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Long-term Stability of Dye-sensitized Solar Cells Utilizing China Doll Leaf-derived Natural Dyes and Electron Dynamics at the TiO_2–dye Interface

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ABSTRACT

Evergreen glossy leaves of the China doll plant were selected as a natural source for the extraction of a photosensitizer. A scalable ethanol-assisted extraction strategy was employed, wherein controlled leaf powder loadings (0.5, 0.8, 1.0, and 1.5 g per 10 mL) enabled systematic tuning of the photosensitizer concentration. The extracted natural dye was characterized using UV–Visible and FTIR spectroscopy to confirm its optical absorption characteristics and functional groups relevant to photosensitization. When employed as a sensitizer in dye-sensitized solar cells, systematic variation of dye loading enabled optimization of device performance. Photovoltaic evaluation revealed a maximum power conversion efficiency of 2.04% with a fill factor of 0.707 for the 1 g per 10 mL ethanol extract, while SEM analysis confirmed a uniform photoanode morphology. The enhanced performance at the optimal dye concentration is attributed to improved light harvesting and charge transport, demonstrating the effectiveness of controlled natural dye loading for DSSC applications.

Keywords: China doll leaf extract; Natural dye; Dye-sensitized solar cells; Microwave-treated leaves.

1. INTRODUCTION

Extensive studies have investigated the performance of dye-sensitized solar cells (DSSCs) employing natural dyes, highlighting their potential advantages in terms of stability, material compatibility, cost efficiency, and environmental sustainability. Among various natural dyes, chlorophyll-rich green pigments extracted from plant leaves have attracted attention due to their favorable light-absorption characteristics and energy-transfer capabilities, which contribute to photo-to-electrical energy conversion. Previous investigations have systematically examined the role of these pigments in electron excitation and charge injection processes following photon absorption. CuO nanoparticles synthesized using Calotropis gigantea leaf extract were deposited on a fluorine-doped tin oxide (FTO) glass substrate to fabricate a DSSC counter electrode (Jitendra *et al.* 2015). With a fill factor of 0.62, the manufactured cell had an efficiency of 3.4% in converting solar energy into electrical energy. A solar cell with a photoanode soaked in chlorophyll pigments made from spinach leaves was created and tested by (Syafinar*et al.* 2015). The fabricated cell exhibited a fill factor of 0.49, an open-circuit voltage of 0.44 V, and a short-circuit current density of 0.35 A cm^{-2}. (Sethu *et al.* 2024) demonstrated the synthesis of herbal extract-based metal oxide materials for use in DSSCs (Premkumar *et al.* 2024; Gottipati *et al.* 2022; Meena *et al.* 2022; Shanmugan *et al.* 2022b; Rajasekar *et al.* 2021; Sethu and Shanmugan, 2024). (Gali *et al.* 2025) enhanced solar design performance using SZNs, achieving 4.5 kg/m^2 yield, faster heat transfer, higher temperature, and 37.2% improved efficiency at 30% concentration. (Durga *et al.* 2025) improved solar design using sugarcane-derived TiO_2 nanofluid, achieving 8.4 kg/m^2/day yield, 58.7% efficiency, enhanced heat absorption, larger evaporation area, and reduced water cost. (Sowmya *et al.* 2025) enhanced solar still performance using TiO_2–dragon fruit peel coated absorber, achieving 9.02 kg/m^2/day yield, 178.9% productivity improvement, higher photothermal efficiency, reduced carbon emissions, and lower water cost. (Murugesan and Shanmugan, 2025) enhanced M-shaped solar still using paraffin–silver nanoparticle PCM, achieving 8.1 kg/m^2/day yield, 54.7% efficiency, 26% productivity increase, reduced cost, and environmental benefits. (Balaji *et al.* 2025) enhanced single-basin solar still using Grewia extract–$MgCl_2{\cdot}6H_2O$ PCM, achieving 57% higher productivity, 59.5% efficiency, lower water cost, improved exergy performance, and reduced annual CO_2 emissions. (Nashmi *et al.* 2025b) improved solar distiller

