

Green Fabrication of Dye-Sensitized Solar Cells Using TiO₂ and Four Natural Leaf Dye Extracts

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ABSTRACT

Different natural dye extracts were prepared and used as sensitizers in the fabrication of dye-sensitized solar cells (DSSCs). A dense TiO₂ blocking layer was deposited by spray pyrolysis and characterized by profilometry, while a mesoporous TiO₂ layer was subsequently deposited by screen printing. DSSCs sensitized with extracts from *Dacryodes edulis*, *Mimosa pudica*, *Delonix regia*, and *Xanthosoma sagittifolium* were fabricated and characterized under an illumination intensity of 100 mW/cm². The short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF), and conversion efficiency (η) were determined. *Delonix regia* exhibited the highest conversion efficiency of 0.0555%, with J_{sc} = 0.196 mA/cm², V_{oc} = 0.483 V, and FF = 0.586. *Dacryodes edulis*, *Mimosa pudica*, and *Xanthosoma sagittifolium* recorded efficiencies of 0.0106%, 0.0044%, and 0.0113%, respectively. The results demonstrate measurable photovoltaic responses from all four natural dye extracts, with *Delonix regia* producing the highest response under the experimental conditions. However, the relatively low conversion efficiencies indicate the need for further optimization of dye extraction, sensitization, TiO₂ film characteristics, and cell fabrication conditions.

Keywords:

Dye-sensitized solar cells (DSSCs),
Titanium dioxide (TiO₂) semiconductor,
Natural Dyes Sensitizers,
Photovoltaic performance.

INTRODUCTION

Dye-sensitized solar cells (DSSCs), first demonstrated by O'Regan and Grätzel, (1991), have gained significant attention as a promising alternative photovoltaic technology. Unlike conventional silicon-based solar cells, DSSCs separate the processes of light absorption and charge transport by using dye molecules immobilized on nanocrystalline metal oxide semiconductors such as TiO₂. This biomimetic design replicates natural photosynthesis and enables the use of abundant, wide-bandgap oxide materials instead of expensive semiconductor wafers.

Metal-organic sensitizers, particularly ruthenium and osmium polypyridyl complexes, have been shown to exhibit excellent light-harvesting and electron injection properties (Rahman *et al.*, 2023; Malhotra *et al.*, 2024; Velpuri, 2024 and Nurul *et al.*, 2025). However, their synthesis typically involves multistep ligand preparation, purification using chromatography, and high production costs. As global research trends increasingly move toward sustainable and environmentally benign materials, natural pigments have emerged as viable sensitizers for DSSCs.

Plant-based dyes such as anthocyanins, chlorophylls, tannins, and carotenoids are abundant, non-toxic, and biodegradable, making them attractive for green photovoltaic applications (Suyitno, 2024; Suyitno *et al.*, 2022; Erande *et al.*, 2020, Aung *et al.*, 2016 and Liu *et al.*, 2021). Recent studies also report improved dye extraction methods, enhanced photo stability, and notable power conversion efficiencies when natural dyes are properly purified or chemically modified (Khan, 2024; Kraidy *et al.*, 2024 and Lai *et al.*, 2021).

Despite these advances, a key research gap remains: only limited comparative studies have evaluated the photovoltaic performance of multiple indigenous West African plant dyes under identical fabrication and testing conditions. Most reported works focus on single-plant sensitizers or species investigated under different experimental conditions, making direct comparison of their photovoltaic performance difficult. Consequently, there is limited information on the comparative photovoltaic sensitizing performance of locally available Nigerian plant extracts. Evaluating different natural dyes

using the same TiO_2 structure, comparable extraction conditions, cell-fabrication procedure, and illumination conditions provides a more consistent basis for assessing their relative photovoltaic responses. The present study therefore focuses on *Mimosa pudica*, *Xanthosoma sagittifolium*, *Dacryodes edulis*, and *Delonix regia*, which were selected based on their availability in Nigeria, their natural pigment constituents, and their potential as plant-derived sensitizers. Such a controlled comparison provides baseline information on the photovoltaic responses of these selected Nigerian plant extracts and may help identify areas requiring further optimization for low-cost DSSC applications.

