International Journal of Design & Nature and Ecodynamics
Vol. 20, No. 6, June, 2025, pp. 1235-1242
Journal homepage: http://iieta.org/journals/ijdne

Fabrication and Characterization of DSSC Using Natural Dyes from Purple Sweet Potato (*Ipomoea Batatas L.*) and Papaya Leaf (*Carica Papaya*) with Cocktail Dye Method

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https://doi.org/10.18280/ijdne.200605

Received: 3 February 2025
Revised: 17 June 2025
Accepted: 23 June 2025
Available online: 30 June 2025

Keywords:
dye-sensitized, natural dye, anthocyanin, chlorophyll, immersion time, cocktail, purple sweet potato, papaya leaf

ABSTRACT

The abundance of natural dyes combined with their non-toxic and biodegradable properties results in lower environmental impact throughout the entire Dye-Sensitized Solar Cells (DSSCs) life cycle. The extraction methods for these dyes are simple and inexpensive, which reduces the total manufacturing expenses of solar cells. Fabrication and characterization of DSSCs utilizing natural dyes of purple sweet potato, *Ipomoea Batatas* L. (PSP), and papaya leaf, *Carica Papaya* (PL) have been investigated. The inherent absorption range limitations of single natural dyes are addressed in this work through co-sensitization methods, which combine different natural dyes as a promising solution. A single dye and a mixture of the two dyes (Cocktail) were used to make solar cells. The sandwich structure of DSSC consists of dye-coated semiconductor TiO_2 deposited on conductive glass by doctor blades as the working electrode and a carbon layer on the conductive glass as the counter electrode. The research was conducted to determine the effect of the ratio of the mixture (3:1, 1:1, 1:3) and the effect of immersion time (12, 24, 48 hours) of the dye mixture between purple sweet potato anthocyanin dye and papaya leaf chlorophyll dye on the efficiency of DSSC. The final test was carried out to simulate indoor applications by measuring voltage and current using a multimeter while illuminating the cell with a 20-Watt LED (wavelength, 380 nm to 740 nm) as a light source calibrated with a Solar meter to match 100mW/cm^2. The test results obtained higher and more stable voltage and current on DSSC with dye cocktail PSP: PL ratio 1:3 with a 48-hour immersion. In conclusion, a 133% efficiency improvement over single-dye DSSCs was obtained in the variation of the mixture ratio of PSP: PL 1:3 with an immersion time of 48 hours and 0.607% of efficiency.

1. INTRODUCTION

Energy is an inseparable aspect of human existence today, given our heavy reliance on it. Among the various sources of renewable energy, solar energy holds immense potential. In particular, solar cells are considered a highly promising means of generating renewable power in the future. Organic solar cells, also referred to as Dye-Sensitized Solar Cells (DSSC), utilize organic dyes to facilitate light absorption. In the DSSC process, light absorption and electric charge separation occur as distinct mechanisms. The absorption of light is facilitated by dye molecules, while nanocrystalline organic ion semiconductors facilitate charge separation [1]. Functioning through a photoelectrochemical system, DSSC is capable of converting visible light into electricity [2].

