FABRICATION OF SOLAR CELLS WITH TiO_2 NANOPARTICLES SENSITIZED USING NATURAL DYE EXTRACTED FROM MANGOSTEEN PERICARPS

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ABSTRACT

With the increasingly shrinking reserves of fossil-based energy, as well as the damaging effects of the use of fossil-based energy sources such as the greenhouse effect and global warming, efforts are needed to find energy alternatives. One device being developed as an alternative source of renewable energy by utilizing solar energy source is dye-sensitized solar cells (DSSC) that works using simple photosynthetic-electrochemical principle in the molecular level. In this device, the inorganic oxide semiconductors such as TiO_2 has a great potential for the absorption of the photon energy from the solar, especially in the form of TiO_2 nanoparticle structure. In this study, a commercial TiO_2 nanoparticle was used. The as-received TiO_2 nanoparticle was characterized using X-ray Diffraction (XRD) and scanning electron microscope (SEM). For sensitizer, natural dye extracted from mangosteen (*Garcinia mangostana L.*) pericarps was used. The extracted natural dye was characterized using Fourier transform infrared (FTIR) for the functional groups, whereas the ultra violet-visible (UV-Vis) was used to examined the absorption activity of the extracted natural dye. The performance of DSSC was analyzed through a precision current versus potential different (I-V) curve analyzer. The maximum power conversion efficiency (PCE) of the mangosteen natural dye was given by yhe one extracted using ethanol containing 20% distilled water as compared to the commercial organic dye with PCE of 4.02%. This result is convincing and promising for the next development.

Keywords: Anthocyanin; Dye-sensitized solar cell; Hydrothermal method; Mangosteen pericarp; TiO_2 nanoparticle.

1. INTRODUCTION

One of the renewable energy alternatives with enormous potential to be developed in Indonesia is solar cells. This is mainly because Indonesia has an abundant source of solar energy that is relatively constant throughout the year. As a country located in the equatorial region, Indonesia has the potential of solar energy with an estimated capacity of 4.800 kWh per square meter per day (Tharakan, 2015).

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Solar cells, or photovoltaic cells, is an electronic device with the ability to convert light energy into electrical energy directly using photovoltaic effect. A new type of solar cell known as dye-sensitized solar cells (DSSC) has been developed using a simple electrochemical principle that mimics the effects of photosynthesis, i.e. by capturing photon energy at the molecular level and convert it into electrical energy (O'Regan & Grätzel, 1991). The system consists of a semiconductor oxide layer with a wide band gap energy enclosed by a molecular layer of organic dyes and placed in contact with a redox electrolyte.

Dye-sensitized solar cell performance in converting light energy into electrical energy is determined by one of which the oxide used. Oxide semiconductor is commonly used in photoelectrochemical for its stability against photocorrosion (Kalyanasundaram & Grätzel, 1998) and relatively low band gap energy (< 3.2 eV) is required for more photon energy absorption in the visible light spectrum (Dette et al., 2014). Apart from that, the ability of DSSC-based materials is also influenced by the structure of the oxide layer associated with characteristic of high surface area to absorb sensitizing dyes so that system performance can be maximized. In this case, the semiconductor layers most often used in DSSC are usually metal oxide materials such as TiO_2, ZnO, CdSe, CdS, WO_3, Fe_2O_3, SnO_2, Nb_2O_5, and Ta_2O_5 (Wang et al., 2014; Sholehah et al., 2017).

Titanium dioxide (TiO_2) or titania is one of the inorganic oxide metals included in the transition class known as transitional metal oxides (TMO) (Liu et al., 2013). This material has long been the subject of in-depth research and industry in the world because of the wide range of fascinating properties and its technological applications such as photovoltaic cells, batteries, sensors chemicals (Varghese et al., 2003), optical emission, photonic crystals, catalysts, photocatalysts (Livraghi et al., 2005), and purification of environment (Ikezawa et al., 2001). Titanium oxide exists in several crustal structures, namely anatase, brookite, and rutile; however, the most use for DSSC is the anatase TiO_2 (Al-Alwani et al., 2015). In addition, the anatase TiO_2 are also used in lithium batteries and electrochemical devices (Hagfeldt & Grätzel, 1995; Kavana et al., 2000; Grätzel, 2001).

