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***Piper Ornatum* and *Piper Betle* as Organic Dyes for TiO₂ and SnO₂ Dye Sensitized Solar Cells**

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Abstract. Dye sensitized solar cell (DSSC) mimics the principle of natural photosynthesis are now currently investigated due to low manufacturing cost as compared to silicon based solar cells. In this report, we utilized Piper ornatum (PO) and Piper betle (PB) as sensitizer to fabricate low cost DSSCs. We compared the photovoltaic performance of both sensitizers with Titanium dioxide (TiO₂) and Tin dioxide (SnO₂) semiconductors. The results show that PO and PB dyes have higher Short circuit current (J_{sc}) when applied in SnO₂ compared to standard TiO₂ photo-anode film even though the Open circuit voltage (V_{oc}) was hampered on SnO₂ device. In conclusion, from the result, higher electron injections can be achieved by choosing appropriate semiconductors with band gap that match with dyes energy level as one of strategy for further low cost solar cell.

1. Introduction

Solar energy is one of the greenest types of energy resources. It is free and abundant that makes this energy form one of candidate to replace fossil fuel. Silicon based solar cells have dominated the installed solar cell today. However, Silicon based solar cell required huge energy resources to purify the silicon and high production cost in order to achieve high efficiency cell. Low cost solar cells, such as Dye sensitized solar cells (DSSCs) are gaining more and more attention owing to their high efficiency and relatively lower cost of production [1]. A DSSC consist of two sandwiched electrodes, working and counter electrode, made from transparent conducting glass which mainly made of Fluorine doped tin oxide (FTO). Working electrode is made of thin layer of semiconducting material such as titanium dioxide (TiO₂), tin dioxide (SnO₂) Zinc oxide (ZnO) etc, coated with dyes as photo anode on top of FTO [2-9]. Counter electrode is made of FTO with thin layer of platinum or carbon [10-13]. Electrolyte is placed between both electrodes to connect and maintain electrons transfer. One of the main components to enhance the Photo-conversion efficiency (PCE) is dye sensitizer. The most widely used in DSSCs research area are Ruthenium based sensitizer. In the recent past Ruthenium based as well as metal free dyes have been reported having nearly quantitative photon harvesting mainly in the visible region of solar spectrum [14-17]. High quality ruthenium dyes are produced in sophisticated ways that increase the production cost of the DSSC. Most of sensitized dyes need anchoring groups in order to attach in nanoporous surface. Although attachment dyes on the nanoporous oxide semiconductor via a suitable anchoring group is common practice in DSSCs but



introduction of such anchoring groups in dye structure at suitable position is synthetically cumbersome. In order to attach dyes on nanoporous oxide semiconductor surface, it could be achieved by several types of bonding, for example hydrophobic or electrostatic interactions and co-valent bonding [18]. Natural dye has widely used in textiles industry and recently in DSSC as sensitizer due to easier to prepare and low in cost [19-23]. In this works, the extracts of Piper ornatum and Piper betle as dye sensitizer in DSSC. The absorption spectra of PO and PB dyes solution in ethanol was measured with UV-VIS Spectrophotometer. The DSSC was made using two types of semiconductors which are TiO_2 and SnO_2 . The current-voltage (I-V) characteristics were measured along with Incident photon-to-current Efficiency (IPCE) to determine the photo-conversion efficiency and internal quantum efficiency.

2. Experimental

2.1. Dye preparation

The natural dyes used in this work extracted from Piper Ornatum (PO) and Piper Betle (PB) leafs. PO and PB fresh leafs was cleaned with distilled water and dried in shaded place for more than 3 weeks. After dried, the dried leaf were crushed with Food grinder FTC-Z100 with output fineness 20-180 mesh. Powdered PO and PB were diluted in ethanol. 1 gr of powdered leafs was immersed in 10 ml of ethanol and stored for 48 hours in dark place. After that, dye solution was filtered from remained powder using vacuum pump. Dye solution was kept in dark container to protect it from direct light exposure.

