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Review/Research Article

Effect of Solution pH and Concentration of Dewandaru Fruit Extract as a Natural Dye on Optical Characteristics and Performance of Dye-Sensitized Solar Cells

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Abstract. Given growing global energy demand and the need for sustainable solutions, solar energy is a promising alternative. Dye-Sensitized Solar Cells (DSSC) represent a cost-effective photovoltaic technology, and natural dyes as sensitizers have significant potential to enhance their efficiency. This study explores the use of natural dye from Dewandaru fruit (Eugenia uniflora) in DSSCs by varying pH levels and concentrations. Natural dyes offer an environmentally friendly and sustainable alternative compared to synthetic dyes, which are often expensive and not eco-friendly. In this research, Dewandaru fruit extract was used to sensitize TiO2 photoanodes, and the effects of varying pH (1.00, 1.66, 2.27, and 3.00) and dye concentrations (2.5, 5.0, 7.5, and 10%) on optical properties and performance were analyzed. UV-Vis spectroscopy, FTIR, cyclic voltammetry, and J-V measurements assessed absorbance, energy levels, and efficiency. Optical characterization results showed that changes in pH and concentration can affect the dye's absorbance and energy band gap. The optimal DSSC performance for the dye was found at pH 3.00, yielding a Voc of 0.425 V, Jsc of 0.053 mA/cm², fill factor of 68.7%, and efficiency of 0.1548%. The best dye concentration was 10%, achieving a Voc of 0.45 V, Jsc of 0.105 mA/cm², fill factor of 69.0%, and efficiency of 0.322%. The statistical analysis indicates that pH substantially and positively influences DSSC efficiency and band gap energy. Moreover, dye concentration significantly affects DSSC efficiency, light harvesting efficiency, Jsc, and fill factor, with the most substantial impacts observed on efficiency and Jsc. However, concentration appears to have less effect on dye absorbance, band gap energy, and the HOMO/LUMO values. The study indicates that Dewandaru fruit extract has potential as a natural dye for DSSCs, with varying performance based on pH and concentration.

Keywords: Dye sensitized solar cells; Natural dye, *Eugenia uniflora,* pH solvent, Concentration

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1. Introduction

Over the past few years, the world's population has increased massively, rapidly increasing energy demand. As a result, fossil fuel reserves on Earth are quickly being depleted. Energy consumption is estimated to grow from 19.2 TW per year to 24.6 TW per year from 2015 to 2040 (Carella et al. 2018; Sukarno et al. 2015). Using this template will enable you to prepare your manuscript by the instructions for authors for International Journal of Renewable Energy Development (IJRED) papers with a minimal amount of manual styling and formatting. In addition to depleting supplies due to massive use, fossil fuels are a source of electrical energy that produces greenhouse gas emissions such as carbon dioxide, carbon monoxide, and sulfur dioxide, which result in global warming (Omer 2012; Singh et al. 2013).

Technological sources that are abundant and do not cause adverse effects on the environment are needed to realize a sustainable life (Kabeyi and Olanrewaju 2022). Solar energy can be

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a promising answer because it is a clean energy source, considered inexhaustible, and does not incur costs (O'Regan and Grätzel 1991). Solar energy can be converted directly into heat, so researchers use solar energy as a power source (Rahman, S et al. 2023). Dye-sensitized Solar Cells (DSSC) are one of the most cost-effective technologies researched in recent years (Yan et al. 2025). Manufacturing these solar cells requires relatively low cost because the cells are made of low-cost materials, and the assembly is easy (Rajkumar et al. 2025).

Third-generation cells aim to surpass the efficiency limits of previous generations while being cost-effective (Sharma et al. 2022). The DSSC, first developed in 1991 by O'Regan and Grätzel, had an energy conversion efficiency of 7% (O'Regan & Grätzel, 1991). Nanostructured metal oxide films, which form the basis of DSSC devices, are sensitized by adsorbed dyes to harvest visible light. The components for DSSC assembly consist of transparent conductive glass as a substrate, a semiconductor with a wide band gap, an electrolyte, and a counter electrode (Prima, Eka Cahya et al. 2024). Transparent conductive glasses commonly used as substrates are Fluorine-doped Tin Oxide (F:SnO_2) and Indium Tin Oxide (In: SnO_2) (Bekele and Sintayehu 2022). For semiconductor materials, the widely used material in these devices is TiO2. TCO (Transparent Conducting Oxide) coating on the substrate is the electrode and counter electrode. The counter electrode is coated with an electrolyte commonly used as I^-/I_3^- (iodide/triiodide) to accelerate the redox reaction (Prima, Eka Cahya et al. 2020). The dye is oxidized because it absorbs photons, and energized electrons are injected into the semiconductor conduction band. After being injected, the electrolyte will work to collect electrons due to redox reactions, resulting in a cycle of light conversion into electricity (Bhogaita et al. 2016).

The pigment in dye-sensitized solar cells (DSSCs) plays a crucial role in determining the efficiency of the cells. The efficiency of DSSCs is significantly influenced by the ability of the sensitizer to absorb a wide range of the solar spectrum (Alessa et al. 2024). Generally, Ruthenium-bipyridyl is used as a dye in DSSC devices, producing high energy conversion efficiencies of around 11-12% (Saud et al. 2024; Singh, Shruti et al. 2021). Ruthenium (Ru) is a suitable sensitizer for DSSCs because of its strong charge transfer absorption and efficiency in visible light. Due to the high price of Ruthenium (Ru), researchers began to develop metal-free organic sensitizers. Natural dyes are a good choice to replace Ruthenium (Ru) as a sensitizer. Natural dyes can be found in fruits, flowers, leaves, roots, etc. (Calogero et al. 2015). The choice of natural dyes as sensitizers is not only because of their low cost but also because of their ease of manufacture, making them a practical solution. Pigments such as anthocyanins and carotenoids in natural dyes make the pigments photoactive and can be adsorbed on the surface of TiO_2 (Eka et al. 2013). Although these natural dyes are suitable for DSSCs, their chemical structure is a significant problem because they are difficult to change in vitro. This is because plants evolve naturally (Prima, E. C. et al. 2018).