The presence of the oxide semiconductor layer plays a very important role in the process of converting light energy into electrical energy. Further, the interaction between the dye and semiconductor oxide particles will be very instrumental in determining the process of converting light energy into electrical energy (Narayan, 2012). Meanwhile, it has been known that with the small size of the material down to the nanometer scale, the ratio between the surface area to volume ratio (surface to volume ratio) will be greater so that the possibility of surface interactions with the environment will be higher (Yuwono et al., 2012). It is expected that the use of TiO_2 nanomaterial in the form of nanoparticles, nanotubes and or mesoporous as semiconductor oxide layers in a DSSC will improve its system performance (O'Regan & Grätzel, 1991).

Dye sensitivity is one important thing in DSSC since it acts as a photon absorber and starts the process of converting solar energy into electrical energy. There are many kinds of natural dye that can be used in DSSC such as chlorophyll (Kumara et al., 2006), betalains (Zhang et al., 2008), and anthocyanin (Fernando & Senadeera, 2008). The latter has the most promising results in maximizing the efficiency of DSSC (Calogero et al., 2012).

Mangosteen is an indigenous plant to Indonesia (Morton, 1987) and has long been cultivated in many areas. The fruit of the plant is also called mangosteen, with purplish red when ripen, although there are also variants whose skin is red. In general, this fruit is utilized for its white and sweet contents, whereas the skin or pericarp will be wasted. Based on the research, it has been known that the skin of the mangosteen also contains anthocyanin, a flavonoid compound known responsible for the purplish red color on the skin of ripe fruit (Du & Francis, 1977; Chaovanalikit et al., 2012). Up to the present time, this mangosteen pericarp has not been known to be used for sensitizing solar cells devices. Therefore, in this study, the pigment contained in the mangosteen pericarps was used as a sensitizing dye in the TiO_2 nanomaterial-based DSSC. The focus is on the extraction of the mangosteen natural dye using various solvents and used it as sensitizer in DSSC device fabricated with a commercial TiO_2 nanoparticles.

2. EXPERIMENTAL SETUP

2.1 Dye Extraction from Mangosteen Pericarp

Some fresh ripen mangosteen from local market were emptied by taking the sweet white flesh of edible parts out. The deepest white layer of skin was scraped and the pericarps were washed thoroughly under running water and allowed to stand for some time so that the remaining water evaporated. The mangosteen pericarps were chopped in a blender and air dried for 48 hours. The dried chopped pericarps were ground using a blender to obtain a very fine powder. The powder was sieved to obtain a fine homogeneous powder.

Five Erlenmeyer flasks were prepared and each was filled with 10 grams of the fine mangosteen pericarp powder. Each Erlenmeyer flask was added with 100 mL of different solvent, i.e. pure ethanol (Merck KGaA, Germany), ethanol containing 20% distilled water, ethanol containing 1% HCl, and ethanol containing 1% acetic acid. The mix was agitated by using a magnetic stirrer at room temperature for 6 hours. After the stirring process, the supernatant was passed through a filter paper to separate the insoluble powder with the solvent. The filtered supernatant was allowed to stand for some time at ambient conditions to evaporate the solvent and to obtain a concentrated dye of about 10 mL. The steps in this dye extraction are given schematically in Fig. 1.

Figure 1. Natural dye extraction from mangosteen pericarps

The solution was then ready for characterization by using attenuated reflectance Fourier transform infrared (ATR FTIR, PerkinElmer Spectrum 2) and ultra violet-visible (UV-Vis, PerkinElmer Lambda 25) equipment. As a comparison, 2.5 mg commercial organic dye (Sensidizer RK1, Solaronix) dissolved in 100 mL ethanol was also examined.

