

Dye Sensitized Solar Cell (DSSC) Efficiency Derived from Natural Source

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Abstract: Dye Sensitized Solar Cell (DSSC) with natural dye from Leunca fruit (*Solanum Nigrum L*) and Jamblang fruit (*Syzygium Cumini L*) extract as sensitizer has been created. This DSSC is composed of ZnO doping TiO₂ using milling tool for 30 minutes which would be used as photoelectrodes. This study used two natural dyes which are Leunca and Jamblang fruit. The characterizations were performed to examine crystal structure of ZnO-TiO₂ with XRD, to measure ZnO-TiO₂ particle size with SEM, to examine optical properties from the dye using UV-Vis spectrophotometer, and to run an electrical test to find the efficiency from DSSC. The results indicate that the use of Jamblang fruit as sensitizer is better than Leunca fruit. This is because Jamblang fruit extract has light absorption area on the range of 250 - 800 nm which is higher absorbance than that of Leunca fruit. In addition, the milling time applied was found to be not long enough to produce semiconductor with smaller crystal size. The electrical test result shows Jamblang fruit based DSSC performance is better than Leunca fruit. The maximum power output values are 4.01×10^{-8} Watt with the efficiency of 22.57×10^{-4} % and 2.16×10^{-7} Watt with efficiency of 6.02×10^{-4} % when radiated with Halogen lamp and sunlight, respectively. Aside from the fact that the preparation technique is relatively easy, natural dye material can be found abundantly in nature and its price is also cheap, hence this study is very promising. However, several changes are needed to gain better results. The brief of this research is to develop alternative energy from solar energy, so that its use can be maximized as environmentally-friendly energy source.

Keywords: DSSC; Jamblang Fruit; Leunca Fruit; ZnO-TiO₂.

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I. INTRODUCTION

Nowadays people are very dependent on electrical energy in performing every activity. However due to the large number of human needs, resources demand is also increasing, but the availability is very limited. Therefore, currently there are many alternatives that can help human in energy that have been developed. The utilization of sun energy is one of the alternatives that has been developed. Solar energy emitted from the sun, received by the earth only 69% from the total energy emitted. Meanwhile, solar energy supply absorbed by the Earth's surface reaches 3×10^{24} J every year, this is equals to 2×10^{17} Watt, or can be said as the total energy is equals to 10,000 times of energy consumed currently in the whole world [1]. Thus, solar energy has the chance in its development as an alternative energy in the future.

One of the development of solar energy is Dye-Sensitized Solar Cell (DSSC) which is one kind of the solar cells developed by Prof. Gretzel and Brian O'Regan in 1991 [2]. DSSC can change light into electrical energy based on sensitization of wide band gap semiconductor materials [3]. DSSC gathers global attention because of the cheap and easy production. Silicone based solar cell had been developed before. The difference between DSSC with silicon solar cell is charge separation and light absorption performed separately. Light absorption performed with coloring molecule while charge sep-

aration performed with nanocrystal semiconductor materials [4]. Inside DSSC, there are several important components, including a pair of electrodes (counter and working electrode), electrolytes, and coloring solution [5].

Particle size from semiconductor material affects efficiency of DSSC. This is because the smaller the particle size, the wider surface area of semiconductor layered on TCO glass is. Therefore, it would increase absorbed dyes and electrons that have been through excitation.

Based on several previous studies, this study will make DSSC with photoanode pieces layered by TiO₂ semiconductor along with ZnO. Although TiO₂ is more expensive than ZnO, but the bandgap from TiO₂ is good enough for DSSC. By employing TiO₂ along with ZnO, we expect that the bandgap is not too wide to obtain good photocatalysts effect. The composition that would be used for ZnO:TiO₂ in this study is 20%:80%. This refers to previous study where the best efficiency was found at 20%:80% ratio of ZnO:TiO₂ with 0.277% efficiency [6].