DSSCs have emerged as a promising alternative to traditional photovoltaic devices due to their cost-effective production and high conversion efficiency [3]. The fundamental structure of a DSSC comprises two electrodes - a working electrode and a counter electrode - which are separated by a liquid electrolyte containing a redox mediator. The working electrode is composed of a transparent conductive oxide (TCO), such as fluorine-doped tin oxide (FTO), that is coated with a thin film of nanocrystalline semiconductor materials, including titanium dioxide (TiO_2) [4], tin dioxide (SnO_2) [5], or zinc oxide (ZnO) [6]. This semiconductor film is infused with a dye molecule that functions as both a light absorber and an electron donor [7]. The counter electrode is also a TCO coated with a catalyst layer, such as carbon [8], graphene [9], or platinum [10]. The catalyst's role is to facilitate the reduction of the oxidized redox mediator, thereby regenerating the dye molecule. The most commonly employed redox mediator in DSSCs is the iodide/triiodide couple [11], although alternative systems like Co(II)/Co(III) have also been investigated [12]. The performance of a DSSC largely depends on the choice of dye sensitizer, which should possess a broad absorption spectrum in the visible range, a high molar extinction coefficient, excellent stability under light and heat, and a strong affinity for binding to the semiconductor surface [13]. Ruthenium complexes are the most extensively utilized dyes in DSSCs due to their exceptional photophysical and electrochemical properties [14]. However, their expensive synthesis and limited scalability hinder their widespread application. Consequently, researchers have been exploring alternative dyes, such as organic or inorganic dyes, that can either act as co-sensitizers or completely replace ruthenium-based dyes [15-17]. Co-sensitizers enhance the light-harvesting efficiency of the primary dye by filling in spectral gaps or extending the absorption range. Organic dyes offer the advantage of facile modification through the manipulation of functional groups or conjugation length to fine-tune their optical and electronic properties. However, most organic dyes require anchoring groups, such as carboxylic acid or cyano groups, to attach to the semiconductor surface through hydrophobic or covalent interactions [18]. Another potential source of dyes for DSSCs is natural dyes derived from plants.

In general, the dyes utilized in DSSCs are synthetic or metal-complex dyes, such as ruthenium complex. However, synthetic dyes possess certain drawbacks, including high cost and the presence of heavy metals that are harmful to the environment. Various plant extracts, such as Cosmos caudatus flowers, Blumea balsamifera, banana heart, and senduduk fruit, have been employed as sensitizers in DSSCs [19-21]. Several factors impact the efficiency of DSSCs, one of which is the type of dye employed. While natural dyes are more environmentally friendly and readily available in nature compared to synthetic dyes, their absorption spectra are relatively narrow [22]. Therefore, it is essential to combine or mix multiple types of natural dyes to enhance the operational efficiency of DSSCs. This is because the utilization of mixed dyes can increase the absorbance value of DSSCs [23].

The problems with natural dyes, including their reduced efficiency and limited absorption range, and stability issues, exist as interconnected challenges. The absorption spectrum of dyes has a limited range, which prevents them from capturing photons from the solar spectrum, thus affecting their maximum achievable efficiency. The chemical instability of many natural dyes causes them to degrade quickly when exposed to operational conditions such as high temperatures and continuous illumination, which makes them impractical for extended use [24]. The interconnected nature of these challenges shows why researchers need to develop a comprehensive research strategy that tackles all these performance limitations to create practical, sustainable natural dye DSSCs. The complete potential of eco-friendly materials requires simultaneous improvements in light absorption and electron transfer dynamics, and long-term stability.

Single natural dyes show limited absorption capabilities, which can be overcome by implementing co-sensitization methods that combine various natural pigments [25]. The combination technique provides extended light absorption across multiple spectrums, which enhances photon collection efficiency. The water-soluble pigments Anthocyanins from purple sweet potato contain extensive conjugated π bonds, which lead to strong absorption between 330 nm and 475 nm in the UV to blue-green regions. Papaya leaf chlorophylls absorb light mainly in blue and red wavelengths, with Chlorophyll a showing two main absorption peaks at 430 nm and 662 nm [26]. The absorption properties of anthocyanin and chlorophyll match each other, which makes them suitable for dye combination. The combined absorption pattern of these two pigments provides complete coverage of visible light with high light-harvesting efficiency similar to natural photosynthetic systems [27].

The combination of anthocyanin with chlorophyll produces better DSSC performance than using either dye independently, according to research findings [25]. The optimal combination of 80% anthocyanin with 20% chlorophyll achieves a maximum efficiency of 0.0197% in certain systems [28]. The research shows that a 2:5 ratio of anthocyanin to chlorophyll produces the most favorable conditions for charge transfer driving force and energy loss minimization [25]. The research shows that the combined pigments produce a complex synergistic effect that exceeds the simple absorption summation. The device can process the complete solar spectrum through this mechanism to reach higher efficiency in natural dye DSSCs.

