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MAKING PROTOTYPE DYE-SENSITIZED SOLAR CELLS (DSSC) BASED ON TiO₂ NANOPORY USING EXTRACTION OF MANGOSTEEN PEEL (*Garcinia mangostana*)

Hardani¹, Alpiana Hidayatulloh², Lily M Angraini³

¹ Faculty of Pharmacy, Politeknik Medica Farma Husada Mataram, Indonesia. E-mail: danylastchild07@gmail.com

² Faculty of Technique, University Nusa Tenggara Barat, Indonesia. E-mail: alpianahidayatulloh@yahoo.co.id

³ Faculty of Mathematics and Natural Sciences, University Mataram, Indonesia. E-mail: lilyangraini@gmail.com

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ABSTRACT

Alternative energy sources are available in nature is the Sun's energy. Dye-sensitized solar cell (DSSC) is one of the photochemical electrical cells consisting of a photoelectrode, dye, electrolyte, and counter electrode. The manufacture of prototype Dye-sensitized solar cell (DSSC) utilizes carotene from the dye extract of mangosteen peel pigment (*Garcinia mangostana*). This study aims to create a Dye-sensitized solar cell (DSSC) and know the efficiency it produces. This Dye-sensitized solar cell (DSSC) consists of a pair of FTO (Fluor-doped in oxide) glass intercepts facing each other. The glass acts as an electrode and counter electrode and separated by a redox electrolyte (I⁻ / I³⁻), arranged to flank each other to form a wafer. In the electrode, is deposited a porous nanocrystalline TiO₂ layer, as well as dye extract of mangosteen peel pigment (*Garcinia mangostana*). While on the counter electrode coated with a layer of platinum. This article presents some experimental data on absorbance properties and the conductivity of dye extract of mangosteen peel pigment (*Garcinia mangostana*) as an application in DSSC. Absorbance test using Spectrophotometer UV Visible 1601 PC and electrical properties test using Elkahfi 100 / Meter I-V. DSSC fabrication has been done using dye extract of mangosteen peel pigment (*Garcinia mangostana*) with coating spin coating technique. The results showed that the dye extract of mangosteen peel pigment (*Garcinia mangostana*) had an absorbance spectrum of 380-520 nm range. From the test results using AM 1.5G solar simulator (100 mW/cm²), it was found that the volume of TiO₂ precursors affected the performance of DSSC solar cells and the overall conversion efficiency was 0.092%.

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Introduction

Energy needs in the world are increasing along with the development of technology, in Indonesia even in the world are being incessant-the incessant of the researchers to find alternative energy sources, in lieu of fossil energy. One of the considerable alternative energy

available in nature is solar energy. Solar energy in Indonesia, in particular, the amount is very abundant and is said to be large enough to serve as an alternative energy source. To realize that, then needed a system to convert solar energy into electrical energy. One of the utilization of solar energy is through the use of solar cells (solar cell) because this is a promising alternative[1].

This is what makes energy very important in meeting all the needs of life in the world, so the energy needs in the world increasingly day. Limitations of silicon solar cells are not only expensive, but the absorption spectra are too narrow. The known energy distribution of sunlight comprises about 4% ultraviolet and 96% visible light. The main spectrum of silicon solar cell absorption is ultraviolet and purple[2]. This shows that silicon solar cells cannot use nearly 96% of the energy from sunlight [3]. Attempts to extend the absorption spectrum from the ultraviolet region to the visible light region are now applied as Dye-Sensitized Solar Cell [2], where dyes can assist DSSC to broaden the absorption spectrum [4].

The absorption of light in the range 380-520 nm and the molar extinction coefficient greater than 105 makes the carotenoids as potential sensitizers in photovoltaic solar cells and other artificial photochemical devices [5]. Carrots (*Daucus carota*), melinjo fruit (*Genetum genemon*), and mangosteen peel (*Garcinia mangostana*) are natural ingredients that are widely consumed and contain carotenoids. But the carotenoid content may vary depending on the source.

