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Improving The Efficiency of DSSC with A Novel Multi-dye layers Approach

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Abstract

This study investigates the effect of two different combination methods of natural pigments including anthocyanin from saffron petals and chlorophyll from walnut shell, as sensitizers on the photoelectric conversion efficiency of dye-sensitized solar cells (DSSCs). The photovoltaic properties of chlorophyll and anthocyanin were separately studied and then were blended in a cocktail in equal proportions, by two different combination recipes, to study the photovoltaic properties of the mixed dyes. The first method is the common used method, and in the second one which is proposed in this study, the pigments are layered on top of each other according to their molecular structures. The experimental results showed that the DSSC sensitized with solely anthocyanin exhibited an open-circuit voltage (V_{oc}) of 0.397 V, short circuit current density (J_{sc}) of 2.32 mA/cm^2, fill factor (FF) of 0.71 and conversion efficiency of 0.66%. In the cell sensitized with chlorophyll these parameters were 0.79 V, 0.939 mA/cm^2, 0.71 and 0.528%, respectively. The DSSC sensitized with the common combination method of chlorophyll and anthocyanin exhibited V_{oc} of 0.72 V, J_{sc} of 1.61 mA/cm2, FF of 0.73 and conversion efficiency of 0.85%. While in the cell sensitized with the layered combination of chlorophyll and anthocyanin these parameters were 0.71 V, 1.38 mA/cm2, 0.74 and 0.74%, respectively. The results imply that the combination of pigments can meaningfully improve the cell performance, However the way of pigments combination affects this improvement.

Keywords: Natural pigments, TiO_2 Nanoparticles, DSSCs, hybrid dye

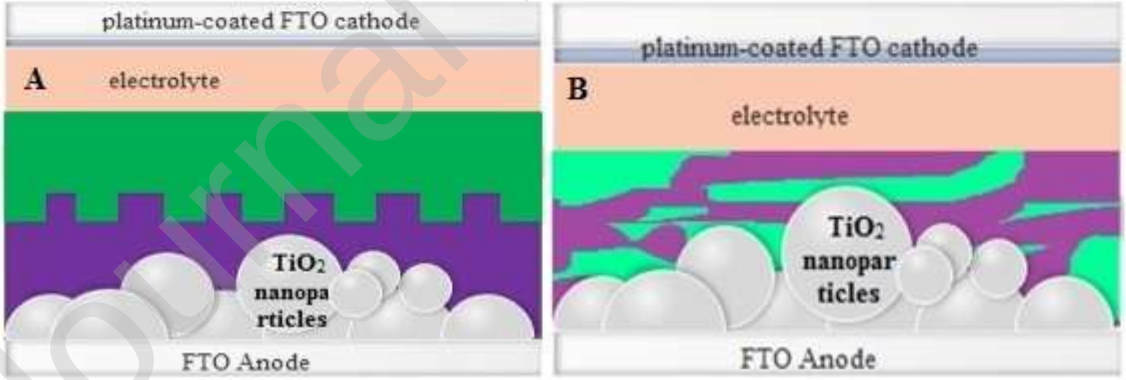
Introduction

Due to the ease of fabrication, cost effective, the ability of generating electricity in cloudy conditions, and also under artificial light (1, 2), the dye sensitized solar cells (DSSCs) have attracted great attention from the scientific community as a potential candidate for alternatives to conventional photovoltaic devices. DSSCs are comprised of an electrode substrate, nonporous TiO_2 film, a dye as a photosensitizer, electrolyte and a counter electrode (3). TiO_2 has a wide band gap energy of about 3.2 eV for anatase structure, the absorption spectra corresponds to 380 nm and thus only a small fraction of the solar spectrum (%5) is absorbed (4-6). So, in DSSCs the photoanode (TiO_2 electrode) is coated with a dye to extend the spectra sensitivity range of the photoelectrode. This approach lets the photoanode to collect the lower energy photons and improves the efficiency of the DSSCs. Therefore, dye plays an important role in the dye-sensitized solar cells for absorbing light. A good dye has to possess the following properties (7):

1- Its absorption spectrum should cover a broad range of wavelengths, ranging from the visible to the near-IR region of the solar spectrum.
2- Efficient electron injection into the photoanode.

