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Research Article

Effect of dye content concentration chlorophyll-anthocyanin on light absorption rate and characteristics of natural dye-sensitized solar cell

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Abstract: Renewable energy materials like Dye-Sensitized Solar Cell (DSSC) using the photoelectrochemical mechanisms have gathered considerable attention worldwide. This study explores the use of natural dyes, specifically anthocyanin extracted from beetroot (Beta vulgaris L.) roots and chlorophyll from alfalfa (Medicago sativa L.) leaves, as sensitizers in natural DSSC. DSSC sensitized solely with anthocyanin exhibited low performance, with an efficiency of η = 0.0131%. To enhance cell efficiency, a combination of anthocyanin and chlorophyll dyes was tested in various volume ratios to determine the optimal mixture for maximum efficiency. The UV absorption spectrum of the combined dyes demonstrated a wider absorption range and greater absorbance in the visible light spectrum compared to anthocyanin alone. The optimized combination of 80% anthocyanin and 20% chlorophyll in the DSSC resulted in parameters *of Isc = 0.276 mA*, *Voc = 0.512 V*, and *FF = 0.313*, leading to a maximum efficiency of *0.0197%*.

Keywords: *Absorption rate, Anthocyanin, Chlorophyll, Dye combination, Dye-sensitized solar cell*

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1. INTRODUCTION

The global demand for energy continues to rise due to population growth and technological advancement, while the availability of fossil energy sources is steadily declining. Moreover, the exploitation and combustion of conventional fossil fuels have contributed significantly to environmental degradation, including greenhouse gas emissions, air pollution, and climate change. In response to these challenges, the development and application of new and renewable energy (NRE) sources have become a global priority. Among various NRE technologies, solar energy holds tremendous potential due to its abundance, sustainability, and environmental friendliness [1]. One promising approach to harness solar energy is through solar cell technologies.

In recent decades, significant efforts have been made to develop efficient, cost-effective, and environmentally friendly photovoltaic (PV) systems. The third generation of solar cells, particularly Dye-Sensitized Solar Cell (DSSC), has gained considerable attention for their unique advantages such as low production costs, ease of fabrication, and potential for large-area applications. Unlike conventional silicon-based solar cells, DSSC operate based on photoelectrochemical principles, utilizing photosensitizer dyes to absorb sunlight and generate electricity [2]. The concept of DSSC was pioneered by B. O'Regan and M. Grätzel in 1991 [3]. They introduced a system using Titanium dioxide (TiO_2) nanoparticles sensitized with a Ruthenium-based dye in an electrolyte medium, achieving an initial photoconversion efficiency of around 7–11%, which has since improved to approximately 14.3% with further optimizations [3]. Despite these advancements, DSSC still face limitations in terms of long-term stability and relatively lower conversion efficiency compared to silicon-based cells, which restrict their commercial application [4]. To address these issues, researchers have explored the use of natural dyes as alternative sensitizers. Natural pigments such as anthocyanins, chlorophylls, betalains, and carotenoids extracted from fruits, vegetables, flowers, and leaves are attractive due to their environmental safety, cost-effectiveness, and ease of availability [5]. Among them, anthocyanins and chlorophylls are widely studied for their strong light absorption capabilities and compatibility with TiO_2 surfaces [6]. However, single natural dyes often have a narrow absorption spectrum and limited photostability, resulting in suboptimal solar cell performance.

An effective strategy to enhance light harvesting and improve efficiency is the co-sensitization of DSSC using multiple natural dyes with complementary absorption spectra. This approach can broaden the absorption range of the solar spectrum, increase light-harvesting efficiency, and improve electron injection dynamics [7]. By combining two or more dyes, it is possible to optimize energy conversion by capturing a wider range of visible light wavelengths.

