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Dye-Sensitized Solar Cell (DSSC) Fabrication Using Methanol Extract of Onion Peel as a Natural Sensitizer

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Abstract: Methanol extracts of onion peel waste (Allium cepa) were applied as natural sensitizer in Dye-Sensitized Solar Cell (DSSC) fabrication. This research investigated the dye characteristics of onion peel extract and its power conversion efficiency. Onion peel extraction was carried out using unacidified methanol extract (UME) and acidified methanol extract (AME). Dye absorption was characterized by UV-Vis Spectrophotometer at a wavelength of 200-800 nm and FT-IR at a wave number of 4000-500 cm^{-1}. The power conversion efficiency of the fabricated DSSC was determined by calculating the voltage and current generated by the multimeter. The red onion peel extracts, both acidified and unacidified methanol, had maximum absorption at UV-Visible range (457, 659 and 662 nm). Onion peel extract has several functional groups such as –OH, C=O, C-O-C, C=C, and C-H aromatics- showing specific absorption corresponding to the anthocyanin structure. A solar energy conversion efficiency of η= 0.0413% was obtained from UME extract with a short circuit current of up to Jsc=0.6031 $mAcm^{-2}$ and a fill factor of 0.2764.

Keywords: Anthocyanin, methanol extract, natural sensitizer, red onion peel, solar cell.

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INTRODUCTION

Indonesia has several potential sources of renewable energy, for example, hydropower (94.3 GW), geothermal (28.5 GW), bioenergy (32.6 GW), wind energy (60.6 GW), marine energy (17.9 GW), and solar energy (207.8 GWh) (1). The highest potential of solar energy encourages researchers to transform this into an electrical energy source. They expend a lot of effort on developing solar cell devices; one commonly used today to harvest solar radiation is the silicon-based solar cell. Silicon-based solar cell for photon conversion into electricity is currently faced with several disadvantages. It is inflexible, more expensive to fabricate, requires a sterile processing technology, and has a specific band gap that is uncontrollable (2,3,4). Organic solar cells, quantum dots solar cells, perovskite solar cells and dye-sensitized solar cells (DSSCs) are the third generation of solar cells that have been largely developed in order to produce the economical alternative of conventional Silicon-based solar cells (5,6,7,8).

The development of DSSC to convert solar radiation is more attractive because of its advantages over silicon-based solar cells. Compared to classic photovoltaic devices, DSSC manufacturing is easier, cheaper, and more flexible to fabricate (6,7). It is more eco-friendly and produces high energy conversion efficiency (11,12). DSSC is a photoelectrochemical device that converts the visible light into electrical power based on the sensitization of wide band gap semiconductors. DSSC is composed of a photoanode (an electrode with wide band gap semiconductor like nanocrystalline TiO_2), sensitizer (a suitable dye molecule), redox electrolyte (iodide/triiodide), and the counter electrode to catalyze electron regeneration (13,14).

Dye sensitizer plays the main roles in affecting the total performance of the solar cell. The dye sensitizer absorbs solar energy and produces excited electrons from HOMO to LUMO orbitals at the semiconductor interface (15,16). It is continually developed to obtain the most stable and effective sensitizers that can promote electron injection into TiO_2 conduction band and improve the electrical conversion of DSSC. Inorganic sensitizers such as polypyridyl ruthenium, porphyrins, and perovskite halides have exhibited high conversion efficiency in harvesting sunlight energy (14). However, these sensitizers are synthetic dyes that are toxic, expensive, and complicated to synthesize (17).

Natural dyes are advised as the best feasible sensitizer to overcome the disadvantages of synthetic sensitizers. Natural sensitizers are cheaper, affordable, abundant, and eco-friendly (18,19). They have multi-pigment options and high absorption coefficient enabling electrons excitation from HOMO to LUMO and electrons injection into the semiconductor efficiently (21). Natural sensitizers can be easily extracted from different plant components such as fruit, flower and leave (20,22). Using natural sensitizers in DSSC needs to consider environmental sustainability, availability of raw materials, season independence, and competing with food products. Biomass waste utilization as a natural pigment source has triggered a powerful interest in developing bio-waste-based natural sensitizers to meet those considerations.