Many different dyes, including inorganic, organic and hybrid dyes have been studied as sensitizers in the DSSCs (8-10). Among the photo sensitizers, the ruthenium polypyridyl complex inorganic dyes could achieve the efficiency as high as 13 % (7). But these dyes contain some heavy metals which are toxic and need to go through a long, tedious and expensive purification processes (11, 12). Therefore, further researches have been alternatively focused on the use of natural dyes. Several natural dyes, such as chlorophyll, anthocyanin and carotene have been studied as photo sensitizer in the DSSCs (13-19). Despite the inherent advantages, the efficiency of the solar cells with natural dyes still remains low. The main reason behind this deficiency is due to their lower optical absorption band (i.e. 400-800 nm), which is much narrower compared to crystalline silicon (i.e. 500-1100 nm) (20). One solution to overcome this limitation is mixing two dyes. The philosophy of this procedure is based on cumulative absorption capability of the dyes and hence providing a wider wavelength regions for absorbing solar radiations. Extensive researches have been carried out on the effect of mixing different natural dyes as photosensitizer on the efficiency of DSSCs (16, 18, 21-24). However, their outcomes are apparently conflicting. Some have reported that the dye mixing reduces efficiency (22, 23), while the others reported an opposing claim (16, 18, 21, 24).

This study, therefore, was conducted to further explore of this contradiction. The main objective was to investigate the effect of two different combining methods of natural dyes on the efficiency of DSSCs. In the first method, two dyes are mixed as one layer (OL[1]) that is a commonly used mixing method. In the second method, which is proposed through this study, the dyes are placed on top of each other, as two distinct layers (TL[2]) (Fig. 1).



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[1] One layer
[2] Two layers

Figure 1. The schematic view of the structure of the DSSCs sensitized by two methods of natural pigments combination. Placing the dyes as two distinct layers (A) the dyes are mixed in one combined layer (B). The green and purple areas represent the dyes.

The dyes include anthocyanin extracted from saffron petal and chlorophyll from walnut shell. Because the anthocyanin of the saffron petal is Delphinidin which has six –OH groups (25), it can make a strong bond with TiO_2 nanoparticles (26). The chlorophyll is responsible for absorbing light in plants (27). Subsequently, the photovoltaic performance of the anthocyanin extracted from saffron petal and the chlorophyll extracted from walnut shell, which are regarded as agricultural wastes, were studied and then the photovoltaic performance of their mixed dyes was investigated. The photovoltaic performance of DSSCs was evaluated by measuring current-voltage characteristics and the incident photon to current efficiency (IPCE). Also, the individual dyes and the mixed dyes were characterized by ultraviolet-visible (UV-Vis) and Fourier transform infrared (FTIR) spectroscopy.

Materials and method

Preparation of the natural dyes

The dyes extracted using acidic ethanol as extracting solvent. For an effective extraction, ultrasonication was carried out using an ultrasonic processor (Sonic VCX 750) at a frequency of 20 kHz. Purification of the anthocyanin and the chlorophyll were performed by Solid Phase Extraction (SPE) and Schertz method (28), respectively. The details of the procedures have been described in the literature (13, 29).

Dye coating and assembly of solar cell

To prepare the transparent electrodes, a Fluorine-doped Tin Oxide (FTO) conductive glass sheet having resistance of 15 Ω/□ was purchased from Dyesol Company (Australia). The FTO glass was first cleaned in a detergent solution and then rinsed with distilled water and ethanol. The FTO glass was cut into some pieces and then were immersed in 100 mL of TiCl4 water solution (40 mM) at 70 °C for 30 min, then rinsed with distilled water and ethanol and dried in an oven at 80 °C. A thin film of titanium dioxide (TiO_2) was coated on the FTO glass plates using “doctor-blading” technique. The coated plates were gradually heated at 325 °C for 5 min, then they were heated up to 375 °C and finally were sintered at 450 °C for 30 min. This procedure enhances the compactness of the internal voids of the film’s structure and eliminates the organic impurities. Consequently, the inter-connection of the TiO_2 nanoparticles is improved and hence the absorption performance of TiO_2 film is enhanced (30) . Subsequently, the TiO_2 film was treated in 40 mM TiCl4 solution at 70 °C for 30 min, then rinsed with water and ethanol and sintered at 500 °C. The films prepared by this method attained a thickness of 10 μm. To prepare the electrodes the sintered TiO_2 films were cooled down to room temperature and then immersed in the pre prepared solutions of the natural dyes that extracted from saffron petals, walnut shell and their mixture (chlorophyll and anthocyanin) in volumetric proportion of 1:1 for 24 h.

With this method three solar cells were prepared with three different dye configurations. The fourth solar cell was prepared based on the method suggested in the current study (TL), comprising the mixed dye of chlorophyll and anthocyanin with a layered configuration. The philosophy of this method was behind the aforementioned properties of a good dye. To fabricate this cell: firstly, the TiO_2 film was immersed in the anthocyanin dye solution for 24 h and then immersed in the chlorophyll solution for further 24 h. The outcome was a two-layer dye.