To address this gap, the present study fabricates DSSCs using dyes extracted from *Mimosa pudica*, *Xanthosoma sagittifolium*, *Dacryodes edulis*, and *Delonix regia*-plants that are readily available in Nigeria but are underreported in DSSC research. Their sensitization performance was evaluated by recording the I-V characteristics under simulated sunlight (100 mW/cm²) using a Model 4200-SCS Semiconductor Characterization System at the Sheda Science and Technology Complex (SHESTCO), Abuja. This comparative approach aims to evaluate the photovoltaic responses of the selected natural dyes under a common experimental protocol and to provide baseline data for their potential application and further optimization as natural sensitizers in DSSCs.

MATERIALS AND METHODS

Selection of Plant Samples

Four Nigerian plant species, *Mimosa pudica*, *Delonix regia*, *Dacryodes edulis*, and *Xanthosoma sagittifolium*, were selected as sources of natural dye sensitizers for the fabrication of dye-sensitized solar cells (DSSCs). The plants were selected based on their reported pigment content, availability and accessibility in Nigeria, and previous reports on the potential application of plant-derived pigments as low-cost sensitizers for DSSCs. Fresh, healthy leaves without visible signs of damage or deterioration were collected for dye extraction.

Preparation of Mesoporous TiO_2 Photoanode

FTO-coated glass substrates were cleaned with sodium lauryl sulfate, rinsed with distilled water, and masked to define the deposition area. A dense TiO_2 blocking layer was deposited by spray pyrolysis using a precursor containing 0.015 M Titanium (IV) isopropoxide, 0.03 M acetylacetone, and 1.7 mL water. The precursor was aged for 24 h and sprayed using a U-Spray machine (USP 1500, Regal Scientific) at 400 °C, with an atomizing voltage of 7000 V, substrate-to-nozzle distance of 21 mm, and a deposition time of 3 min. Profilometric analysis showed TiO_2 blocking-layer thicknesses of 40, 60, and 100 nm for deposition volumes of 0.3, 1.0, and 2.0 mL, respectively, at 400 °C. Preliminary deposition at 480 °C produced

thicknesses of 200 and 400 nm for 0.5 and 1.0 mL deposition volumes, respectively.

A mesoporous TiO_2 layer was subsequently deposited by screen printing using commercial TiO_2 paste (Titanium Nanoxide D/SP, Ti-20; Solaronix) and a 70-mesh screen. The paste was uniformly applied to obtain an active area of 4 cm² and a film thickness of 10 μm . The printed electrodes were heated at 450 °C for 30 min and subsequently at 500 °C for 5 min, followed by cooling to room temperature before sensitization.

Extraction of Natural Dye Sensitizers

Freshly collected leaves were washed thoroughly with distilled water to remove adhering dust and surface contaminants, cut into smaller pieces, and homogenized. A measured mass of 200 g of each plant sample was separately immersed in 80 mL of distilled water and allowed to stand for 30 min to facilitate extraction of the water-soluble pigments and other dye constituents. The resulting mixtures were filtered to remove residual plant material, and the filtrates were collected as the respective aqueous natural dye extracts. The extraction procedure was adapted from the plant-pigment extraction approach reported by Boyer (1990), with modifications to suit the preparation of aqueous natural dye sensitizers for DSSCs. In the present study, distilled water was used instead of acetone as the extraction solvent, the plant material was homogenized prior to extraction, and 200 g of fresh leaves was extracted with 80 mL of distilled water for 30 min. The resulting aqueous filtrates were used directly as natural dye sensitizers for TiO_2 -based DSSCs. The obtained filtrates were transferred into airtight amber-coloured bottles to minimize exposure to light and stored at room temperature until sensitization.

Preparation of Counter Electrode

Platinum-coated FTO counter electrode were prepared using Platisol platinum catalyst paste (Solaronix). The FTO substrates were initially cleaned with a sodium lauryl sulphate solution and rinsed thoroughly with distilled water. A mask was applied to one end of the FTO to define the deposition area and preheated at 200 °C for 15 minutes. Platinum paste was then deposited using screen printing and subsequently annealed at 400 °C for 30 minutes to form a catalytic Pt layer suitable for the reduction of triiodide species in the electrolyte. The platinum counter electrode was prepared with a coated area sufficient to cover the active region of the TiO_2 photoanode.