Because of these limitations, alternatives are needed to maximize the function of natural dyes in DSSCs, such as changes in pH and concentration (Kumara, N et al. 2017). The type of solvent is essential in DSSC fabrication because the acidity level can affect the absorption spectrum of the dye as well as the bonding of the dye with TiO2, which will have an impact on the optical characteristics and performance of the DSSC (Bekele and Sintayehu 2022; Teja et al. 2023). The same thing will also happen if there is a change in the concentration of the extractor in the solvent (Faqih et al. 2019; Muryani et al. 2019).

Dewandaru fruit (Eugenia uniflora), also known as pitanga, is rare in Indonesia (Seidemann 2005). This fruit, belonging to the Myrtaceae family, is native to Central and South America, particularly in countries like Brazil, Argentina, and Paraguay (da Silva Diniz et al. 2014). Although it is appreciated for its health benefits and ornamental potential, it is not widely known or cultivated in Indonesia. When used as a sensitizer, this is one of the first systematic investigations of its optical and photovoltaic characteristics (Magalhães et al. 2025). While several other natural dyes, such as anthocyanins from blackberries, mangosteen, and hibiscus, have been previously explored (Maiaugree et al. 2015; Rajkumar et al. 2025; Raturi and Fepuleai 2010), Dewandaru remains underreported despite its high anthocyanin and phenolic content (Santoso et al. 2020)

This fruit is also called Brazilian cherry because it mainly grows in Brazil. Pitanga fruit contains phenolic compounds, including 69 mg/100 g of anthocyanins. Where the content is more excellent than frozen grapes (Vitis vinifera), which is 30.9 mg/100 g, and strawberries (Fragaria vesca), which is 22.8 mg/100 g (Bagetti et al. 2011). The high reactivity of anthocyanins depends on their structure and several factors such as pH, temperature, light, oxygen, and enzymes. pH is an essential factor affecting anthocyanin stability. Generally, anthocyanins are more stable in acidic environments with a pH <7. The advantage of anthocyanin as a natural colorant in DSSC fabrication lies in its structure's carbonyl and hydroxyl groups, which facilitate attachment to the TiO2 layer, increasing energy conversion efficiency (Pramananda et al. 2021).

This research investigates the potential of Dewandaru fruit-derived natural dye in DSSCs by analyzing the impact of varying pH levels and dye concentrations on the resulting optical properties and device performance, aiming to provide a sustainable alternative to synthetic dyes. We report the first-ever use of Dewandaru (Eugenia uniflora) fruit extract as a dye in DSSC applications. Additionally, our research simultaneously investigates two influential parameters: pH and concentration, which have not been commonly done in previous literature. We provide more understanding of dye optimization by analyzing how these parameters affect the optical properties, HOMO-LUMO energy levels, and photovoltaic performance. We also compare direct performance with a commercial dye (N719) under the same conditions. We believe this comprehensive approach represents a meaningful advancement in the exploration of low-cost, eco-friendly photosensitizers for DSSCs and provides new directions for utilizing underutilized tropical fruits in green energy technologies.

2. MATERIALS AND METHODS

2.1 Dye Extraction

A total of 1 kilogram of dewandaru fruit samples was washed and cleaned; then the seeds were removed. A total of 750 grams of dewandaru fruit removed from the seeds was then dried using a vacuum oven for three days and crushed until smooth using a blender (Cosmos, 450W). After pulverizing the dried dewandaru fruit, the results were 245 grams of powder samples. To prepare variations in solution pH, methanol solvent was mixed with acetic acid in a ratio of 95:5 (%v/v) with a total volume of 200 ml divided into four equal parts, and then HCl (Merck) was added as an acid control (López et al. 2017). The four solutions had a pH of 1.00, 1.66, 2.27, and 3.00. The fine powder was put into each solvent in a 1:20 (w/v) ratio, stirred using a magnetic stirrer at 500 rpm until evenly mixed, then isolated for seven days at room temperature. After that, the solution was filtered using Whatman filter paper (Merck, 11 µm pore size) with the help of the Filtration Buncher Vacuum Kit at 0.1 bar. The thick extract obtained from the filtration result was centrifuged at 4000 rpm for 10 minutes. Then, the results were characterized, and one of the dye solutions was prepared in concentrations of 2.5, 7.5, and 10 (%w/v) with the same manufacturing process. For the control pigment, a N719 dye-photosensitizer was prepared in a solution containing 19 mL H2O, 80 mL MeOH, and 1 mL HCl, resulting in a 2.2 mM pigment concentration (Prima, Eka Cahya et al. 2024)

2.2 Film Preparation

TiO2 nanoparticle electrode on FTO (Pilkington, TEC-8, 8Ω/sq, received from Yingkou OPV Tech New Energy Co., Ltd., China) was immersed in 10 ml dye solution. The FTO substrate, in this case, Pilkington TEC-8 glass, serves as a transparent conductive oxide, enabling light to pass through while also providing an electrical contact for collecting the generated electrons, and its low sheet resistance (8 Ω/sq) ensures minimal resistive losses, contributing to improved overall cell performance. Similar works on using TiO2-adsorbed FTO substrate were reported by Chen et al. (2020); Chen et al. (2023), and Zhang et al. (2020).

The maceration was carried out in a refrigerator for 7 days to prevent the dye from evaporating. After the immersion, the photoanode was cleaned with 2-propanol and a cotton bud without scratching the thin film, and then allowed to dry. ITO conductive transparent glass (6Ω/sq, 90% transmittance) was cut into a 1×2 cm size for counter electrode preparation. Then, the cut glass was cleaned using an ultrasonic cleaner for 15 minutes. After cleaning, the glass substrate was rinsed with 2-propanol, then dried.

The counter electrode was prepared by dripping 25 µl of 20 mM hexachloroplatinic acid (Aldrich) solution onto the cut-ITO substrate. The solution was evenly applied to the conductive part of the glass substrate. Then, spin coating was carried out at a speed of 4000 rpm for 30 seconds and then heated at 80°C for 5 minutes. When the temperature had returned to normal, the platinum electrode was annealed at 380°C for 30 minutes. The annealed counter electrode was allowed to dry.