The novelty of the current work lies in its systematic investigation of the synergistic effects of various concentration combinations of pure natural dyes, anthocyanin derived from beetroot (Beta vulgaris L.) and chlorophyll extracted from alfalfa (Medicago sativa L.), on the optical absorption and photovoltaic performance of DSSC. In this research, we comprehensively evaluate how these natural dyes, known for their rich pigment content, interact in different ratios to enhance the DSSC efficiency, addressing a relatively unexplored area without relying on synthetic dye mixtures. The natural dyes were extracted and analyzed for their light absorption characteristics using UV-VIS Spectroscopy and functional group compositions using Fourier Transform Infrared Spectroscopy (FTIR). The DSSC have been fabricated by using the standard sandwich cell configuration, and their photovoltaic performance have been tested under a solar simulator to provide the same conditions. This research aims to contribute to the development of low-cost, environmentally sustainable DSSC by utilizing naturally derived sensitizers and exploring the effects of dye co-sensitization on cell performance.

2. MATERIALS AND METHODS

The main materials used in DSSC include Fluorine-doped Tin Oxide (FTO) glass, Titanium dioxide (TiO_2), Silicon dioxide (SiO_2), Distilled water, Ethanol, Potassium iodide (KI), Iodine (I_2), and Polyethylene glycol (PEG) 400. The natural chlorophyll coloring of alfalfa leaves (Medicago sativa L.) is cleaned and cut into small pieces then soaked in ethanol. Natural anthocyanin coloring beetroot (Beta vulgaris L.) cleaned and cut into small pieces. Small pieces of beetroot are then soaked in ethanol. The ratio between dye and solvent is 10 g: 100 mL, soaked for 24 hours, then the extract is filtered and used as a source of crude sensitizer. The prepared samples were subsequently analyzed using a UV-VIS Spectrophotometer, Specord 200 Plus model, manufactured by Analytik Jena.

The FTO glass (resistivity = 10 Ω/sq) was thoroughly cleaned and semiconductor oxide paste was applied to the FTO glass with the "Doctor Blade" coating technique. Paste preparation consists of 3.5 g of TiO_2-SiO_2 with 2.975 g of TiO_2 and 0.525 g of SiO_2 each, crushed in a mortar with 15 mL of ethanol for 30 minutes to make a homogeneous mixture. The area of the working electrode is 2×2 cm^2. The TiO_2-SiO_2 coated film was sintered at 450 °C in a muffle furnace for 30 minutes.

Electrolyte preparation by mixing PEG 400 (0.2068 g), KI (0.8 g), and I_2 (0.127 g) with 10 mL of distilled water as a solvent. The electrolyte solution is stored in a closed bottle. The sintered TiO_2-SiO_2 film is then soaked in the dye solution for 24 hours to make the working electrode functional. The resulting samples were then tested using a Fourier Transform Infrared Spectrometer (FTIR), model IR Prestige 21, manufactured by Shimadzu.

The dye absorbed by the TiO_2-SiO_2 film is then dried in air. Comparative electrode preparation, another FTO glass was covered with carbon using the candle nip technique. The photoanode and cathode are then fastened together with paper binder clips and a redox electrolyte solution is injected into the cell. The resulting samples were then tested using a Solar Simulator (100 mW/cm^2), model 10500 SN 370, manufactured by ABET Technologies.

3. RESULTS AND DISCUSSION

Fig. 1 presents the UV-VIS absorption spectra of dyes with varying compositions of anthocyanin (Ant) and chlorophyll (Chl) in the wavelength range of 300–800 nm. The absorption spectrum of 100% Ant (black curve) exhibits three notable features: A broad band centered at approximately 330 nm, a secondary peak around 398 nm with an absorbance of ~0.2805, and a weaker shoulder near 475 nm with an absorbance of ~0.3062.