Natural pigment from onion peels (*Allium cepa*) bio-waste has potential DSSC sensitizing. The samples are easy to gain and found in bulky quantities. Natural pigment that responsible for onion peel colour is anthocyanins. Chemical analysis of the onion peel exposed a high content of anthocyanins (23,24). The acylated and unacylated anthocyanins such as cyanidin mono and diglucoside, petunidin glucoside, peonidin mono and diglucoside, and 5-carboxy pyranocyanidine 3-glycoside are found in onion peels (23). These types of anthocyanins have absorption characteristics in a wider visible spectrum. Carbonyl and hydroxyl groups present in anthocyanins can be bound to the semiconductor surface. The adsorption of anthocyanins onto the TiO_2 surface produces a strong complex in the form of a quinonodial base in which the –OH (or =O) group is bound to the Ti(IV) site on the semiconductor film (25). This binding generates an easy electron transfer from the anthocyanin molecules to the conduction band.

Numerous studies have applied anthocyanin from onion peels as a natural dye-sensitizer. Anthocyanins can be extracted from onion peels using polar solvents, i.e., water, ethanol, and methanol. The previous studies examined the water and ethanolic extract of onion peels sensitizing performance; the power conversion efficiency was written down to 0.065 and 0.049%, respectively (15,26). However, investigating the sensitizing performance of methanol extract from onion peel is still inadequate. In the current study, we measure the power conversion efficiency of fabricated DSSC sensitized by onion peel dyes extracted with acidified and unacidified methanol.

EXPERIMENTAL SECTION

Material

The tools for research are blender, FTIR Bruker Alpha, Genesys 10S Thermo Scientific UV-Vis Spectrophotometer, 200 mesh sieve, glassware, bath, ORIEL S013A solar simulator, and Keithley 2400 I-V meter.

Materials that used are methanol 98%, HCl 37%, TiO_2 (99.5%, Merck), KI (99.7%, Merck), I_2 (99.8%, Merck), 4-pyridinecarboxylic acid hydrazide (97%, Merck) and acetonitrile (99.8%, Merck). Onion peel samples were locally collected from Kefamenanu City, North Central Timor Regency, East Nusa Tenggara Province, Indonesia.

Methods

Preparation of the dye

Onion peels were dried, crushed, and sieved into fine powder. 100 gr of peel powder was dispersed into 250 mL of unacidified methanol and stored in the dark room for 24 hours. The crude extract was filtered through the Whatman Filter paper and used further for dye characterization and solar cell fabrication. The soaking process was repeated by using acidified methanol.

Characterization of the dye

The anchoring groups and absorption bands of the extracts against UV and visible light were analyzed using Fourier Transform Infrared (FTIR) and UV-Vis spectroscopy.

Fabrication and performance of DSSC

The DSSC structure in this work is displayed in Figure 4. It was arranged of a photoanode, onion peel sensitizer, redox electrolyte, and counter electrode. The photoanode was prepared by screen printing TiO_2 paste onto the surface of indium tin oxide (ITO) glass and then dried in a bath at 120 °C for 15 minutes. TiO_2 film on ITO glass was calcined for 1 hour at 500 °C. The photoanode was impregnated in methanol extract of onion peel for 20 hours, then rinsed with isopropyl alcohol and dried in a bath at 80 °C before cell fabrication. The electrolyte solution was prepared by dissolving KI, I_2, and 4-pyridinecarboxylic acid hydrazide in acetonitrile. The counter electrode was prepared by glueing synthetic graphite on a glass slide and then coated with PANI: Graphite (1:3) paste by screen printing. The fabricated film was then dried in a bath at a temperature of 120 °C for 15 minutes. The electrolyte is dripped onto the photoanode and clamped with the opposing electrode using a clip before measuring the DSSC performance. The DSSC performance was measured using an ORIEL S013A solar simulator and an I-V meter Keithley 2400. Cell efficiency was calculated using the equation (27) as follows:

$$\eta(\%)=\frac{(Jsc\ x\ Voc\ x\ FF)}{Pin}\ \text{x}\ 100$$

Where η=cell efficiency (%), J_{SC}=short-circuit current (mA), V_{OC}=open-circuit voltage (V), FF=fill factor, P_{in}=power of solar simulator lamp (100 mW).

