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Cite as: AIP Conference Proceedings 2454, 040003 (2022); <https://doi.org/10.1063/5.0078631>  
Published Online: 09 June 2022

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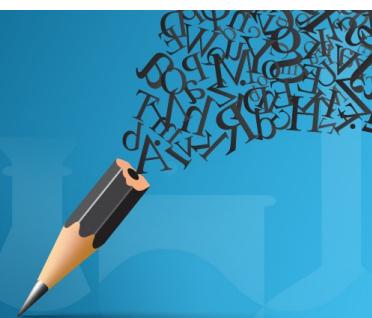


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# Efficient Eco-Friendly Hybrid Natural Dyes As Sensitizer For Dye-Sensitized Solar Cell (DSSC) Application

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**Abstract.** As a result, natural dye-sensitized photovoltaic cells are formed to reduce the shortage of solar cell conversion efficiency. Compared to traditional solar cells, this photovoltaic cell has differential advantages, such as simple processing, often low cost, and integrates environmentally friendly materials. In addition, the use of synthetic dye as a photosensitizing agent such as Di-tetrabutylammonium cis-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato)ruthenium(II), N719 and Ruthenium, Ru complex is a major disadvantage in DSSC. Such materials are highly effective, have incredible chemical stability and also have desirable photo-electrochemical characteristics. However, high fabrication cost due to complicated preparation methods and the presence of heavy metals that pose a threat to the environment, natural dyes were introduced in order to overcome these issues. Ruthenium additives are considered as extremely toxic and carcinogenic, which can spur to cancer. As a consequence, a number of studies have been conducted by many researchers to discover suitable alternative strategies for sensitizers in DSSC using natural pigment dyes such as anthocyanin and flavonoid.

## INTRODUCTION

Dye sensitized solar cell (DSSC) received a lot of recognition due to a relatively minimum cost and green energy photovoltaic device. Silicon-based solar cells needed vast energy resources to sterilize the silicon and huge production costs to attain high performance cell [1]. Therefore, DSSC has many benefits compared to Si-based fuel cells, is not susceptible to deficiencies in semiconductors, and easy transition of energy from photons to the chemical energy can be discovered. DSSC performance is effectively independent of temperature which demonstrates much greater performance in dim sunlight or cloudy conditions than the polycrystalline Si solar cell. The main stream of the scientific work focused on the advancement of materials that would improve conversion efficiency, optimize the consumption of DSSCs and ensure their longevity [2]. In DSSC dye plays a crucial role in capturing sunshine and converting solar power to electricity. The cell's potency focuses significantly on the dye's absorption range and the binding of the pigment to the exterior of the semiconductor. Most metal compounds and organic dyes have been formulated and developed for sensitisation. Transition metal compounds Ru-complex and N719 are commonly employed as efficient sensitizers, owing to their strong charge exchange penetration and extremely stable metal-to-ligand charge transition across the entire detectable spectrum. However, the synthesis process in this complex is very convoluted, costly and consists of toxic metals which mould it unfavourable to its ecological quality [3].

So, natural dyes have been a popular trend in experimentation owing to their cost-effectiveness, harmless and complete biodegradation. Simple methods can be used to extract natural colouring present in seeds, flowers, herbs, vegetables and berries. In this works, several natural pigments, including anthocyanin (*Eleiodoxa conferta*) and flavonoid (*Piper betle*) have been exploited and researched as sensitizers in solar cell application. Various types of dye synthesized from different plant species indicate varying renewable energy-to-electric transformation efficiencies probably depends on the material, the chemical composition of the dye and the extent of the diffusion onto the mesoporous ZnO surface [4]. Such derived dyes have been mixed together to form trihybrid natural dyes which categorized via UV-vis absorption spectra. These extracts have been documented for the photoelectric characteristics

of DSSCs as sensitizers. The composition and some other specifications of these pigments have given rise to varying performance in their efficacy.

