

Article

Exploring *Moringa oleifera* as a Sustainable Chlorophyll Source for Dye-Sensitized Solar Cells (DSSCs)

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Abstract

Chlorophyll, a natural photosynthetic pigment, is gaining interest for its sustainable and eco-friendly applications in renewable energy, particularly as a photosensitizer in dye-sensitized solar cells (DSSCs). This study investigates the feasibility of chlorophyll extracted from *Moringa oleifera* as a natural photosensitizer in DSSCs, building on our previous work demonstrating its high chlorophyll content and long-term stability. Chlorophyll was extracted using acetone under optimal conditions (45 °C, 60 min) and applied in DSSCs comprising a TiO₂ photoanode, iodide/triiodide electrolyte, and platinum counter electrode. The TiO₂ photoanode was characterised using UV-Vis spectroscopy, FE-SEM, XRD, and Raman spectroscopy, confirming the presence of pure anatase phase TiO₂ with uniform spherical nanoparticle morphology. The fabricated DSSCs achieved a short-circuit current density of 0.197 mA cm⁻², an open-circuit voltage of 0.44 V, a fill factor of 32%, and a photoconversion efficiency (PCE) of 0.027%. While this performance is lower than the highest reported chlorophyll-based DSSC efficiency (4.6%), the results demonstrate that *M. oleifera* is a viable and sustainable source of chlorophyll for DSSC applications. The findings highlight the importance of dye-semiconductor interactions and suggest that further optimisation through co-sensitization, TiO₂ surface modification, and improved dye anchoring could enhance device performance.

Keywords: biomass; chlorophyll; moringa; dye-sensitized solar cell (DSSCs); photo-conversion efficiency (PCE)



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1. Introduction

Dye-sensitized solar cells (DSSCs), a third-generation photovoltaic technology, offer a cost-effective and environmentally friendly alternative to silicon-based solar cells [1–3]. Unlike traditional solar cells, DSSCs rely on a photosensitizer dye to harvest sunlight and facilitate electron transfer, converting solar energy into electricity [4]. Over the years, DSSCs have evolved, inspired by the natural process of photosynthesis. This inspiration is not only metaphorical but also functional, as DSSCs mimic the way nature traps light and converts it into energy [2]. These cells perform well under low-light and diffuse-light conditions, making them highly versatile for diverse environments. Their appeal lies in

several attributes: cost-effectiveness, eco-friendliness, compactness, lightweight, and ease of fabrication, all of which have garnered significant attention from researchers [1–3].

The operational principle of DSSCs is elegantly simple yet technically intricate, involving five key components [2,5–7]. The transparent electrode, typically fluorine-doped tin oxide (FTO) on a glass substrate, ensures improved conductivity and minimal energy loss. The photoanode, usually titanium dioxide (TiO_2), features a favorable high bandgap of 3.2 eV and excellent dye adsorption properties [8]. The heart of the DSSC, the dye sensitizer, is responsible for photon absorption, a critical step in converting light to electrical energy [3,9]. An iodide/triiodide electrolyte facilitates charge transport, while the counter electrode, often platinum, catalyzes the reduction of the electrolyte, completing the circuit and ensuring electron flow. Ideal counter electrode materials should combine chemical stability, high conductivity, and low resistance with corrosion resistance [2].

The efficiency of DSSCs depends heavily on the properties of the sensitizer dye. Synthetic dyes, particularly ruthenium-based complexes, have historically achieved the highest power conversion efficiencies (PCEs), exceeding 10% under laboratory conditions [10–12]. However, their widespread use is limited by challenges such as high costs, environmental toxicity, and reliance on rare metals [3,13]. Innovations in synthetic dyes have pushed PCEs even higher, with Ren et al. [14] achieving record efficiencies of 28.4% to 30.2% through advanced co-sensitization techniques and novel dye structures.

Despite these advances, the limitations of synthetic dyes have spurred interest in natural dyes as renewable, cost-effective, and non-toxic alternatives [2,3]. Natural dyes offer unique functional groups that enable stable binding to TiO_2 surfaces, facilitating efficient electron injection [3]. Chlorophyll, the primary pigment responsible for photosynthesis in plants, has shown promise as a DSSC sensitizer due to its strong light absorption in the visible spectrum [2,7]. Chlorophyll, beyond its role in plant coloration, functions as a natural solar panel, capturing solar energy and converting it into chemical energy. This unique property positions it as an organic semiconductor [15].

Numerous studies exploring natural dyes have demonstrated promising but variable success rate in DSSC applications. For example, chlorophyll extracted from spinach leaves achieved a PCE of 0.49% using distilled water as a solvent, outperforming ethanol-based extraction, which yielded 0.36% [16]. Chlorophyll from seaweed (*Sargassum* sp.), extracted with acetone, achieved a higher PCE of 1.50% [17]. These findings highlight the crucial role of solvent selection, as it influences not only the polarity-dependent optical properties of chlorophyll, but also its stability, aggregation behaviour, and interaction with the semiconductor surface, all of which ultimately affect DSSC performance.

In addition to studying individual plant sources, researchers have explored co-sensitization strategies to enhance DSSC performance. Shaikh et al. [18] blended chlorophyll with anthocyanins and betacyanins from blackberries and beetroot, achieving a PCE of 0.221%. Similarly, Srivastava et al. [19] evaluated *Codiaeum variegatum* leaves and *Delonix regia* flowers, obtaining PCEs of 0.12% and 0.17%, respectively, while their mixture yielded a higher PCE of 0.24%. This approach aims to broaden the absorption spectrum and enhance solar cell efficiency.

Although natural dyes are abundant and environmentally friendly, they face limitations in photoconversion efficiency, stability, and compatibility with DSSC components [2]. These co-sensitization strategies highlight the potential for complementary absorption spectra to address the inherent limitations of natural dyes, such as low photoconversion efficiency. For instance, Kakiage et al. [20] achieved a PCE of 14.34% by combining multiple dyes to leverage their complementary absorption spectra.

Among natural dye sources, *Moringa oleifera* stands out as a particularly promising plant for DSSCs. Commonly known as the “Miracle Tree” or “Horseradish Tree,” *M.*

oleifera is renowned for its exceptional growth rate, large biomass, and ability to thrive in drought-prone regions [21–23]. A single tree can reach heights of 4 m in just 6 months to a year and 6 to 15 m at maturity, with leaves yielding approximately 4.2 to 8.3 tons per hectare every 40 days [22,23]. These characteristics position *M. oleifera* as a scalable and sustainable resource for renewable energy applications, particularly in regions with limited water resources.

The current study builds on the author's previous research that assessed the chlorophyll content, degradation, and stability of *M. oleifera* under various postharvest storage conditions. Remarkably, even after two years of storage, *M. oleifera* leaves retained chlorophyll levels surpassing those of many freshly harvested plants [23]. This high chlorophyll stability, along with its compatibility with widely used plants in the food and pharmaceutical sectors, underscores its versatility and industrial potential. This stability and longevity, coupled with its high chlorophyll content, underline *M. oleifera*'s unique potential as a source of chlorophyll for industrial and renewable energy applications.

To the best of our knowledge, this is one of the first studies to explore *M. oleifera*'s chlorophyll as a sensitizer in DSSC applications, addressing critical gaps in both the solar energy and natural dye research domain. This study seeks to address this gap, evaluating its application in DSSCs while leveraging its scalability, stability, and sustainability. By optimizing chlorophyll extraction processes and assessing its photovoltaic performance, the study aims to contribute to the ongoing pursuit of efficient, green, and sustainable solar energy solutions.

2. Materials and Methods

2.1. Materials

This study utilized *Moringa oleifera* leaves, following the method outlined by Ngcobo et al. [23]. The leaves were harvested from Hammanskraal, Gauteng Province, South Africa (25.4132° S, 28.2578° E), during the summer month of January. Mature leaves were collected from 15 randomly selected trees to ensure a representative sample. After harvesting, the leaves were washed to remove any dirt or debris and air-dried at 45 °C for 120 h. This drying process was chosen to preserve the plant material, prevent pigment degradation, and maintain the integrity of the biomass, as supported by Manolopoulou et al. [24]. Figure 1 illustrates the drying setup used in this study.