RESULTS AND DISCUSSION

Dye Extraction

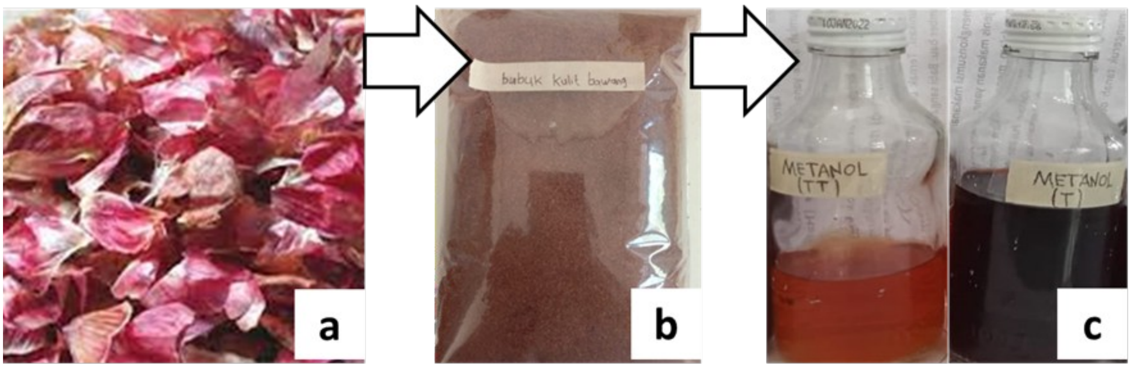

Figure 1: Onion peel sample (a), onion peel powder (b), and crude extracts (c).

Onion peels were soaked into unacidified methanol 95% and acidified methanol with hydrochloric acid. Maceration was chosen as the extraction method because of its effectiveness in onion peel lysis. Solvent and solute interaction during the maceration process is more extended than in other extraction methods. It consumes less extraction solvent, leading to economic loss and environmental problems. Maceration has been reported to recover higher total anthocyanins in onion peel extraction than other methods, such as percolation and soxhletation (28). Initial extraction was performed using methanol to obtain natural dyes from onion peels. The choice of applying methanol is based on the high polarity of the solvent, which makes the extracted material more soluble. Maceration procedures involving hydrochloric acid denature cell tissue membranes and simultaneously dissolve anthocyanin pigments. The acid tends to hold steady anthocyanin pigments, but it may also modify the native form of the dye in the tissue cell. Moreover, combining methanol with acid prevents flavonoid oxidation and promotes solvent polarity. Acidified methanol can increase the percentage retention of anthocyanin extraction from onion peel up to 99.17% and produce a higher amount of extracted anthocyanin than acidified water (29). The colour difference between the acidified and unacidified extract was visualized in Figure 1. The acidified extract is more concentrated than the unacidified extract. The deep red colour of the acidified extract indicates the stabilizing of flavilyium cations by the acid. The addition of acid into the extraction solvent can create an acidic atmosphere during the maceration process, which favours anthocyanin's stability (30).

Dye Absorption

The primary performance of a DSSC device is lean on the light absorption capacity of the dye sensitizer and diffusion of the expelled electron through the mesoporous TiO_2 band. Dye's capability to absorb light is quantified as absorbance. It was determined before being used as a sensitizer in DSSC. The optical characteristics of the pigments extracted from onion peel were determined using a UV-Vis spectrophotometer at 200-800 nm and exposed in Figure 2.