Sensitization of TiO_2 Photoanodes

The cooled TiO_2 electrodes were immersed in the respective dye solutions and kept overnight to ensure adequate adsorption of pigments onto the mesoporous structure. After sensitization, the films were gently rinsed with distilled water to remove unbound dye molecules and

dried on a hot plate at 60 °C for 20 minutes. Prepared electrodes were stored in sealed, light-protected containers until assembly.

Assembly of Dye-Sensitized Solar Cells

The DSSCs were assembled in a sandwich configuration. The dye-loaded TiO₂ photoanode and Pt counter electrode were placed face-to-face with their active areas slightly offset. The edges were sealed with Amosil sealant to prevent electrolyte leakage. A 1 mm hole had been pre-drilled into the counter electrode to allow electrolyte introduction. The redox electrolyte was injected through the hole using vacuum backfilling, after which the opening was resealed with Amosil. The assembled cell was secured using binder clips to maintain tight contact between the electrodes and minimize leakage during measurement.

Characterization of DSSCs

The fabricated dye-sensitized solar cells (DSSCs), using natural dye extracts as sensitizers, were characterized to

determine their photovoltaic performance. Current-voltage (I-V) measurements were conducted under simulated AM 1.5 white light irradiation (100 mWcm⁻²) using a solar simulator interfaced with a 4200-SCS Semiconductor Characterization System at the Sheda Science and Technology Complex (SHESTCO), Abuja. The key performance parameters evaluated include short-circuit current (*I*_{sc}), open-circuit voltage (*V*_{oc}), maximum power point (*P*_{max}), fill factor (FF), and overall power conversion efficiency (*η*). Output power (*P*) at any point on the I-V curve (shown in Fig. 1) was determined. Relevant relations applied in the calculations (Murakami *et al.*, 2003) include:

$$FF = \frac{J_{mp} \times V_{mp}}{J_{sc} \times V_{oc}} \quad (1)$$

$$\eta (\%) = \frac{J_{sc} \times V_{oc} \times FF}{P_{in}} \quad (2)$$

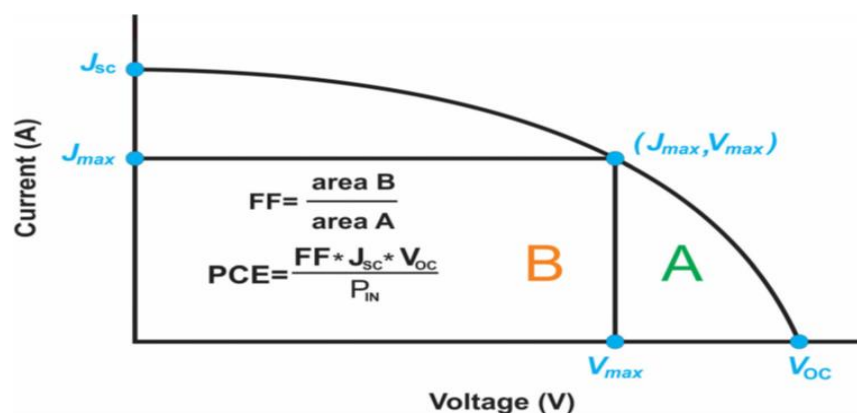


Figure 1: I-V curve to evaluate the cells performance (Halme, 2002).

Interpretation of the I-V Curve for DSSC Performance Evaluation

The I-V curve shown is a graphical representation of the electrical performance of a dye-sensitized solar cell (DSSC) under illumination. From this curve, several important photovoltaic parameters are extracted:

Short-Circuit Current density (*J*_{sc})

This is the current per area where the voltage is zero. It indicates the maximum photocurrent the cell can generate and is directly related to the light absorption and charge-transport efficiency of the dye and semiconductor.

Open-Circuit Voltage (*V*_{oc})

This is the voltage where the current becomes zero. It reflects the energy difference between the semiconductor conduction band and the redox potential of the electrolyte, indicating the cell's ability to drive electrons through an external circuit.

Maximum Power Point (*V*_{mp}, *I*_{mp})

The point on the curve where the product of voltage and current is maximized (*P*_{max}). It is represented by the rectangular area labeled B, this defines the actual usable output power of the DSSC.