Anthocyanin has –OH group which it makes a strong bond with TiO_2 nanoparticles (7). On the other hand, chlorophyll is responsible for light absorbance in plants (28). In other words, this cell configuration which is proposed in the current study, uses chlorophyll for the purpose of light absorption (collection) and employs anthocyanin for makeing a strong bond with TiO_2 for efficient electron injection into the photoanode.

To prepare the counter-electrode a hole of 1.5 mm in diameter, was drilled in each FTO-glass plates. The perforated plates (as substrates) were washed and cleaned with water and ethanol in order to remove any residual of glass powder or organic contaminants. The Pt catalyst was deposited on the conductive face of the FTO plates by dropping hexachloroplatinic acid (H_2PtCl_6) solution (5 mM in isopropanol) followed by heating at 500 °C for 30 min. They were then sintered in air at 350-400 °C for 1 h in a furnace and finally cooled down to room temperature (31). The anode and the Pt counter-electrode were assembled into a sandwich type cell and sealed using a thermopress with a hot melt gasket of 25 μm thickness made of the Surlyn ionomer (Solaronix). The aperture of the Surlyn frame was larger than the TiO_2 area. A drop of electrolyte solution was placed on the hole in the back of the cathode. The electrolyte (LiI 0.8 M, I2 0.05 M, in acetonitrile) was introduced into the cell by means of vacuum backfilling. The cell was placed in a small vacuum chamber to remove inside air. Again by exposing the cell to ambient pressure, the electrolyte was driven into the cell. Finally, the hole was sealed and closed by heating another Surlyn film and putting a thin cover glass onto the hole (17).

The instrumentations and measurements

The UV–Vis absorption spectra of the dyes were recorded by a Perkin Elmer L25 spectrophotometer (in the range of 200-1100 nm). The Perkin Elmer Spectrum RX I FT-IR instrument was used to identify the functional groups of samples. The current-voltage curves were provided using an analyzer (potentiostat/galvanostat, Palmsens) under the AM 1.5 (100 mW/cm^2) illumination from a solar simulator (Luzchem, Canada). The incident photon to current conversion efficiency (IPCE) was obtained by measuring short-circuit photocurrent under incident monochromatic light irradiated by monochromatic incident light of 1 × 1016 photon/cm^2 under 100 mW/cm^2 with bias light in DC mode (model Jarrel Ash monochromator), using a 100W halogen lamp and a Thorlabs calibrated photodiode.

Result and discussion

The dyes absorption spectra

In order to understand the visible-light response of the different dyes (including extracted from saffron petal, from walnut shell and the combined dyes), (gray curves in Fig. 2) and the dyes after adsorption on the TiO_2 surface (black curve), the UV–Vis absorption spectra were recorded. In the UV-Vis spectra of chlorophyll (gray curve in Fig. 2(B)) a maximum peak is seen at 461 nm, in comparison to UV-Vis the spectrum of anthocyanin and the combined dye (the gray curve in Fig. 2 (A) and (C)). This is mainly due to good absorption of chlorophyll in 400 to 700 nm. However, when the chlorophyll adsorbs on TiO_2, the peak has a sharp drop (the black curve in Fig. 2 (B)). This may be due to the ability of chlorophyll in making complex with metal. In other words, a complex between chlorophyll and TiO_2 is probably created, consequently the conjugated bonds are decreased. In the anthocyanin, when dye adsorbed on TiO_2 surface, the red shifting from 510 nm to 530 nm (Fig. 2 A) might be due to the six –OH group in its structure (25). The UV–Vis absorption spectrum of the combined dye after adsorption on TiO_2 has two peaks at 520 nm and 680 nm (the black curve in Fig. 2 (C)). This is more suitable than the dye with solely chlorophyll or anthocyanin. The red shifting from 500 nm to 520 nm and 660 nm to 680 nm is due to the –OH groups in its structure and its absorption spectrum cover a broad range of wavelengths. So it seems that the absorption spectrum of the combined dye is better than the absorption spectrum of the dye with only anthocyanin or chlorophyll, because it has both aforementioned properties of a good dye.