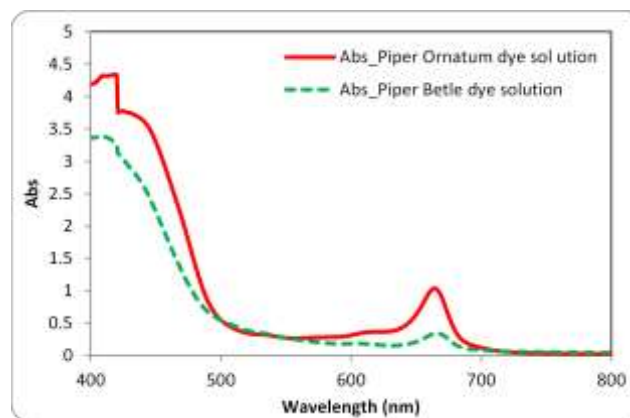


Figure 1. Absorption spectra of Natural dyes: Piper Ornatum in solution (red solid lines) and Piper Betle in solution (green dashed lines).

2.2. Preparation of dye sensitized solar cell

DSSCs were fabricated with standard sandwich-like structure. Working electrode was prepared using Fluorine doped Tin Oxide (FTO) glass coated with $7 \mu\text{m}$ thick TiO_2 (30NRD) and SnO_2 followed by backing at 450°C for 30 minutes. Baked working electrode immersed in dye solution for 20 hours (figure 2). The cell area was 0.25 cm^2 . The thickness of the spacer used to create a gap between working electrode and counter electrode was $25 \mu\text{m}$ (Solaronix Spacer). Counter electrode using Platinum (Pt) sputtered on FTO glass and electrolyte containing 500 mM LiI, 50 mM I_2 , 600 mM Ethylmethylimidazolium dicyanoimide (MeEtIm-DCA), and 580 mM *tert*-butylpyridine (TBP) in Acetonitrile was used to fabricate the DSSCs. Natural dye solution in ethanol was used for sensitizer on the TiO_2 and SnO_2 surface. Photovoltaic characterization the DSSCs were made using a solar simulator (Bunko-Keiki Co. Ltd., Model Solar Simulator CEP-2000SRR) and black metal mask used to avoid the effect of optical reflection on photovoltaic measurement.



Figure 2. Immersion of semiconductor films on PO and PB dye.

3. Results and Discussions

Two kinds of Natural dyes (PO and PB) have been utilized as sensitizers in the present work. Two kind of semiconducting materials (TiO_2 and SnO_2) were utilized to accommodate energy level of the dyes. Both PO and PB dyes used in the present work exhibits intense light absorption having absorption maximum at 667 nm and 671 nm in ethanol solution for the PB and PO dyes respectively. Light absorption spectra of dyes were characterized using UV-VIS spectrophotometer (Jusco V-670) to confirm the dye absorption (figure 1). Due to lower solubility of the natural dyes, it has been found that 48 hours are required to immerse in ethanol solution before proper usage in DSSC fabrication.

Table 1. I-V Characteristic of DSSCs with PO and PB dyes.

	PO- TiO_2	PO- SnO_2	PB- TiO_2	PB- SnO_2
Efficiency [%]	0.032	0.022	0.034	0.014
FF	0.553	0.357	0.598	0.325
Voc[V]	0.463	0.238	0.455	0.202
Jsc[mA/cm ²]	0.124	0.261	0.125	0.207

Figure 3 exhibits the current-voltage characteristics of the DSSCs fabricated using these two dyes as sensitizer. It can be clearly seen that PO and PB exhibits relatively same PCE on TiO_2 as compared to the SnO_2 fabricated under similar experimental conditions. This phenomenon occurred maybe due to deeper LUMO (Low Unoccupied Molecular Orbital) of these dyes that suppress the electron injection to semiconductor conduction band. The main factor controlling the efficiency is the short circuit density (Jsc) since open circuit voltage (Voc) and fill factors (FF) are nearly the same for PO and PB for each semiconductor. Table 1 shows result of PO- TiO_2 have Jsc of 0.124 mA/cm², Voc 0.463 V, FF 0.553 and PCE 0.032% and PB- TiO_2 have Jsc of 0.125 mA/cm², Voc 0.455 V, FF 0.598 and PCE 0.034% for device with TiO_2 semiconductor. The results were comparatively higher that dye from same genus reported before utilizing Piper Nigrum which was exhibit only 0.009% of PCE [24].