Figure 2 shows the absorption spectra of the two extracts. The unacidified extract maximally absorbs in the UV-visible range (457 and 662 nm); otherwise, the acidified extract absorbs in the visible zone (659 nm). The two noticeable peaks in the electromagnetic spectrum of higher wavelengths than 400 nm are observed in acidified and unacidified extracts. Identical absorptions at the visible range indicate the similar compounds found in the two extracts. There is no absorption observed in the UV zone for acidified extract. The peak's absence is thought to be due to colour degradation by acid addition. The wide spectrum absorption of the unacidified extract produces more excited electrons to the TiO_2, which can increase the photocurrent strength of the fabricated solar cell. Similar absorption characteristics have been reported by previous researchers (31). The other researcher has further emphasized that the presence of aromatic anthocyanin groups will exhibit absorption at a wavelength of 340 nm for the benzoyl group and 400-500 nm for the cinnamoyl group (32). Another researcher stated that anthocyanin absorption was observed at 550 nm and the main absorption range is 450-650 nm (33).

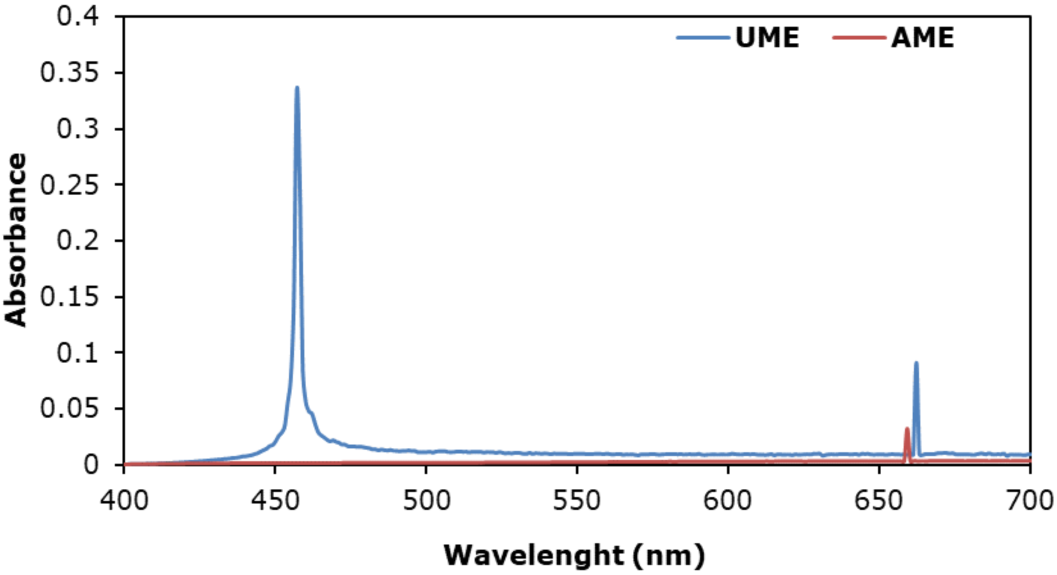

Figure 2: UV-Vis absorption spectra of extracted onion peel dyes.

Functional Groups Characteristics

TiO_2 photoelectrode used in DSSC fabrication requires dye sensitizers possessing several specific functional groups binding to the film surface. Therefore, the functional groups in onion peel extract need to be characterized. The functional groups of the acidified and unacidified extract were analyzed by FT-IR instrument and figured out through FT-IR spectra in Figure 3.

FT-IR spectra confirmed the chemical structure of the onion peel extracts by showing several peaks in the wave number range of 4000-500 cm^{-1}. There is a significant peak at 3419 and 3423 cm^{-1} for the –OH strain vibration. Aliphatic C-H strains appear at 2917-2923 cm^{-1} and 2849-2864 cm^{-1} for $-CH_2$ and $-CH_3$, respectively. The other peaks were observed at 1639 and 1642 cm^{-1} for the C=O group, while another peak that appeared at 1454 cm^{-1} confirmed the presence of C–N groups for the chlorophyll group. C–O group implying the presence of carbohydrates, was detected at 1086 – 1054 cm^{-1}. Aromatic groups was demonstrated by two typical peaks at wave numbers of 1502-1502 cm^{-1} and 650-655 cm^{-1} for aromatic C=C and C-H bonds, respectively. A specific band detected in the 1211-1210 cm^{-1} was contributed to the pyran ring of flavonoid (34). The typical peaks of functional groups in the FT-IR spectra confirmed that the onion peel extract contained anthocyanin compounds. These characteristics are similar to the previous findings (15). There is another highlight that the presence of functional groups such as O-H, C=O, C=C, and C-O-C in the FT-IR spectra confirmed the absorption of anthocyanin compounds (34). The carbonyl and hydroxyl groups that are bound to the semiconductor (TiO_2) surface provide electron transfer from the sensitizer (anthocyanin molecules) to the conduction band of porous semiconducting (TiO_2) film. Anthocyanins absorb light and transfer that light energy by resonance energy transfers to the anthocyanin pair in the reaction centre of the photosystems. In addition to anthocyanins, the observed C-N groups confirm that onion peel extract contains chlorophyll compounds (35).