2.2 DSSC Fabrication

The anode was prepared using commercial TiO_2 nanoparticles (Degussa P25). One gram of TiO_2 nanoparticle powder was added with 10 mL of ethanol and stirred. Two drops of TRITON X-100 (Sigma Aldrich) were added into the paste and stirred until homogeneous phase was obtained.

Two fluorine-doped tin oxide (FTO, Solaronix) conductive glass substrates were prepared. One of the glass substrates was drilled to create two tiny perforations at one side and dry cleaned by using methanol. The other FTO glass was also dry cleaned by using methanol and attached to a flat layer. This substrate was tape masked and coated with TiO_2 nanoparticle paste using a doctor-blade method under an area of about 1 cm^2. The paste was dried at 450°C for one hour and allowed to cool down before soaking it in the dye solution prepared from the extraction and air dried. As comparison, the commercial dye (Sensidizer RK1, Solaronix) was also used. The other perforated FTO glass substrate was coated with a platinum paste (Platisol, Solaronix) and dried at 450°C for one hour.

Both of the coated glass substrates were then attached to one another separated by a spacer and sealed to avoid electrolyte leakage. Once dried, the electrolyte (Iodolyte, Solaronix) was then injected through the two tiny holes drilled at one side of the substrate. The holes were then sealed and the cell was ready for characterization. Cell activity was measured by using a simple ammeter, while the cell performance was tested by using a Semiconductor Parameter Analyzer (SPA, Agilent 4155A) with a standard illumination of 100 mW/cm^2.

3. RESULTS AND DISCUSSION

In order to be effectively adsorbed onto TiO_2 layer, a dye need to have specific functional groups. Fourier transform infrared (FTIR) examination was carried out to observe the functional group characteristic presented in the mangosteen extracted dye. The spectrum obtained at wavenumber from 4000 – 400 cm^{-1} are given in Fig. 2. As a comparison, the characteristic of pure commercial dye is also given.

As seen in Fig. 2, all of the spectra show the appearance of hydrogen bonded OH stretching bands ranging from 3250 to 3450 cm^{-1} and bands at around 2878 to 2973 represent C-H groups stretching (Pereira Jr et al., 2015). Band at 1640 cm^{-1} indicates C=O stretching vibration; it is small except for the natural dye extracted using ethanol containing 20% distilled water. The small peak at 1450 cm^{-1} belongs to C – N, bands at 1423-1274 cm^{-1} belong to aromatic compound, band at 1045 belongs to C – O, and stretching vibration of C–O–C esters is found at 1046 cm^{-1}. The aldehydes are found in the wavelength between 880 cm^{-1} and 800 cm^{-1} due to the organic base natural dye. In this instant, the presence of carboxyl and hydroxyl group in the dye would interact and strongly bind onto TiO_2 surface (Al-Alwani et al., 2015). The interaction between TiO_2 nanoparticle and functional groups of the dye would then drive the electron transfer from the dye molecules to the conduction band of the semiconductor TiO_2 (Narayan, 2012).

Absorption spectrum of the dye in the ultraviolet-visible region was examined using UV-Vis spectroscopy at wavelength from 400 – 700 nm and the results are given in Fig. 3.