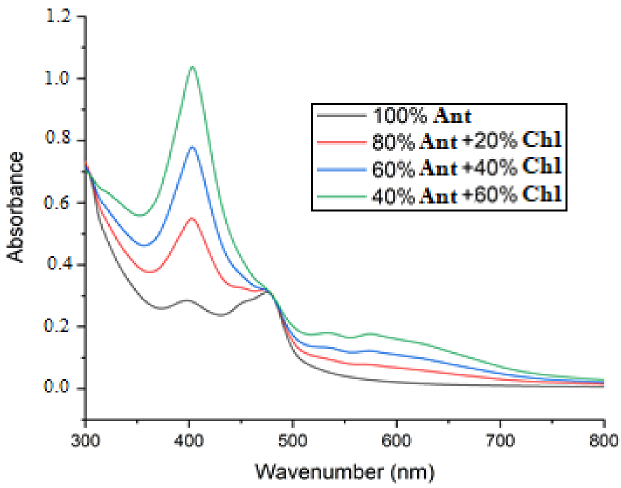

Figure 1. UV-VIS absorption spectrum of Anthocyanin Dye and Dye Combination

These peaks correspond to characteristic π–π* electronic transitions of anthocyanin molecules, primarily in the UV to blue region of the spectrum. For the 80% Ant + 20% Chl dye mixture (red curve), the spectral profile changes significantly, showing increased absorbance at 403 nm (~0.5491), while still retaining a shoulder near 473 nm (~0.3194) and a weaker peak at 567 nm (~0.0788). The emergence of absorption in the green-yellow region reflects the increasing contribution of chlorophyll components. With 60% Ant + 40% Chl (blue curve), the absorption spectrum exhibits stronger absorption at 403 nm (~0.7798) and a broader peak near 574 nm (~0.1222). The higher absorbance and broader peak widths suggest enhanced spectral overlap between Ant and Chl, promoting better light-harvesting capabilities. At 40% Ant + 60% Chl (green curve), the spectrum shows a dominant peak at 403 nm with an absorbance of ~1.0379, along with distinct peaks at 534 nm (~0.1806) and 575 nm (~0.1770). These features indicate significant spectral broadening and enhanced contribution from Chl, particularly in the green to red region of the visible spectrum [8]. Overall, increasing the Chl content enhances not only the absorbance intensity but also the spectral coverage, as evident from the full width at half maximum (FWHM) of key peaks. This broader and more intense absorption across the visible spectrum is advantageous for Dye-Sensitized Solar Cell (DSSC) applications, enabling more efficient photon harvesting. Thus, combining Ant and Chl offers a complementary sensitization effect that can significantly improve photovoltaic performance [9].

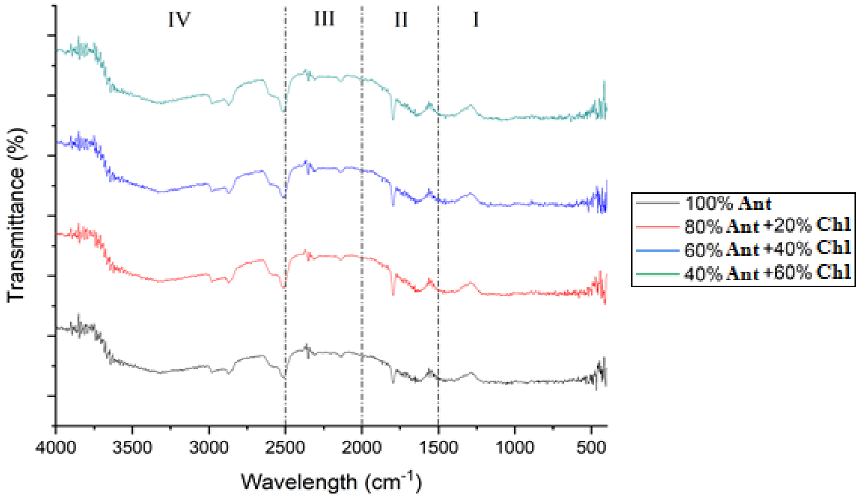

Figure 2. FTIR spectrum of anthocyanin dyes and dye combinations absorbed by TiO_2-SiO_2