Basically, the working principle of DSSC converts light energy into electricity on a molecular scale in the form of a reaction of electron transfer. The first process begins with the excitation of electrons in the dye due to the absorption of photons. This is one of the roles of TiO_2 properties. When photons from sunlight overwrite the working electrode on the DSSC, the photon energy is absorbed by the dye attached to the surface of TiO_2 [6]. So dye gets the energy to excite. The excited dye carries energy and is injected into the conduction band on TiO_2 . TiO_2 acts as an electron acceptor or collector [7]. The dye molecule left behind is then oxidized. The electrons will then be transferred through the outer circuit to the comparative electrode (an electrode containing the platinum layer). Electrolytes (iodide and triiodide pairs) act as electron mediators so they can produce cycle processes in cells. The Triiodide ion captures electrons that originate from the outer sequence with the help of platinum molecules as a catalyst. The excited electrons re-enter the cell and are assisted by platinum to react with the electrolyte causing the addition of iodide ions to the electrons. Then an iodide ion on the electrolyte delivers electrons that bring energy to the oxidized dye. Electrolytes provide electron substitutes for oxidized dye molecules. So the dye returns to its initial state [8].

In general, FIG. 1 shows the DSSC comprising dye-sensitized organic matter, a nanocrystal TiO_2 layer, an electrolyte solution containing the redox pair I^-/I_3^- and the FTO glass substrate as the working electrode. The outside factor of the area and the thickness of the semiconductor layer governs the increase of dye load, then the optical density that results in the efficiency of light absorption [9]. The optical density represents the transmission size of an optical element of a certain wavelength. When connected with radiation on an object, the optical density is the ratio between the initial intensity and the intensity of the transmission.

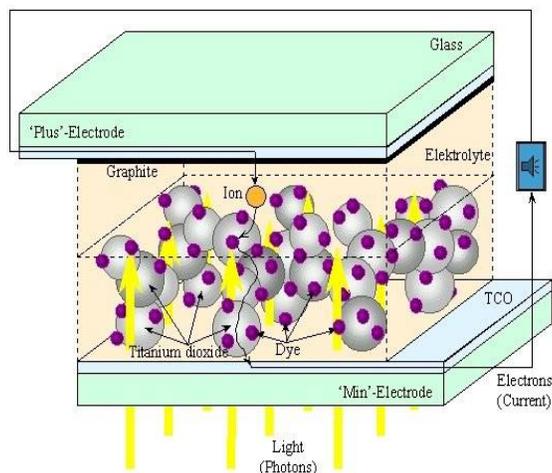


Figure 1 DSSC Scheme [10]

DSSC is a sandwich-shaped structure, in which two electrodes ie TiO_2 electrode with dye and a comparative electrode made of FTO-plated glass flattened by electrolyte form a photoelectrochemical cell system. The reference electrode is made of FTO glass coated with platinum because it has sufficient conductivity and heat resistance and electrocatalytic activity of triiodide reduction.

TiO_2 is a photocatalyst material that has strong oxidizing power, high photostability and redox selectivity [11]. An important requirement for increasing the catalyst activity of TiO_2 is to increase the surface area of TiO_2 depending on the size of the crystal.

The physical and chemical properties of TiO_2 depend on their size, morphology and crystal structure[12]. TiO_2 has three crystalline forms namely anatase, rutile, and brookite. Anatase phase TiO_2 crystals have a more active ability than rutile. Anatase is considered to be the most favorable phase of photocatalysis and solar energy conversion[13]. TiO_2 is only capable of absorbing ultraviolet light (350-380nm). To increase the absorption of the TiO_2 spectra in the visible region, a dye layer is required that will absorb visible light. The dye serves as a sensitizer [14].

This study presents some experimental data of the carotenoid content of the mangosteen fruit peel pigment that can be used as a sensitizer. A material analysis was performed on the optical and electrical properties of organic matter from mangosteen fruit peel extract (*Garcinia mangostana*). The extracts from the natural ingredients used in the study showed a similar absorbance of β -carotene in the 380-520 nm range. While the value of absorbance and optimum conductivity on the peel of mangosteen fruit. This study aims to analyze the optical properties and to know the electrical properties of the mangosteen peel pigment consisting of 3 stages: the extraction of the mangosteen peel pigment, the measurement of the absorption of the spectrum, and the measurement of the conductivity of the extraction[15]. Various studies about DSSC using natural colors of plant extracts has been conducted and the research proves that the substances natural color can give the effect of photovoltaics. On

this research resulted in higher efficiency compared to previous research that has been done by Basitoh Djaelani (2014)[20]. What is new in this study is located on the use of TiO₂ coating technique that uses the techniques of spin coating coating. While previous research still uses the technique of the doctor blade.