Generally, the two extracts have absorption peaks at the same wave number but are slightly different in absorption band intensity. It can be seen in Figure 3 that functional groups of the unacidified extract absorb more intensely than the acidified extract. The decrease in the absorption intensity of the acidified extract was related to the pigment reduction due to the molecule destruction by adding acid to the methanol. It has been reported that evaporation of methanol solvent in the anthocyanin extracts consequences of hydrolysis of the labile acyl bonds in the presence of HCl (36). Functional groups and characteristic absorption regions for anthocyanins in onion peel extract are summarized in Table 1.

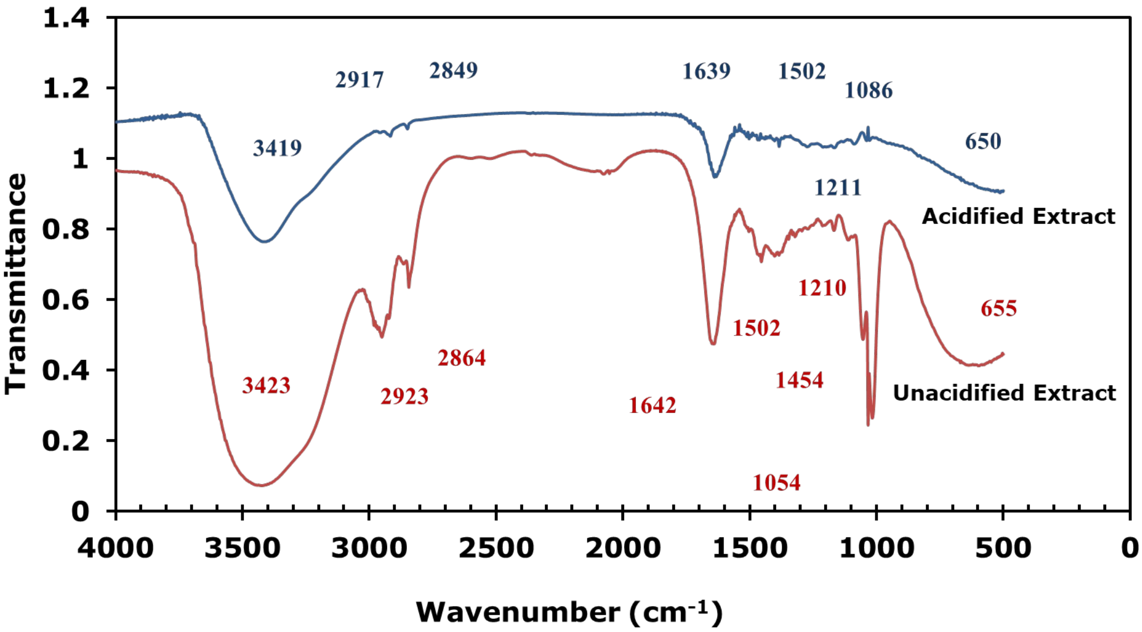

Figure 3: FTIR spectra of extracted dyes.

Table 1: FT-IR spectra of Extracted Onion Dyes.