2.3 Fabrications

Around the TiO2 thin film, Low Temperature Thermoplastic Sealant (25µm thickness, Dyesol) was applied, and then EL-HPE electrolyte (Dyesol, mixture of solvents, a redox couple (triiodide/iodide), and additives including acetonitrile and valeronitrile as solvents. The redox couple is I3-/I-, and additives included inorganic iodide salts, organic iodide salts, and pyridine derivatives, which were dripped about 20 µl. Then the two electrodes were joined and clamped with a paper clip. After that, the DSSC sandwich was heated using an oven at 80°C for 5 minutes. This was done to melt the thermoplastic sealant that will bind the DSSC sandwich.

2.4 Characterizations

Fourier Transform Infrared (Shimadzu with model FTIR-8400) was used to identify chemical bonds by analyzing the vibrational spectra produced by compounds at specific wave numbers. In addition, FTIR was also used for quantitative analysis to calculate the intensity value of the absorption peak. The characterization process with FTIR involved analyzing the resulting spectrum according to the peaks formed by certain functional groups. The compound could absorb electromagnetic radiation in the infrared region with wave numbers between 500 to 4000 cm-1.

A UV-Vis spectrophotometer (UV-1600PC) was used to determine the dye's absorbance ability and the TiO2 that has been colored. Characterization was done by testing the dye sample photoanode at 200-800 nm wavelengths. The characterization aimed at obtaining absorbance values and wavelengths that could be absorbed. Then this absorbance value could be used to calculate the percentage of Light Harvesting Efficiency (LHE) and the band gap value of the sample (Liu et al. 2019). Meanwhile, to determine the morphology of the TiO2 thin layer, the HitachiTM SU-3500 Scanning Electron Microscope (SEM) was used at PPNN ITB.

The data obtained was plotted into a graph depicting the wavelengths absorbed by this layer. Then, a graph plot of αhv2 against photon energy after determining the absorption coefficient can be used to determine the band gap value (Mursyalaat et al. 2023).

$$h\nu = 1240/\lambda \quad (1)$$

$$\alpha = A/l \quad (2)$$

$$\alpha h\nu^2 = K(h\nu - E_g) \quad (3)$$

λ is the wavelength, A is the absorbance, and l is the surface area. The band gap value can be determined by projecting a tangent to the curve, carried out with the x-axis intersecting point αhv2 = 0 (Tauc, Jan 1968; Tauc, J. et al. 1966). To find the percentage of LHE, we must first determine the absorbance coefficient value of each sample using Equation 2, which is then substituted into Equation 4.

$$\text{LHE}\ (\%) = (1\text{-}10\text{-}\alpha) \times 100\% \quad (4)$$

Cyclic voltammetry was used to determine the LUMO and HOMO energy levels of the dye, performed using a CS-310M electrochemical workstation (potentiostat/galvanostat). The test was conducted using three electrodes: glassy carbon was used as the working electrode, the counter electrode was platinum, and Ag/AgCl was the reference electrode. The electrolyte solution was 10 mL of 0.1 M KNO3, which was dripped with dye—data generated in the range -1.8 V to +1.8 V (Prima, Eka Cahya et al. 2024).

The data obtained from cyclic voltammetry, specifically the current and voltage values, played a crucial role in calculating the band gap of the dye. These values were processed to determine the amount of reduction and oxidation potential energy. The resulting values were then utilized in Equations 5 and 6, underscoring the importance of cyclic voltammetry in our research (Heinze 1984; Nicholson 1965).

$$\text{HOMO} = -(E_{ox} + 4.4) \quad (5)$$

$$\text{LUMO} = \text{HOMO} - E_g \quad (6)$$

The J-V characterization of the DSSC was conducted using the advanced Solar Standard Simulator AM 1.5G filter with ADCMT 6241A Source Meter, which provided precise measurements at 100 mW/cm2 (Touihri et al. 2021). The results of this characterization were processed to obtain data on short-circuit current density (Jsc), open-circuit voltage (Voc), fill factor (FF), and DSSC efficiency (η) using Equations 7 and 8 (Atia and Ahmed 2023).

$$FF = (V_{max} \times J_{max}) / (V_{oc} \times J_{sc}) \quad (7)$$

$$H = (V_{oc} \times J_{sc}) / P_{in} \times FF \quad (8)$$

The Jsc value was determined by isolating the current value when the voltage was zero and then dividing it by the surface area of the layer. Similarly, the Voc value was determined by identifying the magnitude of the voltage when the current was close to zero. The Vmax and Jmax values, representing the most significant power the DSSC can generate, are then determined (Ghann et al. 2017; Pathak et al. 2019)

3. Results and Discussion

3.1 TiO2 Microstructure Analysis

Scanning Electron Microscope (SEM) analysis was employed to examine the surface morphology of the TiO_2 layer. As shown in Figure 1, the SEM image revealed that the **TiO_2 surface was composed of mesoporous granules.** The size of the granules was verified using ImageJ software, resulting in an analysis of distributed particles with an average size of 64 nm.

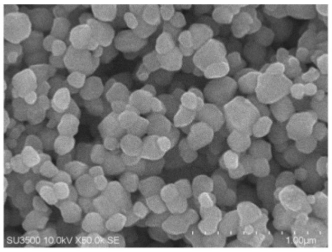

Figure 1. TiO_2 film's Scanning Electron Microscope

Specifically, the uniformity and small size of the grains help minimize aggregation, reducing the dye adsorption on the TiO_2 surface. On the other hand, the crystalline structure of the nanoparticle TiO_2 surface can increase the adsorption of dye molecules. Apart from the pores, the thickness of the TiO_2 layer also plays a role in dye molecule absorption. In this study, the TiO_2 layer thickness was 9.1 µm, which is within the typical range for TiO_2 thin films, generally between 7-20 µm (Al-Bat'Hi et al. 2018). Thicker TiO_2 layers generally lead to increased dye adsorption. This is because a thicker layer provides a larger surface area for the dye molecules to attach, enhancing the overall dye loading capacity (Rahman, MF et al. 2020). Moreover, thicker TiO_2 layers can improve light absorption by increasing the path length of light within the layer, enhancing the probability of photon absorption by the dye molecules (Dissanayake et al. 2020).