Experimental Method

A. Tools and Materials

The materials used in this study include glass substrate Fluorine Doped Tin Oxide (FTO), Titanium (IV) dioxide (TiO₂) nanopowder 21 nm, Polyethylene Glycol (PEG) 400, Pottasium Iodide (KI), Iodine (I₂), Ethanol , Pt (Hexachloroplatinic (IV) acid 10%), Isopropanol, 70% Alcohol, and Mangosteen Peel. Equipment used include digital multimeter, hot plate with magnetic stirrer, hairdryer, ultrasonic cleaner, 10 ml and 50 ml beaker, dropper dropper, 5 ml glass bottle, digital scales, filter paper whatman no.42, mortar chromatography column, knife, furnace, and spin coater.

B. Preparation

This preparation stage includes cleaning tools for the extraction and preparation of TiO₂ paste. The preparation process for extraction is done by cleaning the tool in the form of mortar, Fluorine Doped Tin Oxide (FTO) glass, glass bottle, beaker, and dropper with ethanol solution and using ultrasonic cleaner to be free from materials that can not be cleaned with water only. Clean glass affects the test results of samples to be superimposed on the glass substrate.

C. FTO glass cleanup (Fluorine Doped Tin Oxide)

Alcohol 70% poured on glass of chemical as much as 100 ml. The 2.5 x 2.5 cm FTO glass to be cleaned is inserted in a glass containing chemicals. Ultrasonic cleaner filled aquades to the specified limits. Chemical glass containing alcohol and FTO glass is inserted into ultrasonic cleaner at 30 minutes. After 30 minutes, the glass is dried using a hair dryer. Then measured resistance to the FTO glass using a digital multimeter.

D. Making of TiO₂ Nano Pasta

TiO₂ 0.5 gram of nano powder dissolved in 2 ml ethanol is then stirred using a stirrer vortex with a speed of 200-300 rpm for 30 minutes. The already formed TiO₂ paste is fed into aluminum foil-covered bottles and stored in a spot that avoids direct sunlight to reduce the evaporation process.

E. Mangosteen Peel Extraction (*Garcinia mangostana*)

Mangosteen peel weighed using 25 grams of digital scales. Furthermore the peel of mangosteen fruit crushed and mashed using mortar. The finely ground mangosteen peel was dissolved in 125 ml of ethanol solvent with ratio (1: 5) and then stirred for 60 minutes using a stirrer vortex with a rotation speed of 300 rpm in 60°C. After the solvent is dissolved for 24 hours and filtered with filter paper whatman no.42. The extraction results were then

chromatographed by pouring in chromatographic columns and waited until dark red extraction.

F. Making Working Electrode

The working electrode is made of FTO conductive glass on which the TiO₂ nano paste is deposited by spin coating technique. In FTO glass measuring 2.5 x 2.5 cm formed an area for the deposition of TiO₂ measuring 2 x 1.5 cm above the conductive surface. The FTO side taped the tape as a barrier. The TiO₂ paste is dripped on the FTO glass that has been glued in the spinner, then distirrer with a speed of 200-300 rpm with a predetermined time. The coated TiO₂ FTO glass is heated using a hotplate at 500°C for 60 minutes, then cooled to room temperature. The scheme of the TiO₂ paste deposition area is shown in Figure 2.

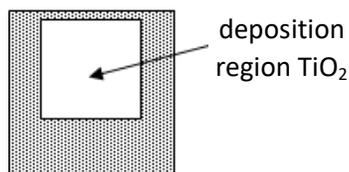


Figure 2. Schematic area of TiO₂ paste deposition

G. Making of Electrolyte Solution

Pottasium iodide (KI) of 0.8 grams (0.5 M) in solid form is mixed into 10 ml of polyethylene glycol 400 then stirred. Next to the solution was added Iodine (I₂) of 0.127 grams (0.05 M) then stirred with a stirrer vortex at 300 rpm for 30 min. The finished electrolyte solution is stored in a sealed container coated with aluminum foil.