In this study, the anthocyanin dye mixture derived from purple sweet potato and papaya leaf chlorophyll was varied in terms of composition and immersion time on the active TiO_2 layer to assess its effect on DSSC efficiency. The findings obtained from this study will indicate the most influential dye mixture in enhancing DSSC efficiency.

2. METHODOLOGY

The dye sources utilized in this study were purple sweet potato and papaya leaves, which were further diluted in 96% alcohol to serve as the solvent. Anthocyanins were extracted from purple sweet potato, while chlorophyll was obtained from papaya leaves. The dyes were then combined using the cocktail method, with the composition of the anthocyanin-chlorophyll mixture varying in three different ratios: 1:3 (10 ml anthocyanin: 30 ml chlorophyll), 1:1 (20 ml anthocyanin: 20 ml chlorophyll), and 3:1 (30 ml anthocyanin: 10 ml chlorophyll). This resulted in five different types of dye, as shown in Figure 1. The anthocyanins were extracted from the tubers of the purple sweet potato plants, while the chlorophyll was extracted from the papaya leaves. Both extracts were dissolved in alcohol and left to macerate for a period of seven days. Subsequently, the extraction solutions were filtered using filter paper and transferred into bottles, which were then covered with aluminum foil and stored in a dark place.

The preparation of working electrodes in an academic context involves several steps, namely the production of conductive glass, the formulation of TiO_2 paste, the deposition of the TiO_2 paste, and the immersion in dye. To create conductive glass, a solution of $SnCl_2$ is applied onto the surface of FTO glass. TiO_2 paste is then created by dissolving 6 grams of TiO_2 powder in 25 ml of alcohol, along with a mixture of 1 gram of HEC (Hydroxy Ethyl Cellulose) and 9 ml of acetic acid added little by little. Acetic acid functions as a multifunctional component in TiO_2 paste preparation, which extends beyond its role as a solvent. The peptizing properties of acetic acid affect both the stability of colloids and the rheological behavior of TiO_2 suspensions. The optimum acetic acid content is crucial because it directly affects the particle aggregation and, consequently, the final porosity and uniformity of the TiO_2 film [29]. The resulting paste is deposited onto an FTO substrate measuring 3 cm × 3 cm, with a deposition size of 1 cm × 1 cm. The doctor blade method is employed for the paste deposition, followed by heating the substrate with a heat gun at a temperature of 450°C for a duration of 10 minutes.

