natural dyes.

Figure 3 shows the process of transferring electrons in the DSSC by sunlight. Photons in the sunlight will initiate the electron transfer or oscillation from dyes to semiconductors, and electrons will be regenerated with electrolytes in dyes. Comprehensive electron transfer from dyes to semiconductors occurs when the E_{LUMO} level is higher than the energy level of conduction band from semiconductor (TiO_2) material. Furthermore, efficient electron regeneration also occurs when the E_{HOMO} level is lower than the potential energy of the electrolyte redox. As a result, the wide bandgap energy between HOMO and LUMO can produce a larger photocurrent.

Table 1 shows the results of the cyclic voltammetry testing of each dye sample tested. The difference in the LUMO and HOMO energy levels occurred due to the composition of the mixed dyes, as shown in Figure 3 and Table 1. The results also indicated that, the higher the absorbance value of dyes, the greater the bandgap energy. The wider bandgap energy will increase the electron injection from the conduction band to the valence band. Therefore, chlorophyll, curcumin, and a mixture of chlorophyll–curcumin can be used as a photosensitizer for DSSC because it had lower LUMO energy than did the conduction band of TiO_2 (-4.5 eV) and the HOMO energy, which was higher than the electrolyte redox energy (-4.8 eV). As a result, it will affect the electric current and voltage of the solar cells.

Table 1. Cyclic voltammetry for chlorophyll–curcumin mixed natural dyes.

Dye composition (chlorophyll: curcumin)	E_{ox} (eV)	E_{red} (eV)	E_{HOMO} (eV)	E_{LUMO} (eV)	$E_{band\ gap}$ (eV)
100:0	1.10	-0.19	-5.50	-4.21	1.29
75:25	1.29	-0.31	-5.69	-4.09	1.60
50:50	1.19	-0.30	-5.59	-4.10	1.49
25:75	1.09	-0.22	-5.49	-4.18	1.31
0:100	1.04	-0.26	-5.50	-4.14	1.30

3.2 Performance of DSSC

Figure 4 depicts the curves of photocurrent density versus photovoltage (I–V) for each of the DSSC samples. The output characteristics of DSSC that can be produced from I-V curve are V_{OC}, I_{SC}, FF, and η. The V_{OC} is maximum voltage of solar cell that will supply under conditions of infinite-resistance electrical load. The I_{SC} is maximum current of solar cell under conditions of a zero-resistance electrical load. In addition, the I_{SC} divided by the solar cell active area is known as the J_{SC}. The FF is the ratio between the maximum power (P_{MPP}) and the product of V_{OC} and I_{SC}, as shown in equation 3. The most importance parameter from I–V curve is the power conversion, η (i.e., the ratio of generated electricity to incoming light energy), as shown in equation 4.

$$FF = \frac{P_{MPP}}{I_{SC} \times V_{OC}} \quad (3)$$

$$\eta = \frac{P_{MPP}}{P_{light}} = \frac{P_{MPP}}{I_{rad} \times A} = \frac{I_{SC} \times V_{OC} \times FF}{I_{rad} \times A} \quad (4)$$

In the variation of the dye sensitizer, the main factors that influence the characteristics of the solar cells are the E_{HOMO} and E_{LUMO}. Figure 3 shows how the electron injection activity from the dye to the semiconductor operates, as well as the electrolyte dye regeneration process. The electron injection process occurs when the LUMO dye energy level must be lower than the semiconductor (TiO_2) energy level. The electrolyte dye regeneration process occurs when the HOMO energy level of the dye is higher than the redox electrolyte energy level[21]. Another factor that affects the characteristics of the solar cells is the absorbance value of the dyes. The increasing composition of chlorophyll in the mixed dyes will also increase the efficiency of the DSSC since chlorophyll has wider absorbance in the area of 500–600 nm. The wider the absorbance area, the greater the energy of the absorbed photons. By increasing the absorbed photons, the electrons will be more excited to initiate the flow current in the circuit.

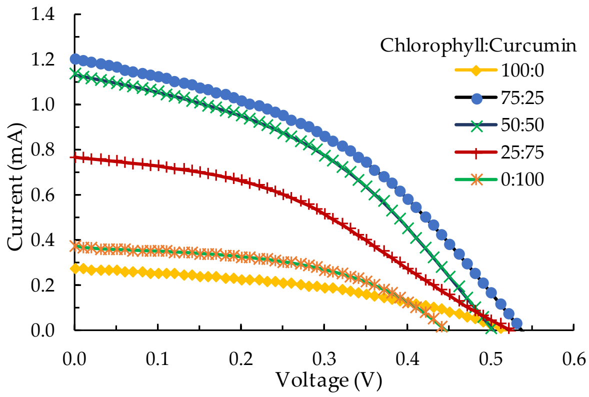

Fig. 4: I–V curves of the DSSC under an irradiation intensity of 100 mW/cm².

Table 2. Performance parameter of the DSSC for chlorophyll–curcumin mixed natural dyes.

Dye composition (chlorophyll: curcumin)	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	η (%)
100:0	0.513	0.271	0.408	0.056
75:25	0.535	1.200	0.405	0.261
50:50	0.502	1.130	0.387	0.232
25:75	0.523	0.766	0.482	0.155
0:100	0.445	0.372	0.408	0.080

Table 2 depicts the electrical characteristic of the DSSC in terms of the values of the V_{OC}, J_{SC}, FF, and efficiency for all sample variations. Factors that affect the FF are the internal barriers and electrical resistance of the photoanode, counter electrode, and conductive glass. In accordance with electrical potential, V_{OC} value is influenced by the bandgap energy level; the higher the bandgap energy, the greater the V_{OC} gained [21)]. Related with electrical current, I_{SC} is influenced by the bandgap energy and absorbance. The wider the bandgap energy, the greater the number of electrons flowing from the conduction band to the valence band. When many electrons are flowing, the photocurrent resulting from the photoexcitation becomes larger.

3. Conclusions

A DSSC with various natural dye mixtures was successfully fabricated. Mixing of the natural dyes using natural curcumin dyes extracted from turmeric and natural chlorophyll dyes extracted from spinach leaves was successfully conducted, and the mixture was used as a photosensitizer for a DSSC. The highest DSSC efficiency was achieved with a 75:25 mixture of chlorophyll and curcumin with η, V_{OC}, I_{SC}, and FF values of 0.261%, 0.535 V, 1.20 mA/cm^2, and 0.405, respectively. It confirms that our proposed mixed-natural dyes have a good potency for the DSSC.

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Nomenclature

DSSC	dye-sensitized solar cell
$E_{band\ gap}$	band gap energy (eV)
E_{HOMO}	HOMO energy (eV)
E_{LUMO}	LUMO energy (eV)
E_{ox}	oxidation energy (eV)
E_{red}	reduction energy (eV)
FF	fill factor
FTO	fluorine-doped tin oxide
HOMO	highest occupied molecular orbital
HPA	heteropolyacid
I–V	current–voltage
I_{SC}	short circuit current (A)
LUMO	lowest unoccupied molecular orbital
P_{MPP}	maximum power point (W)
V_{OC}	open circuit voltage (V)

Greek symbols

η	efficiency (%)

Subscripts

ox	oxidation
rad	radiation
red	reduction

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