Fill Factor (FF)

Is calculated as the ratio of Area B to Area A (*I*_{sc} × *V*_{oc} rectangle). It describes how "square" the I-V curve is. A higher FF indicates lower internal resistance and better charge collection efficiency.

Power Conversion Efficiency (PCE or *η*)

Is determined using *I*_{sc}, *V*_{oc}, FF, and the incident light power (*P*_{in}) using Equations (1) and (2).

RESULTS AND DISCUSSION

Results on Solar Simulation

The current-voltage characteristics curves of all cells were measured and recorded under illumination with white light of intensity 100 mW/cm^2 from a high-pressure mercury arc lamp. The I-V characteristic of the DSSCs prepared using *Dacryodes edulis*, *Mimosa pudica*, *Delonix regia*, and *Xanthosoma sagittifolium* extracts as the natural dye sensitizers are shown in Figures 2 – 5, respectively. The DSSC output power has been calculated

as $P = I \times V$ using the I-V data corresponding to each cell and plotted as a function of V , as shown in the Figures. The maximum power (P_m) point for each cell is then determined from the Figures, and the photocurrent (J_m) and photovoltage (V_m) corresponding to the maximum power point were then obtained. The values of the fill factor and the cell conversion efficiencies were calculated using equations (1 and 2), respectively. A summarized finding is shown in Table 1.

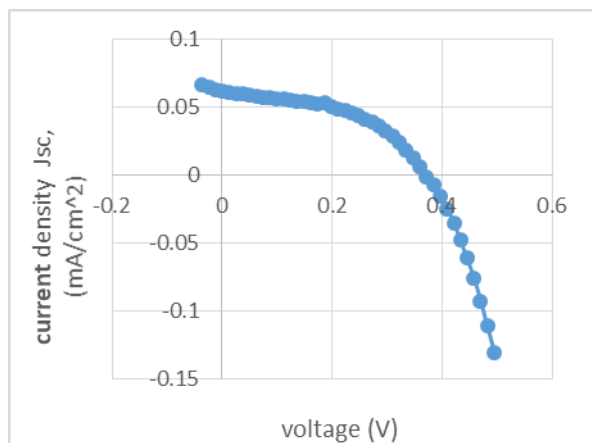


Figure 2: A plot of current density vs voltage of DSSC prepared using DE as sensitizer

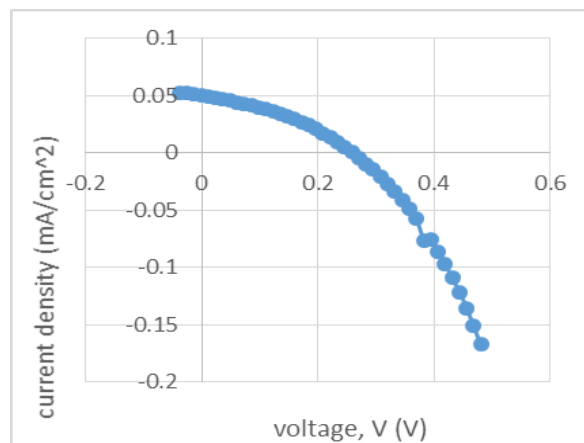


Figure 3: A plot of current density vs voltage of DSSC prepared using MP as sensitizer

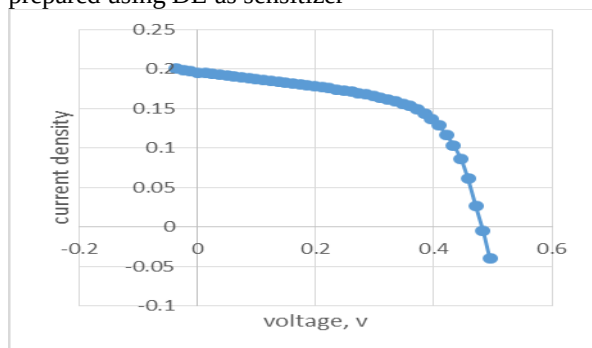


Figure 4: A plot of current density vs voltage of DSSC prepared using DR as sensitizer