H. Making of Opponent Electrode

The counter electrode is a FTO conductive glass which has been coated with a thin layer of Platinum (Hexachloroplatinic (IV) acid 10%). The steps of making the opponent electrode is 1 ml of Hexachloroplatinic (IV) acid 10% mixed with 207 ml of isopropanol and then stirred using vortex stirrer with speed 300 rpm for 30 minutes. The FTO glass was heated using a hotplate at 250°C for 15 minutes then spilled 3 ml of platinum solution onto the surface of the FTO glass substrate by the drop method. The glass that has been dropped platinum then cooled to reach the room temperature. The scheme of the Platinum deposition area is shown in Figure 3.

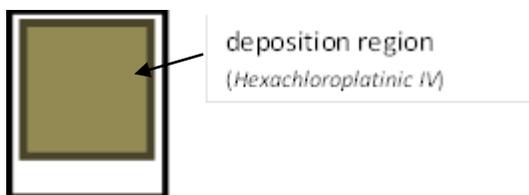


Figure 3. Platinum deposition area scheme

I. Dye Absorbtion On TiO₂ Layer

FTO conductive glass substrate which has been deposited TiO₂ layer then soaked in dye extract of mangosteen peel peel for 24 hours.

J. DSSC Sandwich Making

The arrangement of DSSC layers of FTO glass that has been coated with TiO₂ and has been immersed in dye solution of extraction result is called working electrode. The working electrode is dropped by an electrolyte solution and then covered with a platinum coated glass called the opposing electrode. Then the DSSC arrangement is clamped with a clamp on both sides of the right and left so as not to shift. The finished DSSC results are shown in Figure 4.



Figure 4. Results of DSSC Compilation

K. Natural Dye Extraction

The study used ethanol solvent to dissolve the carotenoid extracted from the natural material of the mangosteen fruit peel pigment. The ingredients to be extracted were cleaned with water, then as much as 25 grams of mangosteen peel peel pigment smoothed and after finely mixed 50 ml of ethanol stirred for 60 minutes 200 rpm using a magnetic stirrer at room temperature. After stirring and then stand for 24 hours and filtered using Whatman no filter paper. 42. After filtration, the solution is stored in a sealed container and protected from sunlight.

L. Absorption Analysis

A spectrophotometric method was used for the simultaneous determination of β -carotene [16]. The spectrophotometric method shows the potential for β -carotene analysis because Pigments can absorb radiation in the visible region [17]. The content of each extracted material was analyzed using Spectrophotometer UV Visible Shimadzu 1601 PC to determine the absorbance properties of the material. The wavelength range of absorption spectrum analysis in visible light is 300-800 nm. from the result of measurement of absorbance characteristic then known the type of dye content from natural material[18].

M. The conductivity of the material

The conductivity measurements using Elkahfi 100 / IV-Meter were performed in a dark state by covering all parts of the container using aluminum foil and under irradiation using a 100 W halogen light source and an energy intensity of 680.3 W / m². Halogen lamps are used because they have a full spectrum that resembles visible light with sunlight[19]. From the result of measurement of I-V then determined conductivity (σ) various material. To determine the conductivity of organic solution can use the equation:

$$\rho = RA / l \quad (1)$$

$$\sigma = 1 / \rho = l / RA \quad (2)$$

Where σ is the conductivity (ohm⁻¹.m⁻¹), R is the resistance (Ohm), l is the distance between the two electrodes (m) and A is the cross-sectional surface area of the electrode (m²).

Result and Discussion

Research using natural ingredients to produce a carotenoid extract from the peel of mangosteen fruit extracted using ethanol with a fixed ratio of 1 gram of natural materials 2 ml of solvent. Then tested the absorbance using Spectrophotometer UV Visible Shimadzu 1601 PC and determined the Voltages using I-V meter / elkahfi 100 of I-V can be seen the value of dye conductivity made from the peel of mangosteen fruit.

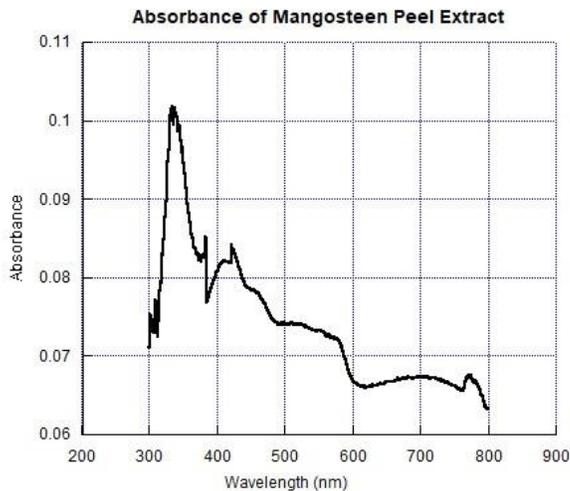


Figure 5 Absorbance of natural pigment extract of mangosteen peel.