performance using microwave-treated MXene–Ag–dye nanofluid, achieving 5.34 L/m² yield, 52% efficiency, enhanced thermal conductivity, energy storage, and superior photothermal conversion at 30% concentration. (Alfred *et al.* 2021) fabricated three solar cells using dyes derived from black chokeberry leaves, pomace juice, and the synthetic N719 dye. A life-cycle assessment-based environmental performance analysis revealed that the N719 dye had the lowest environmental impact among the natural dyes. Charge-transfer mechanisms, charge recombination, and DSSC stability were systematically examined (Durairaj *et al.* 2025; Ravinder *et al.* 2024; Almeshaal and Shanmugan, 2024). In natural dye-based DSSCs, hybrid or co-sensitized configurations are generally associated with efficiencies exceeding 10%, in contrast to systems employing only plant-derived sensitizers (Murugesan *et al.* 2024; Shanmugan *et al.* 2021; Ravinder *et al.* 2023a; Asha *et al.* 2022). In this context, the present work aims to provide mechanistic insights into charge-transfer dynamics (Abdulmohsen *et al.* 2022; Arulraj *et al.* 2022; Kumar *et al.* 2021) and operational stability, along with a practical assessment of device performance (Sangeetha *et al.* 2024; Shanmugan *et al.* 2021; Shanmugan *et al.* 2020; Shafay *et al.* 2025).For photoanode applications, (Gabriel *et al.* 2021) synthesized Ag nanoparticles via a green route, incorporated them into TiO_2 nanoparticles, and subsequently sensitized the composite with dyes extracted from neem, henna, and ruthenium leaves. The resulting DSSCs exhibited power conversion efficiencies of 4.09%, 1.57%, and 1.18% for N719, neem, and henna sensitizers, respectively. The enhanced photovoltaic performance observed at the optimal dye loading in the nanocomposite indicates improved device stability and more efficient interfacial charge transport (Abdulaziz and Shanmugan, 2025; Maruthasalam *et al.* 2025; Abdullah *et al.* 2022; Essa *et al.* 2021). Increases in fill factor and short-circuit current density reflect superior light-harvesting capability and more effective electron injection from the sensitizer into the TiO_2 conduction band (Abdullah *et al.* 2021; Essa *et al.* 2020a and 2020b Palanikumar *et al.* 2020; Bahaa *et al.* 2024). Furthermore, SEM analysis revealed favorable photoanode morphology (Emad *et al.* 2022; Essa *et al.* 2022; Panchal *et al.* 2021a and 2021b) and uniform dye coverage, which are consistent with suppressed interfacial charge recombination and the absence of noticeable performance degradation over repeated measurements.

Building on green leaf-based DSSC studies, the present work explicitly advances understanding of long-term operational stability and interfacial electron dynamics beyond prior reports, while offering a comparative assessment of natural dyes, mixed pigments, and device efficiencies (Thamizharasu *et al.* 2021; Laad *et al.* 2020; Shanmugan *et al.* 2020; Kavitha *et al.* 2021).The performance of solar cells with sensitizers derived from shea and baobab leaves was compared in an experiment conducted by (Urenyang *et al.* 2021). It was found that the shea-based cell had a greater photoconversion efficiency of 0.25% compared to 0.11% for the baobab-based cell. Due to aggregation-induced recombination, natural-dye DSSCs are frequently constrained by poor stability and ineffective interfacial charge dynamics at high dye loadings. Traditional green-leaf sensitizers, which are usually made from generic chlorophyll extracts, offer little control over the interfacial electron transfer and dye concentration. As an alternative sensitizer system, Radermachera sinica (China doll plant) leaf dyes were investigated in this work, demonstrating concentration-dependent electron-injection optimization and recombination suppression.When compared to previously published green-leaf DSSCs, systematic dye-loading control, bolstered by morphological and spectroscopic investigations, results in enhanced stability and interfacial charge transport. (Akula *et al.* 2023) investigated the foliage and flowers of Ixora coccinea and Spinacia oleracea. The co-sensitization of such dyes is stable for 450 hours with a photoconversion efficiency of 2.39%, according to the experimental results. This study shows that dyes made from Radermachera sinica leaves allow for regulated modulation of sensitizer concentration while maintaining effective interfacial charge transfer, in contrast to many DSSCs based on generic chlorophyll extracts. Superior photovoltaic performance is achieved through enhanced light absorption and improved electron injection into the TiO_2 conduction band as a result of optimized dye loading. Unlike conventional green-leaf dyes, which commonly suffer from concentration-induced performance loss, these sensitizers display steady behaviour even at higher dye concentrations. In earlier research on natural dye-sensitized DSSCs, chlorophyll-rich extracts from plant leaves, fruits, and flowers were frequently used; these extracts usually produced modest efficiencies but poor operational stability (Gandhi *et al.* 2022). (Najihah *et al.* 2022) used dyes rich in chlorophyll that were derived from Costuswoodsonii leaves to study the long-term operational viability of DSSCs. In a comparable investigation, a manufactured solar cell achieved an efficiency of 0.65% with a short-circuit current density of 2.25 mA cm^{-2} (Gandhi *et al.* 2021). It is widely known that aggregation-induced recombination and ineffective interfacial electron transport cause performance loss with greater dye loadings. Cross-study comparisons indicate a continual trade-off between efficiency and stability, underlining the paucity of sensitizers that allow regulated dye loading while keeping significant interfacial charge-transfer dynamics. For comparatively under-explored sources such as China doll leaves, systematic investigations of concentration-dependent behaviour and long-term stability remain restricted. While previous research on natural and mixed-pigment DSSCs has