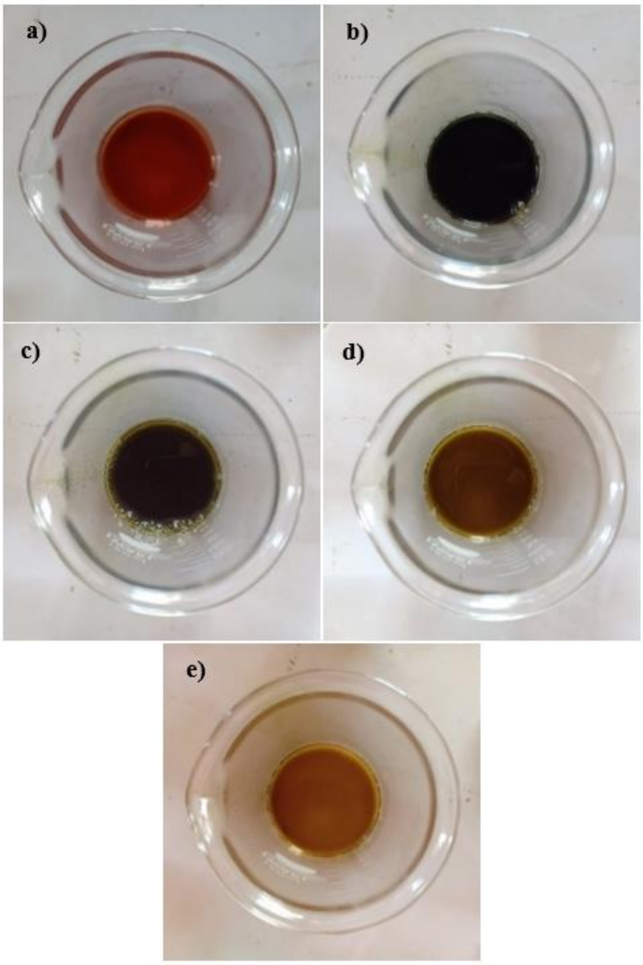


Figure 1. Dye extraction: a) purple sweet potato (PSP) only; b) papaya leaves (PL) only; c) PSP:PL cocktail (1:3); d) PSP:PL cocktail (1:1); and e) PSP:PL cocktail (3:1)

Figure 2. DSSC structure used for I-V measurement

A counter electrode is fashioned using conductive glass coated with carbon from a candle flame, whereby the glass surface is burned until a black layer forms from a few micrometers to around 10 µm [30]. Subsequently, one edge of the glass is rubbed using a cotton bud to create a border. The sandwich structure is then assembled by applying a sealing compound and dripping Iodide/triiodide (I^-/I_3^-) electrolyte (500 mM LiI, 50 mM I_2, in Acetonitrile) onto the working electrode. Finally, the working electrode is affixed to the counter electrode using a paper clip, as shown in Figure 2.

The reproducibility and statistical significance of results were ensured by fabricating multiple replicate devices (n=3) for each experimental condition. The average values of open-circuit voltage (Voc), short-circuit current (Isc), fill factor (FF), and output power (Pout) were used for the analysis.

UV-Vis characterization (Spectrophotometer UV-Vis T60) was conducted to ascertain the absorbance of the dye solution. The current-voltage testing schematic simulates indoor applications, employing a multimeter and 20-Watt LED (wavelength, 380 nm to 740 nm) as a light source calibrated with a Solar meter to match 100mW/cm^2.

3. RESULT AND DISCUSSION

3.1 UV-Vis characterization

Figure 3 illustrates the absorbance profiles of extracts obtained from purple sweet potato and papaya leaves. The obtained dye from purple sweet potato extraction exhibits a pronounced maximum absorbance peak at the wavelength region of 380 nm. This outcome implies the presence of anthocyanins within the purple sweet potato extract. On the other hand, the extraction of papaya leaves yields dyes displaying five distinct peaks of maximum absorbance at wavelengths of 397 nm, 470 nm, 538 nm, 616 nm, and 672 nm. These absorption peaks provide evidence for the presence of chlorophyll within the papaya leaf extract.

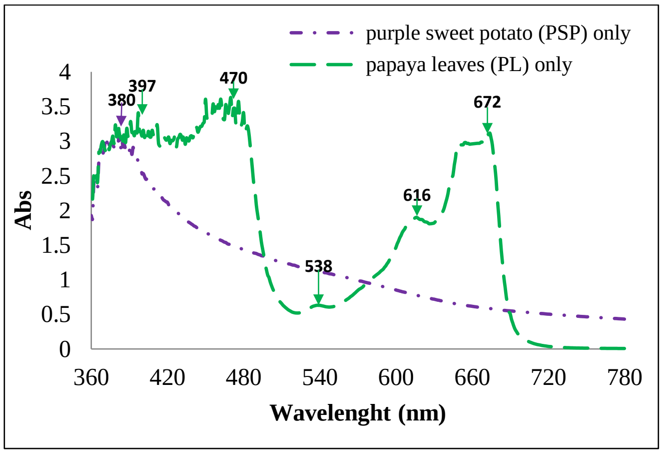


Figure 3. Dye absorption spectra of purple sweet potato dye and papaya leaf dye

The dye mixture exhibits distinct variations in its curve in comparison to the unmixed dye, as illustrated in Figure 4. Notably, there are significant alterations in the peaks corresponding to the region of maximum absorbance across the three dye mixtures. In the case of the mixed dye PSP:PL (1:3), there are 5 prominent peaks of maximum absorbance observed at wavelengths of 398 nm, 451 nm, 537 nm, 613 nm, and 665 nm. On the other hand, in the PSP:PL (1:1) mixture, 5 maximum absorbance peaks are identified at wavelengths of 399 nm, 449 nm, 537 nm, 611 nm, and 664 nm. Conversely, the PSP:PL (3:1) mixture presents only 2 maximum absorbance peaks, occurring at wavelengths of 370 nm and 663 nm.