Based on Figure 5, the mangosteen dye absorbance shows considerable wavelength, with a fairly good absorbance ability, showing the dye spectrum from the peel of the mangosteen fruit having an absorption spectrum similar to β -carotene, which has a major absorption wave at 380-520 nm, with a wave peak range of 350 nm and the amplitude reaches 0.1, so it can be enabled this mangosteen peel to absorb good sunlight energy and is able to maximize the performance of the DSSC.

Table 1. Resistivity (Ohm), Conductivity (Ohm⁻¹m⁻¹), and Flow (mA) of mangosteen peel.

Organic Materials	Condition	R(Ohm)	σ (Ohm ⁻¹ m ⁻¹)	I (mA)
Mangosteen peel	Dark	5.0×10^9	5.20×10^{-6}	3.21×10^{-5}
	Bright	5×10^8	2.34×10^{-5}	3.46×10^{-5}

The conductivity value of the mangosteen peel can be presented in Table 1. Table 1 can be determined that the current in the bright state is greater than in the dark. While the conductivity is greater in the dark than in the light.

DSSC was tested electrically with a Keithley 2602A measurement system. The current and voltage test results are shown in Fig. 6.

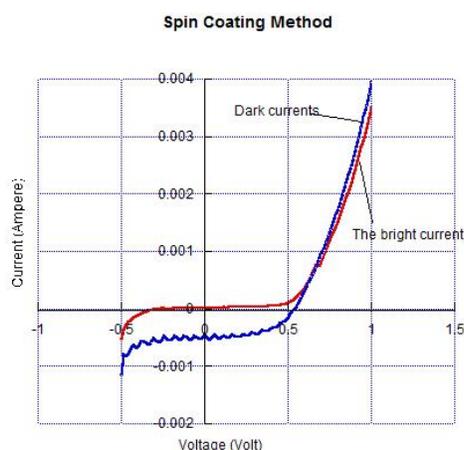


Figure 6 Keithley test results with Spin Coating method

Based on the graph of Keithley test results in Figure 6, we can determine the current (Isc) and voltage (Voc). So based on calculations that have been done the efficiency of pigment peel extract of mangosteen fruit [20], shown in table 2.

Table 2. Specification of Dye Mangosteen Peel

Method	Voc (V)	Isc (A)	Vmax (V)	Imax (A)	Fill Factor	Efficiency (%)
Spin Coating	0.565	0.0003	0.430	0.00048	$1,2 \times 10^{-7}$	0,092

From table 2, it can be seen that the efficiency produced by DSSC for the pigment of mangosteen peel with spin coating method reaches 0,092%. The current generated by

mangosteen peel extract is still very small is 3×10^{-4} resulting in low efficiency. Therefore there needs to be another material to improve its efficiency.

Conclusion

The measurement and analysis of the absorption spectra of natural dye extract of mangosteen peel have been done with the ratio of the mass of natural materials and the volume of solvent is kept steady. The results showed that the dye extracted from the natural material has an absorption spectrum similar to that of β -carotene having absorption at wavelengths between 380 - 520 nm and a wave peak in the range of 450 nm.

Measurements of I-V Meters / Elkahfi used the same voltage source of 9 volts to generate an electric current from the extract of the peel of the larger mangosteen fruit. The current in the dark gives 3.21×10^{-5} mA, while under irradiation gives 3.46×10^{-5} mA.

Measurements of the I-V meter also show the conductivity value of the extract of mangosteen fruit in the dark is 5.20×10^{-6} Ohm⁻¹.m⁻¹. The conductivity under irradiation is 2.34×10^{-5} Ohm⁻¹.m⁻¹.

The results of electrical current measurements and conductivity produced by mangosteen peel extract, this makes the mangosteen fruit needs to be investigated further as a DSSC sensitizer. Efficiency produced by DSSC for the pigment of mangosteen peel with spin coating method reached 0,092%. The mangosteen rind dye so that needs to be done further research with engineering and applications better.

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