

Electrical Performance of Dye-Sensitized Solar Cells Using Trisodium Copper Chlorophyllin and Acidified Chlorophyllin Sensitizers

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ABSTRACT

A dye-sensitized solar cell (DSSC) is an emerging photovoltaic technology classified in the same group as other third-generation solar cells within the thin-film family. Thin-film DSSCs consist of a titanium dioxide (TiO₂) electrode that acts as the photoanode, a dye-sensitizer adsorbed on the TiO₂ film, an electrolyte layer, and a counter electrode. The main purpose of this study is to investigate and compare the performance of trisodium copper chlorophyllin (SCC) and acidified chlorophyllin (AC) as sensitizers in DSSCs. AC was derived from SCC by protonation of the carboxylate group to the carboxylic group using sulphuric acid, and the reaction was confirmed by Fourier Transform Infrared (FT-IR) spectroscopy. From the electrical characterization of the device, SCC showed a power conversion efficiency (PCE) of 0.08% at the optimum concentration of 0.3mM; meanwhile, AC showed a PCE of 0.07% at the optimum concentration of 0.075mM. Furthermore, SCC showed a dye loading of 0.24 g/g TiO₂, and AC showed a dye loading of 0.053 g/g TiO₂ at their respective optimum concentrations. Based on the results, it shows that SCC is a superior sensitizer compared to AC.

Keywords: Chlorophyllin, Dye-sensitized solar cell (DSSC), Fourier Transform Infrared (FTIR), Photosensitizer, Power conversion efficiency (PCE).

1. INTRODUCTION

In recent decades, the significant increase in energy demand has made energy consumption a major concern. The growing concern about the environmental impacts of conventional fossil fuels, particularly global warming, has been the primary impetus for the transition to renewable energy sources. Consequently, it is essential for governments worldwide to continually seek ways to reduce their carbon emissions. Encouraging the implementation and integration of sustainable renewable energy systems serves as the most effective strategy [1]. Several renewable energy options are available, including solar, wind, and hydropower. When compared with solar power, it stands out due to its numerous advantages over other renewable energy sources, such as wind and hydro energy, attracting significant attention as one of the most promising options available. Solar energy is a plentiful, eco-friendly source derived from the sun and can be readily harnessed to produce power. Solar energy has emerged as the favoured energy source because of its lack of

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organic solar cells have undergone substantial improvements over the years, resulting in a marked increase in device PCE.

DSSC represents a revolutionary category of third-generation solar cells that employs a straightforward electrochemical principle, emulating the mechanisms of photosynthesis by enabling molecules to absorb and convert photon energy into electrical energy. DSSC represents a unique category of solar cell, consisting of a wide band gap semiconductor situated within a dye-coated semiconductor oxide layer that interfaces with a redox electrolyte. There has been significant attention to DSSCs recently, as there is growing interest in finding a more affordable and more environmentally friendly alternative to silicon solar cells. The DSSC presents numerous weaknesses that require improvement, providing ample opportunity for people working in the field to explore and enhance the current model of DSSC.

DSSCs offer notable benefits, including streamlined fabrication methods, reduced production expenses, and excellent compatibility with flexible substrates. While the advantages outlined are noteworthy, the effectiveness and enduring stability of the DSSCs system throughout the development of this technology have raised significant concerns from an industrial standpoint [5]. The cell instability of natural dye-based DSSC can be classified into two categories: physical instability, affected by factors like temperature, UV illumination, and humidity; and chemical instabilities, which pertain to the degradation of the dye sensitizer, electrolyte, and counter electrode, along with the influences of temperature, radiation, water, and oxygen [6]. There are various methods to enhance cell stability, including the use of UV filters, sealants, advances in electrolyte formulations, and refinements in counter electrode design. The sensitivity of dyes is vital in dye-sensitized solar cells (DSSCs), as the dye functions as a photon absorber and initiates the conversion of solar energy into electrical energy. Enhancing the dyes shows the most promising outcomes for improving DSSC efficiency.

To fully understand the evolution and chemical foundations of this technology, it is valuable to examine the historical trajectory of DSSC development over the past few decades. These cells, constructed using thin-film technologies, have been extensively researched over the past three decades due to their low cost, relatively easy fabrication, low toxicity, and simple production. Classified firmly as emerging third-generation thin-film systems, DSSCs have surfaced as a practical and economical alternative to traditional p-n junction photovoltaic mechanisms [7]. The core concept dates back to the late 1960s, when it was discovered that electrochemical cells could generate energy through the use of illuminated organic dyes. A pioneering study conducted by the University of California, Berkeley, explored and replicated these primary mechanisms using chlorophyll extracted from spinach, thereby establishing a biomimetic or bionic approach [8]. These investigations were the foundation for the demonstration and discussion of electric power generation through the DSSC principle in 1972 [9], where the initial chlorophyll-sensitized zinc oxide (ZnO) electrode cell was developed, facilitating the injection of electrons from excited dye molecules into a wide band gap of semiconductor, marking the pioneering conversion of photons into electricity.