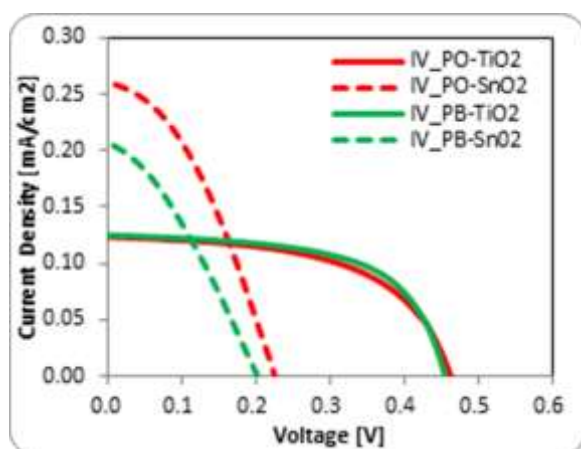


Figure 3. I-V Characteristic of DSSCs.

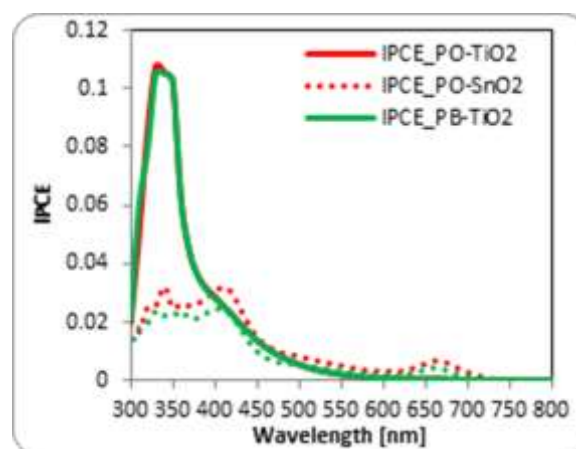


Figure 4. IPCE Spectra of DSSCs.

Device with SnO₂ semiconductor, PO-SnO₂ have J_{sc} of 0.261 mA/cm², Voc 0.238 V, FF 0.357 and PCE 0.022% and PB-SnO₂ have J_{sc} of 0.207 mA/cm², Voc 0.4202 V, FF 0.325 and PCE 0.014%. The results are interpreted in terms for facile dye regeneration in SnO₂ film was higher as compared to TiO₂ film due to its deeper conduction band. IPCE graph shown that photon to current density on SnO₂ film was higher. Higher electron injection was confirmed from peaks shown in 600 to 700 nm wavelength that agreed with absorption peak of the dyes shown in figure 4. Cells fabricated with SnO₂ film have lower Voc due to high traps on SnO₂ semiconductor lead to higher recombination that affect the Voc and FF.

4. Conclusions

DSSCs using Natural dyes Piper Ornatum and Piper Betle were fabricated for with two type semi conducting material Titanium dioxide and Tin dioxide. Fabricated cells produced relatively same PCE on TiO₂ film. Deeper conduction band of SnO₂ compared to TiO₂ lead to higher dye regeneration in SnO₂ film. IPCE graph confirmed that photon to current density on SnO₂ film was higher shown from peaks shown in 600 to 700 nm wavelength. Traps on SnO₂ semiconductor lead to higher recombination that affect the Voc and FF make both parameters lower compared on TiO₂ film. In conclusion, from the result, higher electron injections can be achieved by choosing appropriate semiconductors with band gap that match with dyes energy level as one of strategy for further low cost solar cell.

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