



ENHANCED PERFORMANCE OF DYE-SENSITIZED SOLAR CELLS WITH CO-SENSITIZED NATURAL DYES FROM *Bauhinia variegata* GREEN LEAVES AND *Macaranga peltata* YELLOW LEAVES

*M.U.S. Fernando**, *S. Davisan*, and *V.P.S. Perera*

Department of Physics, The Open University of Sri Lanka, Sri Lanka

This study aims to identify the co-sensitization of different dyes to enhance the overall performance, and efficiency of the Dye-sensitized solar cells (DSSC) by broadening their UV-visible absorption spectrum. The natural dyes extracted from *Bauhinia variegata* (BV) green leaves and *Macaranga peltata* (MP) yellow leaves were combined in a 1:1 ratio, resulting in promising enhancements in the overall performance of the DSSC. The fabrication process of DSSC was done by using the dye-coated TiO₂ film on Fluorine-tin oxide glass as the photoanode and the Platinum (Pt) sputtered glass plate as the counter electrode with iodide/tri-iodide (I⁻/I₃⁻) as the redox electrolyte. Photovoltaic measurements short-circuit current density (J_{SC}), the open-circuit voltage (V_{OC}), the fill factor (ff), and the efficiency (η), were obtained under the illumination intensity of 100 mW/cm² using an I-V analyzer. The displayed performance metrics were for DSSC with BV green dye alone, 0.232 mA/cm², 610.2 mV, 0.711, 0.101%, for DSSC with MP yellow dye alone, 0.710 mA/cm², 481.5 mV, 0.574, 0.196%. In contrast, the DSSC with the 1:1 dye mixture achieved 0.765 mA/cm², 504.5 mV, 0.668, 0.258%. The UV-visible absorption spectra of the dyes extracted with ethanol were measured within the wavelength range of 450 to 800 nm. The resulting spectra exhibit absorption peaks at 540 nm, 610 nm, and 566.7 nm for the 1:1 mixture of BV green and MP yellow dyes. In the extractions, the active pigments are likely to be Chlorophylls and Xanthophyll (Carotenoid). Incident Photon to Current Efficiency (IPCE) measurements conducted over the wavelength range of 450 nm to 750 nm indicated that the DSSC coated with a 1:1 mixture of BV green and MP yellow leaves achieved the highest quantum efficiency within the UV-visible spectrum. The addition of co-pigment evidenced a higher comparative efficiency of 31.63% with MP yellow leaves and 155.45% with BV green leaves. This synergistic effect of co-pigmentation significantly enhances the light-harvesting capabilities and overall cell performance of the solar cells.

Keywords : *Bauhinia variegata*, *Macaranaga peltata*, *Co-sensitization*, *DSSC*

*Corresponding Author: ushanshirantha@gmail.com



ENHANCED PERFORMANCE OF DYE-SENSITIZED SOLAR CELLS WITH CO-SENSITIZED NATURAL DYES FROM *Bauhinia variegata* GREEN LEAVES AND *Macaranga peltata* YELLOW LEAVES

*M.U.S. Fernando**, *S. Davisan*, and *V.P.S. Perera*

Department of Physics, The Open University of Sri Lanka, Sri Lanka

1. INTRODUCTION

The rapid increase in global energy consumption and related ecological problems have paved the path to utilize renewable energy sources such as solar energy, wind energy, tidal energy, etc. effectively that are economically feasible, sustainable, eco-friendly, and safe. Solar energy is a significant renewable energy source to harvest clean energy. The conversion of solar energy into electric energy is done by using photovoltaics, also known as solar cells utilizing the photovoltaic effect.

There are three basic generations of solar cells designated as first, second, and third, and they differ according to their cost, fabrication complexity, and efficiency. Solar cells based on crystalline film technology and thin-film technology are classified as first generation and second generation respectively. The Third-generation solar cells are based on new materials and technologies, such as organic (Wöhrlé & Meissner, 1991), dye-sensitized (O'Regan & Grätzel, 1991), CZTS (Xin et al., 2011), Perovskite (Park, 2014) and quantum dot (Aroutiounian et al., 2001).

A new kind of photoelectrochemical device known as a dye-sensitized solar cell (DSSC) was first developed by Grätzel along with his co-workers in 1991. It converts the visible light and generates electrical power based on the sensitization of wide band gap semiconductors. Subsequently, DSSC has been a captivating field of research and attracted immense attention among scientists because of their low manufacturing cost, eco-friendly production, use of flexible substrate, lightweight, ease of fabrication, and feasibility for efficient light harvesting even at relatively low light intensity (Nowsherwan et al., 2023). The main components of DSSC are a photoanode made from a thin film semiconductor on transparent conducting oxide film-coated glass (FTO or ITO Glass) which helps to capture electrons from the dye, a suitable dye molecule sensitizer to absorb solar energy to generate photons and then convert those photons into energetic electrons, Iodide/tri-iodide redox electrolyte to transport electrons and holes from metal oxide and a counter electrode and to catalyze electron regeneration with platinum counter electrode.