A historic breakthrough occurred in 1991 when researchers discovered a nanoporous titanium dioxide (TiO₂) electrode possessing a roughness factor of roughly ca. 1000, facilitating the development of DSSCs with an efficiency of 7% [10]. Brian O'Regan and Michael Grätzel created a device utilizing a 10-micrometer-thick layer of TiO₂ nanoparticles, resulting in a transparent film with a substantial surface area covered in a charge-transmitting dye layer optimized for effective light absorption. This device attained an impressive 46% conversion of incident solar energy and exceeded 80% efficiency in converting incident photons into electrical current, producing an overall incident photon to current conversion efficiency (IPCE) of 7.1–7.9% under simulated solar light and a 12% IPCE yield under diffuse daylight. By 1993, Grätzel et al. reported a cell efficiency of 9.6%, which reached 10% in 1997 at the National Renewable Energy Laboratory (NREL).

As this technology matures, the role of the sensitizer has remained essential, as it is the exact site where the photo-electrochemical reaction starts. To ensure stable adsorption onto the semiconductor substrate, the sensitizers are generally designed with specific functional groups like carboxylic acid ($-\text{COOH}$), phosphonic acid ($-\text{PO}_3\text{H}_2$), and boronic acid ($-\text{B}(\text{OH})_2$) [11]. In recent years, significant advancements have been made in DSSC, offering an alternative to inorganic solar cells (ISC) with a current efficiency of 13%, a notable increase from the initial 6.3% when it first emerged in solar cell research 30 years ago. Significant efforts have been devoted to enhancing the device's efficiency, with a prevalent approach in the literature being the exploration of novel materials for the photoactive layer. Among the numerous building blocks available in the literature, porphyrin stands out as one of the most promising candidates for synthesizing electron-donor and electron-acceptor materials. Among the numerous building blocks available in the literature, porphyrin stands out as one of the most promising candidates for synthesizing electron-donor and electron-acceptor materials. Porphyrin is a tetrapyrrolic 18 π -electron aromatic macrocycle, as shown in Figure 2, which exhibits remarkable characteristics, including its effectiveness as a light harvester in both the blue (Soret band) and red (Q-band) regions of the visible light spectrum [12]. Additionally, its robust electron-donating capabilities, comprehensive π -conjugated system, and easily adjustable optical and electronic properties via modifications at the periphery (meso- and β -positions) and at the metal centre facilitate highly efficient electron transfer [12].

A prime example of a compound that incorporates this advantageous porphyrin structure is chlorophyll (Chl), commonly found in green plants. Structurally, Chl consists of a porphyrin-like ring containing a Magnesium (Mg) atom that plays a crucial role in transforming solar energy into chemical energy. While five different types of chlorophyll have been identified in nature, chlorophyll *a*, *b*, *c*, *d*, and *f* [13], each of them displays distinct light absorption spectra optimized for specific environmental wavelengths, but their native thermal stability remains a challenge, as the environmental conditions influence it.

To address these stability limitations while exploiting the light-harvesting potential of the macrocycle, this study uses chlorophyllin (Chln) as a basis to develop a modified photoactive material for the solar cell. Chlorophyllin is a modified form of chlorophyll that lacks a phytol group, produced via hydrolysis facilitated by chlorophyllase [14]. The synthesis of chlorophyllin involves substituting the central magnesium atom in the ring with copper, while the phytol esters are replaced with sodium, which will become trisodium copper chlorophyllin (SCC). This specific structural modification leads to excellent water solubility and grants chlorophyllin significantly enhanced stability in comparison to raw, unmodified chlorophyll.

Herein, we present a comparative study to evaluate the potential of chlorophyllin and acidified chlorophyllin as novel photoactive materials for organic-based DSSC devices. By modifying the structural properties of raw chlorophyll through acidification, this research investigates how these chemical alterations influence overall dye performance. The main objective of this study is to analyse and compare the electrical characterization of standard and acidified chlorophyllin dyes, ultimately identifying the superior candidate to enhance charge transport and optimize photovoltaic efficiency in DSSC applications.