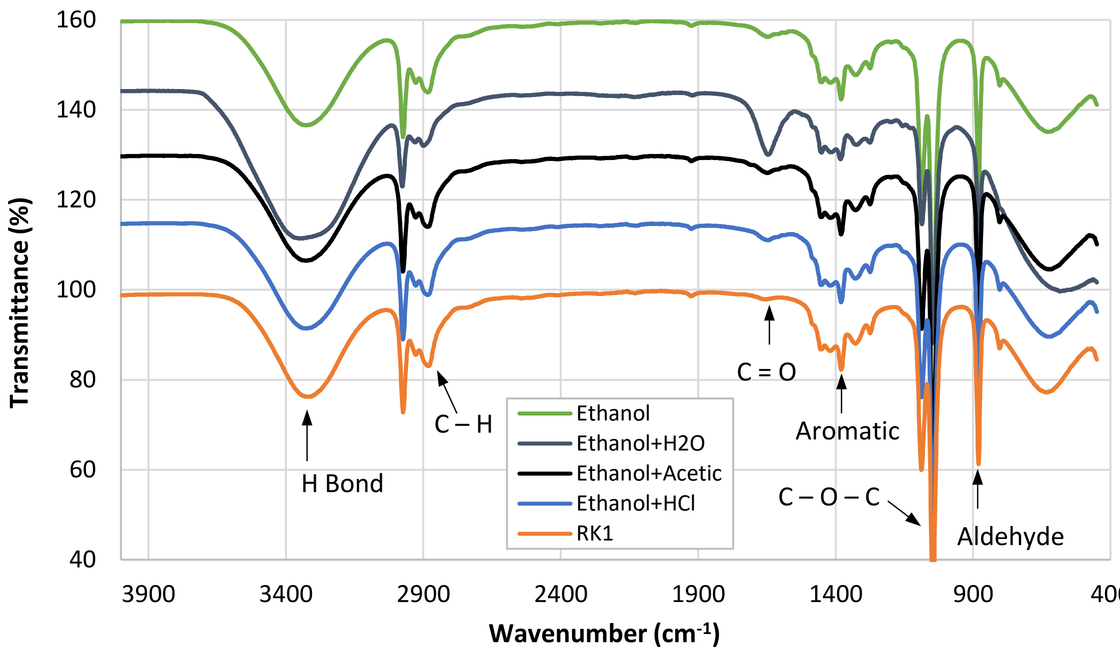

Figure 2. Fourier transform infrared transmittance characteristics of the commercial dye and the mangosteen dye extracted with various solvents at wavenumber 4000 – 400 cm^{-1}