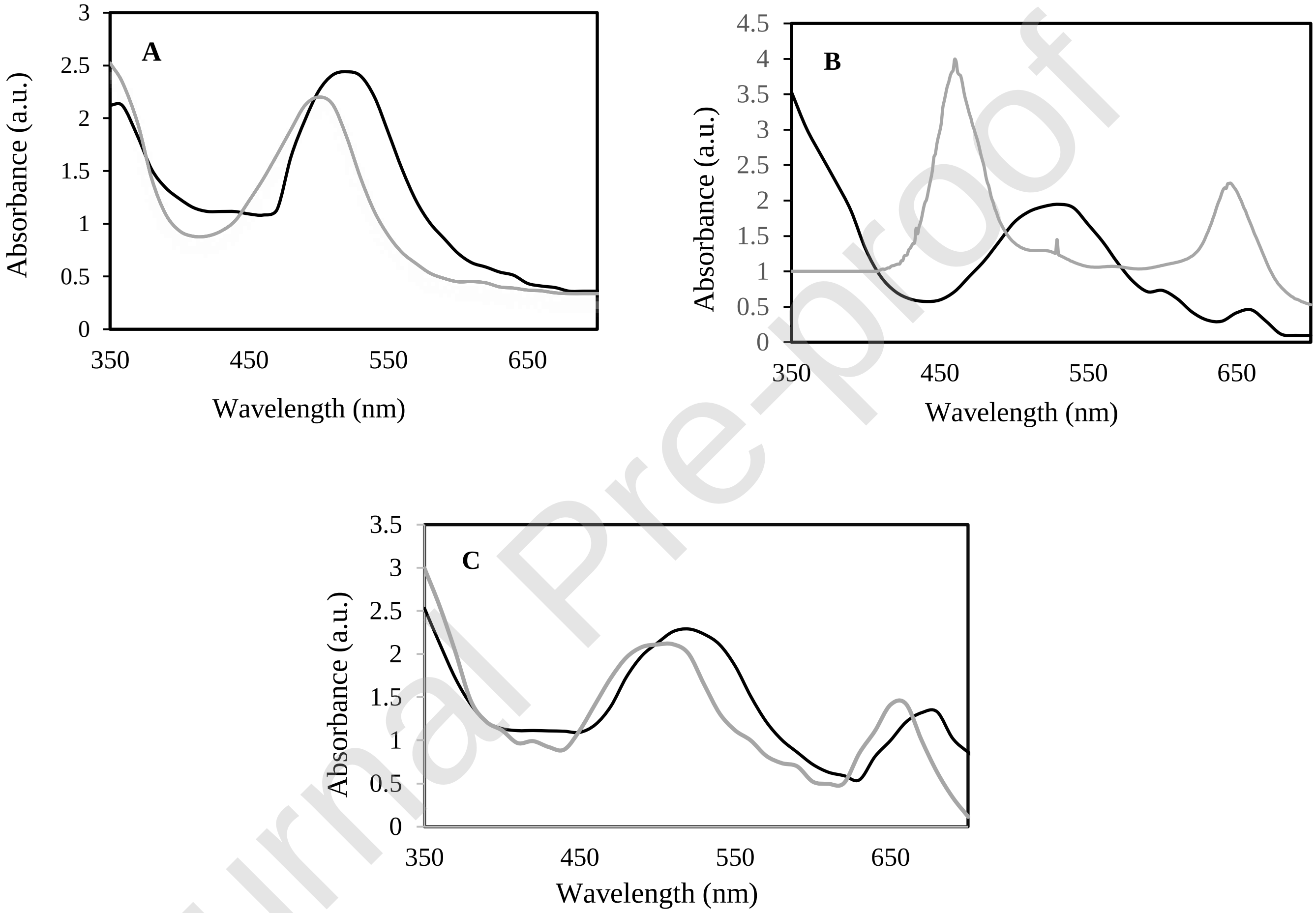

Fig.2. The UV–Vis absorption spectrum of the dye solution (shown as gray curves): (A) saffron petal extraction; (B) chlorophyll from walnut shell; (C) the combined dye and the dyes after adsorption onto the TiO_2 surface (the corresponding black curves).

FTIR Spectroscopy

Because of predetermined structure of natural dyes, the purpose of combining the dyes as layered configuration (TL method), was to have the cumulative properties of the individual dyes in the cell. In other words, in the TL method of layered mixing of the dyes the anthocyanin is responsible for making a strong bond with TiO_2 and the chlorophyll was responsible for light absorbance. But when the dyes are mixed as a single layer, there is no enough information about the structure and inter connection between the dyes. Therefore, the Fourier transform infrared (FTIR) test was conducted to explore the bond created between the dyes.

