



Plant Leaf Chlorophyll Based DSSC Solar Cell with ITO Transparent Nanoparticle Alloy

Muslimin^{1*}, Nurasyah Dewi Napitupulu¹, Nurulhuda Rahman²

¹ Department of Physics Education, Tadulako University, Palu 94118, Central Sulawesi, Indonesia

² Department of Agribusiness, Tadulako University, Palu 94118, Central Sulawesi, Indonesia

Corresponding Author Email: fisikamuslim@gmail.com

<https://doi.org/10.18280/ijht.410222>

ABSTRACT

Received: 2 March 2022

Accepted: 10 February 2023

Keywords:

DSSC performance, dye chlorophyll, annealing, ITO grain size

The optimization of the combination of the optical properties of transparent ITO nanoparticles with the optical and morphological properties of TiO₂ as well as the absorbing properties of chlorophyll dyes plays a very important role in the performance of DSSC solar cells. Therefore, it is necessary to characterize these properties to produce maximum DSSC solar performance. The stages of research are fabrication, characterization, and performance test. TiO₂ film was made by coating TiO₂ on ITO and soaking in chlorophyll dye. Alloy DSSC solar cells are characterized by optical, electrical and morphological properties. Characterization of optical properties, reflectance, absorption of chlorophyll dye using UV-Vis spectrometer. Electrical properties were characterized by measuring the current-voltage in the coating, DSSC solar cell performance and grain size morphology with XRD. Performance optimization of DSSC solar cells was obtained at the grain size of the transparent nanoparticle layer 54.87 at annealing temperature of 50°C and chlorophyll absorption power of 20.900 mg/l efficiency of 0.038%. The grain size is 48.71, annealing temperature is 60°C and chlorophyll absorption is 73.20 mg/l, the efficiency is 0.005%. The grain size is 44.46 the annealing temperature is 50°C and the chlorophyll absorption is 34.6 mg/l and the efficiency is 0.020%. The conclusion is that the grain size of transparent nanoparticles, annealing temperature and chlorophyll absorption affect the efficiency of DSSC solar cells.

1. INTRODUCTION

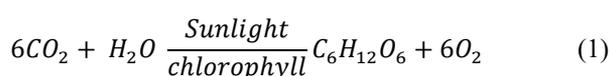
Solar cells are very expensive so they are not affordable by the public. For this reason, it is necessary to think about alternative solutions to produce solar cells in a simple, easy and relatively inexpensive way [1]. Conversion of sunlight energy into electricity can be done through the use of plant leaves as DSSC Dye of chlorophyll leaves [2, 3].

This research is very important considering that its output can contribute greatly to low-cost high efficiency electricity sources so that it can be used to overcome the national energy crisis that is currently happening in Indonesia [4].

Solar cells are a mechanism that works based on the effect of solar cells, photons from radiation are absorbed and then converted into electrical energy [5]. The electric charge in the material is created as a result of the absorption of the material.

A very promising energy source at present is Dye sensitized solar cell (DSSC) which is the most advanced solar cell generation technology [6]. The main ingredients of DSSC that will be made are leaves that contain high chlorophyll as a result of photosynthesis. Solar cells that use natural dyes based on chlorophyll have a higher tendency to degrade [7]. The content of chlorophyll in leaves will cause more electrons to flow, resulting in an electric current.

Chlorophyll absorbs sunlight energy into molecules and releases electrons from water molecules and protons from oxygen [8]. The reactions of photosynthesis are as follows:



When photosynthesis occurs, two electrons will be released, the more electrons will be released [9]. The liberation of electrons due to the photosynthesis process is shown in Figure 1.

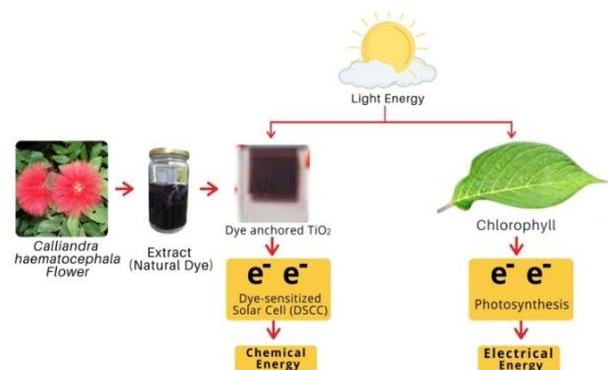


Figure 1. Electron liberation of chlorophyll leaves

The optimization of solar cell performance is not only determined by natural dyes which have high absorption or high chlorophyll content, it is also determined by the combination, namely the electrical and optical properties of the transparent conductor layer used [10, 11]. To optimize the fabrication of solar cells, it is inseparable from the transmittance of the transparent conductor, the Dye absorption power is used [12, 13].