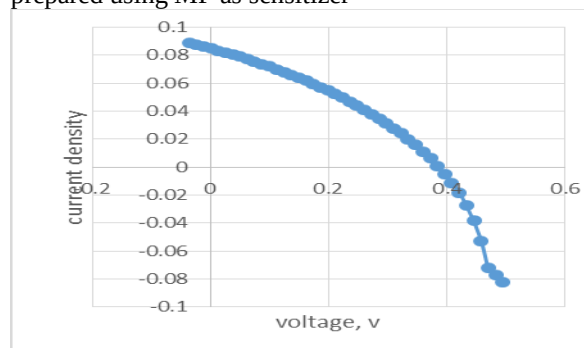


Figure 5: A plot of current density vs voltage of DSSC prepared using XS as sensitizer

Table 1: Shows Photocurrents (J_{sc} , J_m), Photovoltages (V_{oc} , V_m), Fill Factor, and Overall Energy Conversion Efficiency for Four DSSCs Sensitized by Dyes Extracted from Leaves

S/N	Dye	J_{sc} (mA/cm ²)	V_{oc} (V)	J_m (mA/cm ²)	V_m (V)	FF	η (%)
1	<i>Dacryodes edulis</i>	0.062	0.372	0.048	0.211	0.460	0.0106
2	<i>Mimosa pudica</i>	0.050	0.258	0.024	0.185	0.343	0.0044
3	<i>Delonix regia</i>	0.196	0.483	0.149	0.372	0.586	0.0555
4	<i>Xanthosoma sagittifolium</i>	0.084	0.385	0.057	0.199	0.348	0.0113

From Table 1, J_{sc} values ranging from $\cong 0.05 \text{ mA/cm}^2$ (minimum) up to $\cong 0.196 \text{ mA/cm}^2$ (maximum) across the different dye-sensitized cells, V_{oc} varying between a 0.258 V (for the cell sensitized with *Mimosa pudica*) to a

0.483 V (for the cell sensitized with *Delonix regia*). The fill factor (FF) of the cells ranged between a 0.343 and 0.586. The highest conversion efficiency was obtained with the *Delonix regia* dye-sensitized cell, yielding η of

0.0555%. Figure 6 shows the performance summary of DSSCs using natural dye sensitizers.

These results confirm that natural-dye-sensitized DSSCs behave according to expected photovoltaic principles: the

I-V curve shape defines J_{sc} and V_{oc} , while the "squareness" of the curve (area under the maximum-power rectangle vs theoretical maximum rectangle) defines FF and hence the real usable power output.

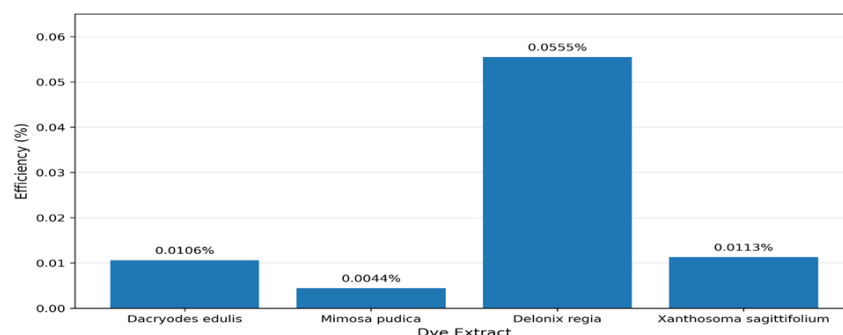


Figure 6: A plot of the performance summary of DSSCs using natural dye sensitizers

Results on Profilometric Analysis

Figures 7 and 8 showed the line profiles of TiO_2 /FTO films prepared by spray pyrolysis for 3 min at a substrate temperature of 480 °C. The profilometric measurements gave TiO_2 layer thicknesses of approximately 200 nm and 400 nm for deposition volumes of 0.5 and 1.0 mL, respectively. The results indicate that increasing the deposition volume from 0.5 to 1.0 mL resulted in an

increase in the TiO_2 layer thickness. Both measured thicknesses exceeded the approximately 60 nm minimum thickness reported for effective TiO_2 blocking layers (Ito *et al.*, 2006). Therefore, the results demonstrate that the spray-pyrolysis conditions were capable of producing blocking layers within the thickness range considered suitable for reducing undesirable electron recombination.