shown that bio-derived sensitizers are feasible, these studies typically yield very modest efficiency. Increasing pigment complexity or dye concentration is a typical way to increase efficiency; however, aggregation-induced recombination often impairs stability. Furthermore, the majority of research focuses on maximal efficiency values with little regard for concentration-dependent effects and interfacial electron dynamics. Therefore, there is still a lack of knowledge regarding charge-transfer and recombination mechanisms in natural dye-sensitized DSSCs, especially those that use less researched colors derived from leaves.The present study addresses these essential gapsby thoroughly analyzing concentration-controlled China doll leaf sensitizers with an emphasis on interfacial charge transport and operational stability.With an emphasis on interfacial electron transport and dye persistence, this study attempts to thoroughly examine Radermacherasinica leaf pigments as natural DSSC sensitizers. This study also looks into how changes in dye concentration affect light harvesting, electron injection, and recombination at the dye–TiO_2 interface. Controlled manipulation of dye loading is intended to enhance interfacial charge transfer, hence enhancing operating stability and efficiency compared with non-optimized green-leaf sensitizers. Following dye adsorption onto TiO_2-coated FTO substrates, the resulting photoanode morphology wasexamined using SEM. Complete solar cells incorporating a graphite counter electrode and an iodide/triiodide electrolyte were fabricated and assessed through current–voltage measurements to evaluate photovoltaic performance.

2. MATERIALS AND PRECURSORS USED

2.1 Preparation of the Dye

The China doll's leaves were collected from the garden of the KL University of Arts and Science, AP, India. To get rid of the dust, the leaves were weighed at 100g and then carefully cleaned with distilled water. After that, it was instantly mashed into a paste without being carefully ground. The powdered leaves were subjected to microwave irradiation, where dielectric heating induced rapid volumetric heating of the material. By causing rapid dielectric heating inside the plant matrix, which breaks down cell walls and speeds up solvent penetration, microwave treatment improves dye extraction. This minimizes extended heat exposure and encourages effective pigment molecule release, which lowers pigment degradation and increases color stability. The procedures used to extract dyes from China doll leaves and treat the powdered leaves with microwaves are depicted in Fig. 1. The microwave-treated powder and its different weight concentrations in 10 mL of ethanol are shown in Fig. 2. After a day, the ethanolic solution containing dye powder was filtered to extract the dye.

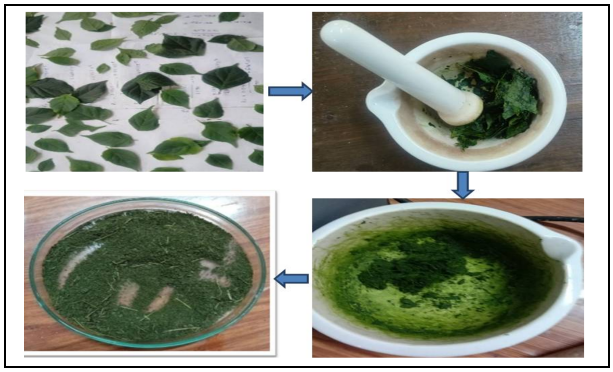


Fig. 1: Powdered leaves from china doll

Sigma Aldrich provided the FTO substrate, which has a broad band gap of 3.6 eV, a high optical transmittance of 85%, and a low surface resistance of 10 ohms/square. TiO_2 nanopowder with the average grain size of 35nm was converted into a paste using the nitric acid,Following the process reported by (Shanmugan *et al.* 2026), TiO_2 paste was made by dispersing TiO_2 nanopowder in deionized water containing 0.1 M acetic acid and magnetically swirling the mixture for 12 h at ambient temperature to obtain a uniform suspension (Ghassan *et al.* 2024).