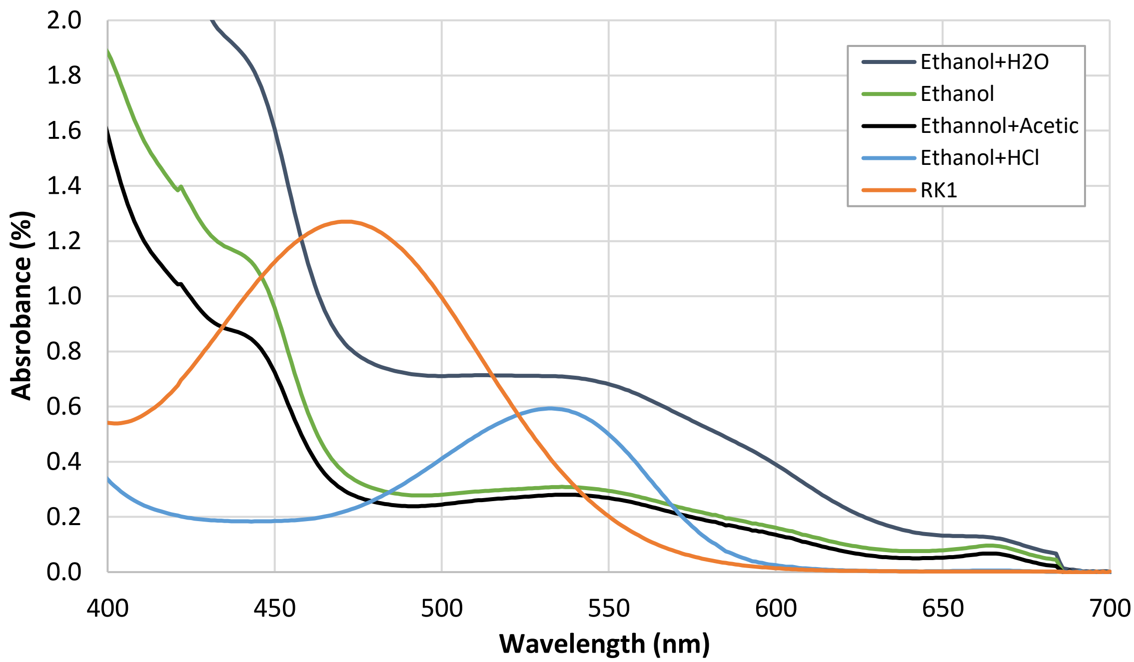

Figure 3. Ultraviolet-visible absorbance characteristics of the commercial dye and the mangosteen dye extracted with various solvents at wavelength from 400 – 700 nm.

As seen in Fig.3, UV-Vis absorbance characteristics of the RK1 commercial dye has a specific absorbance peak at around 474 nm, whereas the mangosteen natural dye extracted using ethanol containing 1% HCl has a specific absorbance peak at around 534 nm. The other three spectra from the mangosteen natural dye extracted using ethanol, ethanol containing 1% acetic acid and ethanol containing 1% HCl have broaden with no specific absorption peak. The absorption peak indicates at which the presence of the dye that absorbs high number of photon energy at the specific wavelength from the visible light source (Fernando & Senadeera, 2008). It can also be noted from the spectra that the absorption from the mangosteen natural dye extracted using ethanol containing 20% distilled water has the most broaden and above the other spectra. This could be an indication that the dye has the widest absorption spectrum in the visible light; a statement that yet to be further confirmed.

The as-received commercial TiO_2 nanoparticle was characterized by using X-ray diffraction (XRD) and scanning electron microscope (SEM). The diffraction patterns from XRD and secondary electron image from SEM are shown in Fig. 4. As seen from Fig.4a, the as-received material confirmed to have a mixed anatase crystal structure (JCPDS file No. 73-1764) and rutile crystal structure ((JCPDS file No. 78-1510). The three distinct diffraction peaks observed at 2θ value of 25.31°, 37.79°, and 48.04° are corresponding to (101), (004), and (200) of the anatase crystal planes (Lekphet et al., 2017). The three distinct diffraction peaks for rutile (♦) are observed at 2θ 27.5°, 36.1° and 41.3° corresponding to (110), (101), and (111) planes, respectively.