3.2 FTIR Analysis

The functional groups that make up the pigments of the dewandaru fruit need to be known to determine whether this fruit can be used as a colorant in DSSCs. Fourier transform infrared (FTIR) characterization produces transmittance graphs at wave numbers 4000 to 500 cm-1, shown in Figure 2. The graph was analyzed to determine the functional groups of the dye constituent molecules.

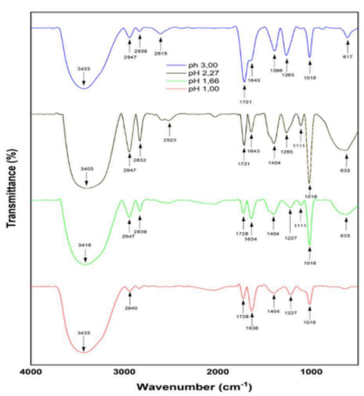

Figure 2. FTIR spectra of dye samples

Table 1, FTIR absorption peak identification

pH 1,00		pH 1,66		pH 2,27		pH 3,00		
k **(*cm*⁻¹)**	T (%)	***k*** **(*cm*⁻¹)**	T (%)	***k*** **(*cm*⁻¹)**	T (%)	***k*** **(*cm*⁻¹)**	T (%)	Functional Group
3433	15	3418	18	3403	6	3433	22	O-H hydroxyl
2940	94	2947	78	2947	52	2947	88	C-H alkane
-		2839	86	2832	66	2839	95	
-		-		2532	92	2615	92	-COOH carboxyl
1728	84	1728	83	1721	60	1721	31	C=O carbonyl
1636	69	1634	77	1643	77	1643	58	C=C aromatic
1404	91	1404	82	1404	62	1396	71	C-H alkane
1227	89	1227	90	1265	76	1265	65	
-		1111	93	1111	85	-		C-O ether
1018	77	1018	43	1018	11	1018	62	
-		633	82	633	54	617	88	C-H aromatic

There are 11 absorption peaks of this dye molecule that can be seen in Figure 2. These peaks, which are summarized and analyzed in Table 1, are significant as they indicate the presence of specific functional groups in the molecule. In the single bond region, which is from wave numbers 2500-4000 cm-1, there are 4 absorption peaks. The hydroxyl group (O-H) is indicated by peaks at wave numbers 3433 cm-1, 3418 cm-1, and 3403 cm-1. The peak at wave numbers 2960-2850 cm-1 indicates a C-H stretch with aliphatic groups, namely alkanes. Acetic acid as a solvent influences the presence of -COOH bonds from carboxyl groups at wave numbers 2532 cm-1 and 2615 cm-1. No absorption peaks were detected for the triple bond region at wave numbers 2000-2500 cm-1. In the double bond region, two groups of peaks were detected, namely at 1721-1728 cm-1 showing carbonyl groups (C=O) and at the peak of 1634-1643 cm-1 showing the stretching of aromatic double bonds (C=C. There are several peaks in the fingerprint area, namely at wave numbers 500-1500 cm-1. At peaks at wave numbers 1404 cm-1 and 1396 cm-1, there are C-H bonds from alkanes (Golshan et al. 2021; Singh, Shalini et al. 2021). C-O bonds of ether groups were detected at 1000-1300 cm-1 wave numbers (Unwakoly et al. 2025). At 677 cm-1 and 633 cm-1, peaks indicate vibrations in aromatic C-H bonds (Nandiyanto et al. 2019; Thummajitsakul and Silprasit 2022).

The statistical analysis of the FTIR absorption data for the Dewandaru fruit pigments across different pH levels (1.00, 1.66, 2.27, and 3.00) reveals several trends in the transmission percentages (T% %) of key functional groups. The C-H alkane group (2940 cm^{-1}) exhibits the highest transmission at pH 1.00 (94%). Still, it shows a significant decrease at pH 2.27 (52%), indicating that the stability of this group decreases as pH increases, though it recovers at pH 3.00 (88%). Similarly, the carbonyl group (1728 cm^{-1}) is most prominent at pH 1.00 (84%) and pH 1.66 (83%) but declines dramatically at pH 3.00 (31%), suggesting that acidic conditions favor this functional group. The carboxyl group (2532 cm^{-1}) remains stable at pH 1.66 and pH 2.27 (both 92%), with no absorption at pH 1.00 and pH 3.00, highlighting its role in the mid-range pH environment. The O-H hydroxyl group (3433 cm^{-1}) shows variability, peaking at pH 3.00 (22%) but dipping at pH 2.27 (6%), suggesting that the hydroxyl group's contribution fluctuates with pH. Other groups, such as the ether (1227 cm^{-1}) and aromatic C-H (633 cm^{-1}), show a decreasing trend in transmission as pH increases, with the ether group reaching its peak at pH 1.66 (90%). The data indicate that functional groups like C-H alkane and carbonyl are more stable in acidic environments. In contrast, the carboxyl group thrives at mid-range pH levels, and the O-H hydroxyl group exhibits pH-dependent variation. These trends suggest that the pH environment plays a significant role in the stability and presence of these functional groups in the Dewandaru pigments.

The functional groups in Table 1 correspond to the main functional groups that make up anthocyanin pigments. Mattioli et al. (2020) mentioned that at pH below 3, anthocyanins primarily exist in the form of red flavylium cations that carry a positive charge on oxygen. This can be from hydroxyl and carbonyl groups that are stable in acidic conditions.

