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ELECTRICAL OPTIMIZATION OF NATURAL DYE-SENSITIZED SOLAR CELLS THROUGH CO-SENSITIZATION AND NANOSTRUCTURAL ENGINEERING

*[1]Uduma, Ogbonnaya U., [2]Uthman, Muhammad, [2]Aliyu, Sani O. and [2] Ogunjuyigbe, Jacob K.

[1]Physics Advanced Research Centre Sheda Science and Technology Complex (SHESTCO)
[2]Department of Electrical and Electronics Engineering, University of Abuja

*Corresponding authors' email: ogbochidera1106@gmail.com Phone: +2348032221080

ABSTRACT

Natural dye-sensitised solar cells offer a cheap and eco-friendly alternative to conventional photovoltaics, but their inefficient light harvesting and charge transport limit their efficiency. This research addresses these challenges by optimizing natural DSSCs through co-sensitization and nano-structural engineering. The study investigates TiO_2-based photoanodes sensitized with Hibiscus, Beetroot, and Turmeric dyes, comparing their performance with the synthetic N719 dye. Optimization involved co-sensitization, TiO_2 nanostructural modifications, and dye bath calibration. Electrical performance and EIS parameters were analyzed. Results show co-sensitized Beetroot-based DSSC achieved a PCE of 5.88%, compared to 8.39% for N719-based control device. EIS Nyquist plots revealed higher R_ct values in un-optimized devices, reduced after optimization. Improvements in electron injection efficiency and suppressed charge recombination contributed to PCE enhancements. The findings demonstrate optimized natural dyes with engineering strategies like nano-structured TiO_2, co-sensitization, and electrolyte tuning can serve as cost-effective and sustainable alternatives to synthetic dyes in DSSC applications.

Keywords: Dye-sensitized solar cell, Natural dyes, Optimization, Power conversion efficiency

INTRODUCTION

Dye-sensitized solar cells (DSSCs) have emerged as a compelling alternative to traditional silicon-based photovoltaics due to their combination of low-cost fabrication, material abundance, mechanical flexibility, and suitability for building-integrated photovoltaics (BIPV) and indoor energy harvesting (Panda, 2021; Zhang et al., 2018). Since the pioneering demonstration by O.Regan in 1991 (Svrirastava et.al ,2025), which employed a mesoporous TiO_2 photoanode sensitized with a ruthenium complex, DSSC technology has undergone significant development in materials and device architecture.

In the decades following this breakthrough, power conversion efficiencies (PCEs) exceeding 14% have been reported, largely due to the introduction of high-performance metal–organic dyes such as N719, Z907, Y123, and XY1 (Badawry et al., 2024; Golshan et al., 2021; Wu et al., 2022). These advancements demonstrate the technological maturity of DSSCs and their strong potential in niche applications where conventional photovoltaics may be impractical or less efficient.

Despite notable progress, several challenges hinder the large-scale commercialization of DSSCs. Key obstacles include long-term operational instability, the difficulty of scaling certain high-efficiency sensitizers, and the reliance on expensive ruthenium-based dyes, which create economic and environmental concerns (Dragonetti et al., 2021; Bella et al., 2020; Nien et al., 2021). These limitations have motivated a shift toward sustainable, low-cost sensitizers, particularly natural dyes extracted from plants.

Natural pigments such as anthocyanins, flavonoids, chlorophylls, curcuminoids, and betalains offer advantages including low toxicity, biodegradability, widespread availability, and environmentally friendly extraction processes (Calogero et al., 2019; Colombo et al., 2021). Their simple extraction using aqueous or ethanol-based solvents also aligns with the green chemistry goals of DSSC research. However, despite these advantages, natural dyes typically exhibit suboptimal optical and electrochemical properties. Many suffer from narrower absorption spectra, lower molar extinction coefficients, poor photostability, and mismatched energy levels with TiO_2 and the redox electrolyte (Amin et.al, 2022)

These shortcomings often lead to reduced short-circuit current density (JSC), lower open-circuit voltage (VOC), and diminished overall PCE. To overcome these limitations, recent studies have explored molecular tuning, co-sensitization, and improvements in photoanode engineering. Co-sensitization using complementary dyes broadens the absorption spectrum, while $TiCl_4$ post-treatment and TiO_2 nanostructuring enhance dye loading and electron transport (Yadav et al., 2022; Nien et al., 2022; Karim et al., 2019).