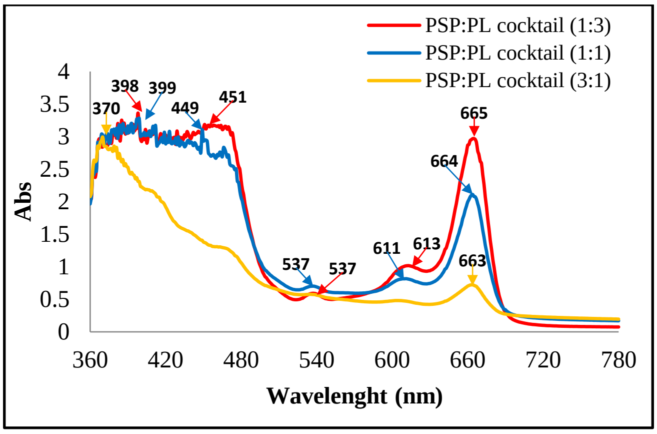
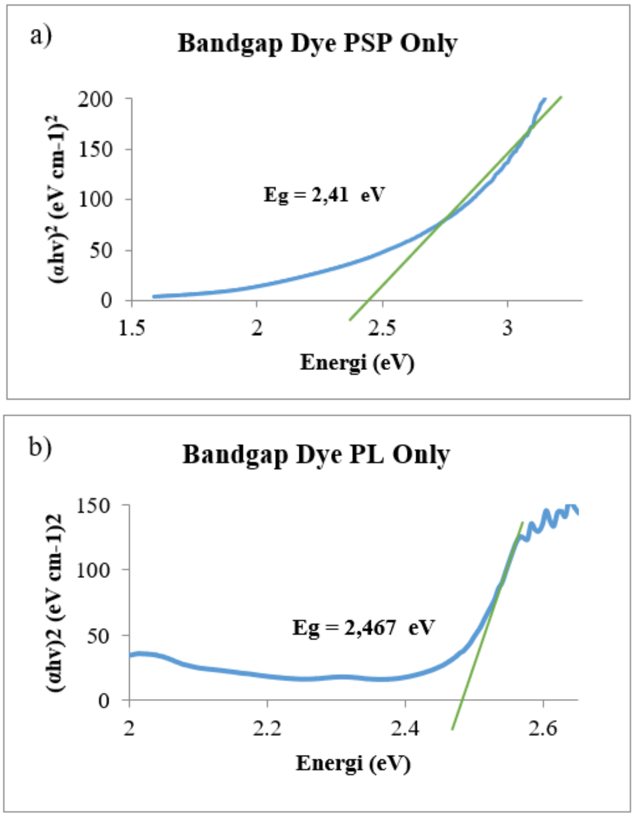


Figure 4. Dye absorption spectra of mixed dyes PSP:PL (1:3), PSP:PL (1:1), and PSP:PL (3:1)

The absorbance data can be utilized for the determination of the energy band gap value through the Tauc Plot method. As shown in Figure 5, the energy band gap value of a single dye or a dye cocktail is found to be smaller compared to the energy band gap of the TiO_2 semiconductor in its anatase phase, which is measured at 3.2 eV. Consequently, it demonstrates potential for application as a photosensitizer in Dye-Sensitized Solar Cell (DSSC) [31]. Based on the energy band gap calculation results, it can be inferred that the anthocyanins extracted from the purple sweet potato exhibit higher efficiency in comparison to the chlorophyll extracted from papaya leaves. This is attributed to the fact that the energy band gap of the anthocyanin extract from purple sweet potato is narrower than that of the chlorophyll extract from papaya leaves.