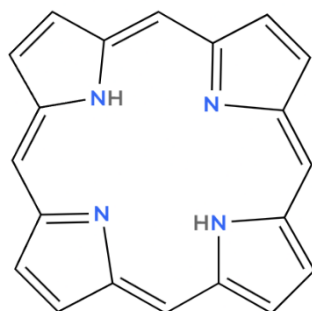


Figure 2: Chemical structure of porphyrin.

2. METHODOLOGY

2.1 Preparation of Standard and Acidified Chlorophyllin Dyes

Trisodium copper chlorophyllin powder was dissolved in distilled water to make concentrations in the range between 0.05 mM to 1.0 mM.

For the synthesis of the acidified chlorophyllin, approximately 500 mg of trisodium copper chlorophyllin powder was dissolved in 100 mL of distilled water, followed by the addition of 1.25 mL of 1 mol/dm³ sulfuric acid to the solution. The reaction resulted in the protonation of the carboxylate group of trisodium copper chlorophyllin, converting it to a carboxylic group [15,16]. This process resulted in a dark green solid precipitate (crude acidified chlorophyllin), which was in turn filtered and rinsed with distilled water. The crude acidified chlorophyllin was subsequently dried in a desiccator for a minimum of three days, while maintained in a cold, dark environment. To acquire purified acidified chlorophyllin, the crude product was dissolved in diethyl ether and filtered to isolate the undissolved solid product. Subsequently, the undissolved solid product was treated with methanol to facilitate the extraction of acidified chlorophyllin, then filtered to remove any residual solid. The acidified chlorophyllin solution in methanol was further processed to yield acidified chlorophyllin powder by removing methanol with a rotary evaporator.

2.2 Component Preparation and DSSC Device Fabrication

A substrate composed of titanium dioxide (TiO₂) nanoparticle powder (particle size ≤ 25nm) was fabricated. This porous TiO₂ layer plays a critical role in the photoanode performance, as its porosity dictates the amount of dye adsorbed onto the matrix to facilitate efficient electron transport. To prepare the paste, approximately 6 g of TiO₂ powder was mixed with 9 mL of 0.06 M acetic acid solution (pH 3), 2 mL of Triton X-100, and 0.2 mL of distilled water. The solution was ground in a mortar and pestle until a smooth white paste formed, with no visible granules. The paste was then transferred into a syringe for storage and ease of use.

Concurrently, the iodide electrolyte, which serves as the medium for the redox reaction within the cell, was prepared by dissolving lithium iodide powder and iodine prill in methoxypropionitrile to achieve an electrolyte concentration of 0.7 mol/L iodide and 0.05 mol/L iodine. The solution was transferred to a 50 mL volumetric flask, and additional solvent was added to bring the volume to the calibration mark.

For the counter electrode, it is essential to utilize a chemically stable material, as most metals tend to react with iodide. The preparation of the carbon electrode involved a straightforward technique for gathering soot produced by candle combustion; a clean ITO glass substrate was exposed directly to the flames to ensure complete coverage of the glass with carbon.

For final device fabrication, commercial Indium Tin Oxide (ITO)-coated glass substrates (20 mm × 20 mm × 1.1 mm, sheet resistance < 10 Ω/sq, transmittance > 83%) were used. Prior to film deposition, the ITO glass substrate underwent a thorough cleaning process. The prepared TiO₂ paste was then applied to the conductive surface of the ITO glass using the doctor-blade technique. Adhesive tape with a thickness of 16 μm was used as a masking template to define an active cell area of 1 cm × 1.5 cm while controlling the resulting film thickness. The substrate was then dried at 180°C, followed by high-temperature furnace sintering at 450°C for 30 min. The sintered TiO₂ substrate was subsequently immersed for approximately 20 hours in the prepared chlorophyllin-based dye solutions to ensure that a significant amount of dye adhered to the semiconductor layer. To ensure reliable dye loading and prevent contamination or degradation, this immersion process was strictly conducted in an airtight beaker within a completely dark environment.

2.3 Solar Simulation and Electrical Characterization

To evaluate the structural functional groups of the prepared dyes, Fourier Transform Infrared Spectroscopy (FTIR) analysis was performed on both the standard chlorophyllin and acidified chlorophyllin samples.

