



(RESEARCH ARTICLE)



ENHANCING DSSC EFFICIENCY WITH CONDUCTING PPY/GO - DECORATED COUNTER ELECTRODES AND PHOTOANODE SENSITIZED BY NATURAL DYES

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ABSTRACT

This research is to study the preparation of conducting polymer decorated Ppy/GO composite material for uses as hybrid counter electrode in dye sensitized solar cell. It started with the synthesis of Ppy/GO composite and X ray, SEM characterization has been studied, followed by the preparation of counter electrode, exaction of dyes from the the seeds of Bixa Orellana, flower of Cannal lily, Fruits of Basella Alba for preparation of photoanode. Using UV- vis spectrometer, optical properties has been studied out for all the extracted dyes. DSSC is fabricated by assembling extracted dye sensitized photoanode and Ppy/GO coated counter electrode. The photovoltaic characteristics had been studied and efficiency of DSSC is calculated, results obtained for dyes from the fruits of Basella Alba, flower of Cannal lily, seeds of Bixa Orellana respectively as an open circuit voltage (Voc) of 17mV, 18mV, 18mV, Short circuit current 167mA, 183 mA, 173mA, Fill factor 0.42, 0.54, 0.55 and conversion efficiency 0.164, 0.246, 0.24.

Keywords: Dye sensitized solar cell, Ppy/Go composite counter electrode, Polypyrrole, Polymer composite, Natural dye

1 INTRODUCTION

Among all other renewable energy sources, solar energy is safer and inexhaustible source of energy and it has greatest potential[1] . Photovoltaic's convert sunlight directly into electricity [2]. From the last three decades, the solar cell market is dominated by silicon-based devices[3]. However, recently DSSC have gained increasing interest due to its simple and low-cost manufacturing process [4]. A DSSC constitutes a photoelectrode and a catalytic electrode with an electrolyte between them. In 1991, O'Regan and Gratzel developed DSSC that work on the principle of plant photosynthesis and the efficiency of this cell was reported as 7.1% and 7.9%[5]. Usually, dye sensitized solar cells

consist of a mesoporous titanium dioxide film, which is sensitized by a dye, in combination with a liquid or solid-state material. In hybrid solar cells the acceptor-type material is replaced by inorganic semiconductors like TiO_2 , CdS, CdSe, PbS, PbSe, ZnO, or SnO_2 [6–12]. The DSSC sensitized by Ru compound absorbed onto nanoscale TiO_2 is most efficient and has efficiency 11-12% [13].

But ruthenium dye, including N3 and N719 are costly and hazardous to environment, thus other numerous metal free organic dyes have been used in DSSC [14]. Thus, the natural dyes extracted from different parts of plant like leaves, fruits, flower, root, seed have been used as a sensitizer for DSSCs.

G. Calogero et al.[15] reported that Betalaine pigment containing red turnip has highest efficiency of 1.70%. Azira Khairudinet al. [16] have studied mangosteen pericarp as a sensitizer with efficiency 1.17%. Yang C-S et al. have used *Rhoeo* Spathacea as a natural sensitizer and reported its efficiency as 1.02%. Bayron Cerda et al. have used Maqui, Black Myrtle, Spinach and spinach black myrtle as natural sensitizer to increase the efficiency of DSSC and concluded that black myrtle has more efficiency as 0.040% among them. Lawrence Amadi et al. [17] have studied the creation of natural dye sensitizer solar cell using vegetable dye from Red Cabbage, Green Cabbage, Spinach, Red Potato, Radish and concluded that green cabbage gives the cell efficiency as 0.1% while red cabbage as 0.09% and radish and Potato as 0.06%. S.Rajkumar and K. Suguna [18] used Beet root and Pomegranate fruit to enhance the efficiency of DSSC. Monzir S et al. [19] used fifteen different dyes to study DSSC and found that schinus *Terebinthifolius* Leaves shows best performance for DSSC with photoelectrochemical parameter of $J_{sc} = 2.40 \text{ mA/cm}^2$, $V_{oc} = 0.68$, $FF = 0.44$ and efficiency $\eta = 0.73\%$.