Fig. 2 presents the FTIR spectra of dye samples within the spectral range of 4000–450 cm⁻¹, divided into four spectral Zones (*I–IV*). This analysis highlights the functional groups responsible for electron injection and anchoring of dye molecules onto semiconductor surfaces. Efficient electron injection into semiconductor oxides requires strong absorbance, which can be achieved through anchoring groups such as carboxyl, carbonyl, and hydroxyl present on the dye molecules [10]. In Zone *I* (400–1500 cm^{-1}), multiple absorption peaks corresponding to C–O stretching vibrations were observed. The 100% anthocyanin (Ant) sample shows distinct peaks at 1016 cm^{-1} and 1095 cm^{-1}, which can be attributed to ether and alcohol groups. For the 80% anthocyanin (Ant) + 20% chlorophyll (Chl) sample, a strong peak appears at 1315 cm^{-1}, while the 60% Ant + 40% Chl and 40% Ant + 60% Chl samples exhibit peaks at 1313 cm^{-1} and 1199 cm^{-1}, respectively. These peaks are associated with phenolic and flavonoid structures, which are key constituents of anthocyanin-based dyes, indicating C–O bond stretching [11].

In Zone *II* (1500–2000 cm^{-1}), significant peaks related to C=O stretching in conjugated aromatic rings were found. The 100% Ant sample displays a peak at 1614 cm^{-1}, corresponding to aromatic C=C and carbonyl group stretching. The 80% Ant + 20% Chl and 60% Ant + 40% Chl samples both show peaks at 1795 cm^{-1}, with the former also exhibiting an additional peak at 1959 cm^{-1}. The 40% Ant + 60% Chl sample features two distinctive peaks at 1791 cm^{-1} and 1961 cm^{-1}. These variations reflect the increased contribution of chlorophyll functional groups, particularly carbonyl groups, with growing Chl content. Such spectral shifts suggest changes in molecular conjugation and ring structure, which may influence dye–semiconductor interaction strength. Zone *III* (2000–2700 cm^{-1}) covers a region typically characterized by overtone and combination vibrations appearing weaker and broader. While some shoulders or broad features can be observed, this region lacks sharp, well-defined peaks, and thus, it was not subjected to further detailed quantitative analysis [12]. In Zone *IV* (2700–4000 cm^{-1}), strong absorptions corresponding to C–H and O–H stretching vibrations are observed. The 100% Ant sample shows peaks at 2872 cm^{-1} and 2843 cm^{-1}, indicating symmetric and asymmetric stretching of alkane C–H bonds. The 80% Ant + 20% Chl and 60% Ant + 40% Chl samples display peaks at 2858 cm^{-1} and 2981 cm^{-1}, and at 2868 cm^{-1} and 2980 cm^{-1}, respectively. The 40% Ant + 60% Chl sample exhibits a peak at 2868 cm^{-1} and a broad, intense band at 3278 cm^{-1}, corresponding to O–H stretching from alcohol or phenol groups in chlorophyll. The breadth and intensity of this O–H peak suggest strong hydrogen bonding, which may enhance dye–surface interactions in photovoltaic applications [13]. Overall, the variation in the number, position, and intensity of FTIR peaks across samples indicates the progressive incorporation of functional groups such as hydroxyl and carbonyl with increasing chlorophyll content. These anchoring groups play a critical role in improving dye sensitization efficiency and facilitating effective charge transfer to semiconductor surfaces [14].

Table 1. DSSC performance measured with solar simulator lighting (100 mW/cm^2).