At first, both semiconductor materials are combined using milling technique with High Energy Milling (HEM) to obtain nanocrystal structure with wider band gap so that the electron mobility would be more free. This semiconductor material will also be tested by us with XRD and SEM to view the phase from the material, crystal structure, and chemical composition. Meanwhile, the electrode counter part would

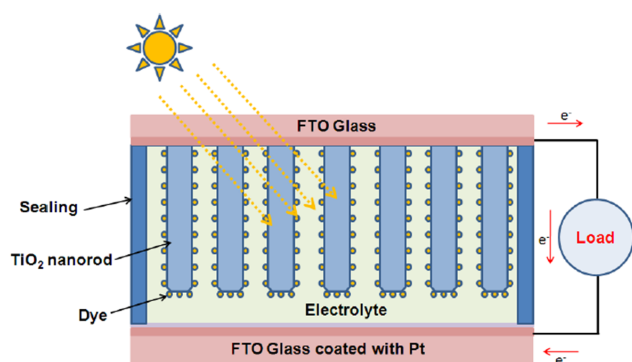


FIG. 1: DSSC structure and work principle [5].

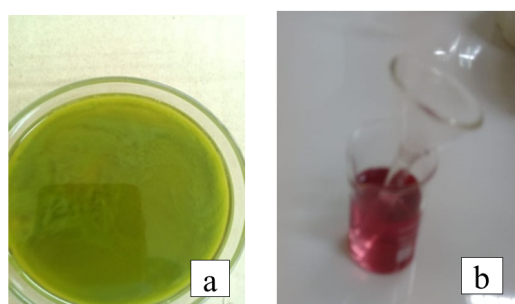


FIG. 2: Finished dye product of (a) Leunca Fruit and (b) Jamblang Fruit.

be made from candle soot. As this study employs natural material-based dye, thus it is expected to generate environmentally friendly energy.

Materials that would be used for natural dye in this study are Leunca and Jamblang fruit. The reasons to use these materials are because their chlorophyll composition. Moreover, to the best of our knowledge there are not so many DSSC studies using natural dye with basic ingredients such as Jamblang and Leunca fruit in past. This study is expected to give an insight of the efficiency of Jamblang and Leunca fruit based sensitizer on DSSC. Here, their dye would be used to prepare sensitizer of DSSC. On the previous study conducted by M. Gretzel using ruthenium complex material resulting 11-12% efficiency [7]. However, this material supply in nature is extremely rare and not environmentally friendly. So, the alternative dye using material from nature, like fruits, vegetables, flowers, and leaves are sought.

The dye is characterized with UV Vis spectrophotometer to measure wavelength and absorption value, and also to estimate gap energy value of the dye. After DSSC successfully assembled and circuit is arranged, several characterizations are carried out, namely voltage and current testing. The ZnO-TiO₂ paste is prepared using milling technique which is expected to create better efficiency when applied into DSSC compared to ZnO-TiO₂ without milling technique reported in previous study [6]. The novelty here is creating DSSC using dye materials from Jamblang fruit and Leunca fruit with

photoelectrode materials using ZnO-TiO₂ made from milling technique.

II. METHOD

A. Tools and Materials

In this study, tools used were beaker glass, glass stirring rod, petri dish, measuring cup, magnetic stirrer, spatula, mortar, digital balance, blender, weighing paper, filter papers, oven, measuring and dropping pipettes, digital multimeter, scotch tape, paper clips, scissor, cutter, halogen lamp 1000 W, potentiometer, luxmeter, hygrometer, x-ray diffraction, UV-Vis spectrophotometer, and spex shaker mills.

Meanwhile materials used were Indium Tin Oxide (ITO) glass, TiO₂ (Merck), ZnO (Merck), PolyEtilen Glicol liquid (PEG) 400 (Merck), Poly Venil Aldehyde (PVA) 500 (Merck), metanol (CH₃OH) (Merck), Alcohol 96%, Potassium Iodida (KI) (Merck), iodium (Merck), aquades, Jamblang fruit, and Leunca fruit.

B. Preparation of Natural Dye Sensitizers

Fresh Jamblang and Leunca fruit were extracted by mashing it first using blender into 40 ml aquades which is then filter using filter paper. The resulted extract of Leunca fruit is then added with 100 ml methanol and that of Jamblang fruit with 100 ml aquades. The resulted dye prepared from Leunca and Jamblang fruit can be seen in Fig. 2.

C. Preparation of ZnO-TiO₂ Paste

In ZnO-TiO₂ paste production, there are two materials that needed to be made, which are ZnO-TiO₂ powder and gel solution. ZnO and TiO₂ were milled at first with composition ratio of 80% (ZnO) : 20% (TiO₂) for 30 minutes. This composition is based on that acquire the best efficiency in previous study [6]. Furthermore, gel production from the mixture of 0.4 gr PVA and 8 ml aquades was prepared using magnetic stirrer for an hour, with 80°C temperature and at the speed of 300 rpm. After ZnO-TiO₂ powder was already milled and gel had been made, both of those materials were mixed using mortar for 30 minutes until it turned into paste.