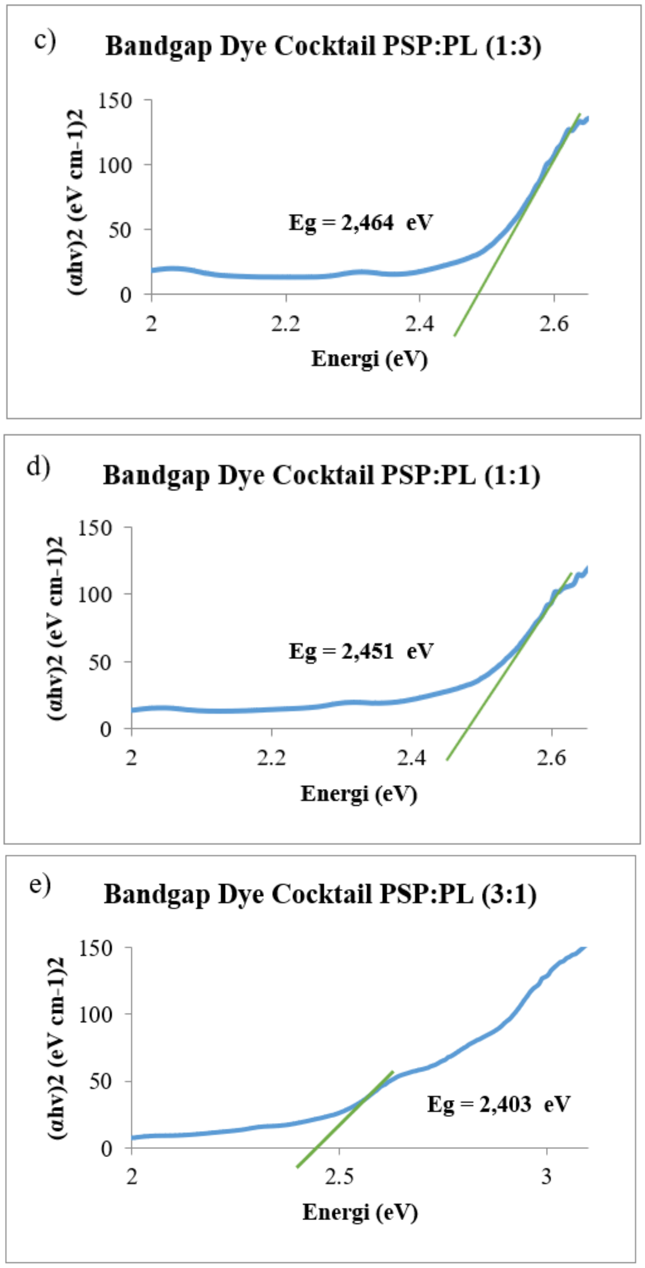


Figure 5. Bandgap, a) purple sweet potato (PSP) only; b) papaya leaves (PL) only; c) PSP:PL cocktail (1:3); d) PSP:PL cocktail (1:1); and e) PSP:PL cocktail (3:1)

3.2 Photovoltaic characterizations

The I-V characterization results are presented in Figure 6 and Figure 7. The determination of DSSC parameters can be derived based on the findings of the I-V characterization, which are illustrated in Table 1, Table 2, and Table 3.

Based on Tables 1 and 2, the findings indicate that the performance or efficiency of the Dye-Sensitized Solar Cells (DSSCs) fabricated using purple sweet potato anthocyanin was marginally superior to those made from papaya leaf chlorophyll. Notably, the purple sweet potato DSSCs exhibited a slightly higher output value in terms of voltage and current, thereby exerting a discernible impact on their overall performance.

