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## Low-Cost Solar Energy Harvesting: A Study on Dye-Sensitized Solar Cells Using Inthanin Leaf Extract as a Natural Photosensitizer

Maria Onyemowo<sup>1,2,</sup> Yuwalee Unpaprom<sup>2,3</sup> and Rameshprabu Ramaraj<sup>1,2\*</sup>.

- <sup>1</sup> School of Renewable Energy, Maejo University, Chiang Mai, 50290, Thailand
- <sup>2</sup> Sustainable Resources and Sustainable Engineering Research Lab, Maejo University, Chiang Mai, 50290, Thailand
- <sup>3</sup>Program in Biotechnology, Faculty of Science, Maejo University, Chiang Mai 50290, Thailand

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#### CORRESPONDING AUTHOR

\*E-mail: rrameshprabu@gmail.com

#### ABSTRACT

The world is actively pursuing renewable energy resources to combat environmental pollution and greenhouse gas emissions caused by the burning of fossil fuels. Solar energy is the most abundant, continuously available form of renewable energy, and dyesensitized solar cells (DSSCs) are being explored as an alternative to traditional siliconbased solar cells due to their low cost of materials, ease of production, and efficiency. The photoanode is a crucial component in DSSCs, and the main challenges faced by these solar cells are charge recombination and low light harvesting ability. This study aims to investigate the efficacy of natural dyes as the sensitizer for the fabrication of DSSCs. Natural pigments were extracted from Inthanin leaves using the solvent extraction technique. The concentration and composition of the pigment were analyzed through pigment analysis utilizing UV a spectrophotometer. Inthanin has a higher chlorophyll a to b ratio. A higher chlorophyll a to b ratio has been shown to be associated with improved light harvesting and electron transport efficiency. This is because chlorophyll a is more efficient at transferring electrons to the electron transport chain than chlorophyll b. The higher chlorophyll a to b ratio could lead to improved electron transfer to the photoanode. The photoelectric parameters of the DSSC were evaluated using I-V characterization. DSSCs coated with Inthanin dye extract produced a short circuit current density (J<sub>sc</sub>) of  $0.02 \text{ mA/cm}^2$ , open circuit voltage ( $V_{oc}$ ) of 0.07 V, fill factor (FF) of 33% and an efficiency ( $\eta$ ) of 0.1%.

#### 1. INTRODUCTION

#### 1.1. Research Background

Solar energy has emerged as a promising solution to address the current global energy crisis. Solar cell research aims to create affordable, safe, and highly efficient materials that transform solar energy into electricity [1]. While the first generation of solar cells was made of expensive monocrystalline silicon, the newer generation of solar photovoltaics, Dye-sensitized solar cells (DSSCs), offers a more cost-effective and temperature-insensitive alternative [2,3]. DSSCs are made of nanostructured semiconducting metal oxide materials and sensitized to visible light by a dye molecule. When the dye molecule absorbs visible light, it injects electrons into the conduction band of the nano TiO<sub>2</sub> layer [4]. The electrons pass through the semiconductor film

through an external circuit to the counter electrode, and the electrolyte in the system helps to regenerate the dye [5]. The fundamental components of DSSCs are the photoanode, sensitizer, electrolyte, and counter electrode. The photoanode is the most significant element in determining the photoconversion efficiency of the solar cell. Nanocrystalline TiO<sub>2</sub> is the most commonly used photoanode material in DSSCs because of its exceptional optoelectronic characteristics [6]. The use of TiO<sub>2</sub> as a photoanode in DSSCs is prevalent due to its cost-effectiveness, abundance, lack of toxicity, stability, biocompatibility, and environmental friendliness. Photoanodes are created by adsorbing the dye onto the surface of the TiO<sub>2</sub> layer [7,8].