Wavenumber (cm^{-1})		Functional Groups	Reference	Literature Source
AME	UME			
3419	3423	-OH	3500-3000	(37)
2917	2923	-CH3	2926	(38)
2849	2864	-CH2	2855	
1639	1642	C=O	1639	(39)
-	1454	-C-N	1447	(40)
1086	1054	C-O-C	1072	
1502	1502	C=C	1516	(41)
650	655	C-H aromatics	675	

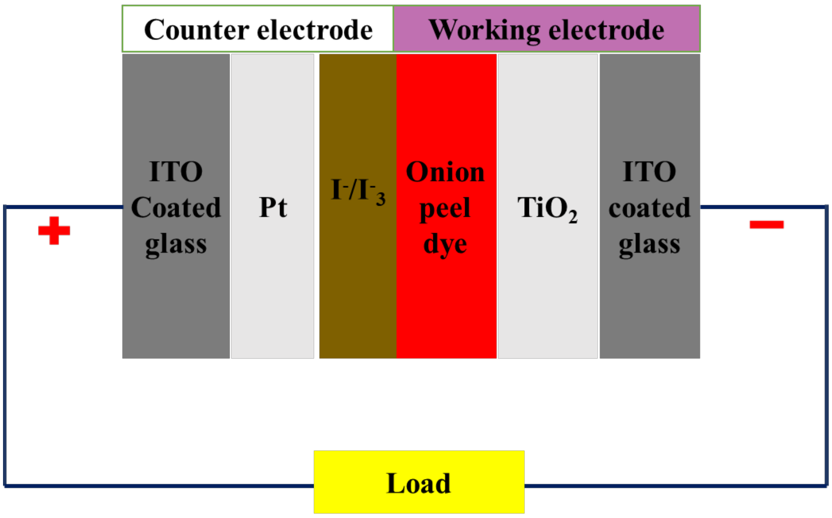

Figure 4: Schematic Diagram of The Prepared DSSC.

DSSC Performance

In this work, the fabricated dye-sensitized solar cell is illustrated in Figure 4. Its structure consists of a titanium dioxide layer (semiconductor) coated photo anode electrode, a counter electrode used as a cathode, a sensitizer, and an electrolyte. The dye-sensitizer is a key parameter that affects DSSC performance in converting solar energy into electrical power. Electron excitation from the HOMO to LUMO energy level can occur when the sensitizer molecule absorbs photons. Electrons from sensitizer molecules move to the TiO_2 conduction band and then pass through the external circuit to the counter electrode. The flow of electrons triggers an I^- /I_3^- redox reaction at the opposite electrode. After receiving electrons from oxidized I^- ions, the oxidized molecule of sensitizer was regenerated. This process continues as long as the sensitizer absorbs solar energy, generating an electric current (16). Electrical current production in the DSSC does not cause a permanent chemical transformation or change. The schematic process is expressed in the following equation:

$$S + hv \rightarrow S^*$$
$$S^* + TiO_2 \rightarrow S^+ + e^- (TiO_2)$$
$$e^- (TiO_2) + Load \rightarrow TiO_2 + e^- (Load) + Current$$
$$2S^+ + 3I^- \rightarrow 2S + I^-_3$$
$$2e^-(Load) + I^-_3 \rightarrow I^-$$

The J-V characteristic curves of the DSSC sensitized with methanol extracts of the onion peel are demonstrated in Figure 5. At the same time, the power conversion efficiency of the DSSCs is summarized in Table 2.

The short-circuit current (J_{SC}) and fill factor (FF) values differ quite significantly, while the open-circuit voltage (V_{OC}) shows the same value as shown in Table 2. Open circuit voltage is determined by the difference between the semiconductors' fermi energy and redox potential. Hence, using the same redox and semiconductor pairs will produce the same V_{OC} value. The different types of onion peel extracts as natural sensitizers only affected the J_{SC} and FF values, affecting the cell's power conversion efficiency.