The combination of the liberation of chlorophyll electrons with the transmittance level of the transparent conductor alloy

and the level of crystallinity of the formed layer greatly determines the performance of the solar cell. The grain size and the level of crystals formed greatly affect the rate of diffusion of the natural dye of chlorophyll.

The liberation of electrons in solar cells to improve the performance of solar cells is also largely determined by the transparent conductor used. A good transparent conductor based on the results of research on solar cells is a thin films that has high reflection. Anti-reflection on ITO used in previous studies was 87.67% with a grain size of 48.71Å nanoparticles. The photovoltaic performance obtained on papaya leaves with an efficiency of 0.69% [14]. In addition to transparent conductivity as well as concentration, grain size and duration of immersion in TiO₂, these parameters will change the pore size characteristic of the surface area so that it will increase the amount of dye absorbed, which implies an increase in the amount of light absorbed [15, 16]. As a result, pigment absorbance gives hope for chlorophyll plant pigments as good absorbent materials [17]. The dye proving some progress is DSSC TiO₂ [18]. To produce a DSSC that has high performance, it is necessary to optimize several constituent components including optimization of anti-reflection ITO, widening of the energy band and enlargement of grains formed by TiO₂, concentration of TiO₂, and duration of immersion in TiO₂. In this study, leaves containing high chlorophyll, namely, spinach leaves, cassava leaves and mango leaves will be used, with a reflection coating of Indium Tin Oxide (ITO) and DSSC TiO₂ [19, 20].

The results of preliminary studies that have been carried out by researchers [18] show that this method has the potential to be used in the development of DSSC solar cells. Therefore, optimization of several experimental parameters will be carried out affect the performance of solar cells, namely optical properties, electrical properties, dye concentration. Naturally, the concentration of electrolytes, annealing of all components that make up the solar cell will also influence the work of the solar cell [21, 22].

To obtain optimal DSSC solar cells, all of its constituent components are optimized related to optical properties, electrical properties, natural dye concentration, electrolyte concentration, annealing of the DSSC solar cells made.

2. LITERATURE REVIEW

Dye Sensitized Solar cell (DSSC) is a new generation of promising performance solar cells because the material is cheap and the manufacturing process is quite simple. The DSSC function is formed due to the interaction of the anode and cathode, TiO₂ is coated with a light-sensitive dye, then surrounded by a sea of electrolytes [21].

Natural dye to absorption photon energy is shown by the results of the study as shown in Figure 2.

Figure 3 and Figure 4 explain the process of electron flow. Photons with different energies in sunlight strike the cells and penetrate into the dye layer because the tin oxide (FTO) layer and TiO₂ nanocrystals are transparent to visible light. The holes produced by photon excitation remain on the molecule during the process because the dye is separated from all other energy levels. The hole is finally filled by electrons from the electrolyte ion. At the same time, the reduction of the oxidized dye by iodide gives tri iodide. The tri iodide diffuses to the counter electrode and accepts electrons from the external load, regenerates the iodide, and then the whole process will provide

a flow of electrons from the working electrode to the outside circuit.