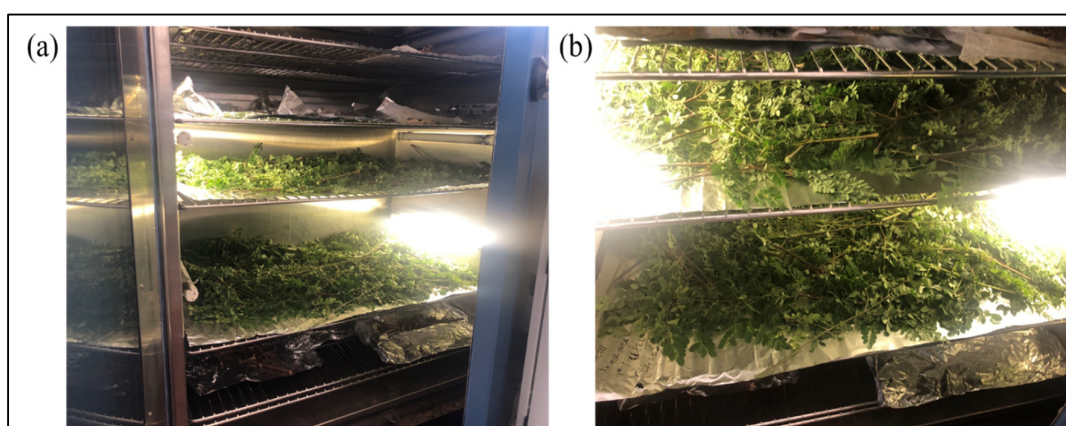


Figure 1. *Moringa oleifera* leaves air-drying process, showing (a) front view and (b) internal view of the drying cabinet at 45 °C, illustrating the multi-shelf drying setup used in this study.

Once dried, the leaves were wrapped in aluminum foil, placed in black polythene bags, and stored in dark conditions at room temperature until needed. To prepare for experiments, the dried leaves were ground into a fine powder using a Naiwang mini

bean electric grinder (Naiwang., YongKang, China), as shown in Figure 2. The particle size distribution of the powdered samples was measured using a Malvern Mastersizer 2000 (Malvern Panalytical Ltd., Malvern, UK), with 80% of the particles passing $<240\ \mu\text{m}$. For the extraction process, high-purity ($\geq 99.9\%$) acetone was used. All chemicals were procured from Sigma-Aldrich (Merck (Pty) Ltd., Modderfontein, South Africa). Acetone was selected as the extraction solvent based on our previous study [23], which demonstrated that it achieved the highest chlorophyll extraction efficiency among the solvents evaluated (ethanol, methanol, and DMF). While ethanol represents a greener alternative, acetone was preferred due to its superior extraction performance and its established use in chlorophyll-based DSSC research [16,17]. DMF, despite its high efficiency, was excluded due to its environmental toxicity [25].



Figure 2. *Moringa oleifera* leaves processed into a fine powder using a Naiwang mini bean electric grinder.

For the DSSC fabrication, the extracted chlorophyll served as the photosensitizer. A range of high-quality reagents was procured from Sigma-Aldrich (South Africa), including ethanol ($\geq 99.9\%$), methanol ($\geq 99.9\%$), mesoporous titania (TiO_2 paste, active opaque), iodine ($\geq 99.8\%$), potassium iodide ($\geq 99\%$), FTO glass substrates, ethylene glycol ($\geq 99\%$), and platinum paste (Greatcell Solar PT1, Greatcell Solar Materials Pty Ltd., Queanbeyan, Australia). The FTO glass substrates, with a sheet resistance of $7\ \text{ohm}/\text{cm}^2$, were specifically chosen for their high conductivity and low energy loss, ensuring efficient electron transfer. All chemicals were used without further purification, as received from Sigma-Aldrich (Merck (Pty) Ltd., Modderfontein, South Africa).

2.2. Extraction Method

The chlorophyll extraction process began after the pulverization of the *Moringa oleifera* leaf material. A precise 0.5 g of powdered *Moringa oleifera* leaves was mixed with 50 mL of acetone. This mixture was stirred for five minutes using a magnetic stirrer to ensure the leaf powder was evenly dispersed. The extraction process was performed in a water bath maintained at a constant temperature of $45\ ^\circ\text{C}$, with the duration standardized at 60 min to identify optimal conditions for chlorophyll yield. Following the extraction period, the solution was centrifuged at 2000 rpm for five minutes to separate the extracted chlorophyll from residual leaf material. The supernatant, containing the extracted chlorophyll, was carefully collected for subsequent analysis. Figure 3 provides a visual representation of the chlorophyll solution post-centrifugation, where the sandy residue at the bottom of the centrifuge tube indicates the leftover plant material. This distinct separation highlights the success of the extraction process.



Figure 3. Extracted chlorophyll post-centrifugation (the letters represent specific parts of the labelling sequence used to identify the photoelectrodes).

2.3. Preparation of the Fluorine-Doped Tin-Oxide Glass

The preparation of FTO glass substrates for DSSC fabrication began with the sectioning and etching of glass slides coated with a fluorine-doped tin-oxide conductive layer. Each slide was cut into $20 \times 20 \text{ mm}^2$ pieces using a precision cutter on the non-conductive side to maintain the integrity of the conductive coating.

Post-sectioning, the substrates underwent a multi-step cleaning process to remove contaminants and ensure optimal adhesion of the photoanode material. The substrates were sequentially immersed in 250 mL of deionized water, ethanol, and acetone, with each solution subjected to 10 min of ultrasonication at 25°C . This process leveraged cavitation effects generated by ultrasonic waves to effectively dislodge surface impurities.

Following ultrasonication, the substrates were dried in a laboratory oven at 45°C for three hours to eliminate residual moisture and volatiles. The conductive side of the FTO glass was further treated with ethanol to enhance surface cleanliness. The final step involved measuring the sheet resistance of the conductive layer using a multimeter to ensure uniformity and reliability of the electrical properties of the substrates. The methodology for substrate preparation was based on the procedure outlined by Sabarikirishwaran et al. [26], with modifications tailored to the specific requirements of this study.

2.4. Preparation of the Electrodes

The photoanode was prepared using the doctor blade method (tape casting). The process began with securing the FTO glass substrate to the laboratory counter using adhesive tape, leaving the conductive side exposed. A layer of mesoporous titania paste was then evenly applied onto the conductive surface. Following the application, the coated substrate was air-dried for five minutes to ensure uniform adhesion of the paste. Sintering of the coated substrate was performed in a furnace with a gradual temperature increase, applying a heating ramp as per the protocol described by Agrawal et al. [27], reaching 470°C and maintained for 30 min. This step facilitated the formation of a robust mesoporous titania layer with effective electron transport properties. Based on the use of a single masking tape layer ($\sim 50\text{--}60 \mu\text{m}$) as the spacer and sintering at 470°C , the TiO_2 film thickness is estimated to be in the range of $5\text{--}10 \mu\text{m}$, which is consistent with values reported in the literature for comparable doctor blade-deposited TiO_2 films [28,29]. After sintering, the substrates were gradually cooled to room temperature inside the furnace.

The counter electrode was prepared using a similar procedure. Platinum paste was applied to the conductive side of a separate FTO glass substrate. This assembly was sintered at 470 °C for 15 min, as per the methodology described by Shah et al. [2]. This shorter sintering duration ensured optimal conductivity and adhesion of the platinum layer without compromising its structural integrity. Figure 4 provides a visual representation of the various stages in the preparation of the photoanode. It illustrates the coating process of the mesoporous titania paste, the sintering process, and the appearance of the coated mesoporous titania after sintering.