The fully assembled solar cells were evaluated using a solar simulator. Electrical characterization, specifically the current-voltage (I-V) behavior of the devices, was recorded using computer-controlled KickStart Software paired with a Keithley 2450 SourceMeter Unit (Keithley SMU 2450). The devices were illuminated under AM 1.5G simulated sunlight (conforming to the ASTM G-173-03 standard) at an incident light intensity (P_{in}) of 1000 W/m² (equivalent to 100 mW/cm²). The power conversion efficiency (PCE) was calculated using Equation 1. To ensure experimental reproducibility and data reliability, a total of 5 sample devices were fabricated and tested for each device configuration, with each individual device undergoing 15 consecutive testing cycles.

$$PCE(\%) = \left(\frac{J_{sc} \times V_{oc} \times FF}{P_{in}} \right) \times 100 \quad (1)$$

To provide a unified view of the entire fabrication and characterization process, the overall experimental workflow is schematically depicted in Figure 3.

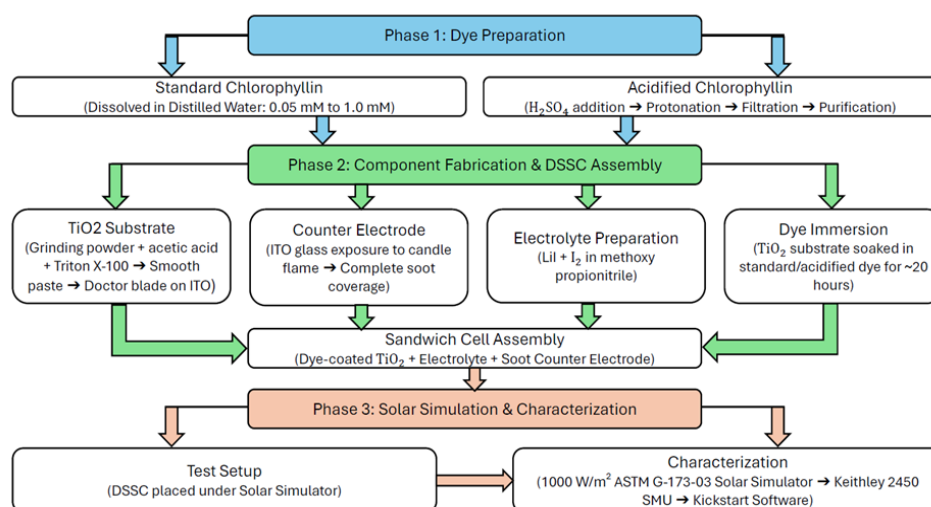


Figure 3: Schematic flowchart detailing the experimental workflow, including standard/acidified dye synthesis, individual component preparation, DSSC assembly, and subsequent photovoltaic characterization.

3. RESULTS AND DISCUSSION

3.1 Analysis of Chlorophyllin Dye and Acidified Chlorophyllin Dye

After synthesizing the photoactive layer, FTIR analysis is performed on both chlorophyllin and acidified chlorophyllin dye to evaluate their spectra. Figure 4 shows the FTIR spectra of chlorophyllin and acidified chlorophyllin dye.

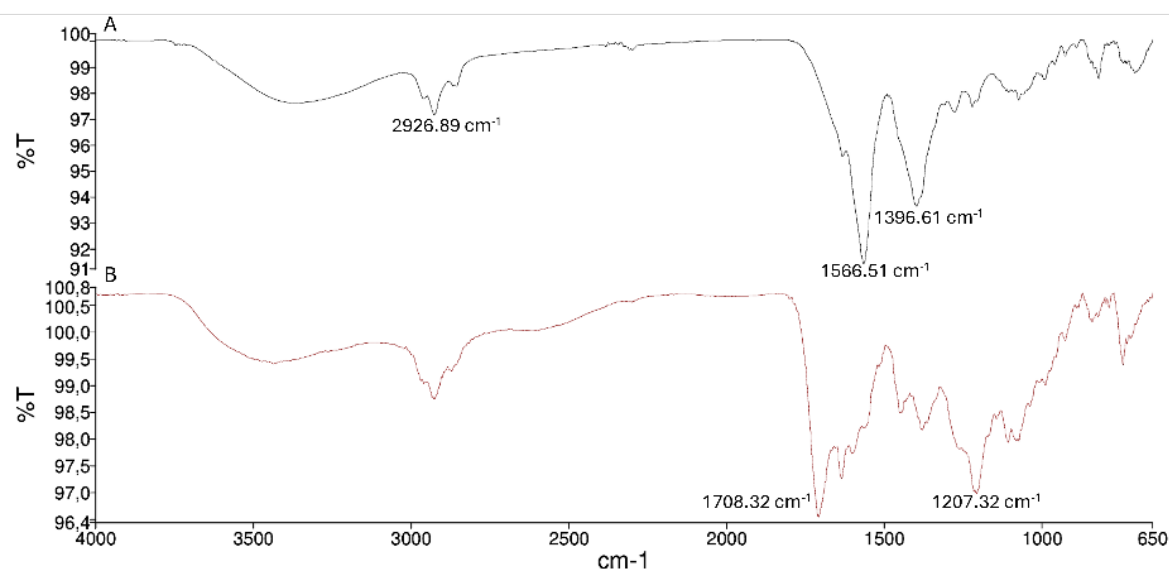


Figure 4: FTIR spectra of chlorophyllin (top) and acidified chlorophyllin (bottom).