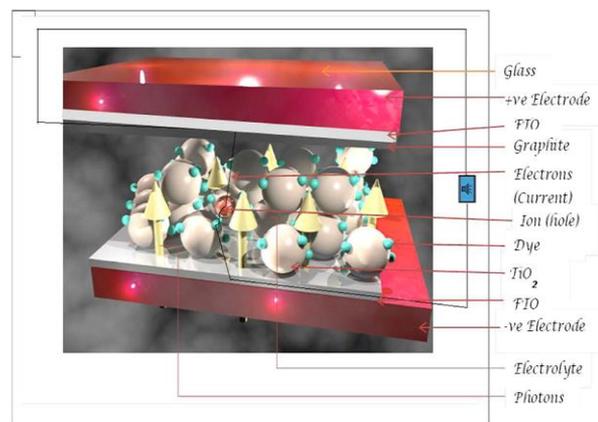


Figure 2. An enlarged cross-section of a dye sensitized solar cell

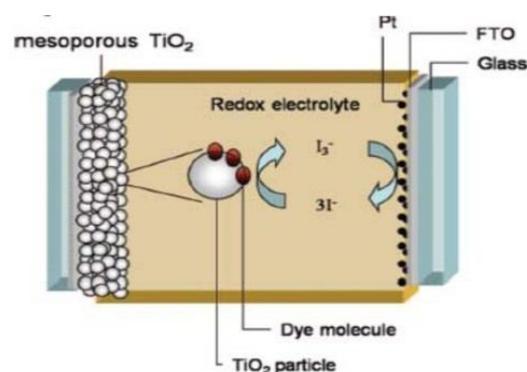


Figure 3. The working mechanism of DSSC

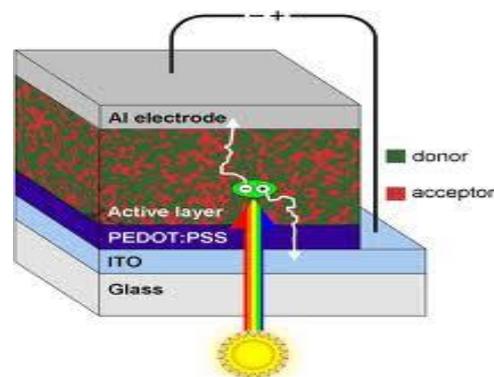


Figure 4. The working mechanism of DSSC

The electric current in the DSSC is generated from the injection of electrons ($S^*/TiO_2 \rightarrow S-/TiO_2 + e^- (TiO_2)$) after photo excitation of the dye molecules electrons by light radiation ($S + h\nu \rightarrow S^*$). by electron capture by the acceptor compound in the electrolyte ($3S^-/TiO_2 + I_3^- \rightarrow S/TiO_2 + 3I^-$). The mechanism of recombination (electron interception) occurs between electrons in the conduction band and compounds I_3^- in the electrolyte thereby reducing the value of the photon current produced by DSSC [23]. The absorption of the mulberry fruit dye extract solution that has been carried out is as shown in Figure 5. The best ability of the dye extract to absorb 510 nm type of light with a reddish purple color while the absorbed light is green light.

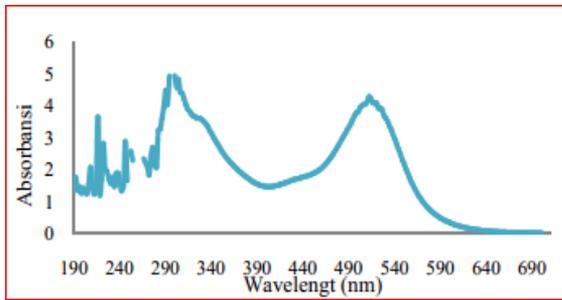


Figure 5. Mulberry fruit extract dye absorption

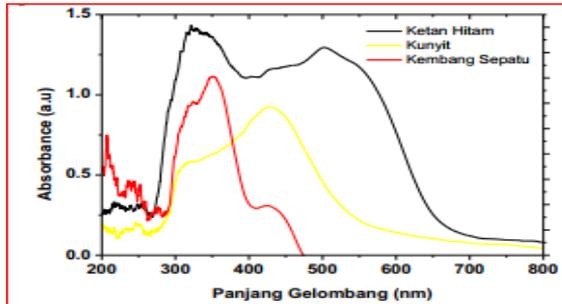


Figure 6. Andong dye absorption

The absorption spectrum of the dye is shown in Figure 5. The absorption spectrum of chlorophyll leaf extract is in wavelength 190 to 690 nm with a maximum absorption peak of chlorophyll a value at 300 nm and chlorophyll b at 520 nm. It is observed that the best dye absorption photo at a wavelength of about 300 nm, whereas TiO₂ absorbs best in the range of 300 nm to 450 nm and 520 nm.

The results of the absorption study in Figure 6 on black sticky rice, turmeric and hibiscus extracts at a wavelength of 200 nm to 800 nm. The maximum absorption was black sticky rice at a wavelength of 310 nm, turmeric 350 nm and hibiscus flower at 420 nm.

The results of research that have been carried out on the performance of DSSC solar cells from papaya leaf chlorophyll, the highest efficiency was obtained at the grain size of the substrate ITO 44.46, namely 0.00071% [24].