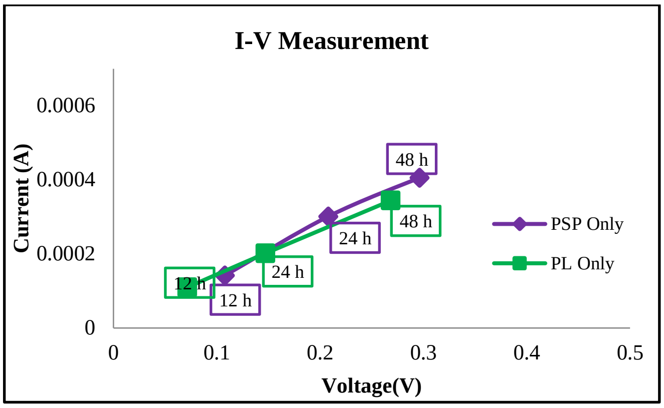


Figure 6. Single dye I-V measurement

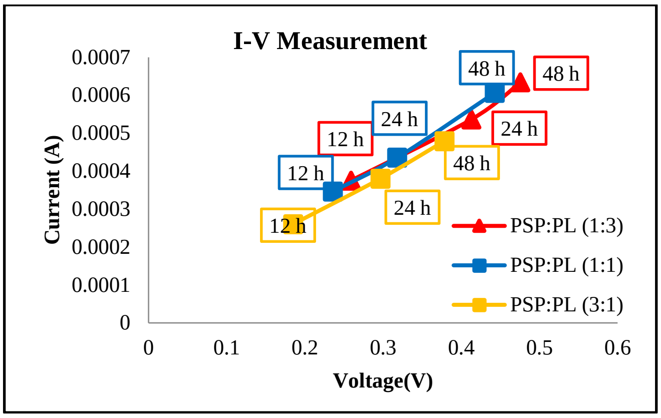


Figure 7. Cocktail dye I-V measurement

The data presented in Table 3 demonstrates that the incorporation of purple sweet potato anthocyanins and papaya leaf chlorophyll, in various composition variations, can enhance the performance or efficiency of DSSC. Notably, the highest efficiency is achieved when using a PSP:PL mixture in a ratio of 3:1 with 48h time immersion. This shows an efficiency improvement 133% better than PSP single-dye and 197% better than PL single-dye. It can be concluded that an increase in the amount of chlorophyll used is accompanied by a corresponding increase in efficiency [32]. Conversely, an increase in the amount of anthocyanins used leads to a decrease in efficiency [33]. This phenomenon can be attributed to the interplay between the purple sweet potato anthocyanins and papaya leaf chlorophyll, as both compounds exhibit different effectiveness at varying pH levels. Consequently, the mixing process has an impact on the absorbance value [33].

Table 4 reveals that the composition of purple sweet potato anthocyanin dye and papaya leaf chlorophyll dye, with various mixture ratios (1:3, 1:1, and 3:1), influences the resulting DSSC efficiency. The highest efficiency was observed in the ratio of PSP:PL mixture (1:3). Furthermore, the immersion time of the active TiO_2 layer in the dye also affects DSSC efficiency when mixing two different types of dye. Longer immersion durations result in increased efficiency.

Table 1. Photo-conversion efficiency of DSSC with purple sweet potato dye

Immersion Time	Voc (V)	Isc (mA)	FF	Pout (mW)	η (%)
12 Hours	0.108	1.4	0.268	0. 004	0.041
24 Hours	0.208	3.0	0.234	0. 015	0.147
48 Hours	0.296	4.1	0.216	0. 026	0.260

Table 2. Photo-conversion efficiency of DSSC with papaya leaf dye

Immersion Time	Voc (V)	Isc (mA)	FF	Pout (mW)	η (%)
12 Hours	0.072	1.1	0.285	0. 002	0.022
24 Hours	0.147	2.0	0.253	0.008	0.075
48 Hours	0.268	3.4	0.221	0.020	0.204

Table 3. Photo-conversion efficiency of DSSC with dye cocktail

Dye Cocktail PSP:PL	Immersion Time	Voc (V)	Isc (mA)	FF	Pout (mW)	η (%)
	12 Hours	0.26	3.8	0.223	0.022	0.216
1:3	24 Hours	0.41	5.4	0.204	0.045	0.451
	48 Hours	0.48	6.3	0.201	0.061	0.607
	12 Hours	0.24	3.5	0.227	0.019	0.186
1:1	24 Hours	0.32	4.4	0.213	0.030	0.295
	48 Hours	0.44	6.1	0.202	0.054	0.544
	12 Hours	0.19	2.6	0.240	0.012	0.116
3:1	24 Hours	0.30	3.8	0.216	0.024	0.243
	48 Hours	0.38	4.8	0.206	0.038	0.375