The FTIR spectra in Figure 4 confirm the chemical change between the two dyes. Spectrum A shows the standard chlorophyllin dye used directly without changes, while Spectrum B shows the acidified chlorophyllin dye.

In Spectrum A, the major peak at 1566.51 cm⁻¹ represents the carboxylate salt group (COO⁻) from the trisodium copper chlorophyllin, with another related peak at 1396.61 cm⁻¹. An alkane peak is also visible at 2926.89 cm⁻¹.

In Spectrum B, the chemical modification is confirmed by the appearance of new carboxylic acid peaks at 1708.32 cm⁻¹ (corresponding to C=O carbonyl stretching) and 1207.32 cm⁻¹ (corresponding to C-O stretching). This occurs because the trisodium copper chlorophyllin anions were successfully protonated into neutral carboxylic acid groups (COOH) when sulfuric acid was added. This modification is further verified by the sharp drop in the original salt peak at 1566.51 cm⁻¹, indicating that the salt terminals were converted into acid terminals.

3.2 Dye Loading Characterization

Figure 5 and Figure 6 show the dye loading of trisodium copper chlorophyllin and acidified chlorophyllin on the TiO₂ photoanode, respectively. Both the dye loading of trisodium copper chlorophyllin and acidified chlorophyllin showed a similar trend, where the amount of adsorbed dye increased with increasing dye concentration.

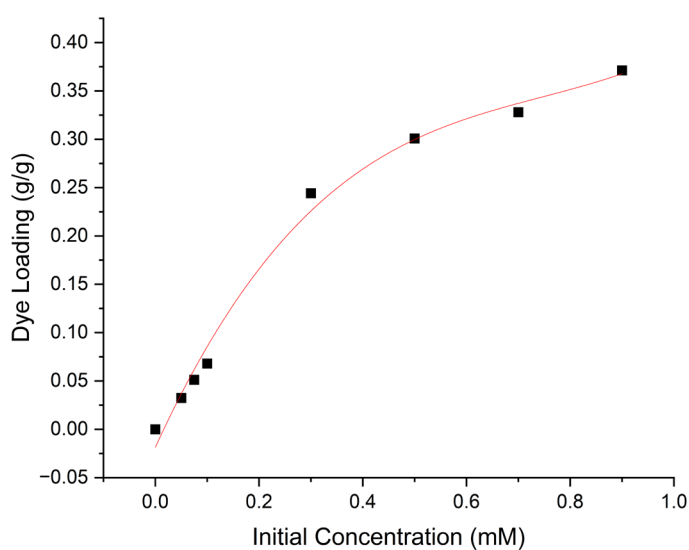


Figure 5: Dye loading of chlorophyllin.

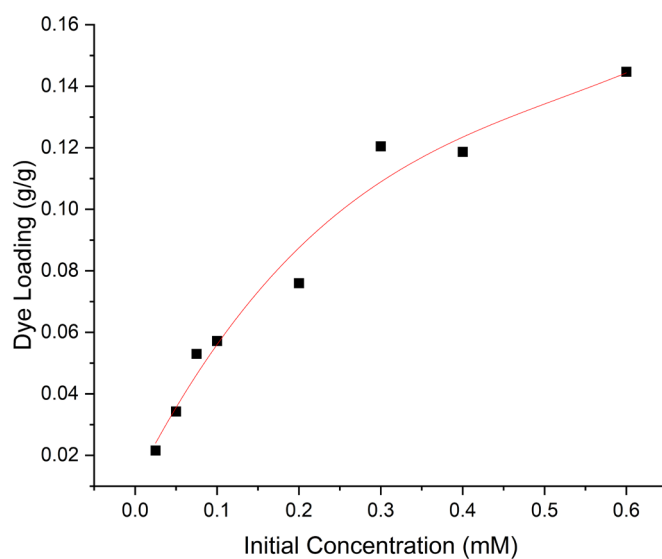


Figure 6: Dye loading of acidified chlorophyllin.