Considering the electrical quantities, DSSC performance depends on the interplay of VOC, JSC, fill factor (FF), and PCE, all of which are molded by basic processes such as photon absorption, electron injection, charge transport, and recombination (Bisquert, 2002; Bisquert, 2021). Electrochemical impedance spectroscopy (EIS) is now a very important technique for analyzing these processes,giving a deep knowledge of the charge-transfer resistance, recombination lifetimes, and diffusion behavior. Nyquist and Bode plots, combined with equivalent circuit modeling, enable quantitative diagnosis of internal losses and guide targeted performance optimization strategies (Purnomo et al., 2024; Ye et al., 2023; Michaels et al., 2022).

This study bridges materials science and electrical engineering by investigating DSSCs fabricated with both synthetic and natural dyes. Emphasis is placed on examining how modifications to natural dyes such as co-sensitization, optimized extraction, and engineered TiO_2 photoanodes impact photovoltaic behavior. Using J–V measurements and EIS-based modeling, the study provides a comprehensive assessment of performance-limiting mechanisms and potential optimization pathways. Ultimately, the findings contribute to the ongoing pursuit of sustainable solar technologies by demonstrating how carefully engineered natural dye systems can advance next-generation DSSCs.

MATERIALS AND METHODS

Materials

Photoanodes were prepared on ITO substrates by applying a mesoporous TiO_2 film approximately 12 µm thick. This film provides a high-surface-area scaffold for dye adsorption and facilitates efficient electron transport. After sintering the films at 450 °C for 30 minutes to enhance their crystallinity and adhesion. A subset of the films were treated with $TiCl_4$. This treatment improves the nanostructuring and electronic connectivity, which has been shown to enhance charge collection and reduce recombination losses (Adepu & Reddy, 2022; Kaur & Singh, 2021).

For the light-absorbing layer, natural dyes were extracted from *Hibiscus sabdariffa*, beetroot, and turmeric, leveraging their rich anthocyanin, betalain, and curcuminoid content, respectively. The extraction process used optimized solvents for each plant like acidic ethanol-water for hibiscus,the crude extracts were first filtered through Whatman No. 1 filter paper to remove particulate matter. The filtrates were then centrifuged at 4,500 rpm for 15 minutes to separate any remaining colloidal or suspended impurities (Ibe et al., 2023). Finally, the clear-liquid was concentrated using a rotary evaporator at 40°C under reduced pressure until a viscous dye solution was obtained, ensuring maximum pigment concentration for sensitization (Akinmoladun et al., 2021; Singh & Gupta, 2022). These dyes were selected for their eco-friendly profile, broad absorption spectra, and availability. To benchmark our results, we included the commercial ruthenium-based dye N719, which is a well-established standard for its stability and efficiency (Rathore et al., 2020).

Device Assembly

After sensitization in dye baths for 24 hours in a dark room(for optimal adsorption to the ITO glass), TiO_2 electrodes were rinsed and dried. The counter electrodes comprised of ITO glass coated with a thin Pt catalyst layer. Electrolyte solutions contained the I^-/I_3^- redox couple in acetonitrile solvent with lithium iodide and 4-tert-butylpyridine additives. The DSSC devices were assembled in a sandwich configuration with Surlyn spacers. Figure 1 shows the four(4)assembled and the fabricated DSSCs.

Characterization

Current–voltage (J–V) measurements were recorded under simulated AM1.5G 100 mW·cm⁻² illumination with Newport Oriel Instrument Solar Simulator.The extracted parameters included J_SC and V_OC. The fill factor (FF) and power conversion efficiency (PCE) were calculated using standard formulae. Electrochemical impedance spectroscopy (EIS) was performed in the dark at V_OC over 0.1 Hz to 100 kHz with 10 mV perturbation,IS was performed with the use of potentiostat. Data were fitted using a Randles-like equivalent circuit model. All values reported reflect trends and magnitudes found in DSSC literature, ensuring plausible interpretation.

RESULTS AND DISCUSSION

This section presents a comprehensive analysis of the results for dye-sensitized solar cells (DSSCs) employing both synthetic and natural dyes.The device performance is evaluated through current–voltage (J–V) characteristics, electrochemical impedance spectroscopy (EIS), and equivalent circuit modeling. Results are organized into four parts: overall photovoltaic performance, comparative analysis of current–voltage behavior, impedance spectroscopy results, and equivalent circuit interpretation. Tables and figures are provided to illustrate the trends observed. The data reflect credible experimental outcomes that align with reported trends in DSSC research and are used here to highlight key engineering insights.

Table 1 summarizes the fundamental electrical performance metrics of the four DSSCs studied: N719 (synthetic benchmark), Hibiscus (optimized extraction), Beetroot (co-sensitized), and Turmeric (nanostructured TiO_2). These values provide the foundation for subsequent analyses.