#### 1.2. Literature Review

Ruthenium-based compounds have been the top-performing dyes used in DSSCs for an extended period of time, and they are costly,



hazardous, and detrimental to the ecosystem [9]. However, an alternative strategy was developed using natural dyes from fruits, flowers, roots, and leaves. These dyes are simple to extract, require less energy, are more cost-effective, non-toxic, and entirely biodegradable [1,10]. Natural dyes have a more comprehensive range of absorption in the visible region, and they adhere well to the surface of the photoanode, allowing for efficient electron transport. Chlorophyll and carotenoids are two natural dyes extensively studied for their potential use in DSSCs. Chlorophyll is a pigment in the leaves of most green plants, cyanobacteria, and algae and mainly occurs as chlorophyll a. It absorbs light from the red, blue, and violet wavelengths and appears green due to its reflection of this color. Carotenoids, on the other hand, have diverse roles, including light harvesting, photoprotection, and gene expression regulation, and they also facilitate communication within or between species [7,11]. Compared to synthetic dyes, natural dyes used as sensitizers in DSSCs exhibit lower photoconversion efficiency due to their weak binding capacity to the semiconductor oxide layer, resulting in a reduced transfer of excited electrons, ultimately reducing the photoconversion efficiency. Studies have shown that chlorophyll has the potential to be an effective photosensitizer in DSSCs, which makes it an attractive and environmentally friendly alternative to synthetic dyes [1,7]. Since chlorophyll is readily available from natural sources and is biodegradable, it has the potential to become a sustainable and cost-effective solution for developing solar cells.

According to Danladi et al. [12], using natural dyes extracted from roselle flowers and Carica papaya leaves in DSSCs resulted in power conversion efficiencies (PCEs) of 0.046% and 0.022%, respectively. In a similar study, DSSCs that utilized single flame trees and Bougainvillea glabra flowers achieved PCEs of 0.17% and 0.21%, respectively. However, when co-sensitizers extracted with water were added, the PCEs improved to 0.24% [13]. A previous study by Khammee et al. [11], demonstrated that DSSCs using natural dye extracted from *Inthanin bok* achieved a PCE of 1.138%, indicating a significant improvement in the device's performance. This study aims to investigate the photoelectrical properties of DSSCs prepared using natural dyes extracted from Inthanin leaves (Lagerstroemia speciosa) as the photosensitizer. The DSSCs were assembled and tested to evaluate their performance. Therefore, DSSCs sensitized with natural dyes present a promising approach to achieving a more sustainable and eco-friendly energy source. It can potentially reduce dependence on fossil fuels, decrease greenhouse gas emissions, and contribute to a cleaner and healthier environment.

#### 1.3. Research Objective

This study shows that using natural dyes as sensitizers in DSSCs is a promising strategy to overcome the challenges associated with synthetic dyes in terms of cost, toxicity, and environmental impact.

#### 2. METHODOLOGY

#### 2.1. Materials and Methods

This study utilized natural dye sourced from young leaves of *Inthanin* ( also known as *Lagerstroemia speciosa*) collected from

Maejo University, Chiang Mai, Thailand. To fabricate the DSSCs, various chemicals and reagents were utilized, including Ethanol (C<sub>2</sub>H<sub>5</sub>OH) and Acetic acid (CH<sub>3</sub>COOH) from Union Science, nanoparticles TiO<sub>2</sub>, Potassium Iodide (KI), Iodine crystals (I<sub>2</sub>), Acetonitrile (C<sub>2</sub>H<sub>3</sub>N), and Ethylene glycol (C<sub>2</sub>H<sub>6</sub>O<sub>2</sub>) from Sigma-Aldrich. Pigment analysis was carried out using a UV spectrophotometer (Drawell DV-8000). The DSSCs' photochemical performance was evaluated using two digital multimeters, UNI-T (UT61E) and a digital potentiometer (MCP41010), under AM 1.5 G tungsten lamp. To evaluate the DSSCs, I-V characteristic curves were measured at different light intensity levels by exposing the solar cell to white light from a 100-W tungsten lamp at room temperature.