D. Preparation of Electrode and Counter Electrode

ITO glass is cut with 2 cm × 2 cm size for each electrode. The side part of ITO glass is attached with scotch tape, so only the center part of glass with 1.5 cm × 1 cm size remains. The center part of ITO coated with ZnO-TiO₂ using doctor blading method, then heated inside an oven for 15 minutes (See Fig. 3a). Next, the electrode is soaked in a dye for 24 hours. Meanwhile for the counter electrode, the center part of ITO glass is coated with carbon made of candle flame, where

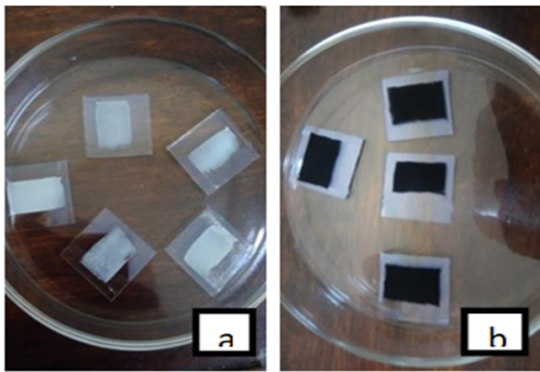


FIG. 3: (a) ZnO-TiO₂ Photoelectrode and (b) counter electrode from the candle flame.

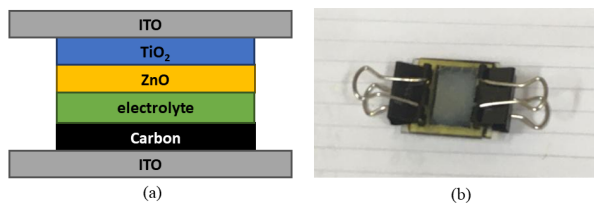


FIG. 4: (a) DSSC Structure and (b) DSSC prototype.

the ITO glass is burned until black color appeared on the ITO glass.

E. DSSC Assembling

Electrolyte solution must be made first before DSSC assembly. Electrolyte is made with 8.3 gr KI, 1.26 gr iodine, and 100 ml PEG mixture [9], which were mixed using magnetic stirrer for 30 minutes with 80°C temperature and at the speed of 300 rpm. Electrolyte solution that had been made was then stored in glass bottle coated with aluminum foil in room temperature to prevent it exposed to light.

DSSC is composed of several components with a structure presented in Fig. 4. The first part is ZnO-TiO₂ photoelectrode which previously soaked in dye, then electrolyte solution for 2 ml was added. After that, the counter electrode is placed above the photoelectrode where the carbon coated part must be attached to the photoelectrode. After being assembled like a sandwich, both electrodes are clamped using paper clip on left and right sides, as shown in Fig. 4.

F. Characterization and Measurement

To know the performance of DSSC, several tests were conducted. Absorption spectrum from dye is examined using UV-Vis spectrophotometer. Crystal structure and crystal size from ZnO-TiO₂ photoelectrode layer examined with X-Ray

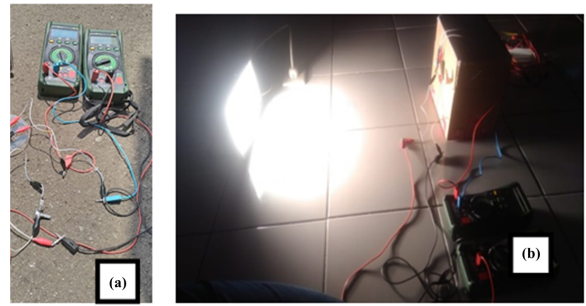


FIG. 5: DSSC electric test on (a) sunlight and (b) halogen lamp.

Diffraction (XRD), and the morphology observed with Scanning Electron Microscope (SEM). DSSC efficiency is measured with different light sources, which are 1000 Watt halogen lamp and sunlight. The DSSC efficiency is estimated using Eq. 1 [9].

$$\eta = \frac{P_{max}}{P_{light} \cdot A} \times 100 \quad (1)$$

Where P_{max} is the maximum power output obtained from multiplication result of maximum current output and maximum voltage output. Then A is an area surface of DSSC coated photoelectrode part and P_{light} is sunlight power measured using luxmeter, where 1 lux value equals to 0.0079 W/m² [8].