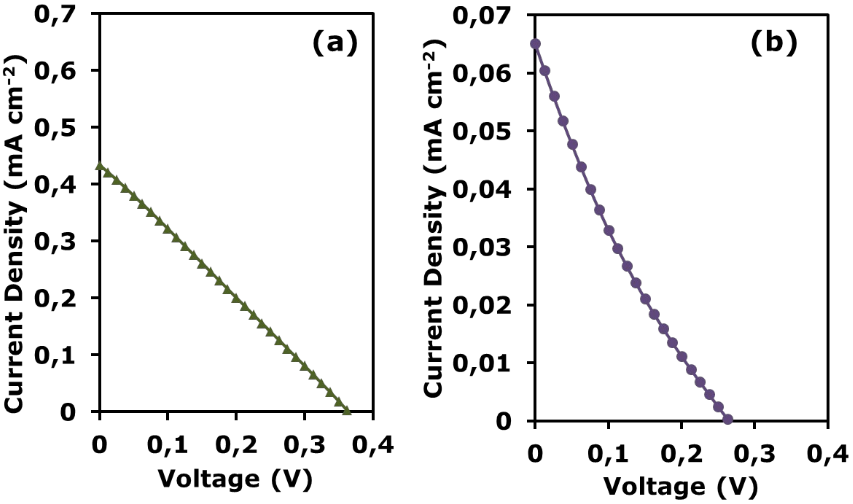

Figure 5: Current-Voltage Density (J-V) Curve of DSSC Sensitized by Onion Peel Extract (a) Unacidified Methanol Extract (b) Acidified Methanol Extract.

Table 2: DSSC Performance Parameters.

Onion peel sensitizers	J_{sc} (mA cm^{-2})	V_{oc} (mV)	FF	Power Conversion Efficiency (%)
Unacidified Methanol	0.4338	338	0.2764	0.0413
Acidified Methanol	0.0619	338	0.1574	0.0033

The J_{SC} value is related to the number of electrons flowing in the DSSC component, which is directly determined by the sensitizer's capability to absorb photon energy and inject electrons into the semiconductor. Onion peel extracts containing dye molecules that undergo electronic transition after absorption of electromagnetic waves in the visible light region can provide greater J_{SC} value and power conversion efficiency. Table 2 shows that extracted onion peel without acidification showed a higher J_{SC} value and cell efficiency than the acidified methanol. The highest power conversion of 0.0413% was obtained from a DSSC cell fabricated using unacidified methanol extract. This is attributed to the higher nondestructive dye molecules contained in the unacidified extract than in the acidified extract. The more the number of extracted dye molecules, the more dye molecules bound to the semiconductor surface; hence the production of electrons increases. As previously discussed, the less quantity of molecules in the acidified extract was due to the dye destruction during acidification, as evidenced by the less intense IR absorbed. This is credited to the poor interaction between the acidified dyes with the semiconductor electrode, restricting the electrons transported from the excited dye molecule to the TiO_2 film.

The power conversion efficiency of solar cells sensitized by methanol extract of onion peel in this study is similar to that of aqueous extract of onion peel (0.065%) as in the previous report, which applies a temperature of 90℃ during maceration (15). Similar findings were reported by the other investigators who also studied the performance of onion peel extract as a natural sensitizer in DSSC and achieved a higher efficiency of 0.29%; the extract was prepared by dispersing sample in a solvent mixture of 96% ethanol: water with the volume ratio of 4:1 at pH 1 (18). The less power conversion efficiency achieved in the present study was caused by the anthocyanin extraction method during the study. It was not selective for anthocyanin; as a result, there are a lot of byproducts that are produced. Some further optimizations must be performed in the future to improve the DSSC performance. Optimizations can be conducted during the dye extraction process by choosing an appropriate solvent, suitable extraction pH range, and purification stages for the extracted compounds.

CONCLUSION

Two extracted dyes from onion peel, both acidified and unacidified methanol, were assessed as natural sensitizers in DSSC. The unacidified extract strongly absorbs UV and visible light at a wavelength of 457 and 662 nm. The acidified extract only absorbs visible light at 659 nm. The two extracts possess carbonyl and hydroxyl groups that are suitable to be used as a natural sensitizer in a solar cell. The power conversion efficiency of DSSC cells fabricated using unacidified extract of onion peel dye exhibited a higher value of 0.0413% than that of acidified dyes.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

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