The dye as a sensitizer in dye-sensitized solar cells plays a crucial role in the performance of DSSC. Hence numerous researchers are keen on using natural pigments extracted from plant leaves, fruits, flowers, seeds, roots, woods, algae, and cyanobacteria as sensitizing dyes because of their high purity form and resemblance to the natural photosynthesis process (Armendáriz-Mireles et al., 2023). An increasing number of natural pigments are used as sensitizers in DSSCs including betalain, chlorophyll, carotenoid, anthocyanin, flavonoid, cyanine, and tannin. Most of the natural dyes are sensitive to the visible and UV range of the solar radiation nevertheless very few are found to be sensitive to the IR region which can be utilized in DSSCs. The dye pigment types, appropriate solvents for dye extraction, and dye absorption duration are the key factors that influence the cell performance.

This research study aims to enhance the overall efficiency and to broaden the UV-visible absorption spectrum by co-sensitization of DSSCs from the dye extracted from *Bauhinia variegata* green leaves and *Macaranga peltata* yellow leaves.



2. METHODOLOGY

2.1 NATURAL DYE EXTRACTION

Bauhinia variegata (Koboleela) and *Macaranga peltata* (Kanda) fresh green and yellow leaves (1g) were collected separately, cut into small pieces and boiled under the temperature of 80 °C with 2.5 ml of ethanol in a beaker placed on a hot plate until the de-colorization of the leaves. Filtered extractions were transferred into sample bottles, covered with aluminum foils, and stored in the refrigerator at 4°C until further use.

2.2 FABRICATION OF PHOTO-ELECTRODE

The FTO glass sheets with 2 cm x 1 cm dimensions were cleaned with water and a few drops of washing liquids in an ultrasonic bath for five minutes and then sonicated again with distilled water and a few drops of conc.H₂SO₄ for 5 minutes. After that, the FTO glass plates were boiled in isopropyl alcohol at 80 °C in a beaker. FTO glass plates were then dried using a hair dryer at mild heat and determined the conducting surface with a conductivity meter.

TiO₂ paste was prepared using 0.25 g of 20 nm TiO₂ powder, 0.1ml of 0.1M HNO₃, one drop of Triton-X 100, and a spec of PEG 400. The doctor blading method was used to apply the prepared TiO₂ on the conductive surface of the FTO glass plates, and the cell was sintered in the furnace for 30 minutes at 450°C. Once the TiO₂ film-coated glass plates were cooled, they were dipped separately in each dye extraction.

2.3 FABRICATION OF THE CELL

The electrolyte was prepared using 0.127g of Iodine (I₂), and 0.83 g of potassium iodide (KI) dissolved in 10 ml of acetonitrile and ethylene carbonate in 8:2 ratio in a volumetric flask (Davisan et al., 2023). Then the solution was stirred for 5 hours to ensure all the solid particles were completely dissolved.

The DSSC fabrication process was done by using the dye-coated TiO₂ film as the working anode and the Pt-sputtered glass as the counter electrode placed side by side clamped with the help of crocodile clips. The liquid electrolyte was filled to the capillary space between the two plates.

3. RESULTS AND DISCUSSION

Plants synthesize a diverse array of pigment molecules, far exceeding the variety found in animals. This diversity is primarily due to plants' dependence on light, which they utilize not only to regulate growth but also as their primary energy source. Additionally, plants produce pigments to attract animals for pollination and seed dispersal. Thus, pigments can serve both physiological and biological roles. In plant leaves, there are three main types of pigments, whose retention or synthesis determines the colors of the leaves before they fall.

Pigments are more than just simple chemical formulas showing the number of atoms of different elements in the molecule. For instance, glucose is a common sugar that can be bought as a sweetener and is one of the two components of sucrose, a disaccharide. More complex diagrams can be used to illustrate the structures of the three types of pigments present during leaf senescence: chlorophylls, carotenoids, and anthocyanins (Davisan et al., 2023).