It has been observed that there is a lot of work to be done in order to improve quantum efficiency of dye-sensitized solar cell. The advantages of natural dyes as photosensitizers are large absorption coefficients, high light-harvesting efficiency, no resource limitations, low cost, simple preparation techniques and no harm to the environment.

In this present work we used the natural dyes extracted from different part of the plant such as seeds of *Bixa Orellana*, Fruits of *Basella Alba*, flower of *Canna Lily*

Basella Alba (Red Vine Spinach) as a sensitizer: - *Basella Alba*, bearing deep-green leaves, is commonly grown and harvested as a novel leafy vegetable in tropical and non tropical region, particularly during the summer season when the production of other leafy vegetables is short. Grown into the fall and winter seasons, the vines produce a substantial quantity of fleshy and dark blue fruits, which are usually discarded by farmers. Value-added use of the natural pigments from the fruits as colorants and bioactive ingredient in related product development deserves research interest.

Basella Alba Fruit Red Pigment Analysis at Various Maturities concluded that three red-violet pigments were detected in ripe *B. alba* fruits. Gomphrenin I was identified as the major pigment. In addition to gomphrenin I, betanidin-dihexose and isobetanindihexose were also detected. *B. alba* fruits could be regarded as a potent source of natural colorant [20].

The Flower of Canna Lily: - The *Canna lily* is the most common garden plants with yellow and red color flower. *Canna lily* are easy to grow and low maintenance, and both flower and foliage offer long lasting color. The dye extracted from the red flower of it, four anthocyanin pigment detected namely Cyanidin-3-O-(6"-O- α -rhamnopyranosyl)- β -glucopyranoside, Cyanidin-3-O- (6"-O- α -rhamnopyranosyl)- β -galactopyranoside, Cyanidin-3-O- β -glucopyraoside and Cyanidin-O- β -galactctopyranoside[21]

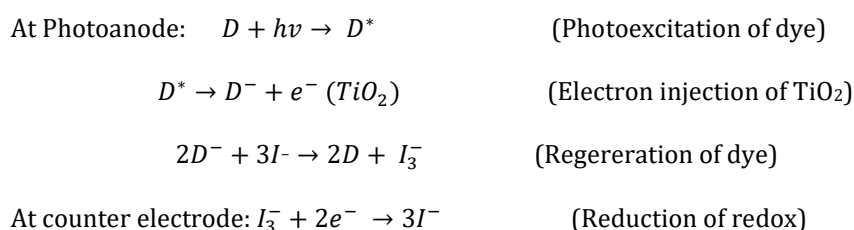
The Bixa Orellana: - *Achiote* (*Bixaorellana*) is a plant native to Brazil but grows in other regions of South and Central America. It is grown in tropical countries such as Peru, Mexico, Ecuador, Indonesia, India, Kenya, and East Africa [22]. From the seeds of this plant, one of the dyes most frequently used worldwide, not only in food products but also in the textile, paint, and cosmetic industries, is produced. It is among one of the dye accepted by World Health Organization (WHO), when other synthetic dye was banned in food and cosmetics and hence its use has been stimulated. It is non-toxic and does not change the food value. Another interesting fact is that 70% of all natural coloring agents consumed worldwide are derived from annatto. Annatto first used in the form of food coloring, also known as paprika, a condiment widely used in cooking to enhance the color of food. However, now its use has spread into many segments of industrial production. Thus, it is now applied on the skin—in the form of makeup like lipstick and sunscreen—and there is research proving that its use brings health benefits [23] *Bixa Orellana* consist of Bixin pigment in high concentration which is a red-colored carotenoid. It is the main substance responsible for the dyeing characteristics of seeds, where its concentration can be as high as 5.0%.[24]. Bixin was isolated for the first time from the seeds of *Bixa Orellana* in 1875 and in 1961 its complete chemical structure and stereochemistry were determined. Bixin belongs to the small class of natural Apo carotenoids, whose formation occurs by the oxidative degradation of carotenoids. Bixin consists of a chain of 25 carbons and has the molecular formula $\text{C}_{25}\text{H}_{30}\text{O}_4$ (MW = 394.51). It has a carboxylic acid and methyl ester group at the ends of the chain. [25]

Operating Principles of Dye Sensitized Solar Cell: - The DSSC differs from other solar cell by its basic construction and the physical processes behind its operation. The principle of operation of a DSSC is quite simple. The basic idea is to separate the light absorption process from the charge collection process, mimicking natural light harvesting procedures in photosynthesis, by combining dye sensitizers with semiconductor.