#### 2.2. Preparation of Photosensitizer

To extract the pigments from *Lagerstroemia speciosa* leaves, commonly known as *Inthanin*, fresh young leaves of *Inthanin* were collected, washed, and separated from the stem. The leaves are then chopped into small pieces, weighed, and blended with ethanol to make a smooth mixture. The mixture was left to react for approximately 10 minutes before being filtered using a vacuum pump to extract the chlorophyll. Afterward, the filtered solution was transferred to a 100 ml volumetric flask and stored in the refrigerator for subsequent analysis [14]. This method of extracting pigments can be used to obtain high-quality pigments suitable for further analysis.



Fig. 1 (a) Fresh Young *Inthanin* Leaves and (b, c, and d)

Dye Extraction Process

#### 2.3. Preparation of TiO<sub>2</sub> Photoanode

Fluorine tin oxide (FTO) as the substrate with sheet resistance measuring around  $20~\Omega.cm^2$  was used as the substrate. The FTO glass was cleaned and subjected to sonication in distilled water and soap. A mix of TiO<sub>2</sub> powder with 10 ml of 5% acetic acid and 0.5g of surfactant was added to obtain the TiO<sub>2</sub> paste with the desired consistency. The paste was applied to the conductive side of the FTO glass using the doctor blade method, followed by drying and sintering at 300°C for 1 h to form a uniform film. This process ensures that the TiO<sub>2</sub> film has good adhesion, high crystallinity, and an optimized morphology for efficient charge transfer and light absorption [15,16].

#### 2.4. Preparation of Counter Electrode

To prepare the counter electrode material, 1g of activated carbon, 5g of latex glue, and 5 ml of ethanol were mixed to form a thick black paste. The paste was applied to an aluminum foil using the doctor blade method and heated at 200°C for 15 mins to enhance its properties. The counter electrode was equal to the photoanode to ensure efficient energy conversion. This process is essential to activate the platinum layer's catalytic properties, facilitating the transport of electrons from the redox electrolyte to the external circuit, resulting in improved energy conversion efficiency in DSSCs. The counter electrode facilitates the movement of electrons from the external circuit to the redox electrolyte [14].

#### 2.5. Electrolyte Preparation

The electrolyte preparation method used in this study was adopted from [16,17]. To prepare the electrolyte, 1 gram of iodine was combined with 5 g of potassium iodide and stirred until the iodine was fully dissolved. Next, 1 g of ethylene glycol and 4 g of acetonitrile were added to the solution. The resulting electrolyte was carefully transferred to a conical flask and stored for further analysis. The quality of the electrolyte solution is important in determining the performance of DSSCs, and a reliable preparation method ensures that the solution is consistent and reproducible. The electrolyte is responsible for transporting the electrons generated by the absorbed light from the dye-sensitized layer to the external circuit.

#### 2.6. DSSC Cell Assembly

DSSC consists of two primary constituents, a photoanode  $TiO_2$  coated with a light-sensitive dye, and a counter electrode made of platinum (Pt), which is typically employed. An electrolyte solution separates the two electrodes. The current study employed platinum-coated aluminum foil as the counter electrode material. The two electrodes were affixed together using adhesive tape, and the spacing between them was adjusted to match the electrolyte's thickness. Next, the electrolyte was interposed between the two electrodes. Finally, the DSSC was constructed, and clips were used to secure it, concluding the DSSC production process. [18].