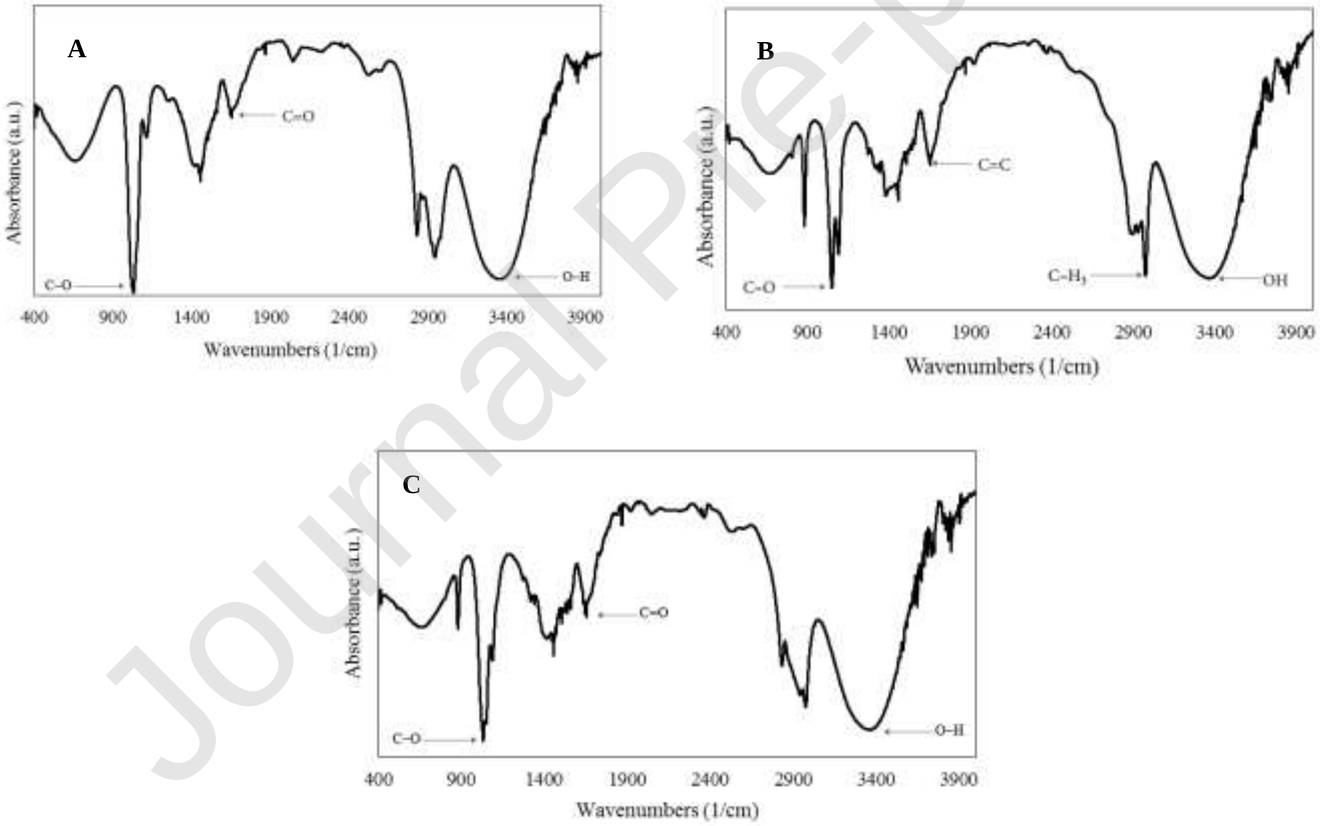

Fig.3. Fourier transform infrared (FTIR) spectra of anthocyanin (A), chlorophyll (B) and the combined dye (C).

The FTIR spectra of the anthocyanin, chlorophyll and combined dyes are shown in Fig.3. The FTIR spectra reveal the existence of –OH, C-O and C=O groups in the molecular structure of the dyes, which can be bound to the surface of TiO_2 film (32). The comparison of the FTIR spectrum of the combined dye (Fig.3 (C)) with the FTIR spectrum of anthocyanin (Fig. 3 (A)) and chlorophyll (Fig. 3 (B)) indicates that there is functional group. As a result, we believe that the physical bonds are the main links between the two dyes in the OL and TL methods.

Incident Photon to Current Efficiency (IPCE)

The IPCE values of the DSSCs sensitized with anthocyanin, chlorophyll, OL combination and TL combination methods are shown in Fig. 4. As seen, the maximum IPCE of anthocyanin is about 22% (Fig. 4(a)). This occurred at 540 nm and red-shifted by 30 nm relative to the maximum absorption peak wavelength. However, for chlorophyll, the maximum value of IPCE occurred at a lower wavelength (Fig. 4(b)). The IPCE curve of chlorophyll shows that only the high-energy electrons could be transmitted to the conduction band of TiO_2. This might be due to the long chain R or a three-dimensional space barrier in the structure of chlorophyll (33-35).