Light absorption and electron ejection: - As sunlight strikes the cell, photons of different energy penetrate into the dye layer and TiO₂ nanoparticle. If the energy gap of dye molecule is comparable with the incident photon energy, then it will be absorbed by the dye promoting one of electron from HOMO to LUMO. This excited electron further injected into the excited level of TiO₂ through the interfacial bond between dye and TiO₂. In this process the charge separation takes place and this process of the electron injection is one of the fastest physical phenomena and it occurs on the femtosecond scale [26]

Electron Transport and Collection In order to generate electrical energy, photo-injected electrons in the conduction band of the semiconductor should be transported through the mesoporous network by diffusion reaching the FTO. The electrons reach the counter electrode through the external load. The time scale at which the transport occurs is in the millisecond range.

Dye Regeneration After the electron injection the dye molecule becomes oxidized, therefore it has to be consequently reduced to ensure the continuous operation of the device. This is done by the reduced form of the redox mediator present in the electrolyte. Therefore, the oxidized dye molecules are regenerated by receiving electrons from the I⁻ ion redox mediator that became oxidized to I₃⁻ (Tri-iodide ions). For efficient operation of DSSC, the oxidized dye should be reduced to its original state after the electron injection as fast as possible. The oxidized form of the redox mediator I₃⁻ - needs to be reduced and this takes place at the counter electrode, which substitutes the internally donated electron with that from the external load and reduced back to I⁻. Therefore, from this cycle, generation of electric power in DSSC causes no permanent chemical change or transformation. The operating cycle of DSSC can be summarized in the following chemical reaction.[27]



2 EXPERIMENTAL SECTION

2.1 The preparation of Ppy/GO counter electrode

To synthesized the polypyrrole by chemical polymerization method. The Polypyrrole was dried for 2 days at room temperature. The Ppy/GO composite was synthesized by in situ oxidative polymerization of monomer. The Ppy/GO composite was dried for 2 days at room temperature. The Counter electrode is composed of conducting glass substrate was coated with Ppy/GO with the help of doctor blade method and dried, and used as counter electrode.

2.2 Preparation of TiO₂ paste

TiO₂ paste was prepared from homogeneous mixture of TiO₂ nanopowder and Ethyl cellulose. TiO₂ nanopowder and ethyl cellulose was mixed in mortar for about 30 min to make the homogeneous mixture and α -terpnol and ethanol was added gradually. Few drops of acetyl acetone were added to the mixture. The slurry obtained is then kept for ultrasonication for 1h.

2.3 Extraction of dye

The sensitizer plays an important role in DSSCs where it harvests the energy from the sun radiation and produces excited electron at the interfaces of the semiconductor. For the present work we used the five different dyes extracted from different parts of plant like leaves, flower, fruits and seeds. In the present work, we have used Basella Alba (Red Vine Spinach) and seeds of Bixa Orellana (Shendri). During the extraction of natural sensitizers, 30g of *Bixa Orellana*, 30 g of *Canna Lily* flower, 30g of Basella Alba were weighed properly and washed with distilled water in order to remove the dust and dirt spread over it. The cleaned plant parts were then immersed in ethanol bath and sealed with aluminum foil for 24 hours. After 24 hours all the solutions so prepared were filtered and the filtrate obtain were stored in a bottle. In addition to these dyes, hybrid dye also called cocktail, was also prepared by mixing the 20 ml of each sensitizer in a beaker. All these sensitizers were then stored in bottle covered with aluminium foil.

2.4 Preparation of TiO₂ photoanode

The photoanode is composed of conducting glass substrate was coated with titanium dioxide with the help of Doctor blade method. The coated film of TiO₂ on FTO glass was annealed for 1 h at 450 °C. After that the glass slide is left dipped into dye for 24 hr so that the adequate adsorption of dye occur into the TiO₂ film. The electrode was then taken out of dye solution and rinsed with distilled water and ethanol, and used as photoanode for solar cell.