3.3 Effect of Dye Solution pH on Optical Characteristics of Active Materials

Figure 3 shows that the absorption rate tested using a UV-VIS spectrophotometer with pH variation is similar, but the intensity is different at some wavelengths. The absorption peak begins at a wavelength of 315-400 nm and increases until the most optimal peak occurs at a wavelength of 400-480 nm, as shown in Table 2. The graph shows a decrease at a wavelength of 500 nm, indicating that the dye absorption ability decreases until the wavelength of 700 nm. **Therefore, the** dye from dewandaru fruit extract can absorb ultraviolet waves and visible light, where the absorption of ultraviolet-A and purple and blue colors is the most optimal wave absorbed by this dye.

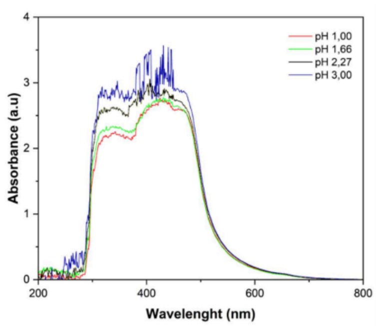

Figure 3. Dye absorption spectrum with pH variation in the UV-Vis range

The absorption intensity in the 300-500 nm wavelength range is best observed by the dye solution at pH 3.00. This suggests that the dye solution's pH can influence the dye's absorption characteristics. While the absorption intensity difference is insignificant at other wavelengths, the dye solution's absorption ability decreases as the solution's acidity increases. This occurs because at low pH, dye molecules undergo protonation, which causes changes in electron structure and causes a reduction in the dye's ability to absorb light at specific wavelengths, resulting in a decrease in absorbance intensity (Alkorta and Picazo 2005). In addition, the dye has a system of double bonds that are close together and interact, known as a conjugated double bond system. Changes in pH can affect this system. For example, adding protons (H^+) at low pH can disrupt the conjugated double bond system, reducing color intensity. Protonation can significantly affect the conjugated π-electron system of dye molecules. This disruption can change the dyes' absorption spectra and color intensity (Mediantsev et al. 2024). Anthocyanin pigment from mangosteen peel is reported to have optimal absorption at a 400-480 nm wavelength with a peak absorption around 430 nm (Pratiwi et al. 2017).

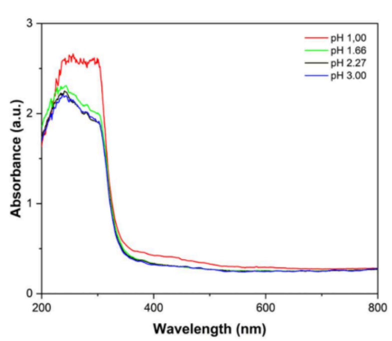

Figure 4. Absorption spectrum of TiO2 with pH variation in the UV-Vis range

The results of UV-VIS Spectroscopy testing on TiO2 thin films that have been soaked in dye solution are shown in Figure 4. The difference in absorption intensity is quite significant, as demonstrated by TiO2 submerged in dye solution at pH 1.00, both in ultraviolet waves and visible light. Absorption in ultraviolet waves strengthened along with the acidity of the dye solution that was applied to TiO2 thin films. While in the visible light range, absorptions on TiO2 submerged in dye solutions at pH 1.66, 2.27, and 3.00 have similar intensities. The absorption peak was detected at a wavelength of 250-350 nm, meaning there is a peak shift and a decrease in absorption intensity in the ultraviolet wavelength and an increase in the visible light range compared to the dye without TiO2. This result shows that TiO2 changes the dye's behavior, improving its ability to absorb light in the UV and visible ranges. The peak shift and changes in absorption suggest that the TiO2 and dye interaction is altering their optical properties (Hegazy 2019).

Yadav et al. (2021) reported that the intensity of visible light absorption increases after dye molecules are adsorbed on the TiO2 surface. This indicates that the interaction between dye molecules and TiO2 can affect the optical properties of the dye, resulting in an increase in absorption intensity and changes in the absorption spectrum. Similarly, the research conducted by Pratiwi et al. (2016), there is an increase in the absorption spectrum of TiO2 thin films in the visible light range, which means that the application of dye on the TiO2 surface can be used as a photosensitizer for DSSC absorption in this range. The absorption spectrum of TiO2 with pH variation provides valuable insights into the potential use of dye-TiO2 systems in DSSCs. Table 2 summarizes the absorption peak data on the solution and TiO2 samples.

Table 2. Peak absorption in dye and TiO2 solution samples with pH variation

pH	Dye sample		TiO2 + Dye sample	
	$\lambda max\ (nm)$	Amax (a.u.)	$\lambda max\ (nm)$	Amax (a.u.)
1.00	425	2.77	256	2.66
1.66	431	2.78	244	2.31
2.27	406	3.06	241	2.25
3.00	431	3.57	238	2.21

Absorption data from UV-Vis testing were analyzed to obtain the energy band value at each pH variation of the dye solution. Figure 5 shows the Tauc's Plot with the value of (αhv)2 on the y-axis and the energy value on the x-axis. Tauc's Plot analysis is done by drawing a line that intersects with the graph and intersects with the x-axis, so that the band gap value of 1.80 eV is obtained for pH 1.00, 1.81 eV for pH 1.66, 1.81 eV for pH 2.27, and 1.82 eV for pH 3.00.

The pH variation in the dye extractor solution affects the band gap value. The band gap will decrease when the pH is more acidic (Saud et al. 2024). Increased acidity can cause dye molecules to break down. This occurs because of the interaction between dye molecules and hydrogen ions (H+) in an acidic environment. As a result, the dye molecules break down into simpler compounds (Lozano-Alvarez et al. 2015).