The IPCE of the DSSC sensitized with OL combination method is shown in Fig. 4(c). The maximum value of IPCE is 25% at 400 nm (Fig. 4(c)). This implies that only the high-energy electrons could transmitted to the conduction band of TiO_2, which is not compatible with the UV-Vis spectrum of OL method (Fig. 2). It may be related to the configuration of the combined dye. When anthocyanin and chlorophyll are mixed in one layer, the chlorophyll covers most of the TiO_2 surface and makes a space barrier in transmit of electrons to the conduction band (CB) of TiO_2. This is one of the main drawbacks of the single layer or one layer (OL) combination method.

Fig. 3(d) shows the IPCE of the DSSC sensitized with TL combination method. The maximum value of IPCE is 22% at 400 nm and it is not compatible with UV-Vis spectrum of the combined dye method (Fig. 2(C)). We believe that this might be due to the thickness of sensitizer layer. In this method the anthocyanin and chlorophyll are placed in two individual layers. Since electrons have a short time of about100 ps to transmit to the conduction band (CB) of TiO_2 (32), most of them cannot reach to the CB of semiconductor. The IPCE curve shows that in this DSSC only high-energy electrons could pass through the layer of sensitizer and reach to CB of TiO_2 and increase the rate of recombination in DSSC. Overall, the higher values of IPCE are seen for both OL and TL combination methods in comparison with the individual dye of anthocyanin and chlorophyll. It could be related to cumulative utilization of the properties of the two dyes in the DSSC.