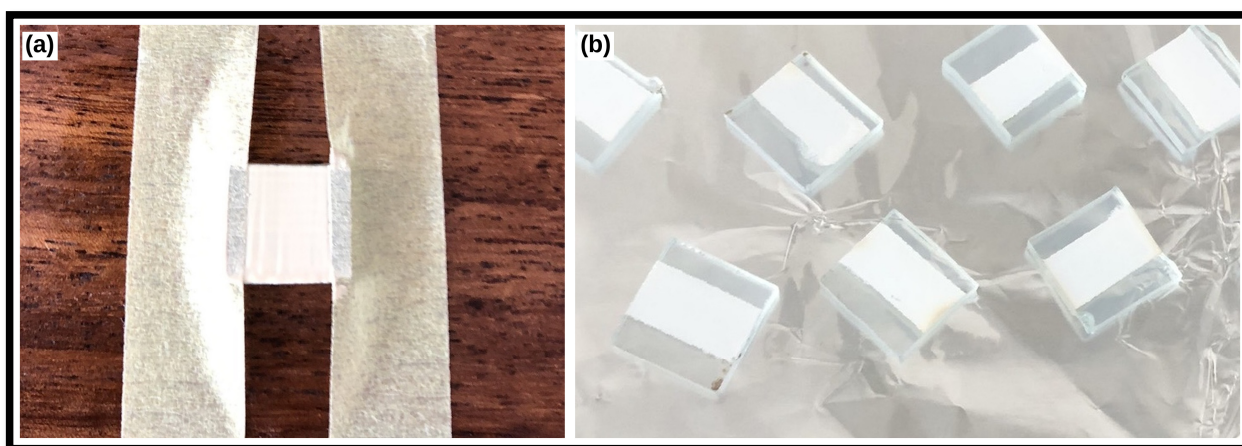


Figure 4. Various stages in the preparation of the photoanode. (a) Coating of the mesoporous titania paste onto the FTO glass substrate using the doctor blade method; (b) appearance of the coated mesoporous titania photoanode after sintering and cooling to room temperature.

2.5. Dye Loading and Electrolyte Preparation

The dye loading process was performed by immersing the sintered TiO₂ photoanodes in 10 mL of the extracted chlorophyll dye solvent for 24 h. The total chlorophyll concentration of the acetone extract was 63 ± 0.524 mg/L, as established in our previous study [23]. This ensured optimal absorption of the dye onto the mesoporous TiO₂ layer. To prevent degradation of the chlorophyll dye, the photoanodes and dye solution were stored in light-proof containers wrapped in aluminum foil and kept in darkness throughout the incubation period. Following the 24-h immersion, the dye-sensitized photoelectrodes were gently rinsed with ethanol and air-dried. This cleaning process was essential to remove any unanchored dye molecules, ensuring efficient sensitization and clean surface interactions for the photoanodes.

The liquid electrolyte was prepared by dissolving 0.05 M iodine and 0.5 M potassium iodide in acetonitrile. The resulting solution was then mixed with ethylene glycol at a ratio of 1:4, following the procedure described by Sabarikirishwaran et al. [26], to form a homogeneous iodide/triiodide electrolyte. The mixture was stirred using a magnetic stirrer until completely dissolved. This process forms a homogeneous iodide/triiodide electrolyte, essential for the operation of the DSSC. Figure 5 provides a visual representation of the dye-sensitised photoelectrodes and the synthesised electrolyte.

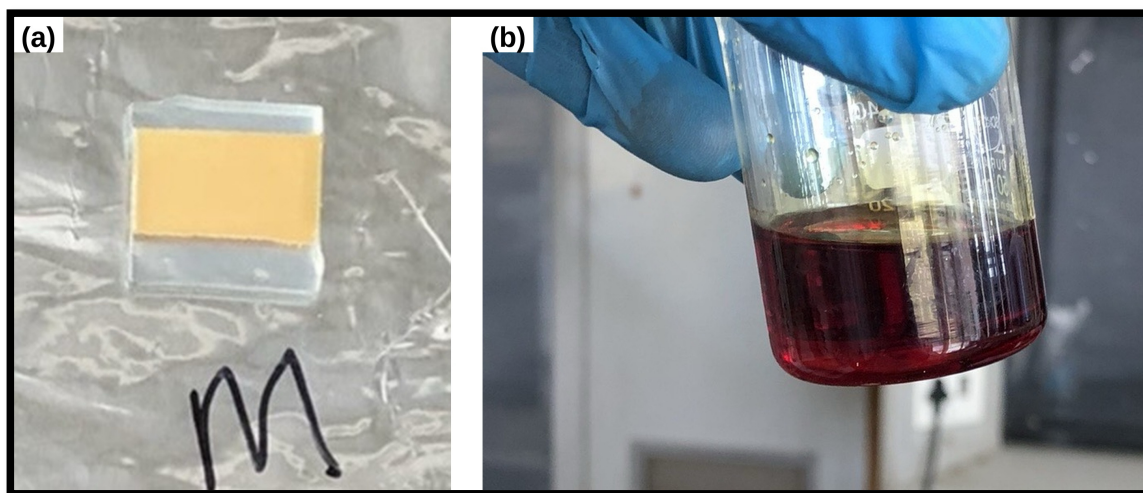


Figure 5. Visual representation of the fabricated DSSC components. (a) Dye-sensitized TiO_2 photo-electrodes after 24-h immersion in *M. oleifera* acetone-extracted chlorophyll dye solution, showing the characteristic amber coloration indicative of successful dye adsorption onto the TiO_2 surface. (b) Synthesized iodide/triiodide liquid electrolyte prepared by dissolving 0.05 M iodine and 0.5 M potassium iodide in acetonitrile, mixed with ethylene glycol at a ratio of 1:4.

2.6. Dye-Sensitized Solar Cell Assembly

The assembly of the DSSC involved carefully positioning the TiO_2 -coated anode facing the platinum-coated cathode. The space between these two electrodes was then filled with the prepared electrolyte mixture, ensuring thorough coverage for optimal device performance. The space between the electrodes remained constant throughout the experiments.

To prevent any leakage of the electrolyte and to maintain the structural integrity of the DSSC, the two electrodes were securely clipped together and sealed using Croxley bulldog clips. This assembly resulted in a device with an active area of 208 mm^2 (2.08 cm^2), defined by the masking tape boundary applied during TiO_2 deposition. Figure 6 provides a visual depiction of the fully assembled DSSC, showcasing the complete setup ready for performance testing.

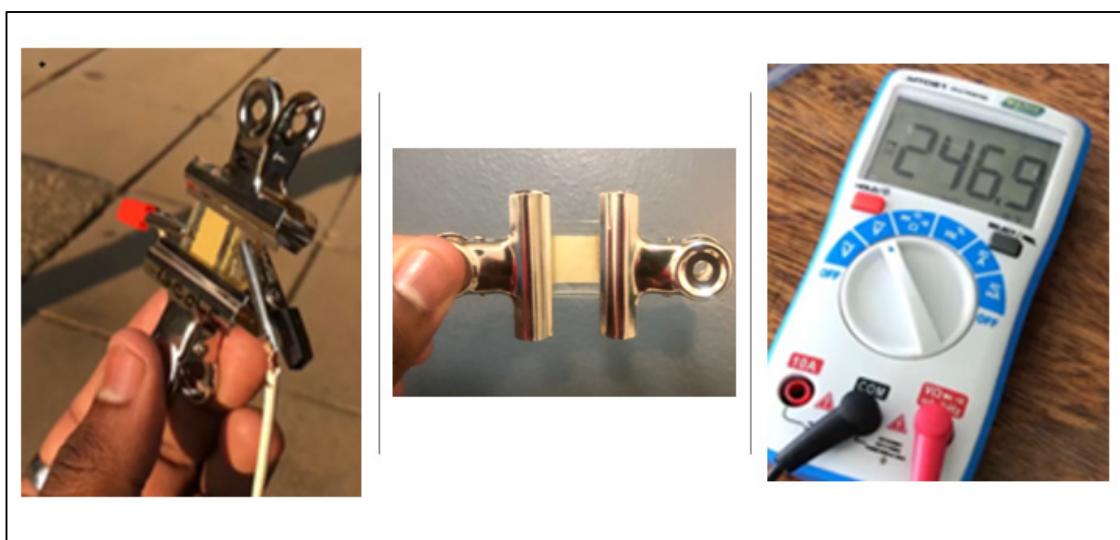


Figure 6. Fully assembled dye-sensitized solar cell device.