2.5 Assembling complete Dye sensitized solar cell

TiO₂ coated glass Slide was used as photoanode while PPy/GO coated FTO glass slide was used as counter electrode. The counter electrode was kept on working electrode and to avoid the direct contact between them, the spacer was inserted between them. The space between working and counter electrode is filled with electrolyte. To couple the electrode the binder clips on opposite sides were used.

3 RESULT AND DISCUSSION

3.1 Structural Characterization

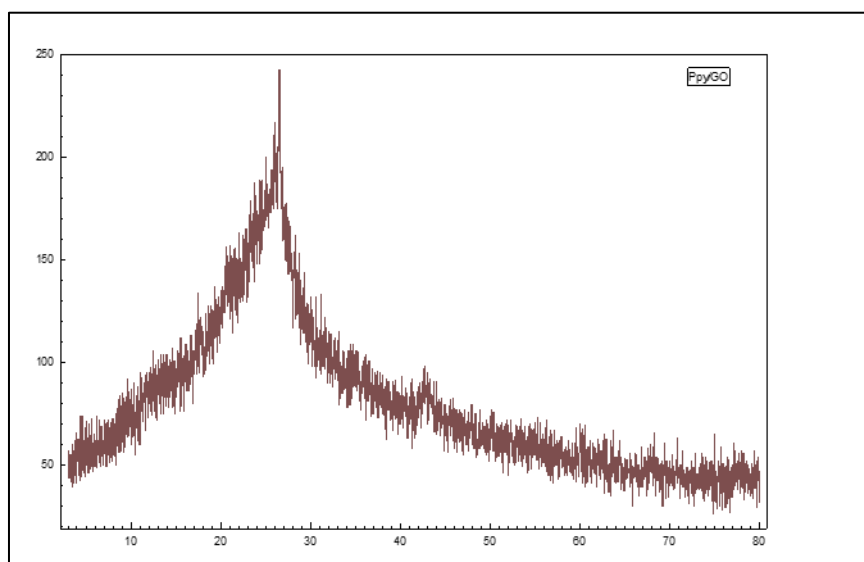


Figure 1: XRD characteristic of Ppy/GO

The crystallographic structure of the PPy/GO composite was examined by X-ray diffraction, and the resulting pattern is dominated by a single broad, low-intensity halo spanning the low- to mid-2θ region, with the broad peak located near of $2\theta \approx 25^\circ$ and a gradual decrease in intensity towards higher angles. No sharp, high-intensity Bragg reflections are resolved across the recorded range, indicating that the composite is predominantly amorphous with only a low degree of crystallinity.

The broad peak at $2\theta \approx 25^\circ$ is characteristic of polypyrrole. Pure PPy exhibits a large diffraction feature in the 22–25° region, confirming its amorphous character, and this amorphous nature is what underlies the flexibility, toughness, and elasticity of the polymer. Also, no reflection attributable to the (001) plane of graphene oxide which would appear near $2\theta = 10^\circ$ is observed. The absence of this peak is the standard XRD of the reduction of GO to rGO, because reduction eliminates the oxygen-containing functional groups and collapses the interlayer spacing. Reduction is accompanied by a decrease in the intensity and an increase in the width of the diffraction peak, reflecting a lower degree of crystallinity and a higher degree of exfoliation in the reduced sheets. The disappearance of the GO (001) reflection therefore indicates that the sheets are exfoliated and distributed within the polymer matrix rather than restacked as a separate crystalline phase.