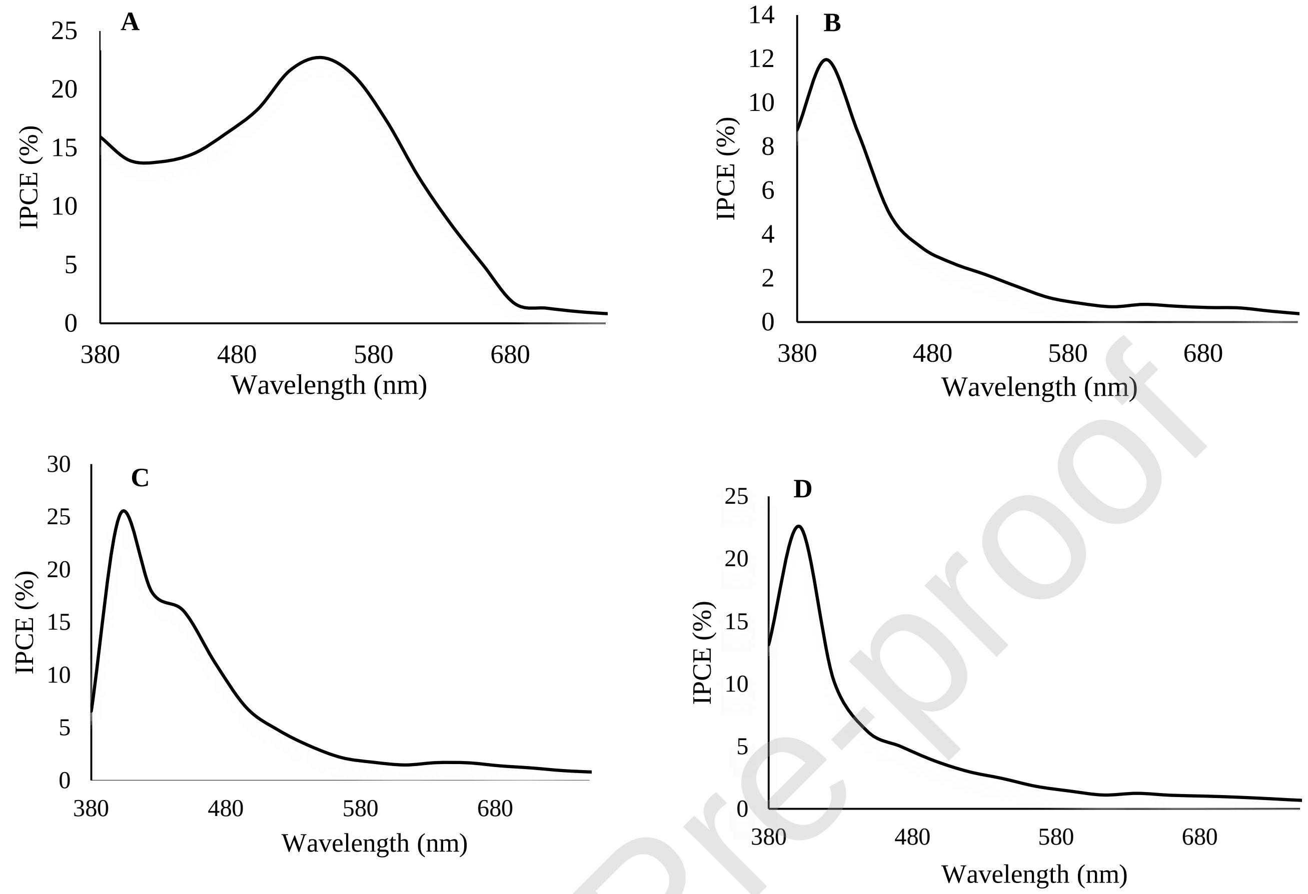

Fig. 4. The IPCE curve photoaction spectra of the DSSCs with: anthocyanin (A), chlorophyll (B), OL combination method (C) and TL combination method (D).

Photovoltaic Performance of the DSSCs

Fig. 5 shows the J–V curves of the TiO_2-based DSSCs sensitized with anthocyanin (a), chlorophyll (b), OL combination method (c) and TL combination method. The photovoltaic performance parameters including fill factor (FF) and power conversion efficiency (%) are defined as follow (7):

$$FF = \frac{J_{max} \times V_{max}}{J_{SC} \times V_{OC}} \tag{1}$$

$$\eta\ (\%) = \frac{J_{SC} \times V_{OC} \times FF}{I_{Inc}} \times 100 \tag{2}$$

where J_{sc} is the short-circuit current density (mA/cm^2), and V_{oc} is the open-circuit voltage (V). J_{max} (mA/cm^2) and V_{max} (V) are the current density and voltage, respectively, in the J–V curve, at the point of maximum power output. The photovoltaic performance of the DSSCs are shown in Table 1.

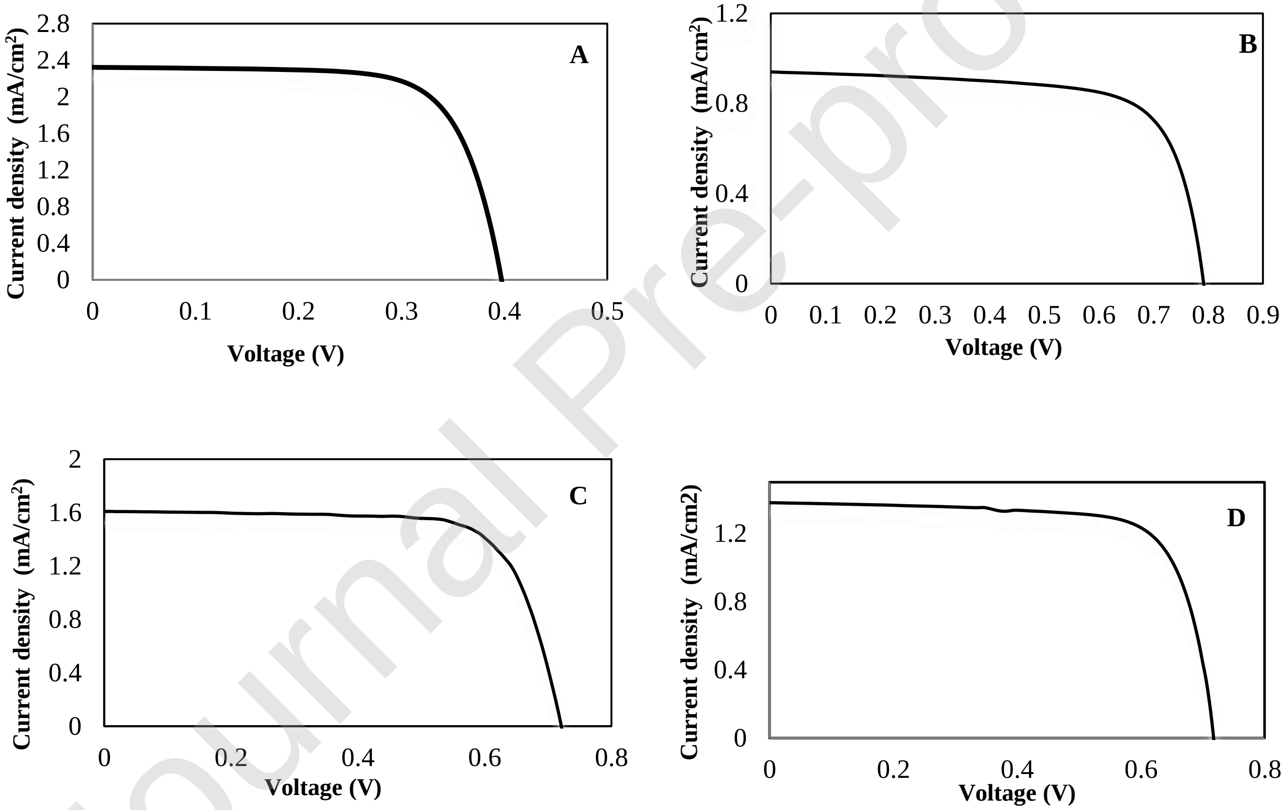

Fig. 5. Photocurrent–voltage (J–V) curve for dye-sensitized solar cell using : anthocyanin (A), chlorophyll (B), OL combination method (C) and TL combination method (D).