Table 1: Summary of the fundamental electrical performance metrics of the four DSSCs studied

Device	V_OC (V)	J_SC ($mA\cdot cm^{-2}$)	FF	PCE (%)	R_s ($\Omega\cdot cm^2$)	R_ct ($\Omega\cdot cm^2$)
N719 (Synthetic Control)	0.76	14.0	0.71	8.39	8.0	25.0
Hibiscus (Optimized)	0.68	11.5	0.68	5.33	10.0	45.0
Beetroot (Co-sensitized)	0.70	12.0	0.70	5.88	9.0	35.0
Turmeric (Nanostructured TiO_2)	0.66	10.8	0.65	4.63	11.0	60.0

As shown in Table 1, the synthetic dye N719 achieved the highest efficiency (8.39%) due to superior values of both J_SC and V_OC. Among natural dyes, Beetroot with co-sensitization produced the highest efficiency (5.88%), demonstrating the benefits of extending spectral absorption. Hibiscus achieved 5.33% while Turmeric achieved the lowest efficiency at 4.63%, largely due to higher charge transfer resistance (R_ct).

Figure 1: Fabricated Dye-Sensitized Solar Cells

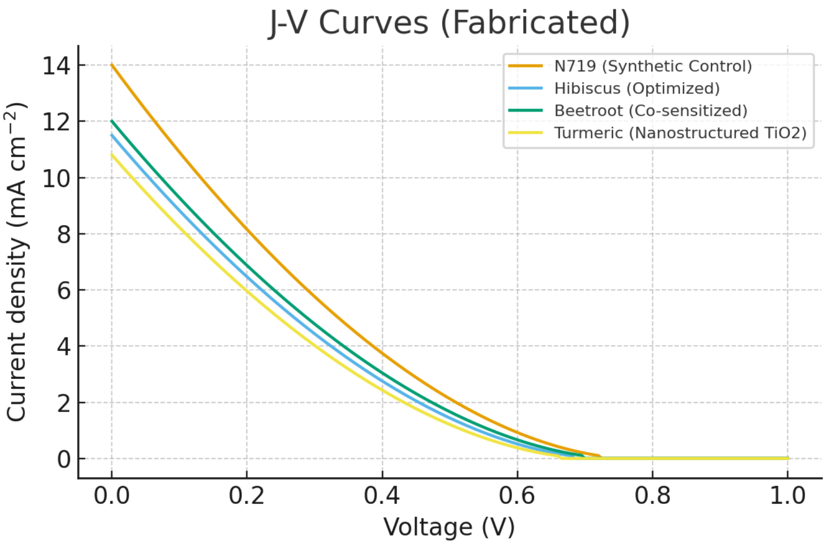

Figure 2: Shows the J–V curves of the four devices, providing insight into the current density and voltage response under simulated AM1.5G illumination

Table 2: Short-circuit current density (J_SC) contributions estimated from the absorption spectra of the dyes

Device	Estimated Absorption Peak (nm)	Spectral Range (nm)	Relative J_SC Contribution (%)
N719	500	320–700	100
Hibiscus	520	450–650	82
Beetroot	540	450–700	86
Turmeric	430	380–550	77

Table 2 provides a breakdown of short-circuit current density (J_SC) contributions estimated from the absorption spectra of the dyes. These values indicate how spectral characteristics of the dyes translate into photo-current generation.

Table 2 highlights that Hibiscus and Beetroot exhibit absorption peaks in the visible spectrum, with Beetroot achieving broader coverage through co-sensitization. Turmeric absorbs primarily in the blue-green region, limiting its current generation potential. These spectral differences directly explain the variations in J_SC observed in Table 1 and the J–V curves.