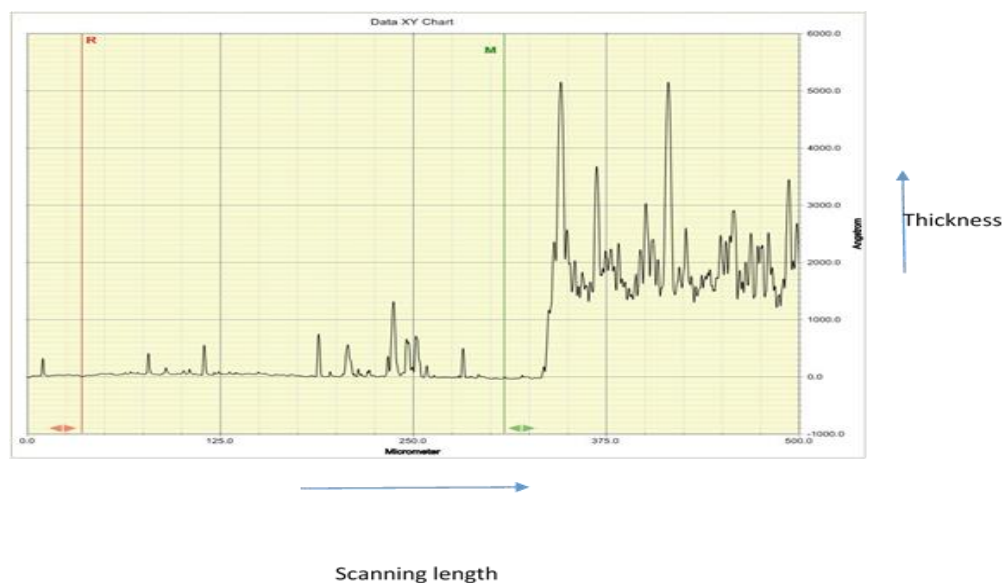


Figure 7: A line Profile for TiO_2 sprayed with 0.5ml for 3 minutes at substrate temperature of 480°C

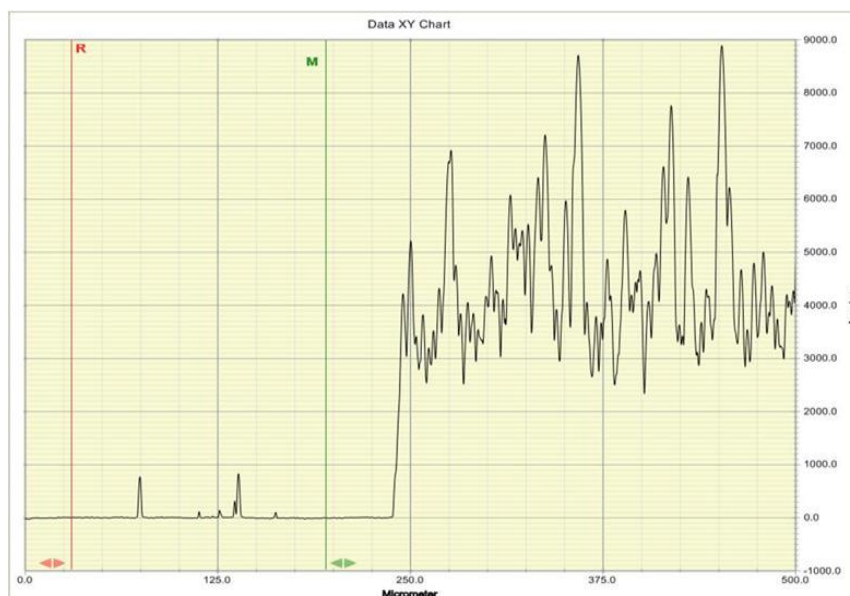


Figure 8: A line Profile for TiO_2 sprayed with 1.0ml for 3minutes at substrate temperature of 480°C

Figure 9 – 11 showed the line profile of TiO_2 / FTO glass prepared by 3 minutes spray pyrolysis at substrate temperature of 400°C . Profilometric results revealed the thicknesses of TiO_2 layers were 40nm, 60nm and 100nm for deposition volume of 0.3ml, 1.0ml and 2ml respectively. The thicknesses obtained were between the

ranges of optimum blocking layer thickness and agrees well with Ito *et al.*, (2006). The TiO_2 / FTO glass prepared by spray pyrolysis with 1.0ml was chosen as the blocking layer to be use in the fabrication of DSSCs since the thickness is approximately greater than 60nm to achieve an increased cell efficiency.