2.7. Device Characterization

To determine the bandgap and absorption analysis of the natural dye, the absorption spectra was measured using a UV-Vis spectrophotometer (Genesys 10S UV-Vis, Thermo Scientific, Waltham, MA, USA). The data were then analysed using SHIMADZU UVProbe version 2.62 software. This step was crucial for determining the light absorption capabilities of the dye across different wavelengths.

The structural properties of TiO₂, a critical component of the DSSC, were analysed using a Bruker-D2 PHASER X-ray diffractometer (Bruker Corporation, Billerica, MA, USA) with Cu K α radiation ($\lambda = 1.789010 \text{ \AA}$). The measurements were conducted over the 2θ range of 10° to 89.996° with a step size of 0.026° . Data analysis was performed using the Match!3 software Crystal Impact, Bonn, Germany). This analysis provided insights into the crystalline structure of the TiO₂ used in the photoanode. Field emission scanning electron microscopy (FE-SEM, JEOL JSM-7610F Plus, JEOL Ltd., Tokyo, Japan) was used to investigate the morphology of the materials. Samples were mounted on aluminium stubs using carbon tape and imaged at an accelerating voltage of 3.0 kV, with a working distance of 3.0 mm, using an InLens detector at a magnification of $100,000\times$ and an aperture size of $30 \mu\text{m}$. Energy-dispersive X-ray spectroscopy (EDX) was performed simultaneously to confirm elemental composition. Raman spectroscopy was performed at room temperature with an excitation wavelength of 532 nm to identify the molecular and crystallographic structure of the TiO₂ used in the DSSC. Finally, the current density-voltage characteristics of the fabricated DSSCs were studied using a standard solar simulator (Oriel-Sol3A Class AAA, Newport Corporation (Oriel Instruments), Irvine, CA, USA) at 1 Sun intensity (AM 1.5G; 100 mW/cm^2) [30] to estimate the DSSCs PCE, a key performance indicator.

2.8. Photochemical Characterization

The DSSCs PV performance was rigorously evaluated using key parameters. These parameters include the I_{sc} , V_{oc} , maximum output power, fill factor, and PCE. The fill factor determines the solar cells maximum power efficiency compared to its theoretical limit. The PCE is another critical metric, which represents the effectiveness of the solar cell in converting absorbed light into electrical energy. Both fill factor and PCE were calculated using Equations (1) and (2), respectively.

$$FF = \frac{P_{max}}{V_{OC} \times I_{SC}} = \frac{V_{max} \times I_{max}}{V_{OC} \times I_{SC}} \quad (1)$$

$$\eta = \frac{V_{OC} \times I_{SC} \times FF}{P_{in}} \quad (2)$$

FF: fill factor.

η : PCE.

P_{max} : maximum power.

I_{max} : maximum current.

V_{max} : maximum voltage.

OC: open circuit.

SC: short circuit.

The PV parameters, as well as the current–voltage curve, were obtained using a solar simulator.

3. Results and Discussion

3.1. Optical Properties Analysis

The optical characteristics of chlorophyll dyes extracted from *Moringa oleifera* leaves were analyzed using UV-Vis spectroscopy. Figure 7 illustrates the absorption spectra of

chlorophyll dyes extracted with acetone, focusing on the visible light range of 400–800 nm. The spectra revealed distinct absorption peaks, confirming the successful extraction of chlorophyll pigments. Prominent absorption bands were observed between 400–500 nm and 600–700 nm, corresponding to the blue and red regions of the spectrum, respectively. These findings align with established absorption characteristics of chlorophyll pigments, particularly chlorophyll *a* and chlorophyll *b* [16].

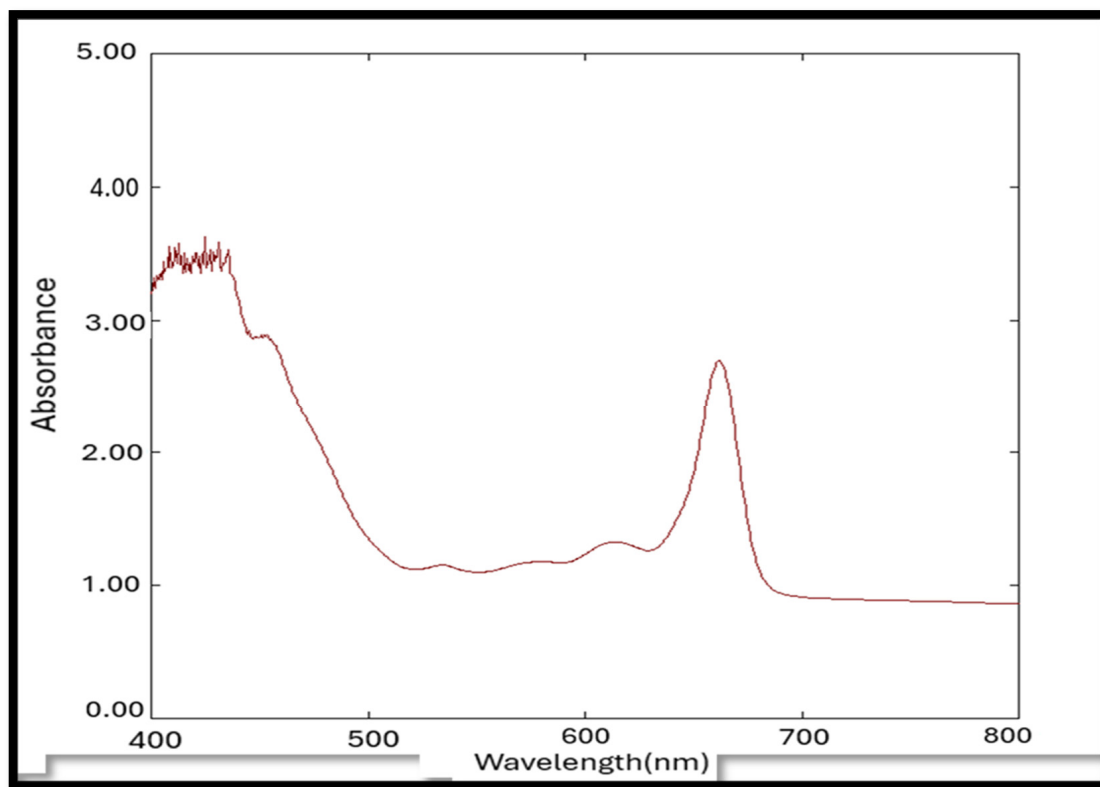


Figure 7. Absorption spectrum of chlorophyll dye extracted from *M. oleifera* leaves using acetone as a solvent.

The *M. oleifera*–acetone dye (Figure 7) exhibited strong absorption peaks at 450 nm and 660 nm. These peaks can be attributed to the chlorophyll pigments present in the extracts, corroborating previous findings that *M. oleifera* is rich in chlorophyll content [16,23]. The spectral distortions observed in Figure 7, particularly between 400 and 450 nm, are attributed to detector saturation resulting from the high chlorophyll concentration in the undiluted acetone extract. At absorbance values exceeding approximately 2.5, the detector response becomes non-linear, leading to the irregular signal observed at shorter wavelengths. The measurements were conducted using a standard 1 cm pathlength cuvette. The strong absorption peaks in the red region (660 to 670 nm) are characteristic Q-bands of chlorophyll. The Q bands originate from $\pi \rightarrow \pi$ transitions within the conjugated macrocycle, as described by the Gouterman model [31], and appear in the red region of the spectrum due to their lower transition energy. This is consistent with the characteristic absorption of chlorophyll *a*, which exhibits a prominent Q_y band around 660–670 nm [12,24,32]. These results confirm that the chlorophyll pigments extracted using acetone maintain their primary absorption properties, consistent with established spectral characteristics reported in the literature [16,23]. The strong absorption at this wavelength is attributed to the conjugated double-bond system in chlorophyll as porphyrin ring, which enables efficient light absorption [33]. These results are consistent with those discussed in our previous publication, where chlorophyll *a* consistently exhibited higher levels with all extraction

solvents than chlorophyll b. This is expected, as chlorophyll a serves as the primary pigment responsible for light absorption and conversion, while chlorophyll b is responsible for transferring photons to chlorophyll a [24,32]. Additionally, the smaller peaks observed in the lower photon wavelength range (below 500 nm) are likely caused by the Soret band. This corresponds to the strong absorption of both chlorophyll a and chlorophyll b in the blue region of the spectrum. Chlorophyll a exhibits prominent absorption in two main regions of the UV-Vis spectrum: the Soret band in the blue region (430 to 450 nm) and the Qy band in the red region (660 to 670 nm) [12].