3.3 Electrical Characterization of Chlorophyllin-based DSSC

Table 1 and Figure 7 present the results obtained from testing the fabricated solar cell using chlorophyllin as the dye for DSSC. To evaluate the electrical performance of the cells, key photovoltaic parameters were monitored, including the short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF), and power conversion efficiency (PCE). All values in Table 1 are reported as the mean $\pm$ standard deviation calculated from replicate measurements. It was found that chlorophyllin at a concentration of 0.3 mM yields the best result at 0.08% PCE. The lower concentration produces a lower PCE, and the results increase with increasing concentration until it reach 0.3 mM. Starting from 0.5 mM, the PCE results began to decrease as the concentration increased. This shows that the optimum concentration of chlorophyllin as dye is approximately 0.3 mM.

Table 1: Performance of DSSC device sensitized with chlorophyllin dye (data are presented as mean $\pm$ standard deviation).

Dye concentration (mM)	Jsc (mA/cm ²)	Voc (V)	FF	PCE (%)
0.05	0.43 $\pm$ 0.03	0.24 $\pm$ 0.01	0.41 $\pm$ 0.02	0.01 $\pm$ 0.001
0.075	0.19 $\pm$ 0.01	0.27 $\pm$ 0.01	0.42 $\pm$ 0.02	0.02 $\pm$ 0.001
0.1	0.33 $\pm$ 0.02	0.36 $\pm$ 0.01	0.46 $\pm$ 0.02	0.05 $\pm$ 0.004
0.3	0.59 $\pm$ 0.04	0.30 $\pm$ 0.01	0.43 $\pm$ 0.02	0.08 $\pm$ 0.006
0.5	0.50 $\pm$ 0.03	0.32 $\pm$ 0.01	0.44 $\pm$ 0.02	0.07 $\pm$ 0.005
0.7	0.43 $\pm$ 0.03	0.31 $\pm$ 0.01	0.39 $\pm$ 0.02	0.04 $\pm$ 0.003
0.9	0.36 $\pm$ 0.02	0.29 $\pm$ 0.01	0.36 $\pm$ 0.02	0.04 $\pm$ 0.003
1.0	0.34 $\pm$ 0.02	0.30 $\pm$ 0.01	0.32 $\pm$ 0.02	0.03 $\pm$ 0.002

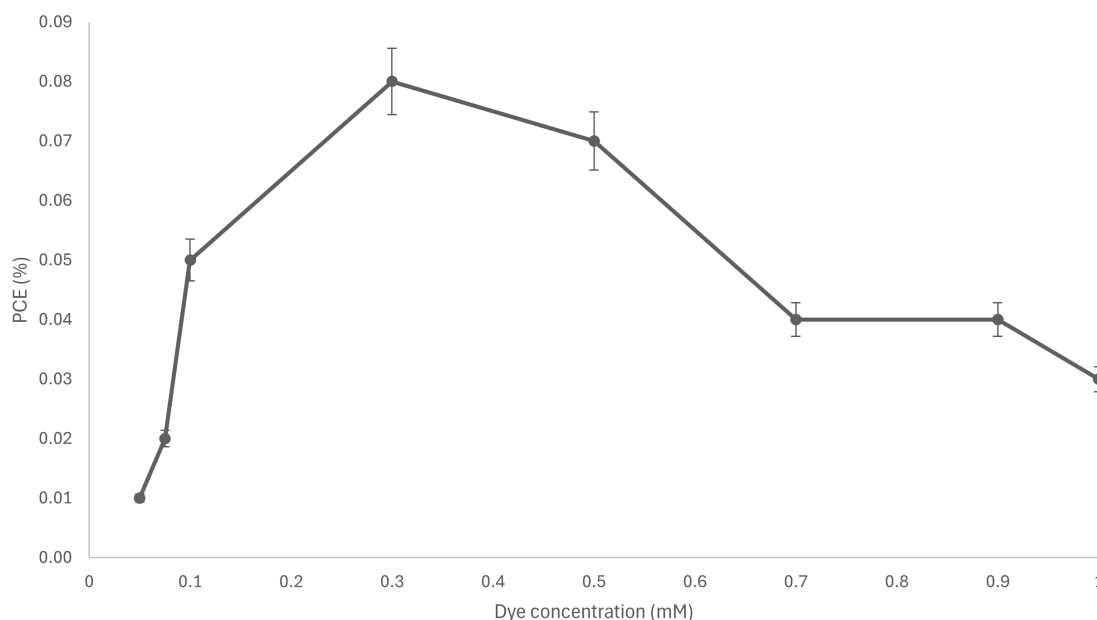
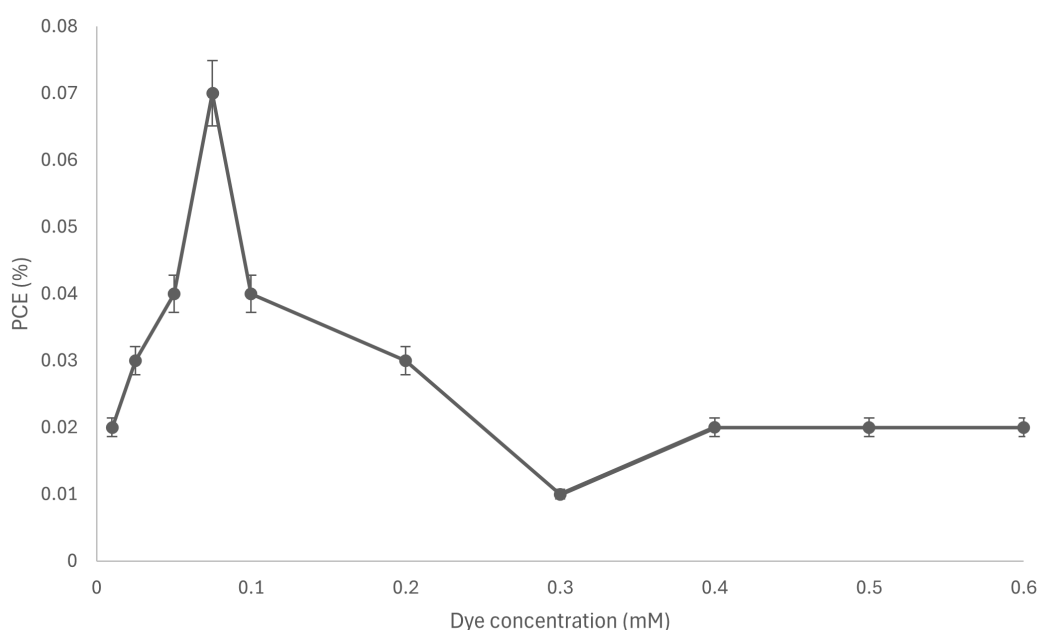
**Figure 7:** Effect of concentration of trisodium copper chlorophyllin on PCE.