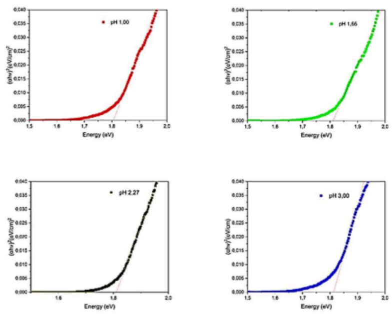

Figure 5. Tauc's plot of the dye sample with pH variation

Light absorption efficiency (LHE) is one of the optical properties that influences DSSC performance. Figure 6 illustrates the light harvesting efficiency (LHE) at each wavelength, calculated using Equation 4. The data reveal that at pH 2.27, the LHE reaches its highest average value of 34.85%, indicating optimal light absorption efficiency under this acidic condition. In comparison, at pH 1.00, 1.66, and 3.00, the average LHE values are slightly lower, with 33.62%, 34.66%, and 33.55%, respectively. While pH 1.66 shows a value close to pH 2.27, the efficiency at pH 1.00 and 3.00 is slightly reduced, suggesting that the pH level of 2.27 may offer the most favorable environment for light absorption. The similar values observed at pH 1.00, 1.66, and 3.00 indicate that while pH affects the LHE, the difference is not drastic, with pH 2.27 standing out as the condition with the highest efficiency.

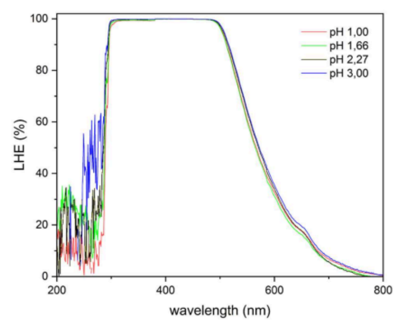

Figure 6. Dye samples' light absorption efficiency (LHE) with pH variation

Figure 7 shows the results of the cyclic voltammetry Characterization test conducted to determine the voltage at which oxidation occurs in the dye molecule. By using Equation 5, the voltage is analyzed to determine the Highest Occupied Molecular Orbital (HOMO) level. At the same time, the Lowest Unoccupied Molecular Orbital (LUMO) level value is obtained by summing the HOMO value with the energy band gap value, as in equation (3.6). The HOMO and LUMO values are summarized in Table 3.

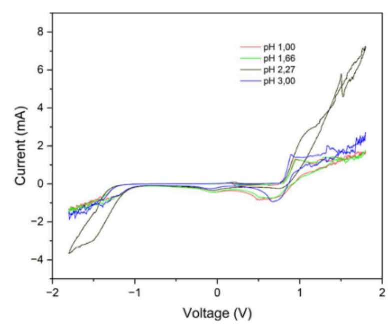

Figure 7. Cyclic Voltammetry characterization test results on dye samples with pH variations

The HOMO and LUMO values are not directly affected by acidity. Variations in pH affect the width of the energy band gap, which will affect the HOMO and LUMO values of the dye sample (Mikheev et al. 2017). Research conducted by Barbinta-Patrascu et al. (2024) experienced similar results on the HOMO and LUMO values of Curcuma Longa extract, where at pH 4.00 the HOMO and LUMO values were -5.87 eV and -4.24 eV, and at pH 3.5 the values were -5.83 eV and -4.28 eV. At pH 3.0, the values were -5.91 eV and -4.22 eV, respectively.

Table 3. Energy level and light absorption efficiency of dye samples with varying solvent pH

pH	HOMO (eV)	LUMO (eV)	Band gap (eV)	LHE (%)
1,00	-5.19	-3.39	1.80	33.62
1,66	-5.25	-3.44	1.81	34.66
2,27	-5.21	-3.40	1.81	34.85
3,00	-5.26	-3.44	1.82	33.55

3.4 Effect of Dye Solution pH on Performance of DSSC

J-V characterization testing at sunlight intensity was conducted to measure the performance of the DSSC. The values of open circuit voltage (Voc) and short circuit current density (Jsc) were obtained from this test. Equation 8 is used to calculate the two values to get the efficiency value of each dye sample. This test was also carried out on a conventional dye sample, N719, as a standard comparison.

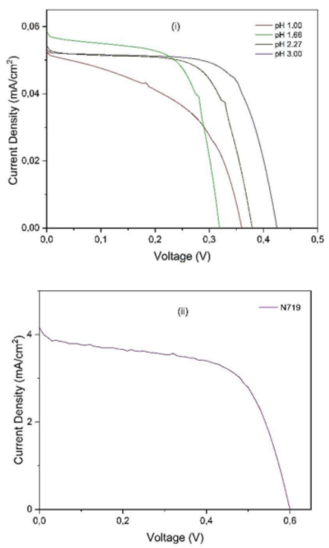

Figure 8. J-V curves of DSSC samples with dye, (i) Eugenia uniflora with pH variation; (ii) N719

DSSC samples with dye from Eugenia uniflora fruit with varying acidity levels produced Voc values between 0.32 and 0.42 Volts and Jsc values between 52 µA/cm2 and 59 µA/cm2. The most significant Voc value is owned by the DSSC sample with pH 3.00, with a value of 0.42 Volts. The most considerable Jsc value is owned by the DSSC sample with pH 1.66. According to the acidity level, the Voc and Jsc values have no consecutive trends. This can occur due to the suitability of HOMO and LUMO energy levels that allow electron injection and dye regeneration to take place efficiently (Conradie, 2024).

Table 4. Performance of DSSC samples with dye from Eugenia uniflora fruit extract with pH variation

pH	Voc (V)	Jsc (µA/cm2)	FF (%)	η (10-3%)
1.00	0.360	52.5	48.4	9.15
1.66	0.319	58.6	63.9	11.93
2.27	0.379	54.4	66.1	13.42
3.00	0.425	53.0	68.7	15.48

Table 4 shows that DSSC samples with pH 3.00 have the best efficiency and decrease as the solution's acidity increases. In addition to the direct influence of Voc and Jsc values, the efficiency value is also indirectly influenced by the fill factor value (Conradie 2024). The efficiency value increases as the fill factor value increases in each DSSC sample with Eugenia uniflora fruit extract dye. Research conducted by Chien and Hsu (2013) found that the efficiency of anthocyanin dye from Brassica oleracea reached its peak at pH 8.00, and there was a decrease in efficiency as the acidity of the solvent increased. For comparison, other research data are summarized in Table 5.