## METHODS

### Fabrication of Dye Sensitizer

The natural dyes used in this analysis was extracted from the *Piper betle* (*P. betle*) and *Eleiodoxa conferta* (*E. conferta*). The raw natural materials were washed with purified water and dried in the oven at 40 ° C before being ground into fine powder with a grinder, respectively. These samples powder was weighed at 20 g. Next, 200 ml of ethanol was applied to beaker (1:10) and stirred respectively. The mixtures of *P. betle* and *E. conferta* were kept in a dark location for 24 hours at room temperature. The extraction was filtered using filter paper in order to remove the solid substances that were contained in the extraction and the filtrate was purified again using liquid-liquid extraction to produce pure natural dye. The natural dye extraction was concentrated using a rotary evaporator at 50 ° C for 4 h. The extraction was stored in a dark bottle to shield it from direct light exposure. The two different pigment solutions were mixed together at 1:1 ratio in order to create hybrid solution.

### Preparation of ZnO Photoanode

The ZnO paste was formulated by adding 1 g of ZnO nano-powder and 0.1 g of polyethylene glycol (G) and then stirring the mixture for half an hour until a homogeneous paste was produced. 5 mL of ethanol was added to the paste in order to achieve an acceptable viscosity for the preparation of the coating. FTO conductive glass (Sigma-Aldrich Chemie, USA) were rinsed with ethanol and then dried. FTO conductive glass was detected for its conducting site using a digital multimeter (All-Sun EM420B digital multimeter). The identified conductive site was stacked up and three sides of conductive glass were taped. The doctor blade method [1] was revised to apply the ZnO paste onto FTO glass layer. The adhesive tape was then carefully discarded and the ethanol was swapped over the taped region to eliminate the remaining sticky adhesive tape. The glass was then sintered on a hotplate at 400 ° C for 30 min until the paste was completely dried and allowed to settle.

### Preparation of Counter Electrode

The carbon paste was prepared by combining the activated carbon powder with ethanol and then stirred for 1 hour. The carbon paste was applied at the conductive site of the FTO glass using the same doctor blade technique [1], referring to the scale of the ZnO paste coated on the glass. The electrode was heated at 250 ° C for 30 minutes to minimize the organic contamination and to strengthen the material.

### Assembly of DSSC

The ZnO coated glass was immersed in hybrid natural dye for 24 h for dye staining. The glass plates were cleaned with ethanol and dried in the air for a few minutes. Lastly, the dye-sensitized solar cells are constructed by sandwiching the redox electrolyte between the dye-adsorbed ZnO film electrodes and the coated counter electrode by binding it tightly.

### Cell Characterization

Phase identification of the nanomaterials were studied using Bruker D8 Advance which was operated in Bragg Brentano geometry and exposed to CuK $\alpha$  radiation ( $\lambda = 1.540\text{\AA}$ ). The X-ray diffraction (XRD) pattern was scanned with step size of 0.02° (2 $\theta$ ) at a fixed counting time of 71.6 s from 10° to 90° 2 $\theta$ . The resulting powder diffraction patterns were analyzed using Highscore Plus software. UV-vis (HP 8453) was used to determine the absorption spectra of all samples. The photocurrent – voltage (I – V) curves of the DSSCs were quantified using a computer-controlled digital source meter (Keithley 2400) with an irradiation exposure of 100 mWcm<sup>-2</sup>.

## RESULTS AND DISCUSSION

Figure 1 shows the XRD patterns of the pure ZnO. The peaks indicate that the complete hexagonal structure of wurtzite (ICDD file no. 98-004-5318). The crystallite size was measured as per Scherer's equation, where the average crystallite size of pure ZnO is 19.1 nm. The mean of crystallite size clearly interpreted that it has larger surface which allowing more pigment added to it and acts as photon dispersers in the prominent place of incident light, resulting in improved productivity of light absorption.