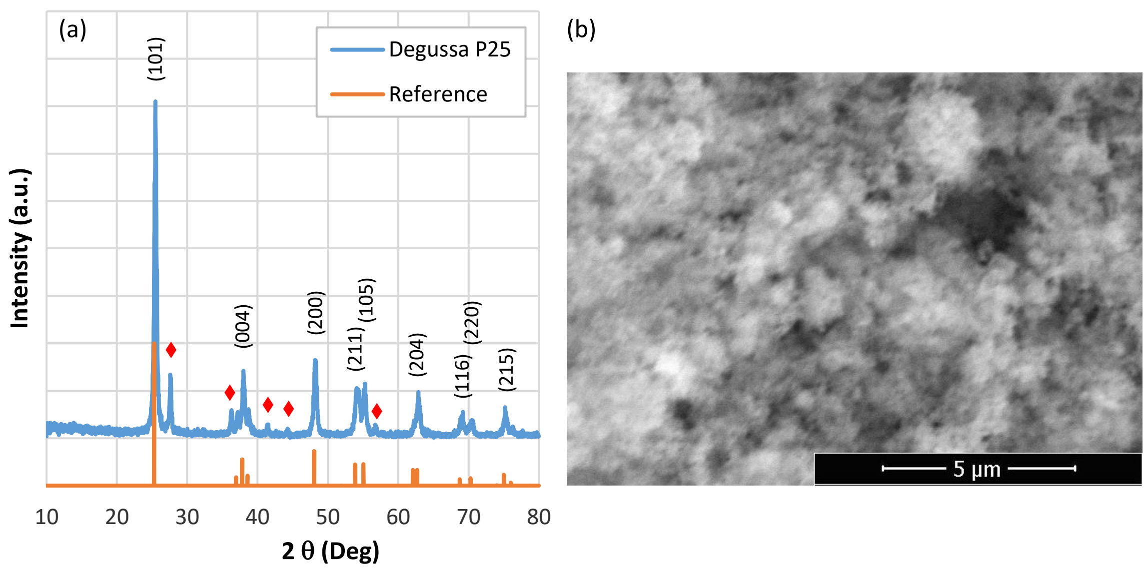

Figure 4. (a) X-Ray diffraction patterns of TiO_2 reference (JCPDS No. 73-1764) and commercial TiO_2 nanoparticle and (b) secondary electron image of the commercial TiO_2 nanoparticle morphology.

The crystallite size of TiO_2 nanoparticles was calculated in according to Debye–Scherrer's equation (Cullity, 1978):

$$c_s = \frac{k\lambda}{B\cos\theta} \quad (1)$$

where c_s is the crystallite size, λ is the wavelength of the X-ray radiation with Cu Kα = 0.15406 nm, k is a constant taken as 0.94, θ is the diffraction angle, and B is the full width at half maximum (FWHM) peak in radians. All of the diffraction peaks were taken into account from which the average crystallite size of 25 nm was obtained.

Scanning electron microscope was used to study the surface morphological features of the as-received TiO_2 nanoparticle and the result is given in 4b. As seen in Fig. 4b, the morphology shows a homogeneous distribution of the nanoparticle. Image analysis revealed that the average particle size is < 80 nm.

Photoactivity of the DSSC device sensitized using mangosteen natural dye extracted with various solvents was examined through a J-V curve characteristic and the results are given in Fig. 6. As seen in Fig. 6, photocurrent-voltage characteristic of the DSSC device sensitized using commercial organic dye and the mangosteen natural dye extracted using ethanol containing 20% distilled water has the highest value with a current density of around 10 mA/cm^2. The other natural dyes extracted using pure ethanol, ethanol containing 1% acetic acid and ethanol containing 1% HCl have only current density below 70 μA/cm^2.