Sample	Isc (mA)	Jsc (mA cm^{-2})	Voc (V)	Fill Factor	Efficiency (%)
100% Ant	0.148	0.0658	0.583	0.342	0.0131
80% Ant + 20% Chl	0.276	0.123	0.512	0.313	0.0197
60% Ant + 40% Chl	0.262	0.116	0.515	0.287	0.0172
40% Ant + 60% Chl	0.146	0.0651	0.844	0.328	0.018

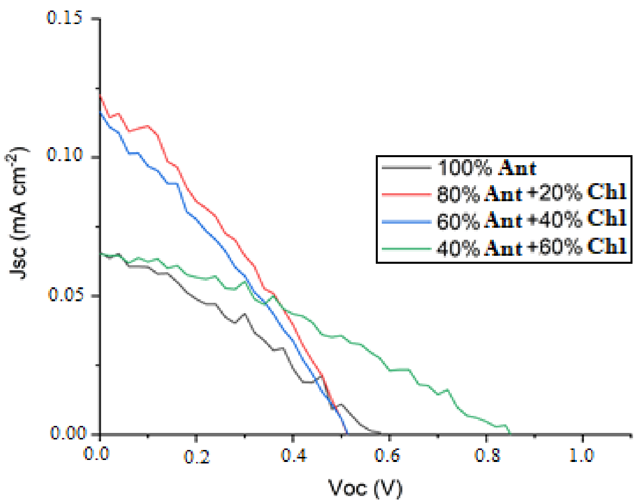

Figure 3. DSSC Current-voltage density (J-V) curve.

The current-voltage (*I-V*) performance of DSSC sensitized with anthocyanin dye (beetroot) and a combination of chlorophyll dye (alfalfa leaves) and anthocyanin (beetroot) was performed by using a simulated sunlight source (100 mW/cm^2). According to Mejica et al., [15], DSSC performance is assessed from open circuit voltage (V_{oc}), short circuit current (I_{sc}) and fill factor (*FF*), as shown in Table 1. The highest current, 0.276 mA, was obtained from a dye combination of 80% anthocyanin and 20% chlorophyll. This value is higher than the current produced by 100% anthocyanin dye, which was 0.148 mA, and the combination of 40% anthocyanin and 60% chlorophyll, which produced amount of 0.146 mA. Fig. 3 shows the current density (J_{sc}) produced by DSSC indicating the dye's ability to collect photons. Different interactions between single dye sensitizers and in combination with semiconductor oxide surfaces impact a direct reduction in the J_{sc} of DSSC devices. The overall absorption of electrons into the conduction band of the semiconductor oxide can be enhanced leading to a higher J_{sc} according to the interaction of the dye with the semiconductor oxide.

Fig. 4 shows that the combination of different dyes increases the photovoltaic impact significantly with single dye sensitization, and increases the absorption of sunlight, and allows more efficient utilization of photon energy. The efficiency of 100% anthocyanin dye is the lowest at 0.0131% compared to the efficiency of other dyes. The characteristics of the dye are directly proportional to cell performance, where the higher the light absorption in the visible range, the higher the cell performance [16].

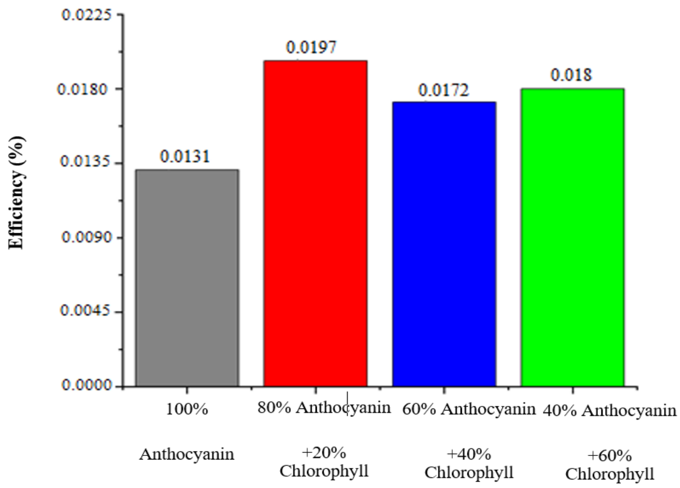

Figure 4. DSSC efficiency with respect to dye concentrations.