Electricity test used six variations of resistors, which are 0, 2, 5, 10, 50, and 250 KΩ. In the test with halogen lamps, the intensity of light used is 1,500 lux, while in sunlight the intensity of light used is in the range of 15,000 - 150,000 lux because sunlight is polychromatic so it is difficult to determine the intensity of light used.

In addition to experimentally calculated efficiency, this study also calculates theoretical efficiency which will later be compared between those two values. The equations used for theoretical calculations are expressed in Eq. 2 and Eq. 3.

$$I = I_L - \frac{V - R_s I}{R_{sh}} - I_0 \left\{ \exp \left[\frac{q(V - R_s I)}{nkT} \right] - 1 \right\} \quad (2)$$

$$I_0 = \frac{I_L}{\exp \left(\frac{V_{oc} q}{nkT} - 1 \right)} \quad (3)$$

Where I is the current that you want to find the value, V is the voltage obtained from the test results, I_L is the current strength whose value is equal to I_{sc} , I_0 is the saturation current of the diode, R_s is the series resistance, R_{sh} is the parallel resistance, q is the electron charge (1.602×10^{-19} C), k is the Boltzmann constant (1.38×10^{-23} J/K), T is the DSSC temperature during the test, and n is the diode ideality factor [10].

The circuit of electricity test on DSSC can be seen on Fig. 5, but for the theoretical calculation here using mathematical model with circuit equivalent to one diode [8]. The use of mathematical modeling like this is due to the standard DSSC circuit using a diode equivalent circuit [10].

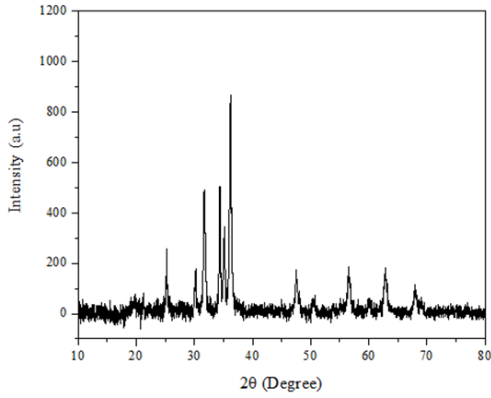


FIG. 6: XRD pattern result on ZnO-TiO₂ paste layer.

TABLE I: XRD characterization data on two representative peaks of ZnO and TiO₂ phase

Phase	2θ(°)	FWHM (rad)	Particle size (nm)
TiO ₂	25.27	0.004915844	30.178
TiO ₂	62.80	0.008518122	19.906
ZnO	34.38	0.005348467	28.330
ZnO	36.21	0.006430022	23.685

III. RESULTS AND DISCUSSION

A. The Phase Composition of the ZnO-TiO₂ Paste Layer

Analysis of XRD data using General Structure Analysis System (GSAS) and Match II software shows that ZnO-TiO₂ paste have three phases, which are ZnO, TiO₂, and In₄O₁₂Sn₃ with the composition of 83.5 %, 11.4 %, and 5.1 %, respectively. ZnO composition is higher than TiO₂ because the ratio of ZnO:TiO₂ is 80%:20% or equal to 2.22 gr ZnO and 0.56 gr TiO₂. The present of In₄O₁₂Sn₃ is because the sample being tested is in the form of a paste coated on ITO glass. Thus the peaks of ITO glass is detected by XRD. The resulted XRD data that was processed using GSAS can be seen in Fig. 6.

The ZnO crystal is detected around diffraction angles of 31.1725, 34.3824, 36.2100, 47.5231, and 67.9515°. Meanwhile for TiO₂ crystal is detected around diffraction angles of 25.2670 and 62.7978°. Crystal structure of ZnO and TiO₂ was found to be hexagonal and tetragonal, respectively.

The crystal size can be calculated using the Scherrer equation as presented in Eq. 4 [11].

$$D = \frac{0.94\lambda}{FWHM \cos \theta} \quad (4)$$

Where D is crystal size (nm), FWHM is highest peak half width size (radian), λ is $CuK\alpha$ wavelength with the value of 0.154 nm, and θ is the used diffraction angle (radian).