### 2.7. Preparation of fabricated DSSC Using Doctor Blade's Method

This process involves spreading TiO<sub>2</sub> paste on a conductive substrate of the FTO using a glass rod blade. The glass rod is used to control the thickness of the TiO<sub>2</sub> film. The TiO<sub>2</sub> paste is then applied to the FTO-coated glass and flattened using the doctor blade technique until a homogenous layer is achieved. The TiO<sub>2</sub> layer is then annealed at 450°C for 30 mins, to improve its adhesion to the substrate. After cooling the TiO<sub>2</sub> film at room temperature, the photosensitive dye is deposited dropwise on the TiO<sub>2</sub> layer of the photoanode. The counter electrode is also prepared by depositing activated carbon on another FTO glass and heating it at 200°C for 15 mins. After that, the two electrodes are sandwiched together and an electrolyte solution is poured between them and sealed with scotch tape to prevent electrolyte leakage, using a binder clip to hold the constructed cell together and then measure its electrical performance [19].

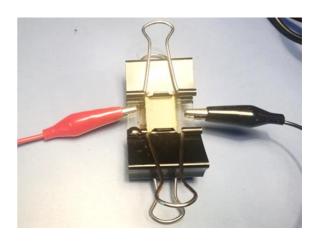


Fig. 2. DSSC Fabricated with Inthanin Dye

#### 2.8. Pigment Analysis

The methodology employed for the analysis of pigments involved the solvent extraction technique, which was carried out sequentially. First, the leaves were carefully washed, separated from the stem, and cut into small pieces before being weighed. Subsequently, the leaves were placed in a mortar with 1g of sand and grinded until they became smooth. Afterward, 20 mL of ethanol was added to the sample, and the mixture was left to react for 5 mins. The resulting mixture was filtered using a vacuum pump, and the filtered solution was transferred to a 100 mL volumetric flask and supplemented with ethanol to the mark. A spectrophotometer recorded the absorbance readings at 470, 649, and 664 nm [14]. Finally, the chlorophyll (a and b) and carotenoid concentrations were calculated using the below equations.

Chlorophyll 
$$a = (13.36xA664 - 5.19xA649) \times DF$$
 (1)

Chlorophyll 
$$b = (27.43xA649 - 8.12xA664) \times DF$$
 (2)

Carotenoid= 
$$(1000xA470 - 2.13xCa - 97.63xCb)$$
 (3)  
/209×DF

Where A = is the amount of light absorbed DF= is referred to as the dilution factor.

#### 2.9. Photoelectric Characterization of DSSC

The J-V characteristics of the DSSCs were examined under simulated illumination with  $100~\text{mW/cm}^2(\text{AM1.5})$ . By analyzing the I-V curve, performance parameters such as short-circuit current density (Jsc), open circuit voltage (Voc), maximum power (Pmax), fill factor (FF), and overall photoconversion efficiency were determined. The fill factor (FF) was computed using the equation below.

Fill Factor (FF) = 
$$((J_{max}V_{max}))/((J_{sc}V_{oc}))$$
 (4)

Jsc and Voc represent short-circuit current density  $(mA/cm^2)$  and open-circuit voltage (V). At the same time,  $J_{max}$  and  $V_{max}$  denote the short-circuit current density and open-circuit voltage at maximum power. Essentially, the fill factor measures how

effectively the cell converts light into electrical energy. Overall photoconversion efficiency  $(\eta)$  was calculated using below.

$$(\eta) = ((Jsc \times Voc \times FF))/Pin \times 100\%$$
 (5)

where Pin is the incident light.

The photoconversion efficiency represents the amount of light energy converted into electrical energy by the DSSCs and is a key performance parameter for solar cells [20].

#### 2.10. Pigment Analysis

The study was conducted with one independent sample in three replicates (n=1), and the results were averaged and presented with their corresponding standard deviations (SD).