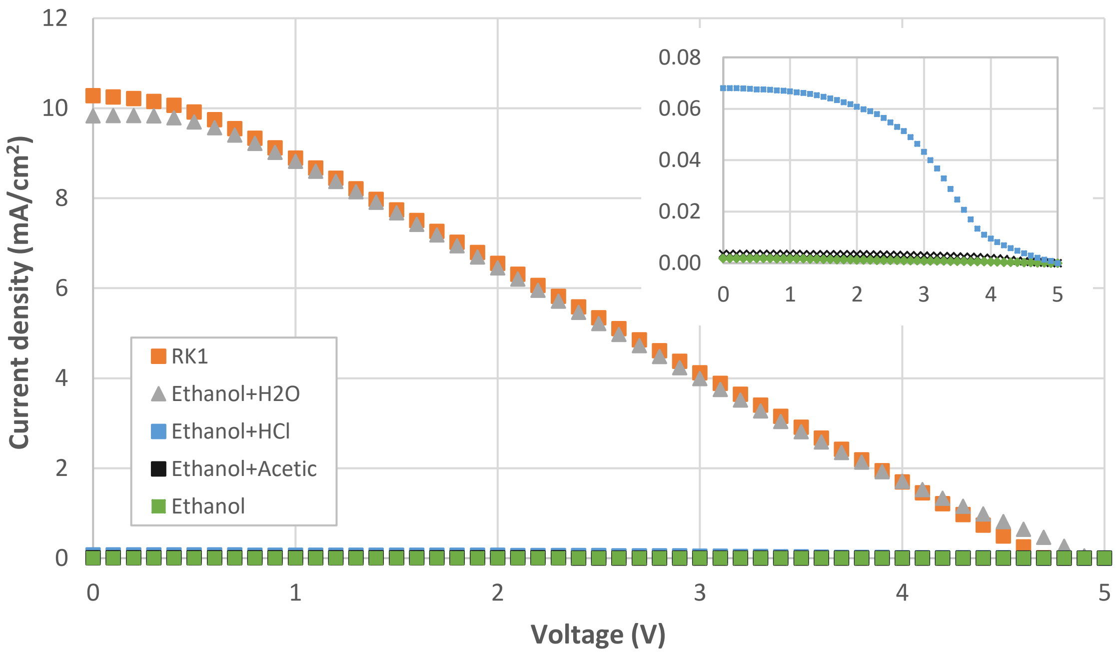

Figure 6. Photocurrent-voltage characteristics of the DSSC device sensitized using the commercial dye and the mangosteen natural dye extracted with various solvents. Inset is the photocurrent-voltage characteristic of the DSSC device sensitized using ethanol containing 1% HCl, ethanol containing 1% acetic acid and pure ethanol.

Based on this J-V curve characteristic, the power conversion efficiency was calculated using the formula (Fernando & Senadeera, 2008):

$$\eta = \frac{FF \times J_{SC} \times V_{OC}}{I_{in}} \times 100 \qquad (2)$$

where J_{sc} is the short-circuit photocurrent density (A cm^{-2}), V_{oc} is the open-circuit voltage (volts), I_{in} is the intensity of the incident light (W cm^{-2}) and FF is the fill factor defined as

$$FF = \frac{i_{\max} V_{\max}}{i_{OC} V_{OC}} \qquad (3)$$

where i_{max} and V_{max} are maximum photocurrent and voltage, respectively. Their values can be extracted from the maximum power of the I-V characteristics, whereas i_{oc} is the open-circuit current (mA). Based on the data obtained from the photocurrent-voltage examination of the DSSC device sensitized using mangosteen natural dye, the maximum power conversion efficiency (PCE) is found to be 3.91% given by the solvent of ethanol containing 20% distilled water. PCE from the other natural mangosteen dyes extracted using pure ethanol, ethanol containing 1% acetic acid and ethanol containing 1% are found to be quite low as seen detail in Table 1. At the same time, PCE for the commercial organic dye is found to be 4.02%, the highest among the others. Compared to the result of the device that used sensitizer from anthocyanin extracted from red skin apple with the efficiency 0.05% (Saputra et al., 2017), the current result is much more convincing and promising for the next development.

Table 1. Power conversion efficiency (PCE) of the DSSC device sensitized using the commercial dye (RK1) and the mangosteen natural dye extracted with various solvents.

	Ethanol	Ethanol+H_2O	Ethanol+Acetic	Ethanol+HCl	RK1
V_{oc} (Volt)	5.000	5.000	5.000	5.000	4.800
I_{oc} (mA)	0.002	9.828	0.004	0.068	10.280
V_{max} (Volt)	0.500	0.400	0.900	1.000	0.400
I_{max} (mA)	0.002	9.784	0.004	0.067	10.060
FF	0.098	0.080	0.176	0.196	0.082
PCE (%)	0.001	3.914	0.003	0.067	4.024

4. CONCLUSION

Extraction of natural dye from mangosteen pericarps has been successful carried out using various solvents. The extracted dyes have been also successfully applied as sensitizer for DSSC fabricated on commercial TiO_2 nanoparticle anode. In this work, the best solvent for extracting natural dye from mangosteen pericarp for using with DSSC device is found to be ethanol containing 20% distilled water with a power conversion efficiency of 3.91%. The dyes extracted using other solvents are found to have low power conversion efficiency, however, the stability of the dyes in the DSSC device yet to be further confirmed.

5. ACKNOWLEDGEMENT

The authors would like to express their gratitude for the funding from the Directorate of Research and Community Services (DRPM), Universitas Indonesia, through Hibah PITTA No. 823/UN2.R3.1/HKP.05.00/2017.

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