Crystal size greatly affects DSSC performance, because the smaller the crystal size the more dye attached to the semiconductor material, and as the result it will absorb more photon. Compared to several previous studies, the crystal size in this

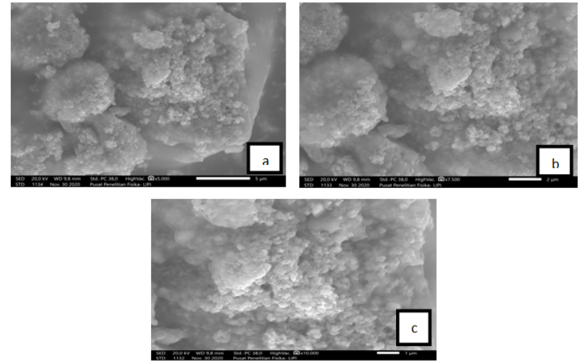


FIG. 7: SEM images of ZnO-TiO₂ surface layer with magnification of (a) 5,000×, (b) 7,500×, and (c) 10,000×.

study is still big. In the study of Aksoy *et al*, the ZnO doped Li had a crystal size of 5-7 nm [5]. Another study by Kumila *et al* with nanosized TiO₂, the crystal size was reported to be 10.5 nm [12]. The milling technique was carried out to get a smaller crystal size, but in this study the crystal size was small enough. This is because the milling time is only 30 minutes. In a study conducted by Puspa *et al*, the effect of milling time on crystal size was that the longer the milling time the smaller the crystal size [13].

B. Morphology of the ZnO-TiO₂ Paste Layer

Thin layer on DSSC has an important role in DSSC circuit, because in this part is where the light absorption occurred which later would be converted into electrical energy. The morphology of this material greatly affects how DSSC works later. Various kinds of morphology on this thin layer would affect the performance. Image result from SEM on ZnO-TiO₂ paste layer with the magnification of 5,000×, 7,500×, and 10,000 × can be seen in Fig. 7.

If we see Fig. 10, the SEM images of ZnO-TiO₂ layer have the form of porous spherical particles. In addition, in this thin layer there is a lot of agglomeration due to the preparation process of semiconductor paste using a binder in the form of a gel from a mixture of PVA and aquades. The amount of binder composition affects the amount of agglomeration formed [14]. As the result of this agglomeration, the efficiency produced by DSSC is not good enough because of the resistance to electron mobility. Therefore, the value of the voltage and current density becomes smaller due to this agglomeration [15].

C. The Absorption Wavelength of Jamblang and Leunca Dye

The absorbance spectrum can be examined with UV Vis spectrophotometer on the range of 250-800 nm wavelength for both Leunca and Jamblang fruit. The peak was obtained at a wavelength of 285 nm for Leunca fruit and 530 nm for Jamblang fruit, while the highest absorbance value for Leunca

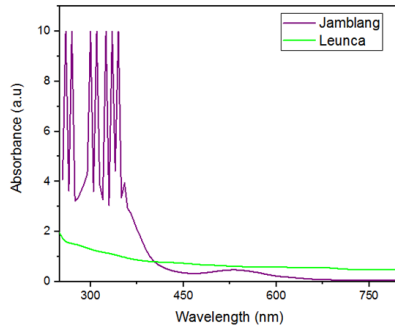


FIG. 8: UV Vis test result chart for Leunca fruit and Jamblang fruit.

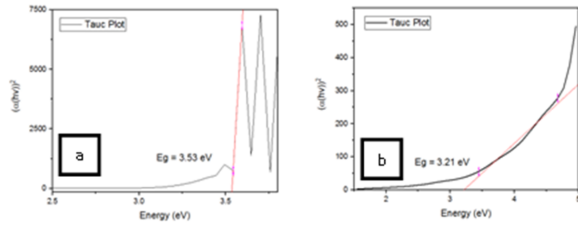


FIG. 9: Tauc Plot Graph for Jamblang fruit (a) and Leunca fruit (b).

fruit is 1.94 *a.u.* and for Jamblang fruit is 10.00 *a.u.* Based on absorption wavelength, Leunca fruit dye can absorb more light on the range of UV spectrum with quite high absorbance value, while Jamblang fruit dye can absorb light on visible light spectrum. Both of these materials have a fairly wide wavelength, so they can absorb a wide range of light spectrum. The results of the UV-Vis spectrophotometer can be seen in Fig. 8.