3. METHODS

3.1 Research flowchart

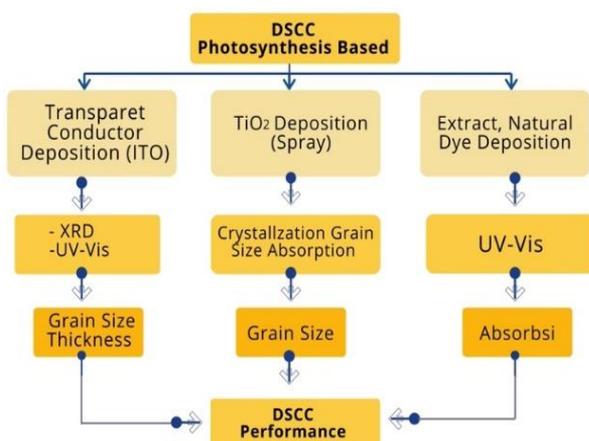


Figure 7. Research flow schematic

Figure 7 research flow, which is as above. Making DSSC based on photosynthesis using transparent conductors from ITO deposition, then coating TiO₂ on top of ITO. The results of coating TiO₂ on ITO will precipitate natural leaf extracts containing chlorophyll. These results were then analyzed using XRD and UV-VIS to see the grain size and layer thickness formed. Then also analyzed the crystal structure and absorption. Based on the results of the analysis above, then the resulting DSSC performance analysis is analyzed.

3.2 Cell panel fabrication process solar DSSC

3.2.1 DSSC layer manufacturing

(1) Dye Making

The principle of chlorophyll extraction from papaya leaves, mango leaves, and orange leaves is used to make dyes. Each leaf is weighed with the same weight of 10 g. Then each leaf was washed using distilled water and then dried at room temperature for 15 minutes. The clean chlorophyll leaves were then ground with a porcelain cup until smooth and then put into 50 ml of 70% alcohol solvent.

(2) Making TiO₂ Paste

In the manufacture of TiO₂ paste, 2.0-gram Polyvinyl Alcohol (PVA) was added to 13.0 ml of distilled water, then the mixture was stirred with magnetic stirring for about 30 minutes until the solution thickened and homogeneous. PVA serves as a binder in the manufacture of TiO₂ paste. Then 0.5 gram of TiO₂ powder was weighed, Then the suspension that has been made is added with TiO₂ powder in a ratio of 5 grams of TiO₂ spatula mixed with 15 drops or 0.75 ml of PVA, then filtered with gauze.

(3) TiO₂ Paste Coating

The TiO₂ layer was coated on the ITO glass measuring 2×2 cm in the form of an area for TiO₂ deposition, which was approximately 10 nm thick on the ITO surface. The sides of the ITO were affixed with tape as a barrier. The previously prepared TiO₂ paste was placed on a non-stick ITO glass surface, and then the paste was smoothed over the ITO surface. The thickness of the TiO₂ deposition layer corresponds to the thickness of the tape used. The TiO₂ layer formed was allowed to stand for 30 minutes, then the tape was removed, and the layer was dried at room temperature to dry. To make the TiO₂ layer more crystallized, annealing was carried out using an electric heater coated with aluminum foil. Annealing was carried out for each dye at a temperature of 50°C. After that cooled to room temperature, each sample with each dye was dripped on the ITO glass deposited with TiO₂ as much as 4 drops until the TiO₂ layer was completely covered with dye, and dried. Then rinsed with NaCl and dried.

(4) Electrolyte Administration

Provision of electrolytes by means of drops using a pipette as much as 4 drops or as much as 0.25 ml. Electrolyte solution is used as electron transport from carbon to dye.

3.2.2 DSSC performance analysis

Testing of DSSC cells was carried out based on the direct lighting method under light. Cell performance and efficiency are obtained when the solar cell object is exposed to light with a certain intensity at the top electrode (anode).

In this study, the light source used was light rays with a fixed intensity. The recorded DSSC output results are the open-circuit voltage (V_{oc}) and short-circuit current (I_{sc}) of the DSSC. DSSC output parameters include open circuit voltage (open circuit) V_{oc}, short circuit current I_{sc} can be known after taking measurements.

4. RESULTS AND DISCUSSION

4.1 Layer characterization

Samples that have been fabricated will be analyzed by UV-Vis and XRD results to obtain coating thickness, reflectance, absorption, grain size and crystallinity level of the coating.