#### 3. RESULT AND DISCUSSION

#### 3.1. Pigment Analysis

Pigments are organic compounds that play a crucial role in the growth and development of plants. Chlorophyll-a is the primary pigment in plant leaves responsible for photosynthesis, converting light energy into chemical energy. Chlorophyll-b, on the other hand, acts as a supplementary pigment, helping absorb light energy that chlorophyll-a cannot absorb. Evaluating the concentration and composition of pigment in plant leaves is important in understanding plant growth and its interaction with the surroundings [21]. The ratio of chlorophyll-a to chlorophyllb in plant leaves is an important indicator of the amount of light the plant receives. A higher ratio of chlorophyll-a to chlorophyllb indicates that the plant is receiving more intense light and can transport electrons more efficiently. In contrast, a lower ratio suggests that the plant is better adapted to shade and can tolerate lower light levels region. Carotenoids, another class of pigments, give plants their bright colors and are responsible for absorbing light in the blue-green region. They also act as antioxidants, protecting the plant from the harmful effects of light [22]. The results from Figure 1 indicate that Inthanin plant leaves had a higher concentration of chlorophyll-a (3.413  $\pm$  0.01  $\mu$ g/ml) compared to chlorophyll-b (1.001  $\pm$  0.01  $\mu$ g/ml) and carotenoids  $(1.050 \pm 0.01 \,\mu\text{g/ml})$ . These results suggest that the *Inthanin* plant was exposed to high light intensities, which could be related to its growth environment. The solvent used to extract the pigments may have influenced the ratio of extracted pigments [23]. The study also revealed that the Inthanin leaves had a more intense shade of green with a lower carotenoid content. This could be due to the genetics of the plant.

#### 3.2. Characterization and Measurements

The photoelectric study was conducted on DSSCs made using a natural dye extract from Inthanin leaves, where the J-V curve of the cell was measured under white light irradiation (100 mW/cm²). The performance of the dye was evaluated using the short circuit current density ( $J_{sc}$ ), open circuit voltage ( $V_{oc}$ ), fill factor (FF), and power conversion efficiency ( $\eta$ ). Fig 1 displays the J-V curve of DSSCs sensitized with Inthanin dye extract. The results indicated that the Inthanin dye significantly influenced the DSSC performance as it can absorb sunlight and convert it into electrical energy [24].

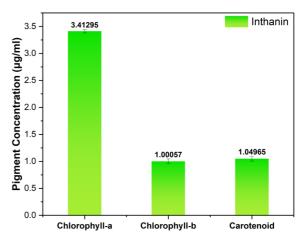


Fig. 3. Pigment Concentration of Inthanin Dye Extract

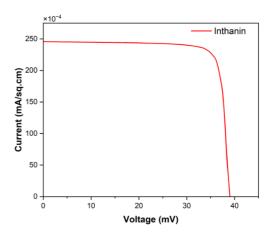


Fig. 4. J-V Curve of DSSC Sensitized with *Inthanin* Dye Extract

Table 1 displays the photoelectric characteristics of the DSSC) sensitized with an extract of *Inthanin* dye. The results reveal that the DSSC possesses an open circuit voltage ( $V_{oc}$ ) of 0.059 V, a short circuit current density ( $J_{sc}$ ) of 0.023 mAcm², a fill factor (FF) of 33%, and an efficiency ( $\eta$ ) of 0.1%. The high efficiency obtained by the DSSC is attributed to the higher values of the photoelectric properties:  $V_{oc}$ ,  $J_{sc}$ , FF, and  $\eta$ . The increase could result from the effective backscattering of light by larger TiO2 particles, leading to increased dye absorption [24]. The light scattering at the TiO2 interface is an essential phenomenon that influences the performance of DSSCs.