The strong absorption peaks in the visible region around 420 nm and 660 nm achieved in this study indicate the potential of the extracted chlorophyll as a natural sensitizer in DSSCs [16]. According to Syafinar et al. [16], chlorophyll absorbs light from the red, blue, and violet regions of the spectrum and reflects green light, resulting in the characteristic colour of plants. In DSSCs, the dyes' ability to absorb sunlight and convert it to electrical energy is crucial [17]. The overall performance of a DSSC depends on the dye sensitizers capacity for light absorption and the diffusion of electrons through the mesoporous TiO₂ layer [9,18]. Dyes regulate both the generation and transfer of photo charges, playing a vital role in DSSC functionality [19]. The UV-Vis absorption spectrum depicted in Figure 7 is similar to that obtained by Ridwan et al. [17]. The author evaluated acetone-extracted chlorophyll pigments from *Sargassum* and obtained a peak at 660 to 670 nm and a chlorophyll concentration of 61.176 mg/L. Furthermore, the investigation revealed a PCE of 1.50%. In another study by Syafinar et al. [16], chlorophyll was extracted from spinach using ethanol and distilled water resulting in absorption peaks at 660 nm and 680 nm, respectively. The PCEs achieved were 0.36% with ethanol and 0.49% with distilled water.

An investigation by Srivastava et al. [19] of *Codiaeum variegatum* leaves resulted in maximum absorption at 440 nm and a peak at 660 nm, which confirms the presence of chlorophyll. The DSSC made with the *Codiaeum variegatum* dye achieved a PCE of 0.12%. Sabarikirishwaran et al. [26] further reported that chlorophyll-based natural dyes typically exhibit absorption maxima between 400 to 550 nm and 600 to 750 nm, consistent with the spectral characteristics observed in the present study.

Overall, solvents affect the electronic structure of dye molecules, causing shifts in their absorbance spectra [26]. In addition to spectral shifts, solvent polarity influences the electronic structure of chlorophyll through interactions with its π -electron system, affecting aggregation behaviour and dye-TiO₂ binding. Polar solvents such as methanol and ethanol can alter these interactions, thereby influencing electron injection efficiency and overall DSSC performance. The current study's findings are in line with the literature and confirm the successful extraction of chlorophyll pigment.

3.2. Morphology of the TiO₂ Semiconductor

The morphology of the TiO₂ photoanode plays a critical role in the performance of DSSCs by influencing the radiation-less electron recombination process [26]. Figure 8 presents the surface morphology of TiO₂ nanoparticles deposited onto an FTO substrate. Figure 8a shows the FE-SEM images of the TiO₂ sample. The image reveals a homogeneous distribution of TiO₂ nanoparticles, which are observed as bright, spherical particles uniformly spread across the entire sample. Figure 8b provides a magnified view of the TiO₂ sample, clearly illustrating the spherical shape of the nanoparticles and demonstrating the aggregation of TiO₂ particles. The average particle size was determined to be approximately 105 nm, with a size distribution ranging from approximately 70 to 130 nm (estimated from SEM analysis in Figure 8b), and with minimal cracks or defects observable on the surface. Figure 8c depicts the Energy-dispersive X-ray spectra of the TiO₂ nanoparticles, showing emission peaks corresponding to titanium and oxygen.

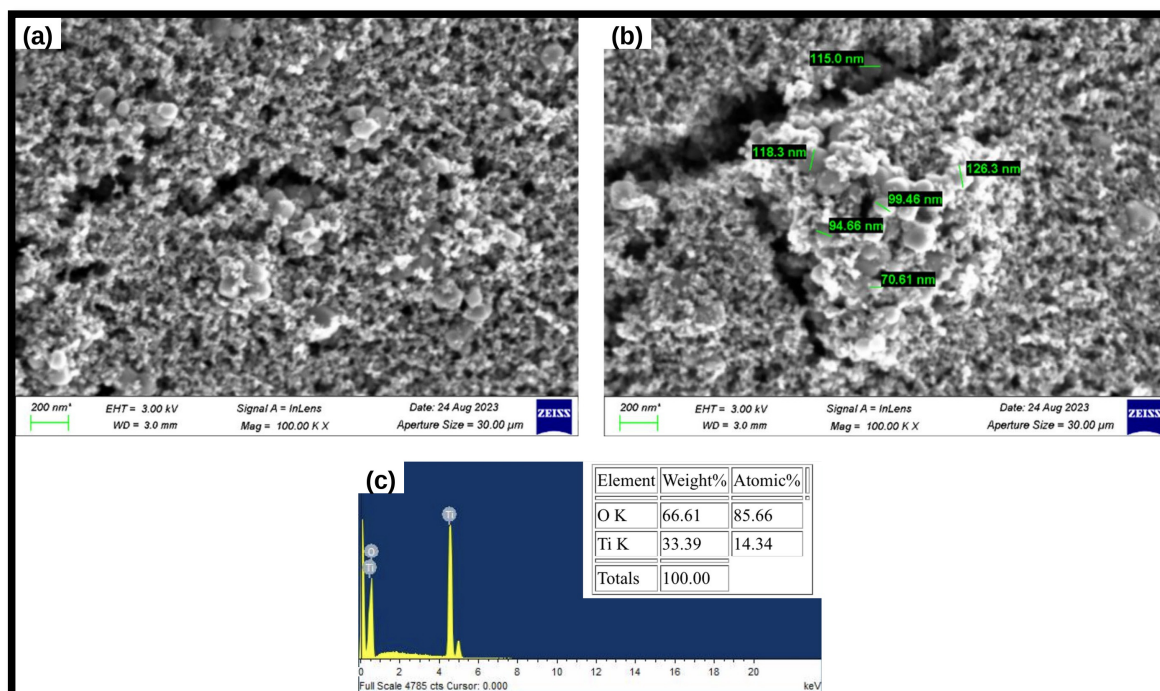


Figure 8. Morphology of titanium dioxide (TiO_2) nanoparticles deposited onto a fluorine-doped tin oxide substrate. (a) Field emission scanning electron microscopy images for TiO_2 , (b) zoomed portion of the TiO_2 sample, and (c) Energy-dispersive X-ray spectra of the TiO_2 nanoparticles. * denotes the scale bar (200 nm).

The results obtained in this study are consistent with those reported in the literature. Sabarikishwaran et al. [26] also observed uniform spherical and semi-spherical TiO_2 nanoparticles coated on an FTO substrate, with an average particle size of 110 nm and some degree of nanoparticle aggregation. This aggregation facilitates the mesoporous morphology essential for dye adsorption [26]. Similarly, Subalakshmi et al. [8] reported quasi-spherical TiO_2 nanoparticles, with energy-dispersive x-ray spectra showing peaks for titanium and oxygen in the same range as observed in Figure 8c. The uniform distribution of the titanium and oxygen elements further confirms the consistency of these results [8]. Srivastava et al. [19] also reported uniform surface coverage of TiO_2 nanorods on the FTO substrate, comparable to the observations in Figure 8a.

Agrawal et al. [27] compared the surface morphology of TiO_2 deposited on FTO substrates using different techniques, including the doctor blade method (as used in the current study) and screen printing. The results showed that both methods produced spherical TiO_2 nanoparticles with similar structures and minimal cracks. The authors attribute these cracks to bubble formation during the doctor blade method. They also noted that porosity can help minimise cracking by accommodating strains in the film during the annealing and sintering processes [27]. The findings of the current study confirm that TiO_2 was successfully coated onto the FTO substrate, optimising it for use in DSSCs, and align well with the observations reported in the literature [34,35].