Table 5. DSSC sample comparison

Dye	Voc (V)	Jsc (mA/cm2)	FF (%)	η(%)	Ref.
Eugenia uniflora, pH 1,00	0.360	0.0525	48.4	0.00915	-
Eugenia uniflora, pH 1,66	0.319	0.0586	63.9	0.01193	-
Eugenia uniflora, pH 2,27	0.379	0.0544	66.1	0.01342	-
Eugenia uniflora, pH 3,00	0.425	0.0530	68.7	0.01548	-
N719	0.694	8.23	61.1	3.49212	-
N719	0.846	17.73	75.0	11.18	(Conradie 2024)
Oryza sativa + N719	0.558	5.69	61.3	1.95	(Prima, Eka Cahya et al. 2020)
Brassica oleracea, pH 2,00	0.533	1.449	71.1	0.555	(Chien and Hsu 2013)
C. xanthorrhiza + A. cordiofolia , pH 1,77	0.320	2.5	-	0.096	(Setiarso et al. 2023)

In DSSC samples with conventional dye N719, the application of the same components produces an efficiency of 3.49%. Far different from the best efficiency value owned by DSSC samples, with dewandaru fruit extract dye, it is 0.01548%. The natural dye (Eugenia uniflora) has lower light absorption and energy conversion efficiency compared to the synthetic N719 dye, which shows higher short-circuit current density (Jsc) and open-circuit voltage (Voc). N719 also has better charge transfer and collection properties, efficiently injecting electrons into the TiO_2 semiconductor. While natural dyes offer environmental benefits, their efficiency lags behind synthetic dyes. Ongoing research aims to bridge this gap through various optimization strategies, including co-sensitization, electrode modifications, and improved dye extraction methods (Kharkwal and Dhawan 2024).

Table 6. Statistical analysis for pH's Dye Solution on DSSC performance

Pearson Correlation		pH	Dye absorbance	BandGap	LHE	Voc	Jsc	FF	HOMO	LUMO	η
η	Correlation Coefficient	0.993**	0.885	0.970*	0.063	0.668	-0.009	0.935	-0.733	-0.620	1**
	Sig. (2-tailed)	0.007	0.115	0.030	0.937	0.332	0.991	0.065	0.267	0.380	0.007
	N	4	4	4	4	4	4	4	4	4	4

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

Table 6 shows the statistical analysis of the effect of pH on the performance of DSSC, revealing significant correlations between the pH of the dye solution and several key optical characteristics and performance parameters. The Pearson correlation coefficient for the pH and the dye's efficiency (η) was 0.993, which is highly significant (p = 0.007), indicating a strong positive correlation between the pH of the dye solution and DSSC efficiency. This suggests that as the pH increases, the efficiency of the DSSC also improves, which is consistent with the observed performance trends in the study, as found by Isah et al. (2015).

The correlation between pH and dye absorbance was also substantial (0.885). However, it was not statistically significant (p = 0.115), indicating that while there is a positive relationship between pH and dye absorbance, the effect is not as prominent as that on efficiency. On the other hand, the pH and band gap energy exhibited a significant positive correlation of 0.970 (p = 0.030), suggesting that changes in pH directly influence the band gap energy of the dye, affecting the DSSC's performance.

Interestingly, the correlation between pH and the light harvesting efficiency (LHE) was weak and statistically insignificant (0.063, p = 0.937), indicating that pH had little impact on this particular optical characteristic. Similarly, while pH showed a moderate positive correlation with Voc (0.668), Jsc (-0.009), and HOMO (0.935), these were not statistically significant (p = 0.332, p = 0.991, and p = 0.065, respectively). This suggests that while pH might have some influence on these parameters, its effect is not as strong or consistent as on the efficiency and band gap energy. Similarly, the band gap of theaflavin in black tea waste extract (BTE) was reduced by 0.17 eV when the pH was lowered (Kumara, NTRN et al. 2015).

The correlations between pH and LUMO (-0.733) and pH and fill factor (FF) (-0.620) were negative, with neither showing statistical significance (p = 0.267 and p = 0.380, respectively). This indicates that changes in pH did not have a significant negative impact on these parameters. The statistical analysis shows that pH substantially and positively influences DSSC efficiency and band gap energy. At the same time, its effects on other parameters such as LHE, HOMO, Jsc, and FF are less pronounced or not statistically significant. These findings underscore the importance of optimizing the pH of the dye solution to enhance the overall performance of DSSC.

3.5 Effect of Dye Solution Concentration on Optical Characteristics

Figure 9 shows the ability of dye absorption against wavelengths in the visible light and ultraviolet wavelength range.

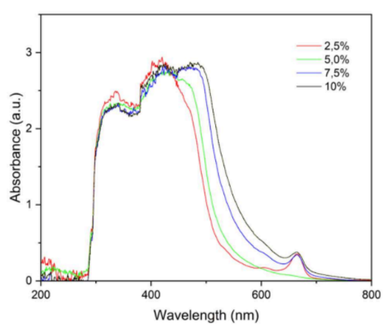

Figure 9. Absorption spectrum of dye with varying concentration in the UV-Vis range

The dye samples in this variation have almost the same absorption intensity, especially at the 380-450 nm peak (Table 7). However, in the 450-650 nm wavelength range, there is a widening of the area and an increase in absorption intensity as the concentration of the dye solution increases. Concentration variations in dragon fruit peel extract reported similar results, where in the visible light range, there was a widening of the area and an increase in absorption intensity (Faqih et al. 2019). Changes in the absorption spectrum due to an increase in dye concentration can occur due to interactions between dye molecules, which most likely cause the aggregation of dye molecules. This aggregation causes a bathochromic effect, or what is known as a red shift. This shift to a larger wavelength occurs due to changes in the electron structure of the dye molecule. (Conradie 2024).

Figure 10 shows that the absorption ability of TiO_2 with variations in pH and solvent concentration produces a relatively similar curve, especially in the UV wavelength range. The absorption peak was detected in the range of 250-350 nm in all variations.