Table 1. The photovoltaic performance of the synthesized DSSCs.

Type of dye DSSCs	V_{oc} (v)	J_{SC} (mA/cm^2)	FF (%)	η(%)
Anthocyanin	0.397	2.32	0.71	0.66
Chlorophyll	0.79	0.939	0.71	0.528
OL combination method	0.72	1.61	0.73	0.85
TL combination method	0.71	1.38	0.74	0.74

As seen in Table 1, the DSSC sensitized with chlorophyll acquired the lowest J_{SC} (0.939 mA/cm^2 and Fig. 2(a)) in comparison with the other solar cells provided in this study. This is probably due to the long chain R or a three-dimensional space barrier in the structure of chlorophyll (32) that reduces the short-circuit current density of this solar cell (Fig. 6).

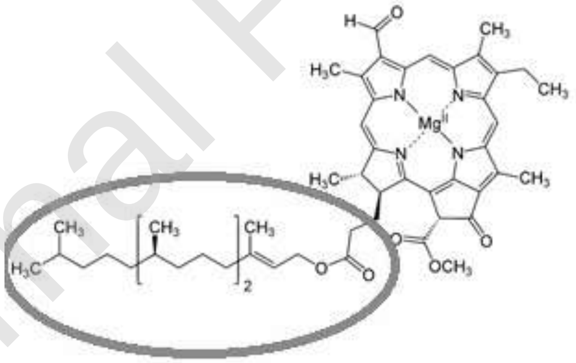

Fig. 6. The chlorophyll structure and long chain the R in its structure (represented in the ellipse).

The DSSC sensitized with anthocyanin has the lowest V_{OC} (0.397 V) among the other cells. In our opinion, this drawback cannot be related to the structure of the dye. Because in this case, the structure of the dye comprises of Delphinidin, which has six –OH group in its structure, can make a strong bond with TiO_2 (25, 26). The high J_{SC} of this cell (2.32 mA/cm^2) is a good evidence of this claim. We believe that the most important factor in voltage drop in this cell is probably due to the incompatibility between the quasi-Fermi level of the semiconductor and the redox potential of charge mediator (7) and especially the incompatibility between redox potential of electrolyte and HOMO level of the dye that reduces the voltage of this cell.

Among all cells considered in this study, the DSSCs sensitized with OL combination method and TL combination method (Fig. 5 (c and d) and Table 1) are ranked as first and second, in terms of efficiency, respectively. The followings can be stated to justify this ranking:

1- The absorption spectrum covers a broad range of wavelengths.
2- Efficient electron injection into the photoanode.

Considering the overall performance parameters of the cells (Table 1), however, a significant difference was not observed between the performance of the cells manufactured with OL and TL combination methods.

Conclusion

The main objective of this study was to investigate the effect of hybridization of different natural dyes, with two method of combinations, on the performance of DSSCs and to find an answer to the existing contradiction. The multi-dye approach has a number of merits.

One can be said is the ability to span the solar spectrum via absorption with a variety of dyes, each of which absorbs over a different spectral region. The results showed that generally the combination of dyes increases the efficiency of cells to some extent. However, this is not a satisfying way of increasing the solar cell efficiency, because according to the IPCE curves of the DSSCs sensitized with OL and TL combination methods, the peaks occurred only around the low wavelength range of the solar spectrum and not for the entire solar wavelength range. Hence, in these solar cells, the high-energy photons are primarily responsible for increasing cell efficiency. In other words, only these photons are mainly converted to current. While most of the light receiving to the earth's surface during the day comprises of low-energy photons. Therefore, it can be concluded that for appropriate efficiency improvement of natural DSSCs, the structure of the dye needs to be changed to a stable form to absorb the energy from wider solar visible to near-IR spectral region and hence facilitate the injection of electrons to the conduction band of semiconductor. In all, we believe that, natural dyes could be a good alternative for dye synthesizing. However, there is still a long way to go, and efforts must be made to ensure that their structures are well engineered.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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