3.3. Phase and Structural Analysis: X-Ray Powder Diffraction

X-ray powder diffraction (XRD) spectroscopy was employed to investigate the crystalline phase and crystallite size of the TiO_2 sample prepared and coated onto an FTO substrate. Figure 9 displays the XRD patterns of the TiO_2 nanoparticles prepared using the doctor blade method. The crystalline phases formation was examined by fitting the data obtained using Match! Software (v4.2), which identified the peaks corresponding

to the (COD 2300113) planes. This indicates a tetrahedral nanocrystal structure of the anatase TiO_2 phase [34]. This phase is known to have a high surface area, which allows for greater dye adsorption, light absorption, and efficiency of the DCCS [36]. In addition, the phase is also responsible for the chemical stability, leading to the longevity of the DSSCs [37]. The observed data and peaks are consistent with the Joint Committee on Powder Diffraction Standards data (PDF No: 21-1272) [38] and align well with previously reported studies [8,19,35,36], all of which confirm the presence of crystalline anatase TiO_2 .

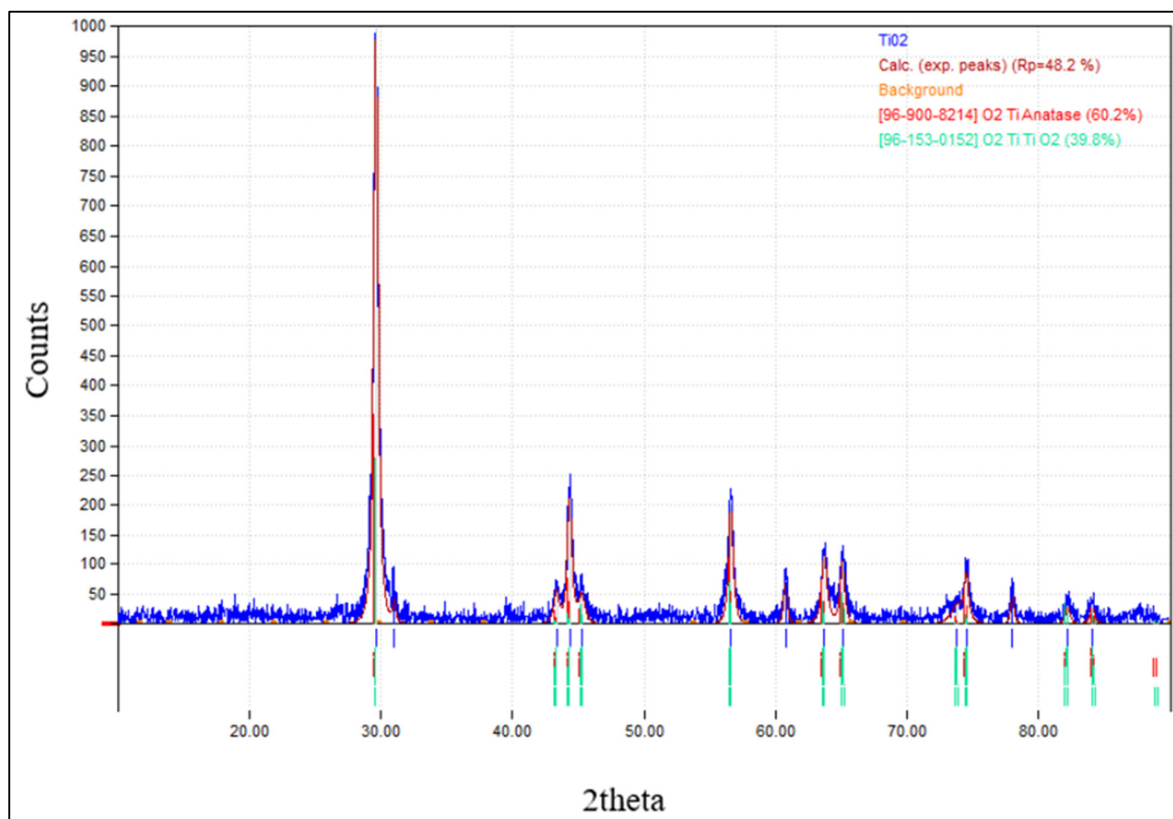


Figure 9. X-ray powder diffraction patterns of the titanium dioxide (TiO_2) nanoparticles prepared by using the doctor blade method.

The sharp and intense peaks observed in the XRD patterns indicate that the synthesised TiO_2 nanoparticles are highly crystalline [8,34]. Each crystalline structure imparts specific physical properties, such as band gap and surface morphology. Among the different phases of TiO_2 , including rutile and brookite, anatase is recognised for its superior charge transport capacity and lower electron-hole recombination rate, making it particularly effective for catalytic and photocatalytic applications [35]. These properties make anatase a preferred phase for PV applications. Notably, other TiO_2 phases, such as rutile and brookite, were absent in the XRD patterns shown in Figure 9, confirming the presence of the pure anatase phase [39]. The absence of impurity peaks further suggests that the TiO_2 phase is composed of pure titanium and oxygen, as indicated by spectroscopy tests [34]. This purity is crucial, as even minor impurities can significantly reduce the efficiency of solar cells [19]. Overall, the experimentally obtained XRD pattern corroborate the successful coating of TiO_2 on the FTO substrate.

3.4. Raman Spectroscopy

Raman spectroscopy was conducted at room temperature with an excitation wavelength of 532 nm to analyse the molecular and crystallographic structure of the TiO_2 used in the DSSC. Figure 10 presents the Raman spectroscopy vibration modes for pure TiO_2

within the range of 96 to 950 cm^{-1} . According to Ohsaka et al. [40], TiO_2 anatase exhibits six active Raman vibration modes ($\Gamma = \text{A}_{1g} + 2\text{B}_{1g} + 3\text{E}_g$). In the current study, Raman peaks were detected at 150, 399, 520, and 629 cm^{-1} , corresponding to the E_g , B_{1g} , A_{1g} , and E_g modes, respectively. The E_g mode represents the symmetric stretching vibration of O-Ti-O in TiO_2 , while the A_{1g} and B_{1g} peaks represent symmetric and anti-symmetric bending vibrations [8].

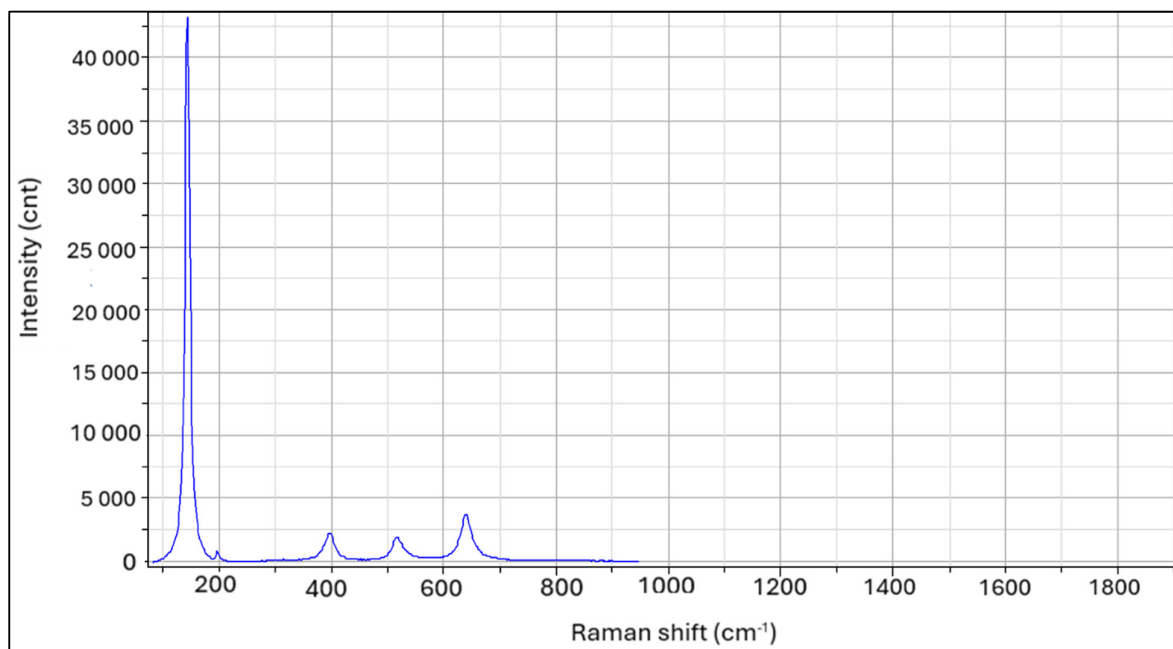


Figure 10. Raman spectroscopy vibration modes for pure titanium dioxide at the range from 96 to 950 cm^{-1} .