Combining anthocyanin dyes with chlorophyll dyes causes DSSC efficiency to increase significantly, as the percentage of chlorophyll dyes in combination increases compared to single dyes. The highest efficiency in the dye combination of 80% anthocyanin + 20% chlorophyll was 0.0197%. The dye combination of 60% anthocyanin + 40% chlorophyll has an efficiency of 0.0172%, while the dye combination of 40% anthocyanin + 60% chlorophyll is 0.018%. DSSC performance also depends on solvent properties, such as polarity, acidity, dye combination and temperature [17]. All dye combinations have higher efficiency than single dyes. Certain coloring pigments can naturally protect other pigments from direct sunlight to maintain the photosynthesis process of plants. One example is a combination of chlorophyll and anthocyanin dyes. Chlorophyll is protected by anthocyanins to prevent chlorophyll damage from direct sunlight. Based on this, the two natural sensitizers with complementary absorption spectra pose a good opportunity to apply a combined sensitization strategy to improve the performance of solar cells, with a wide light absorption range [18]. Although the improvement in DSSC efficiency from the combination of anthocyanin and chlorophyll dyes is relatively small in absolute terms (below 0.02%), the enhancement is significant when compared to individual dye usage, indicating a potential synergistic effect between natural dyes. The low overall efficiency is primarily attributed to the inherent limitations of natural dyes, including their narrow light absorption range and low chemical stability compared to synthetic dyes such as ruthenium complexes [19]. The relatively low values of fill factor and open-circuit voltage (V_{oc}) observed in this study also indicate internal resistance and electron recombination losses within the device, which are further influenced by factors such as dye-solvent interaction, pH, and thermal conditions during fabrication [20]. Despite these limitations, this study provides a valuable early-stage contribution to the application of natural dye combinations in DSSC development. To achieve a more substantial performance improvement, several optimization strategies are recommended. First, the purification and concentration of dye extracts could significantly enhance light absorption and improve dye stability on the TiO_2 surface. Second, selecting solvents with appropriate polarity and acidity can improve dye loading and electron injection efficiency [21]. Third, structural modifications of the photoanode, such as using smaller TiO_2 nanoparticles, incorporating metal doping, or introducing blocking layers, can reduce internal resistance and suppress recombination [22]. Moreover, controlling sensitization time and temperature is critical to ensuring effective dye adsorption and stable operation. With these systematic improvements, the DSSC performance based on natural dye systems could be significantly enhanced. Therefore, despite the currently low efficiency, this article remains worthy of acceptance as it presents original data, a clear analytical framework, and a solid foundation for future research in natural dye sensitized solar cells.

4. CONCLUSION

This study utilizes chlorophyll and anthocyanin pigments as dye sensitizers, both of which can absorb sunlight and convert solar energy into electrochemical energy. Previous research has shown that combining these pigments enhances efficiency. In this experiment, three different combinations of natural dyes were used, with anthocyanin (from beetroot) and chlorophyll (from alfalfa leaves) in ratios ranging from 40% to 80%. These dyes served as sensitizers for the semiconductor oxide TiO_2-SiO_2, acting as an electron conductor. UV-VIS analysis showed that the dye combination resulted in a wider light absorbance and spectrum compared to using a single dye, leading to improved cell performance. FTIR analysis revealed that no new bonds formed between the pigments, suggesting that the two natural pigments coexist without reacting chemically. In terms of DSSC efficiency, 100% anthocyanin achieved a power conversion efficiency (η) of 0.0131%. A 60% anthocyanin + 40% chlorophyll mixture had an efficiency of 0.0172%, while a 40% anthocyanin + 60% chlorophyll blend reached 0.018%. The highest efficiency, 0.0197%, was observed with an 80% anthocyanin + 20% chlorophyll combination. Thus, the optimal dye combination effectively boosts DSSC efficiency.

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