Table 2 and Figure 8 show the results obtained from testing the fabricated solar cell using acidified chlorophyllin as the dye for DSSC. All values in Table 2 are reported as the mean $\pm$ standard deviation calculated from replicate measurements. It was found that acidified chlorophyllin at a concentration of 0.075 mM yields the best result at 0.07% PCE. The lower concentration produces a lower PCE, and the results increase with increasing concentration until it reach 0.075 mM. Starting from there, the PCE results started to decrease with a higher increase of concentration, and it starts to remain constant when reaching 0.4 mM of dye concentration. This shows that the optimum concentration for acidified chlorophyllin is around 0.075 mM.

Table 2: Performance of DSSC device sensitized with acidified chlorophyllin dye (data are presented as mean $\pm$ standard deviation).

Dye concentration (mM)	Jsc (mA/cm ²)	Voc (V)	FF	PCE (%)
0.01	0.15 $\pm$ 0.01	0.24 $\pm$ 0.01	0.52 $\pm$ 0.03	0.02 $\pm$ 0.001
0.025	0.30 $\pm$ 0.02	0.23 $\pm$ 0.01	0.46 $\pm$ 0.02	0.03 $\pm$ 0.002
0.05	0.43 $\pm$ 0.03	0.24 $\pm$ 0.01	0.41 $\pm$ 0.02	0.04 $\pm$ 0.003
0.075	0.61 $\pm$ 0.04	0.26 $\pm$ 0.01	0.43 $\pm$ 0.02	0.07 $\pm$ 0.005
0.1	0.46 $\pm$ 0.03	0.24 $\pm$ 0.01	0.37 $\pm$ 0.02	0.04 $\pm$ 0.003
0.2	0.33 $\pm$ 0.02	0.23 $\pm$ 0.01	0.37 $\pm$ 0.02	0.03 $\pm$ 0.002
0.3	0.24 $\pm$ 0.01	0.25 $\pm$ 0.01	0.26 $\pm$ 0.01	0.01 $\pm$ 0.001
0.4	0.34 $\pm$ 0.02	0.24 $\pm$ 0.01	0.29 $\pm$ 0.02	0.02 $\pm$ 0.001
0.5	0.22 $\pm$ 0.01	0.25 $\pm$ 0.01	0.28 $\pm$ 0.01	0.02 $\pm$ 0.001
0.6	0.29 $\pm$ 0.02	0.24 $\pm$ 0.01	0.24 $\pm$ 0.01	0.02 $\pm$ 0.001