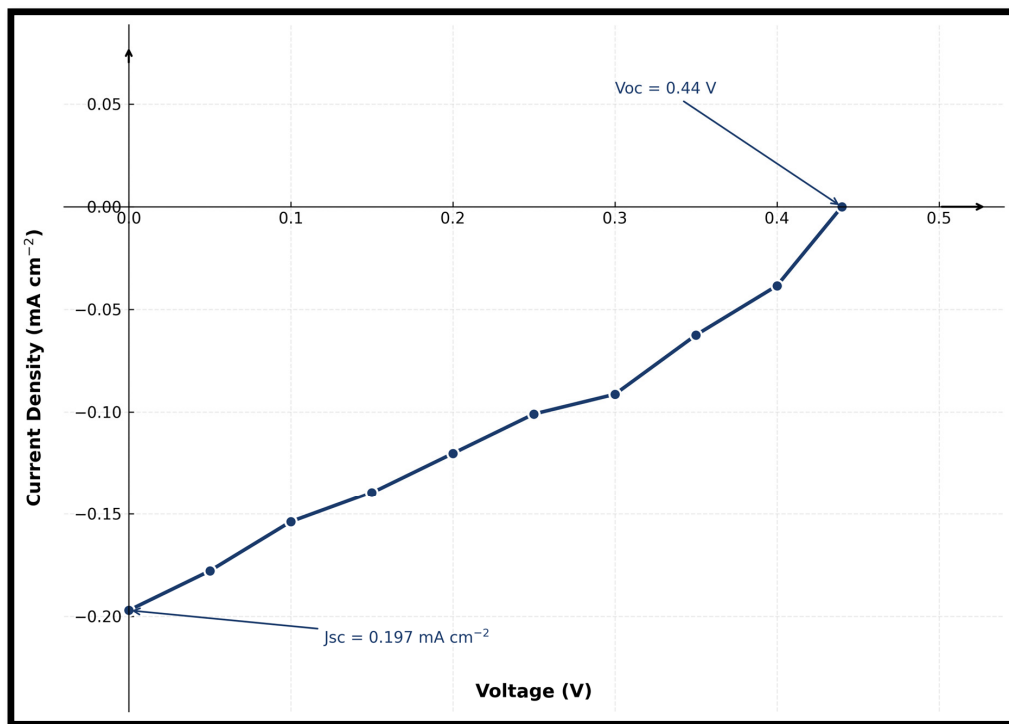
Mirzayev et al. [41] studied the effects of neutron irradiation at different fluences on nanosized anatase TiO_2 and reported that pure anatase showed vibration modes at 147, 398, 520 (combined B_{1g} and A_{1g} modes), and 624 cm^{-1} . The peak positions obtained in the current study closely match those reported by Mirzayev et al. [41], confirming the identification of anatase TiO_2 (Figure 10). Additionally, no active mode corresponding to the rutile phase appears in the Raman spectra (Figure 10), which aligns with the XRD results obtained in this study (Figure 9). The prominent peak observed at 150 cm^{-1} is indicative of the anatase phase formation [42]. This peak is also sensitive to the concentration of oxygen vacancies in TiO_2 , a critical factor influencing its properties [43]. The Raman spectra results in Figure 10 are consistent with those obtained in various studies on pure anatase TiO_2 , further affirming the samples' phase purity [8,40–42,44].

3.5. Power Conversion Efficiency

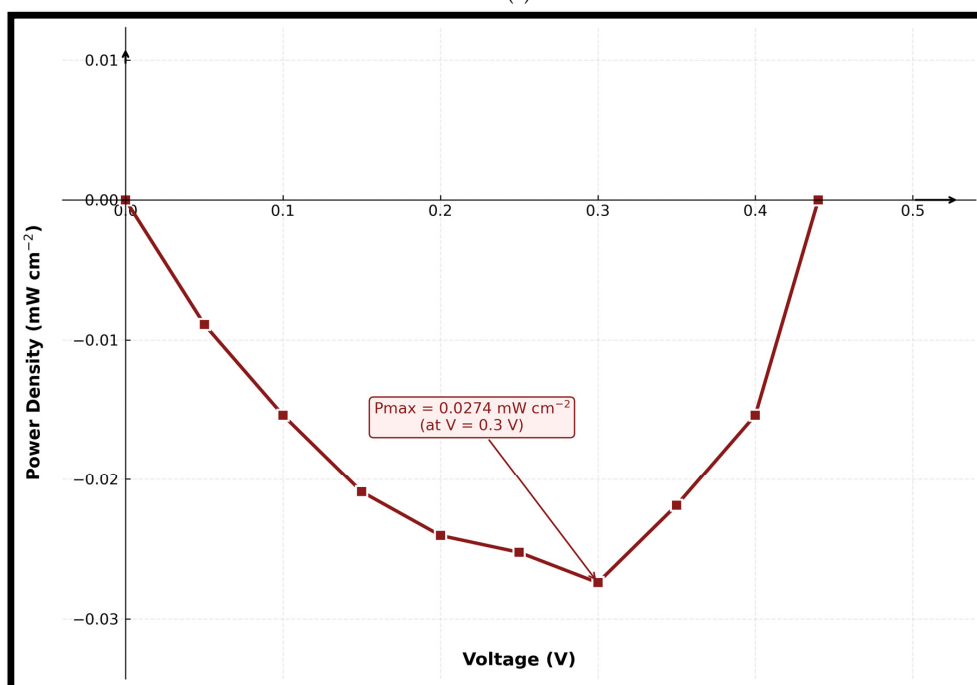
The PV performance of DSSCs using *M. oleifera* chlorophyll dyes was evaluated with a solar simulator. Cells with an active area of 2.08 cm^2 were placed under AM 1.5 illumination with a standard incident light power density of 0.1 W cm^{-2} . The incident power was calculated as 0.208 W. Figure 11a,b, along with Table 1, illustrate the PV performance of the prepared *M. oleifera* chlorophyll–acetone photosensitizer DSSCs.

The short-circuit current density and open-circuit voltage were found to be 0.197 mA cm^{-2} and 0.44 V, respectively (Figure 11a). The maximum power density was recorded as 0.0274 mW cm^{-2} (Figure 11b). The fill factor, a parameter indicating the quality of the solar cell, defined as the ratio of maximum power output to the product of V_{oc} and I_{sc} , was calculated to be 32%. The PCE was 0.027%. These results are summarised in Table 1.

The findings are consistent with similar studies reported in the literature for organic sensitisers [45,46]. To further contextualise these results, Table 2 presents a comparative summary of photovoltaic parameters reported for comparable chlorophyll-based DSSCs from the literature, highlighting the performance of *M. oleifera* chlorophyll relative to other natural dye sources.



(a)



(b)

Figure 11. (a) Current density vs. voltage (J–V) curve for the *M. oleifera* acetone-extracted chlorophyll dye-sensitized solar cell. (b) Power density vs. voltage (P–V) curve for the *M. oleifera* acetone-extracted chlorophyll dye-sensitized solar cell.

Table 1. Summary of photovoltaic parameter results.

Dye	Area (cm ²)	Pin (W/cm ²)	Voc (V)	Vmax (V)	Jsc (mA/cm ²)	Jscmax (mA/cm ²)	FF (%)	PCE (%)
<i>Moringa oleifera</i> —Acetone	2.08	0.1	0.44	0.30	0.197	0.0913	32	0.027

Pin: power input; Voc: open-circuit voltage; Vmax: maximum voltage; Jsc: short-circuit current density; Jscmax: maximum short-circuit current density; FF: fill factor; PCE: power conversion efficiency.

Table 2. Comparison of photovoltaic parameters of chlorophyll-based DSSCs reported in the literature.