Fig. 2: Microwave-treated powdered dye and dye extracts prepared at various weight concentrations in 10ml of ethanol

The paste viscosity was then adjusted for coating and aged for 24 h to promote particle–particle interaction. To increase film adhesion and crystallinity (Sangeetha *et al.* 2022), the conditioned paste was deposited onto FTO substrates using the doctor-blade technique. It was then annealed for two hours at about 450 °C. After cooling, the TiO_2-coated FTO substrates were immersed in dye solutions of different concentrations (0.5, 0.8, 1.0, and 1.5 g diluted in 10 mL of ethanol) to allow effective dye adsorption onto the TiO_2 surface (Ravinder *et al.* 2023b).

2.2 Preparation of Electrolyte and Counter Electrode

Following the procedure outlined by (Jony *et al.* 2026), a liquid iodine/triiodide (I^-/I_3^-) redox electrolyte was created by dissolving 0.67 g of lithium iodide (LiI) in 30 mL of acetonitrile, followed by the addition of nitric acid (0.1 M) to adjust electrolyte acidity. For around three

hours, the solution was magnetically agitated to ensure full dissolution and the creation of a uniform redox electrolyte appropriate for DSSC functioning. Graphite was evenly applied to an FTO substrate for the counter electrode. Then, a binder clip was used to sandwich the dye-sensitized photoanode and graphite-coated counter electrode together. The prepared liquid electrolyte was then added between the electrodes to create an electrolyte layer that promotes redox reactions and allows for continuous dye regeneration while the device is operating. A thin catalytic layer with an average thickness of 8–10 µm was formed by evenly depositing graphite onto the FTO glass substrate to create the counter electrode. Good adhesion between the graphite layer and the FTO substrate was achieved through homogeneous spreading and subsequent heat treatment (Suresh *et al.* 2024). To improve mechanical stability, electrical contact, and catalytic activity towards the I^-/I_3^- redox process, the coated substrates were heat-treated for 30 min at 450 °C.

3. RESULTS AND DISCUSSION

3.1 FTIR Spectroscopy of the Dye

FTIR spectroscopy was used to analyze the dye extracted in ethanol at different concentrations to detect the functional groups present in the dye. The FTIR spectra of the dyes were recorded in ATR mode using an IRAffinity spectrometer fitted with a MIRacle 10 ATR accessory, as illustrated in Fig. 3. In line with the findings of (Poludasu *et al.* 2025), similar significant FTIR peaks were seen for all dye concentrations produced in 10 mL of ethanol, indicating that all four samples contained the same functional groups. The presence of a vinyl monosubstituted group in the dye molecule is indicated by a high absorption peak at 885.57 cm^{-1}, which corresponds to the C–H out-of-plane bending mode. The peak detected at 1039.42 cm^{-1} is ascribed to C–F stretching of an aliphatic fluorochemical, which is coupled with considerable absorption in the visible region. The dye's phenolic group demonstrated O–H bending at 1374.03 cm^{-1}, demonstrating its propensity for coordination bonding with nearby molecules. This interaction shows a high possibility of stable attachment between the dye and the TiO_2 semiconductor oxide. The wide H-bonded O–H stretching band seen at 3326.92 cm^{-1}, along with noticeable C–H asymmetric and symmetric stretching peaks, offers additional proof. This wide band's existence suggests that the dye has a high anchoring affinity for the TiO_2 semiconductor oxide surface. To determine which functional groups were in the extracted dye and to investigate potential interactions with the TiO_2 surface, FTIR spectroscopy was used. The discovered FTIR peaks show the presence of distinctive functional groups responsible for molecule binding and anchoring; nevertheless, FTIR did not provide direct information on visible-light absorption or charge-transfer behaviour. Hence, UV-Visible spectroscopy was utilized to analyze the dye's optical absorption properties, while FTIR analysis was only employed to examine the chemical structure and interfacial compatibility with the TiO_2 semiconductor.