Figure 9: A line Profile for TiO_2 sprayed with 0.3ml for 3minutes at substrate temperature of 400°C

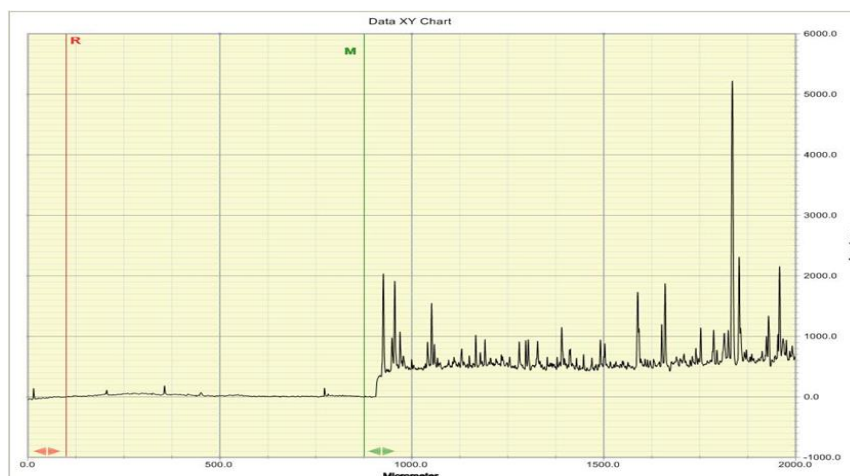


Figure 10: A line Profile for TiO_2 sprayed with 1.0ml for 3minutes at substrate temperature of 400°C

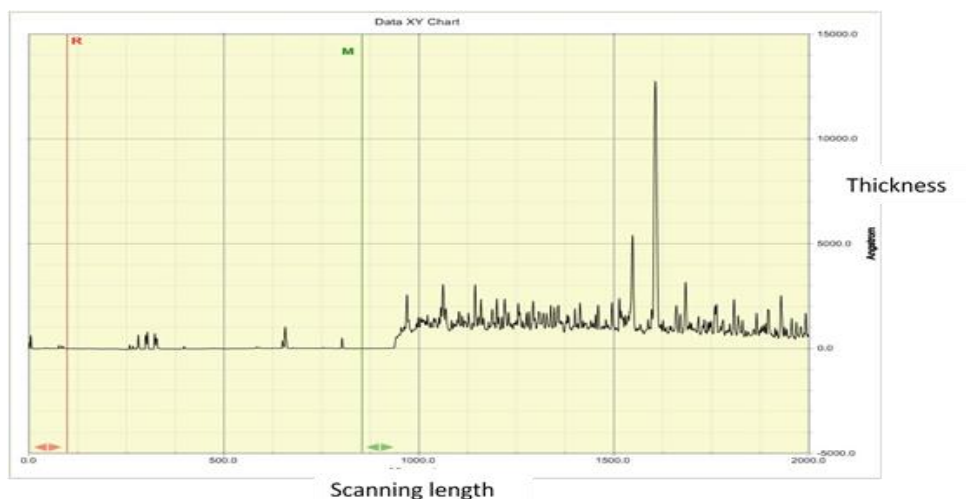


Figure 11: A line Profile for TiO_2 sprayed with 2.0ml for 3minutes at substrate temperature of 400°C

Significance of the Findings

The study demonstrates measurable photovoltaic responses from all four natural dye extracts under the same experimental conditions. *Delonix regia* produced the highest conversion efficiency (0.0555%), followed by *Xanthosoma sagittifolium* (0.0113%), *Dacryodes edulis* (0.0106%), and *Mimosa pudica* (0.0044%). Their corresponding J_{sc} values were 0.196, 0.084, 0.062, and 0.050 mA/cm^2 , respectively, indicating differences in photovoltaic response among the extracts.