4.2 Chlorophyll dye absorption

The chlorophyll absorption test was carried out using a Shimadzu UV-1601 Spectrophotometer at a 300-800 nm wavelength. The absorbance of chlorophyll was measured using a spectrophotometer at λ_{649} nm and λ_{665} nm. Then from these results will be calculated, the amount of chlorophyll *a*, chlorophyll *b*, and total chlorophyll contained in the dye using the equation,

$$\begin{aligned} \text{Chlorophyll } a &= 13.7 (\text{OD}_{665}) - 5.76 (\text{OD}_{649}) \\ \text{Chlorophyll } b &= 25.8 (\text{OD}_{649}) - 7.60 (\text{OD}_{665}) \\ \text{Total chlorophyll} &= 20.0 (\text{OD}_{665}) + 6.10 (\text{OD}_{665}) \end{aligned} \quad (2)$$

Description:
OD = Optical density

Calculation of the adsorption of chlorophyll dye on papaya leaf (sample A), orange leaf dye (sample B) and mango leaf dye (sample C) was carried out by Eq. (2). The results of the analysis of the absorption of chlorophyll dye as shown in Table 1.

Table 1. Absorption of chlorophyll dye

Dye	Absorbance		Chlorophyll mg/l		
	λ_{665}	λ_{649}	<i>a</i>	<i>b</i>	total
Papaya Leaf	2.82	2.79	2.79	49.99	73.20
Lime Leaf	2.56	2.92	18.25	55.29	69.01
Mango Leaf	2.71	3.01	19.27	56.46	72.56

The analysis results showed that the number of leaves that released chlorophyll and then bound to TiO₂ increased in papaya leaves. These results indicate that papaya leaf chlorophyll will absorb more light so that DSSC will produce the most significant power with papaya leaf chlorophyll.

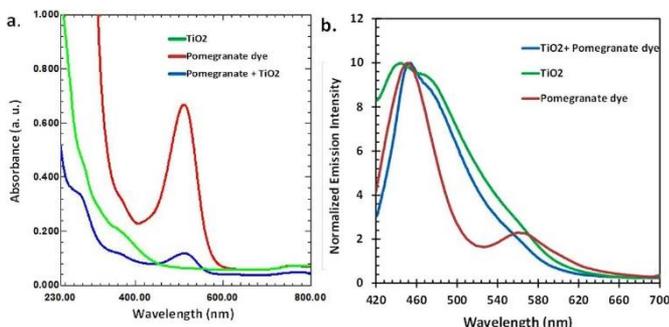


Figure 8. Absorption TiO₂ (green), dye (red) and sensitized TiO₂ film on ITO glass (blue)

The absorption of TiO₂ (green), dye and TiO₂ film (red) on the ITO glass (blue) as shown in Figure 8. In the figure it can be seen that the maximum peak in the formed layer is TiO₂ at a wavelength of 555 nm, or occurs in chlorophyll *b*.

4.3 Reflectance and transmittance ITO transparent coating

Figure 9 shows the results of the UV-Vis spectra of the reflectance and transmittance of one of the transparent layer samples. The transmittance and reflectance are shown in Figure 9. Maximum transmittance is 83% at a wavelength of 670 nm and maximum reflectance is 37% at a wavelength of 572 nm.

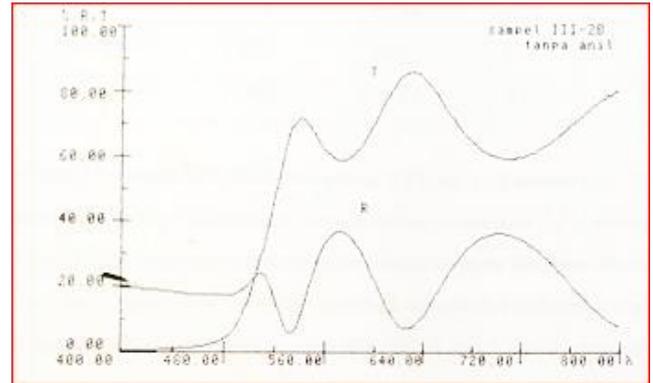


Figure 9. Transmittance and ITO Reflectance

4.4 Reflectance, transmittance and layer absorption

The reflectance (R) and transmittance (T) spectra, as measured by a spectrometer, will first be searched for the layer thickness using the equations $2nd = (m+1/2)\lambda$ and $2nd = m\lambda$. Substituting the two equations, we get:

$$nd = \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_2} \quad (3)$$

With *n* refractive index, *d* thickness, λ_1 the maximum wavelength of the first peak, λ_2 the wavelength of the second peak. The refractive index used in Table 2 is the average refractive index of Table 2, namely the peak of the first wave and the peak of the second wave, which is 2.50. Based on Eq. (3), the thickness of the thin layer is as follows:

$$d = \frac{704 - 572}{2,5 (704 - 572)} = 220,267 \text{ nm}$$

The data needed to determine the layer thickness are as in Table 2. The results of the analysis in the table, it is obtained that the thickness of the transparent layer is 631.22 nm.