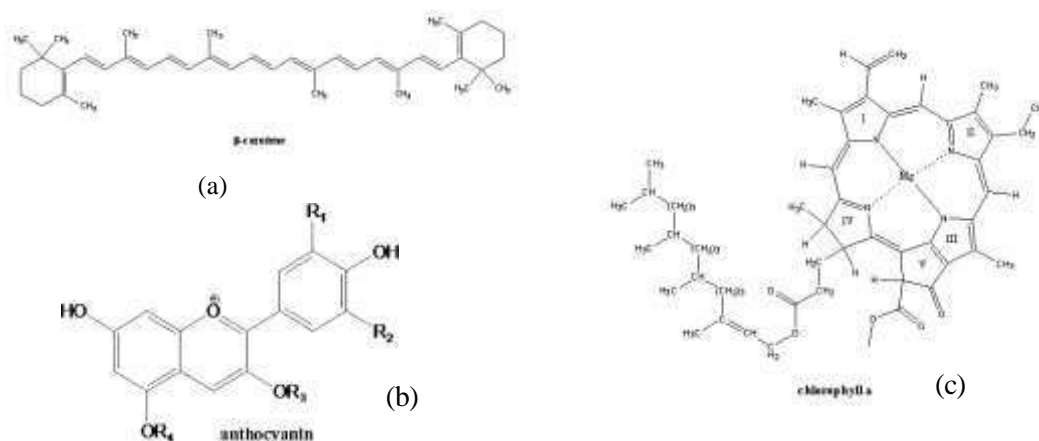


Figure 1: Available pigment structures of leaves (a) Xanthophyll (Carotenoid) (b) Cyanidin (Anthocyanin) (c) Chlorophyll a (Davisan et al., 2023).

Therefore, the green and yellow colour pigments in *Bauhinia variegata* and *Macaranga peltata* leaves are likely Chlorophyll and Xanthophyll facilitated with hydroxyl groups that can chelate to the TiO₂ film.

3.1 Photovoltaic Measurements of the DSSC

The respective photovoltaic measurements related to the DSSC such as open circuit voltage ($V_{(OC)}$), short circuit current ($I_{(SC)}$), short circuit current density ($J_{(JC)}$), fill factor (ff), efficiency (η), series resistance ($R_{(S)}$), and shunt resistance ($R_{(Sh)}$) of the DSSC were evaluated under 100 mWcm⁻² light illumination using the computerized PK-I-V-100 I-V analyzer.

Figure 2 illustrates the J-V characteristic curves for the *Bauhinia variegata* green leaves and *Macaranga peltata* yellow leaves, and their co-sensitized mixture. The respective $V_{(OC)}$, $J_{(JC)}$, ff, and η measurements for the 1:1 dye mixture were 504.5 mV, 0.765 mA/cm², 0.668, and 0.258%.

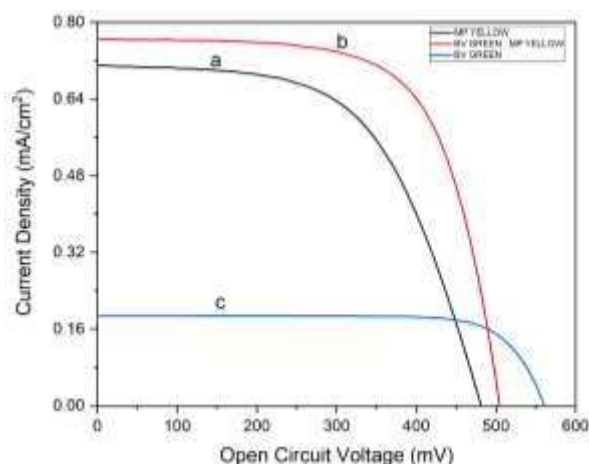


Figure 2: J-V characteristic curves of (a) *Macaranga peltata* yellow leaves (b) *Macaranga peltata* yellow and *Bauhinia variegata* green leaves (1:1 ratio) (c) *Bauhinia variegata* green leaves

Table 1 summarizes the photovoltaic characteristics of *Bauhinia variegata* green leaves, *Macaranga peltata* yellow leaves, and co-sensitized DSSCs in ethanol extracts.



Table 1: Photovoltaic Measurements of the DSSCs

	Open circuit voltage ($V_{(OC)}$) mV	Short circuit current density ($J_{(JC)}$) mA/cm ²	Fill factor (ff)	Efficiency (η)	Series resistance ($R_{(S)}$) Ω	Shunt resistance ($R_{(Sh)}$) Ω
Green Leaf	610.2	0.232	0.711	0.101	382.649	14614.620
Yellow Leaf	481.5	0.710	0.574	0.196	395.267	43839.810
Co-sensitized (1:1)	504.5	0.765	0.668	0.258	238.989	14614.940

3.2 UV- Visible absorption spectrum of the dye

The absorption spectrum of the pigments extracted from MP yellow leaves, BV green leaves, and their 1:1 mixture is represented in Figure 3. These measurements were conducted in the visible range from 450 nm to 720 nm. The corresponding absorbance peaks were found around 480 nm, 620 nm, and 660 nm for MP yellow, while BV green exhibited peaks at 490 nm and 660 nm. The mixed dye showed significant peaks at 480 nm, 530 nm, 610 nm, and 660 nm respectively.