3.2 Morphological Study

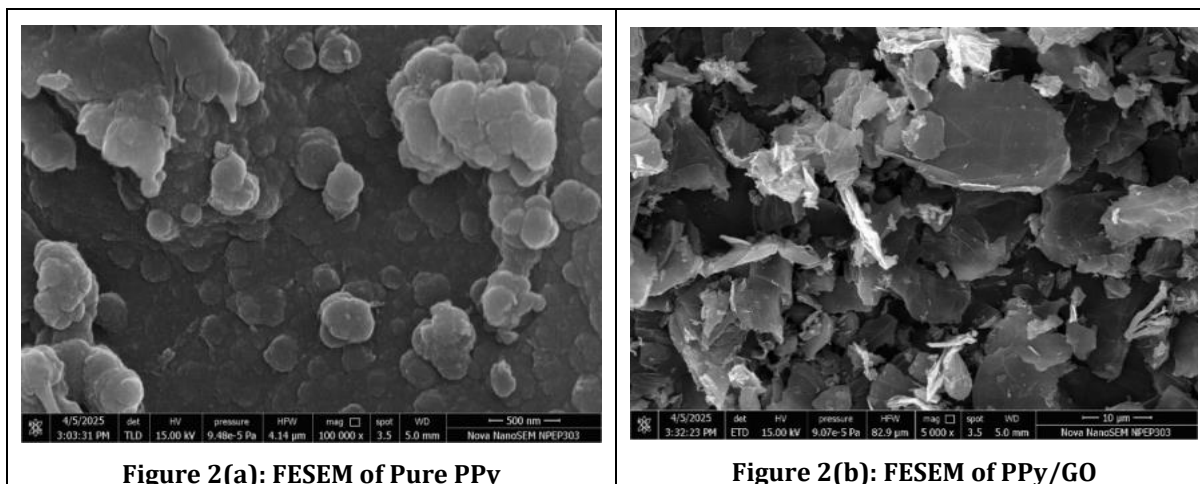


Figure 2(a): FESEM of Pure PPy

Figure 2(b): FESEM of PPy/GO

The surface morphology of pure polypyrrole and PPy/GO composite counter electrodes was studied by field emission scanning electron microscopy, and micrographs obtained are shown in Fig. 2(a, b). Polypyrrole in its pure form (Fig. 2(a)) displays the typical cauliflower morphology of chemically oxidized PPy with globular particles of ~200-600 nm in size, well-defined inter-globule boundaries. Whereas composite with PPy/GO provides the best structure among compositions of this series: a uniformly distributed polymer nanoparticle network supported by exfoliated GO sheets with high surface-to-volume ratio and mesoporosity

3.3 Optical Characterization

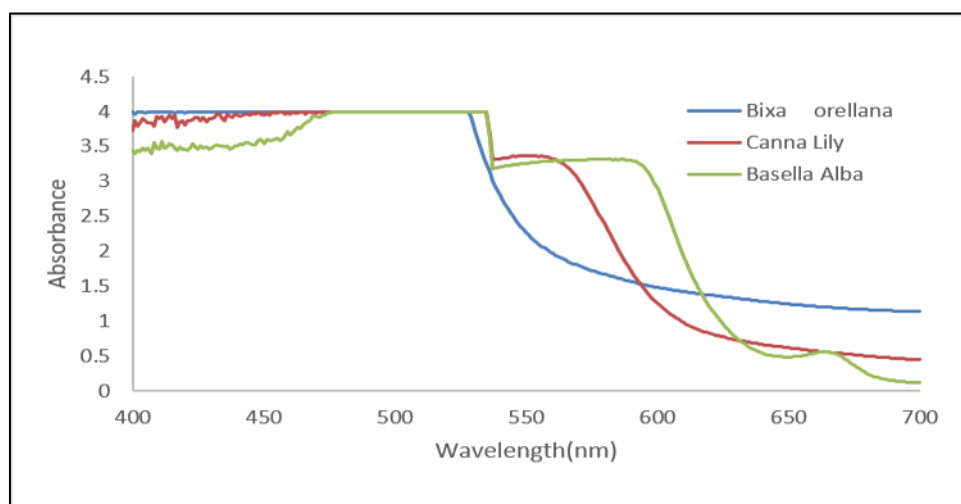


Figure 3: The absorption spectra for the extracts of *Bixa Orellana*, *Canna Lily*, *Basella Alba*

The absorption spectra of colorant were recorded in range of the wavelength 400- 800nm by UV- 1800 SHIMADZU UV spectrophotometer.

Figure 3 shows the absorption spectra of three natural dyes of *Bixa Orellana*, *Canna Lily*, and *Basella Alba*. It is observed that for *Bixa Orellana* shows absorption peaks in range 403 to 527 nm with almost constant absorption. The absorption spectra of *Canna Lily*, shows broad absorption in range 400-535nm with average absorption 3.9342 followed by one more absorption peak at 553 nm with absorption 3.357. The absorption spectra of *Basella Alba* shows a broad peak in the range 470-535nm at average absorption 3.9926, along with this one more peak is appeared at 588nm at absorption 3.314.