3.2 UV-visible Spectroscopy

A Shimadzu 1700 series spectrophotometer was used to perform UV–Visspectroscopy of the extracted dyes at various concentrations in ethanol, as shown in Fig. 4. Powdered dye samples were dissolved in ethanol at concentrations of 0.5 g, 0.8 g, 1.0 g, and 1.5 g in 10 mL solvent. To assess light-harvesting properties, the absorption spectra were captured for every concentration. The 1.0 g/10 mL solution displayed the maximum absorption intensity, with pronounced peaks at 316, 330, 352, 412, 430, 453, 473, 642, and 664 nm, showing considerable light absorption by chlorophyll and carotenoid pigments. Peaks at 430 nm and 662 nm were ascribed to chlorophyll a, while peaks at 453 nm and 642 nm matched chlorophyll b. Carotenoids, which function as supplementary pigments, were identified as the source of an extra band at 480 nm. Although the absorption was relatively weaker at the lower and higher doses (0.5, 0.8, and 1.5 g/10 mL), the peak positions did not change, indicating that the dye composition was constant at all concentrations. In line with (Laxmikant *et al.* 2026), these findings show the dye's potential for visible light harvesting and point to an efficient chemical interaction that can promote electron transfer when employed as a sensitizer in DSSCs.

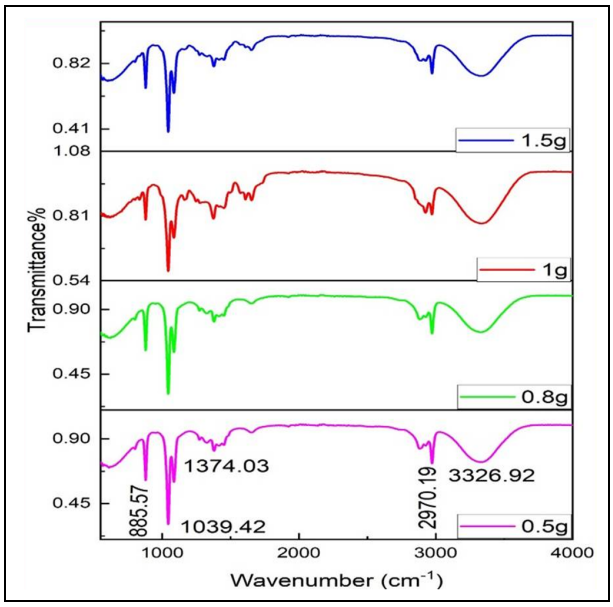


Fig. 3: FTIR for 0.5g, 0.8g, 1g and 1.5g powdered dye in 10ml of ethanol

UV–visible spectroscopy was used to characterize light absorption and pigment composition, while FTIR was utilized to identify functional groups and possible dye anchoring. Electron-transfer mechanisms

were assessed by complementary electrochemical and photovoltaic measurements rather than spectroscopic data. These modifications ensure that every assertion is backed up with relevant data.

3.3 SEM Image of the Photoanode

Fig. 5shows an SEM image of the photoanode. The dye molecules are observed to be coordinated and impregnated into the TiO_2-coated FTO substrate. The morphology exhibits a homogeneous coverage of TiO_2 with heterogeneously distributed dye molecules. Upon exposure to sunlight, the dye molecules on the TiO_2 surface absorb light and generate excitons, which can migrate into the semiconductor oxide material without significant energy loss, in agreement with (Nashmi *et al.* 2025a).The smooth interface between the dye andTiO_2 facilitates efficient charge transfer from the excited state of the dye molecules to the conduction band of the TiO_2.Fig. 5 shows the SEM image of the TiO_2-coated FTO photoanode. The average diameter of the nanoparticles is between 20 and 25 nm, and they form a porous network that makes dye adsorption easier. According to cross-sectional SEM scans, the thickness of the TiO_2 layer is around 10 µm. The porous structure and homogenous film coating give a large surface area for dye loading, which is expected to boost light harvesting. These morphological traits align with thereportedcurrent density–voltage (J–V) characteristics of the DSSCs, wherein optimal film thickness and effective dye coverage lead to increased photocurrentand overall power conversion efficiency.