Dye Source	Photoanode	Electrolyte	CE	Jsc (mA cm ^{−2})	Voc (V)	FF (%)	PCE (%)	Ref.
<i>Moringa oleifera</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	0.197	0.44	32	0.027	This study
<i>Ocimum gratissimum</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	0.044	0.466	40.0	0.021	[47]
<i>Anethum graveolens</i> (fresh)	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	0.965	0.579	40.0	0.220	[47]
<i>Spinach oleracea</i> (dried)	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	1.110	0.583	46.0	0.290	[47]
<i>Sandoricum koetjape</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	0.042	0.049	46.0	0.018	[26]
<i>Strobilanthes cusia</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	-	0.283	25.2	0.012	[46]
<i>Codiaeum variegatum</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	0.902	0.286	42.0	0.120	[19]
<i>Sargassum</i> sp.	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	-	0.241	43.2	1.500	[17]
<i>Ziziphus jujuba</i> (dried)	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	3.180	0.652	51.9	1.077	[47]
Berry leaves (dried)	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	3.573	0.595	44.1	0.939	[47]
<i>Chlorella vulgaris</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	2.530	0.551	65.0	0.900	[47]
<i>Undaria pinnatifida</i>	TiO ₂ /FTO	I [−] /I ₃ [−] LE	Pt/FTO	13.800	0.570	58.0	4.600	[47]

CE: counter electrode; LE: liquid electrolyte; FTO: fluorine-doped tin oxide; Pt: platinum; TiO₂: titanium dioxide; Jsc: short-circuit current density; Voc: open-circuit voltage; FF: fill factor; PCE: photoconversion efficiency. (-): value not reported in source.

In a study by Mejica et al. [46], a PCE of 0.03% using chlorophyll dye extracted from *Strobilanthes cusia* leaves was obtained. Similarly, 25 obtained PCEs of 0.0170% and 0.0155% from chlorophyll dyes extracted from *Sandoricum koetjape* (green pigment) and *Syzygium samarangense* leaves, respectively, using various solvents. The PCE observed in the current study is within the range reported in the literature. However, it is also important to note that this PCE is lower than some values reported in the literature. For instance, Hendi et al. [35] evaluated the PCE of two green dyes, spinach and henna, under similar conditions and achieved PCEs of 0.24% and 2.19%, respectively. The lower PCE observed for spinach was attributed to weak bonding between TiO₂ particles and the dye molecule. Similar to the current study, the low PCE may result from the solvent's effect on the chlorophyll structure, leading to weak bonds with TiO₂ and hindering electron transfer, which ultimately affects recombination in DSSCs [34,45]. Studies by Sarkar et al. [33], Kwartiningsih et al. [48], and Lee et al. [49] further emphasize the importance of aligning the polarity and chemical structure of organic solvents with the phytochemicals being extracted. The polarity of a solvent directly influences its ability to extract chlorophyll without altering its functional structure, which is critical for maintaining strong interactions with TiO₂ particles. In the current study, acetone was used as the extraction solvent, which is known for its efficacy in preserving chlorophyll's structural integrity. Despite this, the observed PCE highlights the inherent limitations of natural dyes, which often exhibit lower performance compared to synthetic alternatives due to their stability and bonding characteristics.

The low PCE of 0.027% observed in this study can be attributed to several interconnected factors. Chlorophyll lacks strong anchoring functional groups, such as carboxylic or hydroxyl moieties, which facilitate robust chemisorption onto TiO₂ surfaces, resulting in weak dye–semiconductor binding and reduced electron injection efficiency [34,45]. In addition, the relatively low fill factor (32%) indicates significant internal resistance and

charge recombination at the TiO_2 –dye–electrolyte interface, limiting the maximum power output. Furthermore, the absence of co-sensitization restricts light harvesting to the absorption bands of chlorophyll a, while chlorophyll aggregation on the TiO_2 surface may quench excited states and further reduce electron injection efficiency. These factors collectively contribute to the limited photovoltaic performance observed. This interpretation is consistent with literature reports, where weak dye–semiconductor interactions and aggregation effects are commonly associated with reduced DSSC efficiency for chlorophyll-based sensitizers [34,45]. Electrochemical impedance spectroscopy (EIS) is expected to provide further insight into charge transfer resistance and recombination dynamics and is therefore identified as a key focus for future work.

Moreover, Hendi et al. [34] note that chlorophyll dye is generally less effective at converting light into electricity in DSSCs compared to other photosynthetic pigments (e.g., flavonoids). This is because chlorophylls efficiency depends on the availability of a viable bond between the dye molecules and nano TiO_2 particles, facilitating electron transport from excited dye molecules to nano TiO_2 . This observation is consistent with the literature, which often notes that chlorophyll alone tends to have lower PCE efficiency compared to other natural sensitizing pigments, such as anthocyanins, betalain, etc. Numerous studies have examined the co-sensitisation of pigments to enhance the overall absorption range in the visible spectrum, which, in turn, enhances the efficiency of solar cells. For instance, Wuryanti [50] combined the red (flavonoids) and green (chlorophyll) leaves of *Syzygium paniculatum*, achieving a PCE of 2.387%. Srivastava et al. [19] evaluated *Codiaeum variegatum* leaves and *Delonix regia* flowers as natural sensitizers, obtaining PCEs of 0.12% and 0.17%, respectively. Their mixture resulted in a higher PCE of 0.24%. These findings collectively suggest that co-sensitization, TiO_2 surface modification, and the use of molecular anchoring agents represent promising strategies for improving PCE in future studies.

The efficiency of DSSCs is influenced by multiple factors, such as the choice of sensitizers, photoanodes, electrolytes, and conducting materials. Given the inherently less stable and shorter lifespan of natural dyes, the calculated efficiencies are often found to be lower [45]. The PCE of *M. oleifera* could potentially be improved by optimising these other factors.

4. Conclusions

The findings of this study highlight the potential of *Moringa oleifera* as a sustainable and accessible source of chlorophyll for industrial applications, including its use as a natural dye in dye-sensitized solar cells (DSSCs). The acetone-extracted chlorophyll dye demonstrated a short-circuit current density (J_{sc}) of 0.197 mA cm^{-2} , an open-circuit voltage (V_{oc}) of 0.44 V, a fill factor of 32%, and a photoconversion efficiency (PCE) of 0.027%. While this efficiency is within the range reported for other organic sensitizers, it underscores the inherent challenges associated with natural dyes, such as limited light absorption and weaker bonding with TiO_2 nanoparticles.

The study also reaffirms that chlorophyll dyes generally exhibit lower photovoltaic performance compared to other natural pigments, such as flavonoids and betalains, which are often characterized by broader absorption ranges and stronger interactions with TiO_2 . However, research has shown that combining chlorophyll with other pigments through co-sensitization can significantly enhance the PCE by broadening the light absorption spectrum and improving charge transfer processes. These strategies offer a promising direction for future studies aimed at optimizing the performance of DSSCs based on *M. oleifera* chlorophyll.

Moreover, *M. oleifera*'s fast growth rate, high biomass yield, and drought tolerance make it a particularly attractive candidate for sustainable energy applications in regions

with limited resources. Its dual potential for industrial dye applications and renewable energy generation further emphasizes the value of this versatile plant.

Future investigations should focus on several key areas to further advance the performance of *M. oleifera*-based DSSCs. Quantification of dye loading via UV-Vis absorbance before and after sensitisation was not conducted in this study and is identified as a priority for future work. A systematic comparison of DSSC performance using chlorophyll extracted with different solvents (e.g., acetone, ethanol, and DMF) should be conducted to establish the relationship between extraction solvent, dye loading, and PCE. In addition, optimisation of dye adsorption conditions, including immersion time and temperature, is required. Profilometric measurement of the TiO₂ film thickness should be incorporated in future work, as this parameter significantly influences dye adsorption, electron transport, and overall DSSC performance. Future work should include long-term stability testing of fabricated DSSCs under varying environmental conditions, including light exposure, elevated temperature, and humidity, to evaluate the operational stability and lifetime of *M. oleifera* chlorophyll as a photosensitizer. Replicate device fabrication and testing, together with full statistical analysis, should be incorporated in future work to ensure reproducibility and improve the reliability of the reported photovoltaic parameters.

Addressing these aspects collectively could lead to the development of more competitive and environmentally sustainable solar energy solutions.

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