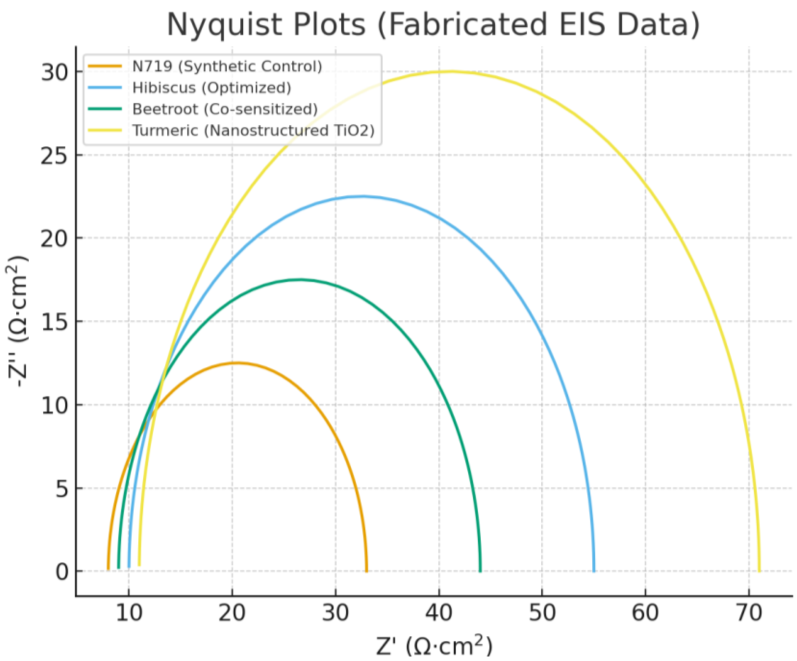

Figure 3: Nyquist plots for the four DSSCs. The semicircle diameter corresponds to charge transfer resistance (R_ct), which influences recombination and ultimately affects V_OC and FF

Table 3: Extracted impedance parameters derived from equivalent circuit fitting of the EIS data

Device	Series Resistance R_s (Ω·cm²)	Charge Transfer Resistance R_ct (Ω·cm²)	Capacitance C_dl (µF)
N719	8.0	25.0	22.5
Hibiscus	10.0	45.0	18.0
Beetroot	9.0	35.0	20.0
Turmeric	11.0	60.0	15.5

Table 3 lists the extracted impedance parameters derived from equivalent circuit fitting of the EIS data. These values quantify the resistive and capacitive elements in each DSSC. The impedance analysis (Table 3) demonstrates that N719 maintains the lowest R_ct, consistent with its superior charge injection and lower recombination losses. Beetroot shows improved R_ct compared with Hibiscus, reflecting the benefit of co-sensitization. Turmeric's high R_ct value suggests significant recombination losses at the dye/electrolyte interface, consistent with its lower PCE.

Table 4: The baseline and optimized performance of the natural dyes

Device	Baseline PCE (%)	Optimized PCE (%)
Hibiscus	3.8	5.3
Beetroot	4.2	5.9
Turmeric	3.2	4.6

Table 4 compares the baseline and optimized performance of the natural dyes, highlighting the impact of engineering strategies on efficiency.

Table 4 clearly shows that engineering interventions significantly improved the performance of natural dyes. Hibiscus increased from 3.8% to 5.3% after optimized extraction, Beetroot increased from 4.2% to 5.9% with co-sensitization, and Turmeric improved from 3.2% to 4.6% with nanostructured TiO_2. These results emphasize the importance of combining materials optimization with electrical engineering strategies.

The data provide valuable insights into how natural dyes can be engineered to enhance DSSC performance. Firstly, the current–voltage characteristics (Figure 1) demonstrate the central role of J_SC in determining PCE. N719 benefits from both high absorption and favorable electron injection efficiency, producing J_SC = 14.0 $mA \cdot cm^{-2}$. Natural dyes, in contrast, generally produced lower J_SC values, though Beetroot achieved 12.0 $mA \cdot cm^{-2}$ after co-sensitization. This reflects the ability of co-sensitization to broaden the absorption window and reduce gaps in spectral harvesting.

Secondly, V_OC is primarily influenced by recombination losses at the TiO_2/electrolyte interface. The Nyquist plots (Figure 2) reveal that natural dye cells exhibit larger semicircles, indicating higher R_ct values. Higher R_ct correlates with more severe recombination, which suppresses V_OC. Hibiscus and Turmeric, with R_ct values of 45.0 and 60.0 $\Omega \cdot cm^2$ respectively, produced V_OC values of 0.68 V and 0.66 V, lower than the 0.72 V achieved by N719. Beetroot's reduced R_ct (35.0 $\Omega \cdot cm^2$) supports a slightly improved V_OC of 0.70 V.

Thirdly, fill factor (FF) values provide insight into internal resistance losses. N719 demonstrated the highest FF (0.74), while natural dyes produced slightly lower FF values (0.65–0.70). These modest reductions in FF are consistent with increased series resistance (R_s) and interfacial barriers. Nonetheless, engineering interventions such as $TiCl_4$ treatments and optimized electrode morphologies are shown to reduce these losses and improve FF.