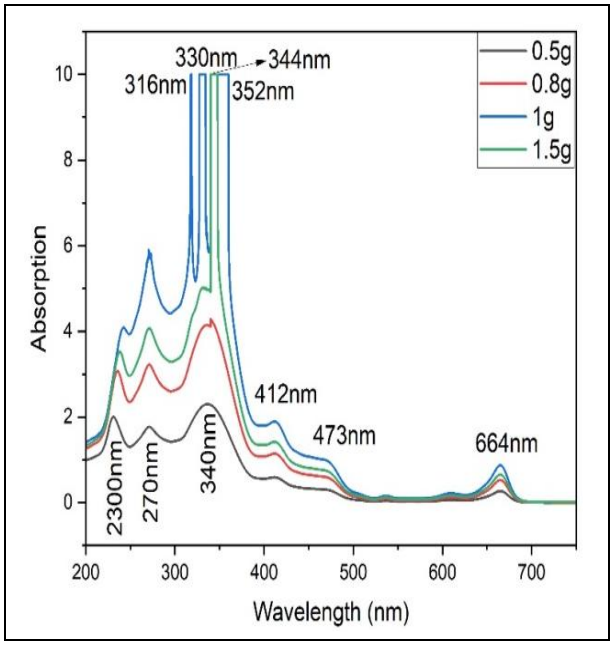


Fig. 4: UV-vis spectroscopy

3.4 J–V Analysis of Solar Cell Fabrication

Photographs of the manufactured DSSCs and their J–V parameters measured using a Keithley 2400 solar simulator are shown in Fig. 6.With a power conversion efficiency of 2.04%, an open-circuit voltage (VOC) of 1.03 V, a short-circuit current density (Jsc) of 2.8 mA/cm², and a fill factor (FF) of 0.7073, the DSSC sensitized with 1 g of powdered leaves in 10 mL of ethanol demonstrated the best performance among the tested dye concentrations. The 1 g/10 mL concentration is ideal for light absorption and device performance under the measured conditions, as evidenced by the relatively lower efficiencies generated by lower dye concentrations. The solar cell properties for different concentrations of powdered leaves in 10 mL of ethanol are presented in Table 1.

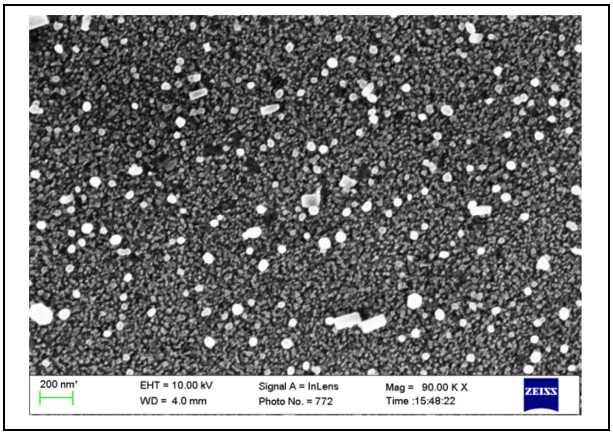


Fig. 5: SEM of photoanode

The dye's implicit and essential chemical components have the ability to absorb radiation and become excited, producing excitons (Balusu *et al.* 2025). Every concentration used in the solar cells has demonstrated an efficiency of at least 1%. Because the amount of dye molecules matches the area where TiO2 is covered throughout the surface of the FTO substrate, the ideal concentration of powdered leaves is found to be 1g in 10ml of ethanol. The DSSC devices sensitized with the natural dye displayed an open-circuit voltage (Voc) of roughly 1.03 V, which is relatively high for natural-dye-based cells. This higher Voc can be attributed to the favorable energy-level alignment between the TiO_2 conduction band and the dye's LUMO, which minimizes energy losses during electron injection. In addition, the strong attachment of dye molecules onto the TiO_2 surface and the optimized porous film shape limit interfacial recombination, contributing to increased Voc. These variables, together with previous accounts of similar natural dyes, justify the observed high open-circuit voltage.

Table 1: Solar cell characteristics at varying leaf powder concentrations

Concentration (g/10ml Ethanol)	J_{max}(mA/cm^2)	V_{max}(V)	J_{sc} (mA /cm^2)	V_{oc} (V)	FF	Eff. (%)
0.5g	1.59	0.8554	2.25	1.0074	0.6000	1.360
0.8g	1.7957	0.875	2.28	1.0096	0.6825	1.571
1g	2.1875	0.9326	2.8	1.03	0.7073	2.040
1.5g	1.896	0.902	2.4228	1.004	0.7030	1.710