Table 2. Results of layer thickness analysis

Peak	Wave (nm)	Reflectance (%)	Refractive index
Max	704	37.69	2.505
Min	627	7.48	2.483
Max	572	37,94	2.492
Min	532	6.05	2.528
Max	508	24.37	1.616

4.5 Large grain layer

Measurement of X-Ray Diffraction (XRD) can be seen the crystal structure and grain size formed on the DSSC layer. To find the crystal orientation obtained by comparing with the

reference taken from the "Joint Committee on Powder Diffraction Standard". XRD results are shown in Figure 6.

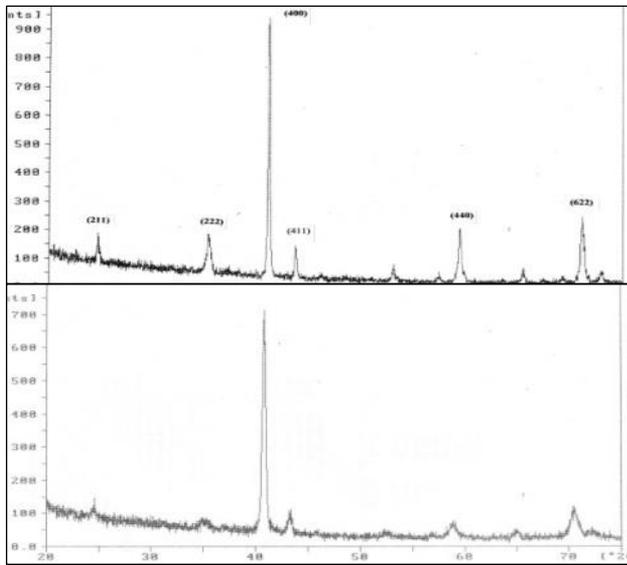


Figure 10. ITO Thin Layer XRD Results

Based on Figure 10. It can be seen that the layer formed has a very thin half peak, so it can be said that the layer is a crystal. The formation of a layer in the form of crystals is caused by annealing at the coating time. Annealing can cause free atoms in the form of oxygen and titanium and other atoms present at the time of coating to bond or undergo diffusion due to heating.

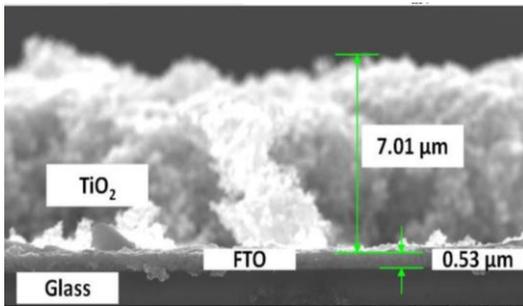


Figure 11. Images of TiO₂ and Dye/TiO₂ samples [21]

The coating scheme of the results of research on DSSC solar cells that has been carried out by researchers is shown in Figure 11. The ITO material is formed on top of ordinary glass, then the layer on the ITO layer forms a TiO₂ layer. In this study also carried out with the same scheme.

Based on the XRD results of the transparent conductor layer, the grain size formed in the thin films can be determined by the equation $D = \frac{0.9\lambda}{\beta \cos \theta}$ [25] where D is the grain size (Å), wavelength, width The diffraction line is measured at half the maximum intensity (radians) and the diffraction angle. XRD data, the grain size of each sample=1.5406 (Å) shown in Table 3.

The results of the grain side analysis showed that the sample with the giant grain was the ITO layer which was used to dye papaya leaves. The size of the grain side can cause the lattice to be more giant so that it is easier for the dye to enter the gaps between ITO grains. The more dye that diffuses in the gaps of the ITO layer will cause more electrons to be liberated, which in turn produces better electrical power.