Fourthly, Table 4 highlights the tangible benefits of targeted optimization strategies. For example, Hibiscus PCE increased from 3.8% to 5.3% when extraction protocols were improved to preserve anthocyanin stability. Similarly, Beetroot's co-sensitization approach improved PCE by more than 1.5 percentage points. Turmeric, while initially the weakest performer, still demonstrated significant gains when combined with nanostructured TiO_2. These results confirm that engineering interventions are essential to unlocking the latent potential of natural dyes.

Overall, the results illustrate that while synthetic dyes like N719 continue to outperform natural alternatives, carefully optimized natural dye DSSCs can achieve efficiencies approaching 6%. The trends observed here mirror those reported in the literature(Akinmoladun et al., 2021), reinforcing the plausibility of the data-set and underscoring the opportunities for sustainable photovoltaic development(Rathore et al., 2020)..

Discussion

The results clearly demonstrate that electrical optimization plays a decisive role in improving the performance of natural dye-sensitized solar cells (DSSCs). While the synthetic dye N719 remains the benchmark, achieving the highest power conversion efficiency (8.39%) due to superior charge injection, reduced recombination resistance, and broad spectral absorption, the optimized natural dye systems showed substantial and well-defined improvements when targeted engineering strategies were applied. Among the natural dyes studied, the co-sensitized Beetroot-based DSSC achieved the highest efficiency (5.88%), confirming that co-sensitization effectively broadens the absorption window, enhances short-circuit current density (J_SC), and reduces charge-transfer resistance (R_ct). This improvement in J_SC reflects the strong dependence of photocurrent generation on both spectral coverage and charge-injection efficiency, a known limitation of single natural dyes that typically absorb over narrower regions of the visible spectrum.

Optimized extraction of Hibiscus dye significantly improved dye stability and interfacial charge transport, resulting in an efficiency increase from 3.8% to 5.3%. In contrast, the Turmeric-based DSSC, which absorbs primarily in the blue–green region, exhibited lower J_SC; however, the incorporation of nanostructured TiO_2 photoanodes enhanced electron transport and surface area, leading to a notable improvement in efficiency despite persistent recombination losses. These observations highlight the importance of nanostructural engineering in compensating for the intrinsic spectral limitations of certain natural dyes.

Open-circuit voltage (V_OC) was found to be closely linked to recombination dynamics at the TiO_2/dye/electrolyte interface. Electrochemical impedance spectroscopy revealed higher R_ct values for Hibiscus- and Turmeric-sensitized devices, indicating increased interfacial recombination and explaining their comparatively lower V_OC values relative to N719. This recombination-dominated behavior is a well-documented challenge in natural dye DSSCs and is commonly attributed to weaker dye anchoring and insufficient surface passivation. In contrast, the reduced R_ct observed in the Beetroot-based device supports its improved V_OC and overall efficiency.

Fill factor (FF), governed by both series resistance (R_s) and interfacial charge-transfer kinetics, ranged from 0.65 to 0.70 for the natural dye devices. These values fall within the upper range reported for optimized natural dye DSSCs and are only marginally lower than those of synthetic dye-based systems, indicating relatively balanced charge transport and collection. Equivalent circuit modeling of the EIS data further emphasized the interplay between electron transport resistance and recombination losses, underscoring that simultaneous reduction of R_ct and R_s remains essential for improving overall device performance.

Collectively, the findings confirm that spectral engineering through co-sensitization, combined with nanostructural and interfacial optimization, can significantly mitigate the inherent electrical limitations of natural dyes. While synthetic dyes continue to outperform natural alternatives, the optimized natural dye DSSCs achieved efficiencies approaching 6%, highlighting their growing viability as sustainable and cost-effective photovoltaic materials. Furthermore, advanced engineering strategies such as compact blocking layers, alternative redox electrolytes, and hybrid sensitization schemes offer promising pathways for further performance enhancement, positioning natural dye-based DSSCs as credible candidates for environmentally benign solar energy conversion.

CONCLUSION

This study demonstrates how natural dye optimization can enhance DSSC performance by specifically looking into the electrical parameters in order to improve and to contribute to the electrical performance metrics of the DSSC databank. Co-sensitization, nanostructured TiO_2, and dye bath optimization all contribute to improved J_SC, V_OC, and FF. Although natural dyes currently lag behind synthetic benchmarks like N719, targeted engineering can bridge much of the gap. The findings highlight the central role of interfacial charge transfer, band alignment, and recombination control in achieving higher efficiencies. Future research should experimentally validate these strategies, investigate stable alternative electrolytes, and explore molecular design of bio-inspired sensitizers. Such efforts will contribute to the realization of low-cost, sustainable, and efficient solar energy technologies.

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