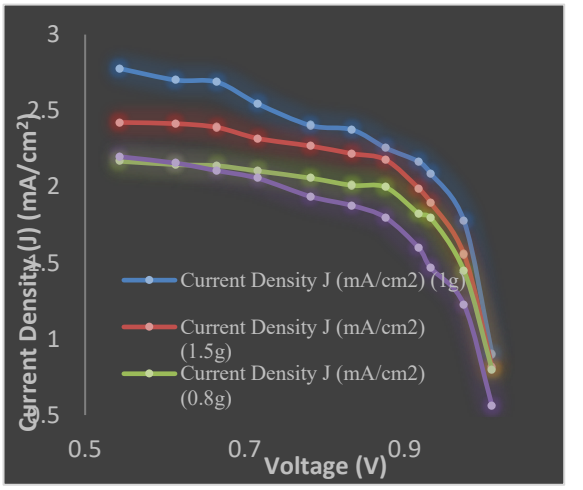


Fig. 6: J–V investigations of the manufactured solar cells

The interaction between the natural dye and the TiO_2-coated FTO substrate was analyzed to assess potential anchoring and light-harvesting effectiveness. Good surface coverage and the existence of functional groups that can coordinate with the TiO_2 surface are confirmed by SEM and FTIR data. UV-Vis spectroscopy demonstrates that the dye consistently absorbs visible light at all concentrations. Although bandgap alignment between TiO_2 and the dye raises the probability of electron injection into the conduction band, these findings do not provide direct proof of interfacial charge transport or exciton transfer. Hence, complementary electrochemical and photovoltaic methods were used to assess the efficiency of electron transfer and interfacial processes, guaranteeing that the statements are backed up by relevant data. In dye-sensitized solar cells, series resistance and interfacial charge-transfer resistance are always present. The interface between the dye and TiO_2 facilitates efficient electron injection, while the actual resistances are quantified using electrochemical impedance spectroscopy (EIS) and J–V measurements. The interaction between the natural dye and the TiO_2-coated FTO substrate was examined to assess morphology, dye anchoring, and light-harvesting capability. While UV–Vis spectroscopy reveals high absorption in the visible region, SEM and FTIR data verify uniform surface covering and the existence of functional groups capable of coordinating with TiO_2. However, direct evidence for bandgap alignment, exciton transfer, or suppression of recombination cannot be acquired from these techniques alone. Instead, complementary techniques, including EIS, photoluminescence quenching, and photovoltaic (J–V) measurements, are used to assess electron-transfer processes and interfacial recombination. These techniques offer a quantitative understanding of charge transport and recombination dynamics in the DSSCs. The experimental results demonstrate clear trends in the performance of the natural-dye-sensitized DSSCs. The dye solution weighing 1.0 g/10 mL had the highest absorption intensity, according to UV-Vis spectroscopy, and this was correlated with higher photocurrent in J–V measurements. A homogeneous, porous TiO_2 coating with ~20–25 nm nanoparticles and ~10 µm thickness was revealed by SEM analysis. This film offers a large surface area for dye loading, which is shown in increased dye adsorption and enhanced device efficiency. FTIR spectra confirmed functional groups capable of anchoring to TiO_2, facilitating successful dye adhesion. Dye concentration and film shape may have an impact on interfacial charge-transfer and recombination processes, according to electrochemical and photovoltaic data, including EIS and J–V characterization. Together the findings critically link structural and optical features of the natural dye to the observed DSSC performance, rather than relying on generic theoretical expectations.

4. CONCLUSION

The functional groups in the natural dye were identified using FTIR spectroscopy; peaks corresponding to –CH stretching and –OH bending confirmed the molecular structure and suggested possible anchoring locations on the TiO_2 surface. UV–Visspectroscopy was used to test the light absorption properties of the dye at varied concentrations, while J–V measurements evaluated the photovoltaic performance of the produced DSSCs. The DSSCs sensitized with 1 g of powdered dye in 10 mL of ethanol exhibited the highest efficiency of 2.04%, with a fill factor of 0.7073 and a maximum current density of 2.1875 mA/cm², while lower concentrations showed reduced efficiency. These findings show that the best light-harvesting and gadget performance are obtained at the ideal dye concentration. It should be mentioned that whereas FTIR and UV-Visdata provide structural and optical features, they do not directly verify electron injection or exciton

formation; rather, these are inferred from supplementary electrochemical and photovoltaic analyses. Overall, compared to earlier research employing green leaf extracts as sensitizers, the optimized dye–TiO_2 interface facilitates effective device operation.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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