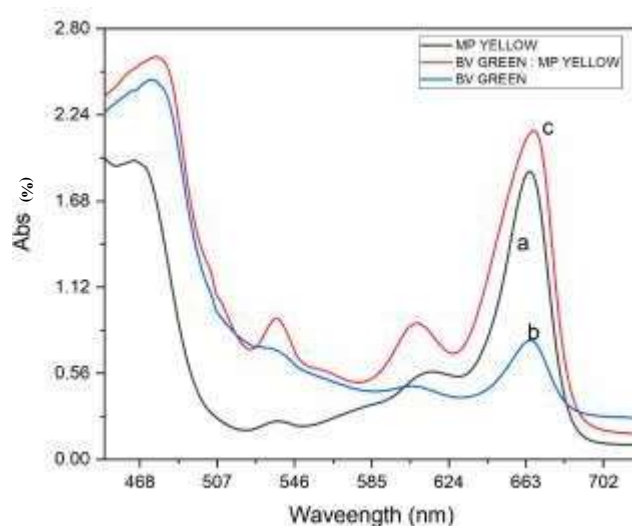


Figure 3: Absorbance spectrum in UV-visible range ethanol extraction in pigments (a) BV green (b) MP yellow (c) BV Green and MP Yellow (1:1)

3.3 IPCE Characteristic of the DSSC

The efficiency of the solar cell is quantified by the ratio of the current generated by the collected carriers at the electrical contacts to the incoming photon flux at a specific wavelength. This measure is known as the Incident-Photon-to-Current Efficiency (IPCE) (Plana et al., 2016). IPCE characteristics were measured by using computerized VK-IPCE-10.

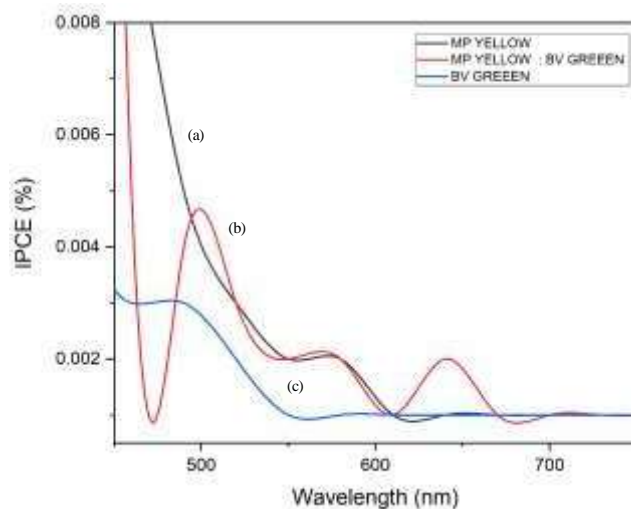


Figure 4: IPCE characteristic curves of DSSCs (a) MP yellow (b) MP yellow and BV green (1:1) (c) BV green dye extracts

Based on the exhibited graph in Figure 4, the functions of this particular DSSC are in the range of 450 nm to 750 nm wavelengths. Notably, there was no quantum yield for BV green cells at 650 nm while the cell with MP yellow shows a marginal response at 650 nm. However, when both the dyes are mixed, IPCE improves significantly than any of the dyes alone. The maximum IPCE of DSSCs was generated for the 1:1 mixture of the BV green and MP yellow dyes in wavelengths around 510 nm, 580 nm, and 630 nm. When referred to photosynthesis, chlorophylls are responsible for the formation of glucose using solar radiation. However, carotenoids and anthocyanin, known as auxiliary or accessory pigments enhance this process which is evident in DSSCs as well when both the dyes are present.