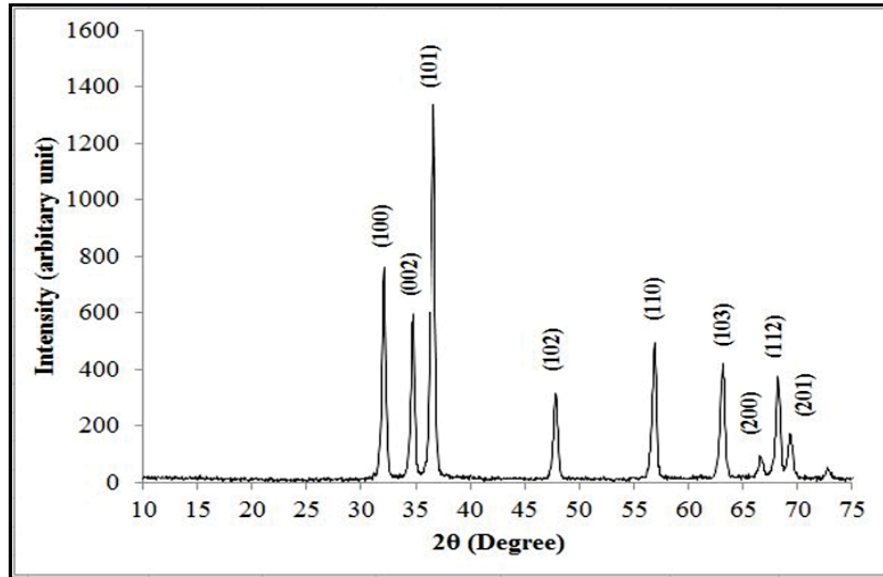


FIGURE 1. XRD patterns of pure ZnO

Fig. 2 shows the absorption spectrum of *E. conferta*, *P. betle*, and dwi-hybrid dye solutions, respectively. The visible absorption band transitions to greater energy states, showing a broad maximum around 530–550 nm with maximum absorption at 542 for *E. conferta* where it absorb more light in the blue-green and red range. The classification of anthocyanin pigments as water-soluble plant substances and pH dependent that do not express their vibrant colours until they are concentrated in the acidic vacuoles and also capture illumination at the highest absorbance [5]. Thus, the variation in the absorption features is owing to the different kind of anthocyanin compound and hue of the pigments [4].

Meanwhile, for *P. betle*, the absorption peak of flavonoid dye is observed at 540 nm where it absorbs more photon in blue-green range. The specific molecules in each flavonoid class are differentiated primarily by the quantity and configuration of the hydroxy and methoxy groups in two benzene rings [6]. Along with flavonoids, the conversion of charge transfers from HOMO to LUMO requiring minimal energy, invigorating dye molecules by visible light, resulting in a huge absorption array in the ultraviolet spectrum [7]. This flavonoid is quickly accumulated to the layer of ZnO by removing the -OH counter ion from the Zinc (Zn) site, which is paired with the proton that is contributed by the flavonoid.

Besides that, the contrasting absorption properties of the anthocyanin and flavonoid pigments were employed in this analysis to combine dwi-hybrid dye, with a set volume proportion (1:1). The dwi-hybrid dye also contains the absorption peaks including both flavonoid and anthocyanin as proven in the Fig. 2. It improved visible light absorption scope about 530-550 nm (green to red zone) with maximum absorption at 539 nm. The spectrum of light absorption of the effective dwi-hybrid extract exhibited bands according to the involvement of the particular samples. Consequently, a greater number of electrons were transferred to ZnO in purpose of strengthening the solar cell's photocurrent power. This intensified productivity of the trihybrid dye is because of the development in ideality element.