Table 3. Calculation results of thin films grain size

Sample	Angle (2θ)	Peak Width (B) (deg)	Grain Size (Å)
A	41.02	0.100	54.87
B	41.03	0.330	44.46
C	41.02	0.318	48.71

Table 4. Power analysis of chlorophyll leaf DSSC function large grain transparent particles

ITO Grain Size (Å)	Dye Absorption (mg/l)	Power (10 ⁻⁹) watt	η (%)
54.87	73.203	0.05064	0.0506
44.46	20.900	0.022994	0.0229
48.71	34.131	0.038368	0.0387

Based on the analysis of each ITO grain size and dye absorption in each extract of papaya leaf, orange leaf and mango leaf, the electrical power and efficiency are obtained as shown in Table 4.

DSSC conversion efficiency value resulted in the highest DSSC efficiency by papaya leaf extract. This result is proportional to the highest absorption for the higher the contribution of the absorbed photons, so the conversion efficiency of the DSSC is also higher. This is also supported by the UV-Vis characteristics of the adsorption dye on the surface of TiO₂ which showed the highest absorption in the orange leaf extract. This is also in line with the grain size of ITO from the XRD characterization of the substrate used for TiO₂ coating. ITO grain size associated with TiO₂ grains can be diffused in the ITO grain gap. The results revealed that TiO₂ is the best semiconductor material for applications in DSSC and its efficiency is related to its properties such as band gap, electron mobility, electron injection rate, and static dielectric constant [24]. Semiconductor thin layers play an important role in the work of DSSC.

Another study reported that the efficiency of DSSC was higher due to higher dye absorption on the TiO₂ surface [19]. The results can be explained that the absorption of photons that are converted into electrons has the most at the highest absorption power. The absorption dye on the surface of TiO₂ dominantly acts in the visible region. The efficiency is high because the output current has the most. This situation has an impact on the high conversion efficiency of papaya leaves.

The overall work of the DSSC depends on the light absorption ability of the dye sensitizer and the diffusion of electrons emitted through the TiO₂ film. Its high absorption coefficient allows adsorption to the surface of TiO₂. This absorption facilitates the transfer of substituted electrons from the dye to the TiO₂ conduction band, which ultimately increases the efficiency of the DSSC [21, 26].

The total efficiency of a DSSC or DSSC performance depends on the properties, optimization, and compatibility of each of the components of the DSSC and, more specifically, the photo-anode, which plays an essential role in the charge generation and transfer process. To improve the power conversion efficiency of DSSC, titanium dioxide nanostructured pores with wide band gaps and large exciton binding energies are commonly used in photo-anode construction. The large surface area or grain size of the nanostructured ITO/TiO₂ ensures the absorption of many dye molecules for efficient harvesting of radiant energy. Strong dye absorption into the nanostructured TiO₂ is required for efficient electron injection into the conduction band of the TiO₂ semiconductor.

5. CONCLUSIONS

Based on the results of the analysis and performance testing of the efficiency of the DSSC solar cell, the optimization of the performance of the DSSC solar cell was obtained at the grain size of the transparent nanoparticle thin films of 54.87 Å with annealing temperature of 50°C and chlorophyll absorption power of 20.900 mg/l, the efficiency of DSSC solar cells was 0.0380%. The grain size of 48.71 Å absorption power of chlorophyll 73.20 mg/l obtained DSSC solar cell efficiency of 0.0050%. Grain size 44.46 absorption power of chlorophyll 34.6 mg/l solar cell efficiency DSSC 0.0239%. So, it can be concluded that the grain size of transparent nanoparticles, and the absorption of chlorophyll dye affect the efficiency of DSSC solar cells.

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NOMENCLATURE

Greek symbols

η	dssc efficiency, %
β	XRD result half peak width, deg
θ	scattering angle, rad
λ	wavelength, nm
Å	layer thickness unit
<i>a</i>	absorption of chlorophyll <i>a</i> dye, mg. l ⁻¹
<i>b</i>	absorption of chlorophyll <i>b</i> dye, mg. l ⁻¹
R	reflectance, %
T	transmittance, %
m	Wavenumber
n	refractive index
D	grain size, nm

Subscripts

ITO	indium tin oxide
TiO ₂	titanium dioxide
OD λ 665	optical density wavelength 665
UV-Vis	ultra violet visible
XRD	X-ray diffraction
Voc	open circuit voltage, mV
Isc	short-circuit current, mA