4. CONCLUSION

The UV-visible absorption spectrum of the ethanol-extracted dye from *Bauhinia variegata* green leaves and *Macaranga peltata* yellow leaves 1:1 mixture exhibited significant improvement of the photovoltaic characteristics. The measured short-circuit current density (J_{SC}), open circuit voltage (V_{OC}), fill factor (ff), and the efficiency (η) of the solar cell are 0.765 mA, 504.5 mV, 0.668 and 0.258% respectively. The overall efficiency of the DSSC using the 1:1 dye mixture increased by 155.45% when compared with *Bauhinia variegata* green leaves and 31.63% with *Macaranga peltata* yellow leaves. The available yellow colour pigment in *Macaranga peltata* is likely to be carotenoids while the green colour pigment in *Bauhinia variegata* is chlorophyll in corresponding extractions. Further studies are needed to identify the active pigments in the dye extractions and to explore methods for enhancing cell performance in a sustainable manner. An Interdisciplinary approach, including the implementation of computerized methods such as machine learning, could significantly benefit the next stage of DSSC development by optimizing pigment ratios based on the availability of various parts of tree anatomy. Furthermore, these techniques can be used in determining the most effective extraction methods, to enhance the overall efficiency and performance of DSSCs.



5. REFERENCES

- Leaf Pigments / Harvard Forest.* (n.d.). <https://harvardforest.fas.harvard.edu/leaves/pigment>
- Plana, D., Bradley, K. A., Tiwari, D., & Fermín, D. J. (2016). Over 75% incident-photon-to-current efficiency without solid electrodes. *Physical Chemistry Chemical Physics/PCCP. Physical Chemistry Chemical Physics*, 18(18), 12428–12433. <https://doi.org/10.1039/c6cp02231f>
- Aroutiounian, V., Petrosyan, S., Khachatryan, A., & Touryan, K. (2001). Quantum dot solar cells. *Journal of Applied Physics*, 89(4), 2268–2271. <https://doi.org/10.1063/1.1339210>
- Cheng, C., Zhang, H., Li, F., Yu, S., & Chen, Y. (2021). High performance ammonia gas detection based on TiO₂/WO₃-H₂O heterojunction sensor. *Materials Chemistry and Physics*, 273, 125098. <https://doi.org/10.1016/j.matchemphys.2021.125098>
- Xin, X., He, M., Han, W., Jung, J., & Lin, Z. (2011). Low-Cost copper zinc tin sulfide counter electrodes for High-Efficiency Dye-Sensitized solar cells. *Angewandte Chemie*, 50(49), 11739–11742. <https://doi.org/10.1002/anie.201104786>
- Wöhrlé, D., & Meissner, D. (1991). Organic solar cells. *Advanced Materials*, 3(3), 129–138. <https://doi.org/10.1002/adma.19910030303>
- Park, N. (2014). Perovskite solar cells: an emerging photovoltaic technology. *Materials Today*, 18(2), 65–72. <https://doi.org/10.1016/j.mattod.2014.07.007>
- O'Regan, B., & Grätzel, M. (1991). A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO₂ films. *Nature*, 353(6346), 737–740. <https://doi.org/10.1038/353737a0>
- Singh, A., & Mukherjee, T. (2022). Application of carotenoids in sustainable energy and green electronics. *Materials Advances*, 3(3), 1341–1358. <https://doi.org/10.1039/d1ma01070k>
- Wickramasinghe, G.C., Jayathilaka, D.L.N. & Perera, V.P.S. (2017). *Construction of Dye Sensitized Solar Cell Using Natural Dye Extraction from Petals of Erabadu Flower.* OURS.
- Hosseinpanahi, K., Golzarian, M. R., Abbaspour-Fard, M. H., & Feizy, J. (2020). Improving The Efficiency of DSSC with A Novel Multi-dye layers Approach. *Optik*, 208, 164068. <https://doi.org/10.1016/j.ijleo.2019.164068>
- Davisan, S., Aponso, G. M. L. P., & Perera, S. (2023). Fabrication of Dye-sensitized solar cell using natural dye extracted from *Elaeocarpus serratus* Red Leaf. *ResearchGate*. <https://doi.org/10.13140/RG.2.2.26829.51689>
- Nowsherwan, G. A., Iqbal, M. A., Rehman, S. U., Zaib, A., Sadiq, M. I., Dogar, M. A., Azhar, M., Maidin, S. S., Hussain, S. S., Morsy, K., & Choi, J. R. (2023). Numerical optimization and performance evaluation of ZnPC:PC70BM based dye-sensitized solar cell. *Scientific Reports*, 13(1). <https://doi.org/10.1038/s41598-023-37486-2>
- Armendáriz-Mireles, E. N., Calles-Arriaga, C. A., Pech-Rodríguez, W., Castillo-Robles, A., & Rocha-Rangel, E. (2023). Alternative sources of natural photosensitizers: Role of algae in Dye-Sensitized Solar Cell. *Colorants*, 2(1), 137–150. <https://doi.org/10.3390/colorants2010010>