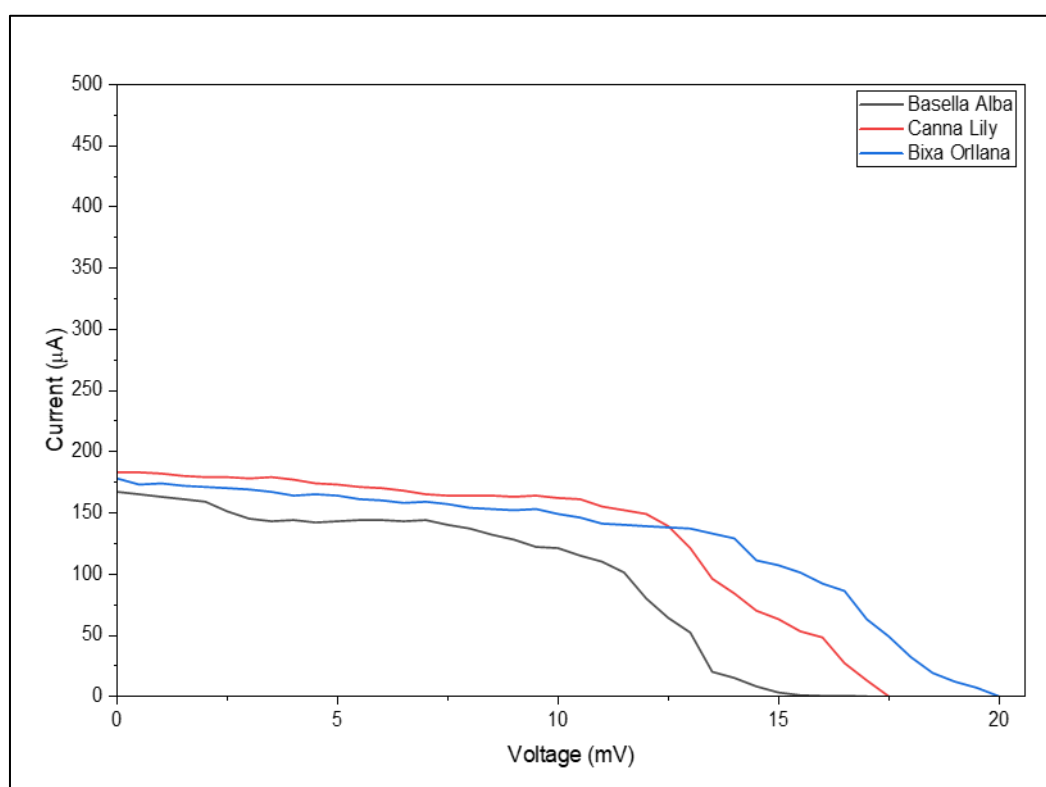
The absorption spectrum of all these colorant, exhibit absorption over 400 to 700nm range of wavelength and hence it can be considered as a good sensitizers to absorb radiation from the visible region of the electromagnetic spectrum. The optical band gap of extracted colorant was calculated using Tauc's equation.

Table 1: Represents the band gap values of these dyes

Sr.no.	Name of Dye	Direct band gap (eV)
1.	<i>Bixa Orellana</i>	2.26
2.	<i>Canna lily</i>	2.09
3.	<i>Basella Alba</i>	2.02

3.4 Photovoltaic performance of fabricated Dye sensitized solar cell

The photovoltaic behavior of the dye-sensitized solar cells assembled with PPy/GO composite counter and Photoanode sensitized by Basella Alba, Canna Lily and Bixa Orellana Sensitizer was obtained by recording the photocurrent–voltage (I–V) characteristics. Figure 4 presents the I–V curves of the devices, and Table2 summarizes the corresponding photovoltaic parameters, namely the short-circuit current (I_{sc}), open-circuit voltage (V_{oc}), maximum current (I_{max}), maximum voltage (V_{max}), fill factor, and power conversion efficiency (η). In DSSC Counter electrode plays Crucial role for facilitating charge transfer from the external circuit to the electrolyte and catalyzes the regeneration of redox couple.