The efficiencies obtained are relatively low compared with reported natural-dye DSSCs, for which efficiencies around 0.5% have been reported as an approximate average. However, direct comparison is influenced by differences in dye composition, extraction procedures, TiO_2 structure, electrolyte, and cell fabrication conditions. The observed differences may be associated with variations in pigment composition and dye- TiO_2

interaction. However, dye loading, interfacial recombination, and photostability were not directly measured in this study and should therefore be regarded as possible factors for the low efficiencies. Nevertheless, the results provide a useful comparative baseline for further optimization and replicate testing of these locally available plant extracts.

Possible Reasons for Low Performance & Critical Analysis

Based on the comparatively low J_{sc} , low FF, and overall low η in these cells, several factors may be contributing: 1. Low dye absorption and/or poor anchoring on TiO_2 : Natural dyes often consist of mixtures of pigments, some of which may not adsorb efficiently, or do not have the ideal energy levels (HOMO/LUMO) for efficient electron injection and regeneration. As discussed in the literature, pigment types (anthocyanins, betalains, chlorophylls,

flavonoids) dramatically influence efficiency. Also, impurities and non-pigmented organics might prevent optimal surface coverage, reducing light harvesting and charge injection.

2. Sub-optimal TiO₂ film morphology / dye loading: Poor film thickness, sub-optimal porosity, or insufficient dye loading can reduce the effective interface area, lowering J_{sc} . Moreso, in many recent studies, performance improvements have required careful engineering of the TiO₂ layer (nanostructuring, optimal annealing, layering) along with precise dye adsorption protocols.

3. Recombination losses and internal resistances: A low fill factor (FF between ~0.34 and ~0.586) suggests significant internal losses - possibly due to recombination of electrons at the TiO₂/dye/electrolyte interface, poor charge transport, or high series resistance. Without optimized counter-electrode, electrolyte, or dye-electrolyte interface, the cell cannot efficiently drive electrons through the external circuit.

4. Dye instability / degradation: Natural dyes are often less photostable than synthetic metal-complex dyes. Over time (or even during light exposure), photobleaching or dye desorption may reduce performance. Many recent studies emphasize stability as a central challenge.

CONCLUSION

DSSCs sensitized with natural dyes from *Mimosa pudica*, *Delonix regia*, and *Xanthosoma sagittifolium* produced J_{sc} between ~0.05 and ~0.196 mA/cm², V_{oc} between ~0.258 and ~0.483 V, and FF between ~0.343 and ~0.586. The highest power conversion efficiency (η) achieved was approximately 0.0555%, in the cell sensitized with *Delonix regia*. Compared to recent literature (Khan, 2024; Kraidy *et al.*, 2024 and Lai *et al.*, 2021), this efficiency is on the low end; many natural-dye DSSCs in recent studies report efficiencies ranging from ~0.27% to ~0.77% (and in some engineered cases even higher).

The low performance is likely due to a combination of sub-optimal dye anchoring or purity, imperfect TiO₂ morphology/dye loading, internal recombination losses, and possibly dye instability. Conclusively, the extracts demonstrated measurable sensitization, with *Delonix regia* giving the highest performance under the stated fabrication conditions.

To improve the performance of the DSSCs, future work should consider: Purifying the natural dye extracts - e.g., via column chromatography or repeated washing to remove impurities that do not contribute to light absorption but hinder dye anchoring. Several studies report improved efficiency after purification. Secondly, optimizing dye adsorption conditions - varying dye concentration, adsorption time, solvent, pH, and temperature, to maximize dye loading and binding to TiO₂. Thirdly, engineering the TiO₂ photoanode - using nanoparticulate TiO₂, multilayer deposition, optimized annealing, or nanostructured electrodes (e.g., nanotubes,

nano-rods, mixed meso/ macroporous films) to increase surface area and improve electron transport. Moreso, Exploring co-sensitization or dye blends - combining two or more natural dyes (e.g., anthocyanin + betalain, anthocyanin + chlorophyll) may broaden the absorption spectrum and improve light harvesting, as suggested by recent studies UNDIP E-Journal. Finally, improving the cell architecture - optimizing the counter electrode material, electrolyte composition, sealing, and minimizing recombination pathways. Some recent DSSC designs even employ green counter electrodes (e.g., graphite, carbon) and eco-friendly electrolytes.

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