**Figure 8:** Effect of concentration of acidified chlorophyllin on PCE.

Based on the results obtained from the electrical characterization of the device, it can be seen that the optimum concentration of trisodium copper chlorophyllin solution needed to sensitise the photoanode is higher (0.3mM) than the concentration of acidified chlorophyllin solution (0.075mM). It can also be seen that the dye loading at the optimum concentration of both dyes is different from each other, where trisodium copper chlorophyllin shows higher dye loading as compared to acidified chlorophyllin. This explains the slightly higher PCE (0.08%) of trisodium copper chlorophyllin at optimum concentration as compared to acidified chlorophyllin (0.07%). Due to a higher amount of sensitizer loaded on the surface, it leads to more electrons being injected into the conduction band of the photoanode, resulting in a higher short circuit current density (Jsc) of 0.59 mA/cm² versus 0.61 mA/cm² [17,18,19,20].

Although the acidified chlorophyllin has superior carboxylic acid (COOH) anchoring groups that improve individual molecular binding to the TiO₂ surface, its lower total dye loading capability limits its maximum photocurrent output. Furthermore, for both dyes, increasing the concentration beyond their optimal thresholds results in a sharp decrease in PCE. This decline at higher dye concentrations is primarily due to dye aggregation on the semiconductor film. The formation of thick dye layers leads to intermolecular quenching and creates a physical barrier that reduces light penetration. Consequently, this leads to poor electron injection efficiency and increased electron recombination losses with the electrolyte, lowering both current density and fill factor values at elevated concentrations. To place these photovoltaic performances in a broader context, the maximum PCE values obtained in this work (0.07%–0.08%) fall within the typical performance range reported for crude natural chlorophyll extracts and simple plant pigments, which generally exhibit low efficiencies (< 0.5%) due to rapid non-radiative decay and unoptimized surface binding [21,22]. While optimized semi-synthetic chlorophyllin derivatives reported in literature can achieve higher efficiencies (~0.5%–2.0%) depending on solvent interactions and co-sensitization [21], they remain significantly lower than engineered synthetic porphyrin dyes, which feature tailored push-pull donor-acceptor architectures and dedicated anchoring groups to exceed 10% efficiency [23]. Nevertheless, the simple, low-cost processing of these chlorophyllin formulations offers a viable, environmentally friendly route to accessible solar cell applications.

4. CONCLUSION

In summary, acidified chlorophyllin was successfully synthesized via the protonation of a trisodium copper chlorophyllin precursor using sulfuric acid. Photovoltaic characterization revealed that standard trisodium copper chlorophyllin yielded a peak power conversion efficiency (PCE) of 0.08% at an optimal concentration of 0.3 mM, whereas the acidified variant achieved a comparable PCE of 0.07% at a significantly lower optimal concentration of 0.075 mM. The major implication of this shift is that, while standard chlorophyllin offers a slightly higher ultimate efficiency, structural acidification lowers the optimal operating concentration threshold by 75%. This provides a viable chemical strategy to drastically reduce organic material consumption during cell assembly without incurring a catastrophic drop in cell performance.

The reported efficiencies (0.07%–0.08%) are relatively low compared to commercial ruthenium complexes, which can be explained by examining the individual photovoltaic parameters extracted from the result. The low photocurrent performance, with a maximum short-circuit current density (J_{sc}) of 0.59 mA/cm² for standard chlorophyllin and 0.61 mA/cm² for the acidified dye, points to limited light absorption and weak dye adsorption on the semiconductor layer. Furthermore, the open-circuit voltage (V_{oc}) consistently remained low (ranging from 0.23 V to 0.36 V), suggesting suboptimal TiO₂ film quality and electrolyte limitations that combined to promote rapid electron recombination at the interfaces. Finally, replacing expensive platinum with a cost-effective, candle-soot carbon counter electrode introduces high internal charge-transfer resistance, which restricts the fill factor (FF) from reaching its maximum potential.

To enhance the performance of these eco-friendly, low-cost DSSCs in future developments, it is recommended to explore co-sensitization techniques, such as blending dyes or using an additional organic dye to broaden the spectral absorption bandwidth, or to apply chemical surface passivation to the TiO₂ film to actively suppress electron recombination. Furthermore, incorporating UV-Vis absorption spectroscopy in future studies is recommended to comprehensively evaluate the optical absorption properties and verify the structural characteristics of acidified chlorophyllin.

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