**Figure 4: IV characteristics of PPy/GO CE material and natural dye sensitized photoanode assembled DSSC****Table 2: Photovoltaic parameter of fabricated DSSCs**

Counter electrode Material	I_{sc} (μA)	V_{oc} (mV)	I_{max} (μA)	V_{max} (mV)	FF	η (%)
PPy/GO+ Basella Alba	167	17	106	11.5	0.42	0.164
PPy/GO + Canna Lily	183	18	149	12	0.54	0.246
PPy/GO+Bixa Orellana	173	18	133	13	0.55	0.24

The efficiency of the DSSC fabricated using the PPy/GO composite counter electrode and the Basella Alba extract sensitized photoanode is the smallest among the three, with the value being 0.164% and the largest one of 0.246% being provided by the Canna Lily sensitizer. The efficiency of Bixa Orellana cell is placed between the two with a value of

0.24%. It should be notice that the values of the short circuit currents for the three devices vary in a narrow range of 167, 173, and 183 μA , their difference is no more than 9.6%. Thus, the main parameter affecting the efficiency is not the amount of absorbed photons but the fill factor, which increases from 0.42 for Basella Alba up to 0.54 for Canna Lily and 0.55 for Bixa Orellana cells, i.e., by 31%. Since the fill factor is an indicator of how close to an ideal cell is the tested device, the differences in the fill factors indicate the differences in the recombination losses and in the dye- TiO_2 interface properties.

The Bixa Orellana device records the highest maximum-power voltage (13 mV) and the highest fill factor (0.55) of the series, which may be attributed to good absorption of Bixa Orellana in the visible region and to favourable interaction with the anchorage sites on the TiO_2 film. Bixin, the principal red carotenoid of Bixa Orellana seeds, carries a carboxylic acid group together with a methyl ester group at the ends of its C_{25} chain, and the presence of such carboxylic groups is known to promote viable adsorption onto the semiconducting oxide and to raise cell performance, while pigments lacking the carboxylic functionality show markedly reduced productivity.

4 CONCLUSION

Effect of the PPy/GO counter electrode: With the PPy/GO counter electrode common to all three cells, the fabricated dye-sensitized solar cells delivered short-circuit currents of 167, 183, and 173 μA , open-circuit voltages of 17, 18, and 18 mV, fill factors of 0.42, 0.54, and 0.55, and conversion efficiencies of 0.164%, 0.246%, and 0.24% respectively. The three devices therefore span an efficiency range of 0.164–0.246% on the same counter electrode, confirming that the PPy/GO electrode supports working photoelectrochemical cells across all three sensitizers and that the residual variation between them originates at the dye TiO_2 interface rather than at the counter electrode.

Effect of the natural dye: Keeping counter electrode same, the three sensitizers produced efficiencies of 0.164%, 0.246%, and 0.24%. The short-circuit currents of the three devices were closely grouped 167, 173, and 183 μA , a spread of only about 9.6%. But, the fill factor was the dominant differentiator, rising from 0.42 for Basella Alba to 0.54 for Canna Lily and 0.55 for Bixa Orellana, an increase of roughly 31%. Because the fill factor reflects how closely the device approaches the ideal rectangular power characteristic, its variation points to differences in the quality of the dye TiO_2 interface and in recombination losses during charge collection rather than in the number of photons absorbed. The strong performance of Bixa Orellana is attributable to its good absorption in the visible region and to favourable interaction with the anchorage sites on the TiO_2 film. In Bixin, principal carotenoid, carries carboxylic acid groups, and the presence of such groups is known to promote viable adsorption onto the semiconducting oxide and to raise cell performance, while pigments lacking the carboxylic functionality show markedly reduced productivity.

STATEMENTS ON COMPLIANCE WITH ETHICAL STANDARDS

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Disclosure of conflict of interest

The authors declare no conflicts of interest regarding this manuscript

Declaration

I declare that the manuscript is original, and its content in some other form has not been published elsewhere previously by any of the authors and/or is not under consideration for publication in any other journal at the time of submission.

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