Table 4. DSSC efficiency

Immersion Time	PSP Only (%)	PL Only (%)	PSP:PL (1:3) (%)	PSP:PL (1:1) (%)	PSP:PL (3:1) (%)
12 Hours	0.041	0.022	0.216	0.186	0.116
24 Hours	0.147	0.075	0.451	0.295	0.243
48 Hours	0.260	0.204	0.607	0.544	0.375

The observed improvement in light absorption results from a synergistic process that includes Förster Resonance Energy Transfer (FRET) rather than simple additive effects of individual dyes [34]. FRET represents a non-radiative energy transfer mechanism through which an excited donor molecule (anthocyanin) transfers its excitation energy to a nearby acceptor molecule (chlorophyll) without photon emission. The process of FRET occurs when the donor emission spectrum matches the acceptor absorption spectrum and the molecules exist within 1-10 nm distance. The energy funneling mechanism of FRET in dye cocktails transfers light energy from one dye (donor) to another dye (acceptor), which has better alignment for TiO_2 conduction band electron injection [35]. The energy transfer process directs absorbed photons to the most efficient electron injector, which optimizes photon utilization and minimizes both radiative decay and unproductive charge recombination. The mechanism enables the cocktail to maximize solar energy utilization, which results in enhanced photovoltaic performance.

The prolonged period of immersion has been found to enhance the absorption of dye by TiO_2, resulting in an increased current and voltage output, provided that the TiO_2 remains insoluble. The duration of TiO_2 immersion in the dye also has an impact on the efficiency of the dye-sensitized solar cell (DSSC), as indicated by the strong current and voltage values [36].

The knowledge of dye adsorption kinetics and isotherm models remains essential because it determines both the best dye loading conditions and the dye-TiO_2 interaction mechanisms. The discovery of anthocyanin chemical adsorption to TiO_2 films and chlorophyll physical adsorption affects both electron transfer efficiency and device longevity. The chemical adsorption of dyes creates stronger bonds with semiconductors, which results in improved electron injection stability and better long-term performance, yet physical adsorption may lead to dye desorption or degradation over time [25]. The knowledge helps determine the best dye concentrations and immersion times to achieve uniform sensitization that optimizes light harvesting and electron injection while preventing harmful dye aggregation.

Moreover, the efficiency of the DSSC is dependent on the wavelength of absorbance utilized. A broader absorbance spectrum allows for the absorption of a wider range of light frequencies by the solar cell [37]. Consequently, a higher level of DSSC absorption leads to a more optimized efficiency.

4. CONCLUSIONS

The combination of purple sweet potato anthocyanin with papaya leaf chlorophyll creates a promising method to build sustainable DSSCs that are economically feasible. The development of practical energy applications for natural dye-based solar cells requires additional research to optimize dye ratios and understand complex interfacial phenomena, and improve long-term stability. Moreover, it is evident that the longer the DSSC is soaked, the greater the efficiency that can be achieved. Specifically, the highest efficiency value of 0.607% was obtained when the ratio of anthocyanin to chlorophyll was 1:3 and the soaking time was 48 hours. This shows an efficiency improvement 133% better than PSP single-dye and 197% better than PL single-dye. Future research should concentrate on creating stronger dye molecules and better electrolyte formulations, and improved encapsulation methods to protect devices from environmental stressors.

ACKNOWLEDGMENT

This research is supported by the Internal Research Grand of Hasanuddin University, Indonesia, with contract number: 00323/UN4.22/PT.01.03/2023.

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NOMENCLATURE

FF	Fill factor
Isc	Short Circuit Current, mA
Pout	Output Power, mW
Voc	Open Circuit Voltage, V