**Table 1.** J-V performance of DSSC Sensitized with *Inthanin* dye extract

Parameters	Values		
V <sub>oc</sub> (V)	0.059		
J <sub>sc</sub> (mA/cm2)	0.023		
I <sub>m</sub> (mA)	0.073		
V <sub>m</sub> (V)	0.054		
FF (%)	33		
Efficiency (η)	0.1%		

The efficiency of converting light into electricity in DSSC sensitized with natural dyes is significantly lower than that of artificial Ruthenium dyes. This is mainly due to the molecular structure of natural dyes, which is dominated by OH and C=O functional groups but lacks COOH like in the case of Ruthenium dyes. The COOH in Ruthenium dyes reacts with the hydroxyl on the TiO<sub>2</sub> particles to form an ester that enhances the coupling effect of electrons on the TiO2 conduction band, thereby facilitating the transport of electrons [26]. Additionally, the poor matching of energy levels between the excited states of natural dyes and the TiO2 conduction band and the poor adsorption of dyes on the TiO2 layer may contribute to the low light-toelectrical conversion efficiency. Furthermore, suppose the TiO<sub>2</sub> photoanode film is too thick, in that case, the incident light cannot penetrate through the entire layer, which limits the amount of dye molecules that can be excited by the light and therefore reduces the efficiency of the cell [27]. On the other hand, if the TiO<sub>2</sub> film is too thin the amount of dye adsorption on the TiO2 layer decreases, leading to lower efficiency. So, there should be an optimal thickness range for the photoanode layer, which balances then the amount of light absorption and the efficient transmission of excited dye molecules to the TiO2 surface. Similarly, acid can help prevent TiO2 nanoparticles from clustering together, leading to better dispersion of TiO2 and creating more places for the dye molecules to be adsorbed. This results in better performance of the DSSCs [28].

**Table 2.** Comparison of Dye Extracts for Solar Cell Performance

Dye Extract	Voc	Jsc	FF	Efficiency	Ref
	(V)	(mA/cm <sup>2</sup> )	(%)	<b>(</b> η)	
Curcumin-					[29]
100	527	0.332	0.552	0.097	
Citrus					[30]
limonum	478	0.90	0.398	0.171	
					[31]
Hibiscus	46.7	0.917	0.27	4.6*10^5	
Pink	0.31	0.55	0.3	0.05	
Frangipani					[5]
Thevetia	0.30	0.26	0.40	0.03	
					[32]
A. dentata	0.54	0.5	56.82	0.15	
Inthanin	0.059	0.023	33	0.1%	This Study

#### 4. CONCLUSION

The study emphasizes the criticality of understanding the roles of pigments in plant leaves for potential use in DSSCs. Chlorophyll mainly converts light into chemical energy, while chlorophyll-b aids in absorbing the energy that chlorophyll-a misses. Besides providing vibrant colors, Carotenoids act as antioxidants protecting the plant from light damage. The chlorophyll-a to chlorophyll-b ratio in leaves indicates the plant's light exposure. The results showed that the Inthanin dye extract had a higher concentration of chlorophyll-a  $(3.413 \pm 0.01 \,\mu\text{g/ml})$  compared to chlorophyll-b (1.001  $\pm$  0.01  $\mu$ g/ml) and carotenoids (1.050  $\pm$  0.01 µg/ml). This indicates that the plant receives more intense light and can transport electrons more efficiently. The study further evaluated the performance of the dye by measuring short circuit current density (Jsc), open circuit voltage (Voc), fill factor (FF), and power conversion efficiency ( $\eta$ ). The results revealed that the DSSC had an open circuit voltage (V<sub>oc</sub>) of 0.059 V, a short circuit current density (Jsc) of 0.023 mA/cm<sup>2</sup>, a fill factor (FF) of 33 %,

and an efficiency ( $\eta$ ) of 0.1%. The increase in efficiency is attributed to the effective backscattering of light by larger TiO<sub>2</sub> particles. Even though the efficiency of DSSCs enriched with natural dyes currently lags behind their synthetic ruthenium dye counterparts, the study offers essential clues towards enhancing performance by employing larger TiO<sub>2</sub> particles for effective light backscattering. Further exploration in this domain may pave the way toward creating more effective, sustainable, and budgetfriendly solar cells, which are also benign to the environment.

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#### CONFLICT OF INTEREST

The authors of this paper assert that they have no conflicts of interest regarding the publication of this article.

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