The energy gap of the two dyes can be determined using the Tauc plot method as represented in Eq. 5.

$$\frac{(\alpha hc)^2}{\lambda} = K \left(\frac{hc}{\lambda} - E_g \right) \quad (5)$$

$$\alpha = \frac{2.303A}{l} \quad (6)$$

Where h is Plancks constant (6.63×10^{-34} J.s), α is absorbance coefficient, A is absorbance, l is the thickness of cuvette used, K is the comparison constant, c is the speed of light (3×10^8 m/s²), λ is the wavelength, and E_g is energy gap [16].

The results of the energy gap calculation using the Tauc plot method can be seen in Fig. 9. There is one point where the energy gap value is taken, but the data used is the interval of the energy gap value at a wavelength of 250-800 nm as shown in Table 2.

The utilization of dye as a sensitizer must have small energy gap than that of the semiconductor material. This is because charge separation happened on DSSC. When emitted photon energy from the light source absorbed by dye, the electrons in

TABLE II: The UV-Vis measurement results of Jamblang and Leunca Dye

Dye	Wavelength (nm)	Energy Gap (eV)
Jamblang	250 - 800	4.96 - 1.55
Leunca	250 - 800	4.95 - 1.54

dye would go through excitation from HOMO (Highest Occupied Molecular Orbital) condition to LUMO (Lowest Unoccupied Molecular Orbital) which then electron would be transferred to conduction band in semiconductor [5].

The semiconductor material used is ZnO-TiO₂, where energy gap from both of those materials is not that much of different, which is 3.2 eV [17]. Characteristic from bandgap greatly affects the value of the absorption coefficient of light and the rate of electron-hole recombination [18].

Based on Table 2 and Fig. 9, one can see that the energy gap of Jamblang and Leunca fruit are not too different and the value is a little bit bigger than those of ZnO and TiO₂. This energy gap value causes the DSSC to be less than optimal to generate electricity. The absorbance of Leunca dye is highest at UV light wavelengths, while Jamblang fruit dye is found in visible light. So that, when radiated with sunlight, Jamblang dye absorbs more light.

D. DSSC Performance

In this study, the DSSC efficiency was calculated experimentally with Eq. 1 and theoretically with Eqs. 2 and 3. Comparison of the obtained results can be seen in Table 3.

By looking at the UV-Vis data and electrical testing obtained, the best dye used is a dye made from Jamblang fruit. Jamblang dye has wavelength absorption from UV to visible light. So that if it is used as a sensitizer in a solar cell, it will be better than Leunca dye. Moreover, the gap energy of Jamblang dye is smaller than Leunca fruit. The photoelectrode that had been made using ZnO doped by TiO₂ produces a fairly good layer with a porous spherical morphology. From the XRD results the photoelectrode crystal size is still not small enough because the milling time used is only 30 minutes.

However, there are still many shortcomings in terms of performance. Where the output voltage and current from the DSSC is still very small when compared to other similar studies. There are several factors that make the efficiency of this DSSC small. First, the liquid electrolyte is volatile and leaks. In the electrolyte gel made without using chloroform, only polyethylene glycol (PEG) was used. Whereas in a study conducted by Kumila *et al* using gel electrolytes that were not easy to evaporate and leak [12]. The second reason is that there is agglomeration in the photoelectrode layer which results in resistance to electron mobility. And the third, the milling time is not long enough.

TABLE III: DSSC efficiency calculated result experimentally and theoretically

Dye	Illumination	Lux	V_{OC} (mV)	I_{SC} (μA)	Efficiency ($\times 10^{-4}$ %)	
					Theoretic	Experiment
Leunca	Sun light	11700	167.76	1.24	8.80	8.60
Leunca	Halogen lamp	1500	13.25	1.45	0.73	2.40
Jamblang	Sun light	30300	142.02	5.08	11.24	6.02
Jamblang	Halogen lamp	1500	39.46	2.16	18.91	11.57

IV. SUMMARY

Dye Sensitized Solar Cell (DSSC) using photoelectrode from ZnO-TiO₂ mixture with sensitizer materials from Jamblang and Leunca fruit was successfully fabricated. Both Jamblang and Leunca fruit can be used as sensitizer on DSSC. Better DSSC performance was found when the sensitizer is prepared from Jamblang fruit. As Jamblang dye has bigger absorbance along with smaller gap energy than that of Leunca dye. Even though the DSSC in this study had been made successfully, it turns out the efficiency is still relatively small and

need further research to improve the deficiencies, such as by increasing the milling time, preventing the agglomeration of the photoelectrode layer and the leakage of the electrolyte gel.

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