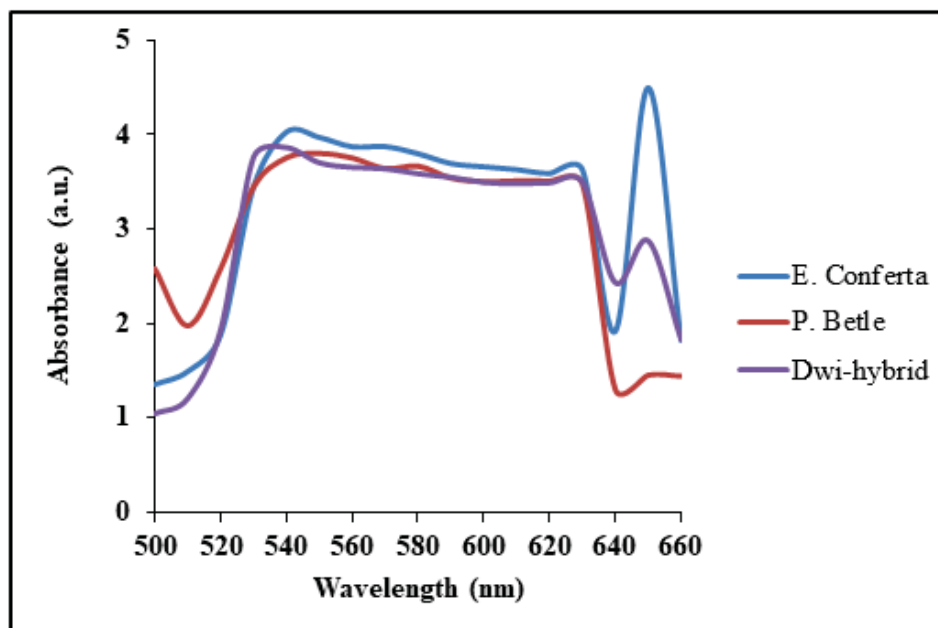


FIGURE 2. UV visible absorption spectra of natural dyes

Table 1 illustrates the photoelectric dimensions for the DSSC sensitised utilizing *E. conferta*, *P. betle* dye and dwi-hybrid dye using ZnO photoanode. Presents the results, *E. conferta* dye generated  $J_{SC} = 5.2 \text{ mA/cm}^2$ ,  $V_{OC} = 0.33 \text{ V}$ ,  $FF = 0.58$  and  $\eta = 1 \%$ . Conversely, dwi-hybrid dye highlighted decent conversion efficiencies where it achieves viable photovoltaic output proving  $J_{SC} = 5.83 \text{ mA/cm}^2$ ,  $V_{OC} = 0.35 \text{ V}$ ,  $FF = 0.63$  and  $\eta = 1.29 \%$  and *P. betle* produced the lowest result which is  $J_{SC} = 4.9 \text{ mA/cm}^2$ ,  $V_{OC} = 0.32 \text{ V}$ ,  $FF = 0.59$  and  $\eta = 0.93 \%$ . While the existence of anthocyanin in *E. conferta* extract is higher on the basis of its absorption peak, but it has weaker efficiency rates relative to dwi-hybrid dye because dwi-hybrid incorporates a mixture of flavonoid and anthocyanin compounds with multiple functional groups, which allow the extract to bind tightly to the oxide surface and improve its light trapping capacity.

Based on Table 1, the cells produced with dwi-hybrid extract gained the maximum conversion efficiency with optimum current density,  $5.83 \text{ mA/cm}^2$ . Natural extracts emitted poor  $V_{OC}$  attributable to potential proactive electron/dye cation recombination mechanisms and acidic dye adsorption conditions.  $H^+$  ions are alternative determinants of ZnO ions, and proton adsorption produces a positive transition in the Fermi level of ZnO, thereby reducing the overall photovoltage that the cells could generate [4]. It has also been shown that the  $V_{OC}$  of organic dye is smaller than that of ruthenium dye, despite of the molecular composition of natural dye, which consists mainly OH ligands and O ligands and lacks  $-COOH$ -ligands that owned by ruthenium dye. The  $-COOH$  ligands may bind with the hydroxyl of the ZnO particles in order to generate ester and improve the interfacial bonding of electrons on the ZnO conduction band in order to obtain a substantial energy transfer level.

TABLE 1. Photovoltaic parameters of DSSCs of dye extractions

Dye source	$J_{SC} (\text{mA/cm}^2)$	$V_{OC} (\text{V})$	FF	$\eta (\%)$
<i>Dwi-hybrid</i>	5.83	0.35	0.63	1.29
<i>E. conferta</i>	5.2	0.33	0.58	1.00
<i>P. betle</i>	4.9	0.32	0.59	0.93

## CONCLUSION

Anthocyanin and flavonoid have been harvested from *E. conferta* and *P. betle* respectively, to represent as natural dyes. In addition, this analysis combined these two pigments in a 1:1 volume ratio to produce dwti-hybrid dye. Through UV visible analysis, the dwti-hybrid dye exhibited the largest absorption wavelength (500-640 nm) accompanied by *E. conferta* in accumulating larger quantities of photon energy, responsible for the formation of more photoelectrons that can strengthen photovoltaic performance. Meanwhile, for photovoltaic performance, the dwti-hybrid recorded the maximum photovoltaic performance compared to others dyes. Dwti-hybrid pigment generated 1.29 % of  $\eta$  with  $V_{oc}$ ,  $J_{sc}$ , and FF values of 0.35V, 5.83 mA/cm<sup>2</sup> and 0.63 respectively.

## ACKNOWLEDGMENTS

The authors would like to thank Faculty of Engineering and Technology, University Malaysia Kelantan for providing all facilities and excellent technical assistance for the completion of this research.

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