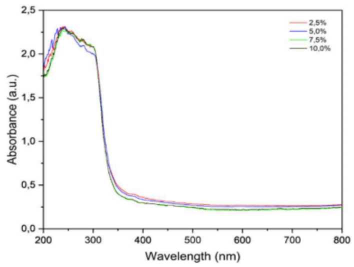

Figure 10. Absorption spectra of TiO2 with varying concentrations in the UV-Vis range

Table 7 summarizes the absorption peaks in dye samples as well as TiO2. Although the absorption curves look similar, in the visible light range, there are differences in absorption intensity that are not so significant in the order from the highest to the lowest, namely at a concentration of 2.5>5.0>10>7.5%. Research on Binahong leaf extract with varying concentrations reported the results of absorption on TiO2, with the order of absorption ability at a concentration of 5>3>1>10% (Muryani et al. 2019). According to Faqih et al. (2019)The ability of TiO2 to bind dye molecules has a limit, so increasing dye concentration does not constantly improve the ability to absorb light.

Table 7. Absorption peaks in dye and TiO2 samples with varying concentrations

Concentration	Dye sample		TiO2 + dye sample	
	λmax (nm)	Amax (a.u.)	λmax (nm)	Amax (a.u.)
2.5%	420	2.94	243	2.32
5.0%	431	2.77	244	2.31
7.5%	426	2.82	239	2.27
10.0%	425	2.87	240	2.30

Tauc's plot curve is made to determine the band gap of the absorbance value of the dye sample (Figure 11). Then, the HOMO and LUMO values can be known after the band gap value is obtained.

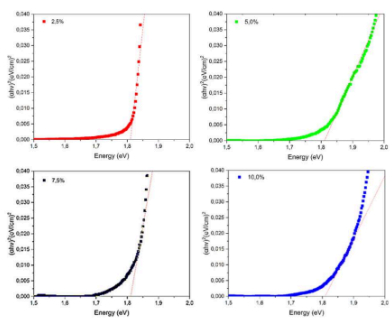

Figure 11. Tauc's plot of the dye sample with concentration variation

Tauc plot analysis shows that the band gap for all variations has the same value, which is 1.81 eV. This indicates that changes in dye concentration do not significantly affect the band gap value.

Figure 12 shows the LHE graph of the dye with various concentration variations. Data processing is done as in the pH variation: averaging the LHE values. The most considerable average LHE value is 40.19%, which is owned by dye samples with a concentration of 10%. At the same time, the concentration variations of 2.5%, 5.0%, and 7.5% have an average value of 36.19%, 34.66%, and 36.29%, respectively. This indicates that while a higher concentration of dye (10%) leads to a significant increase in LHE, lower concentrations result in relatively consistent but lower performance. These variations could be attributed to the efficiency of dye interaction at different concentrations, where a higher concentration may saturate the medium, leading to better light absorption. In contrast, lower concentrations may not be as effective in achieving similar results. Supported by Pradhan et al. (2023), higher dye concentrations can lead to saturation, enhancing light absorption and overall efficiency. For instance, dye loading increases proportionally with concentration until saturation is reached, as observed with a 0.2 mM concentration of D35 dye.

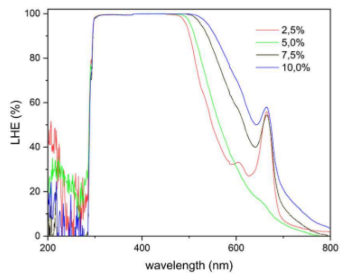

Figure 12. Percentage of light absorption efficiency (LHE) of dye samples with varying concentrations

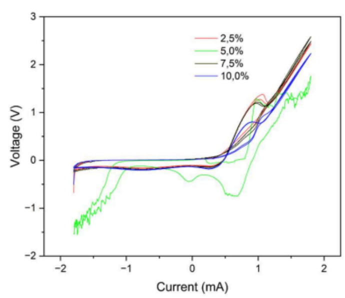

Figure 13. Cyclic Voltammetry characterization test results on dye samples with variations in concentration

The results of the Cyclic voltammetry characterization test at various concentrations are shown in Figure 13. The HOMO level value at pH 1.00 is -5.07 eV; at pH 1.66 is -5.25 eV; at pH 2.27 is -5.12 eV; and at pH 3.00 is -5.19 eV. The LUMO value is obtained from processing the HOMO value using Equation (3.6). The results of the band gap, HOMO, LUMO, and average LHE values are summarized in Table 8.

Table 8. Energy level and light absorption efficiency of dye samples with varying concentrations

Concentration	HOMO (eV)	LUMO (eV)	Band gap (eV)	LHE (%)
2.5%	-5.07	-3.26	1.81	36.19
5.0%	-5.25	-3.44	1.81	34.66
7.5%	-5.12	-3.31	1.81	36.29
10%	-5.19	-3.38	1.81	40.19

3.6 Effect of Dye Solution Concentration on DSSC Performance

Analysis of DSSC performance has been carried out in pH variation experiments, and the same treatment is given to analyze the effect of dye solution concentration on DSSC performance. Voc and Jsc values are the data used to obtain the fill factor value and DSSC efficiency.

The J-V curve shown in Figure 14 results from J-V characterization of DSSC devices with varying concentrations of dewandaru fruit extract dye solution. The Voc value of the four samples was obtained in the 0.32-0.46 Volt range. Meanwhile, the Jsc value ranges from 66.2 to 105 µA/cm2. Voc value with concentration variation has a trend where Voc7.5%>Voc10%>Voc2.5%>Voc5%. At the same time, the trend for Jsc value is Jsc10%>Jsc7.5%>Jsc2.5%>Jsc5%.

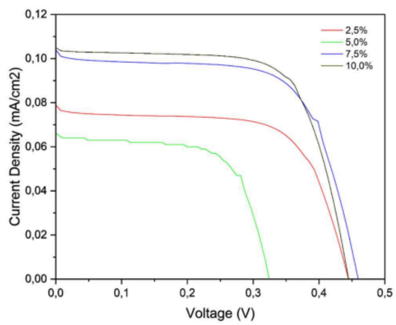

Figure 14. J-V curves of DSSC samples with varying dye solution concentrations

Table 9. Performance of DSSC samples with dye from Eugenia uniflora fruit extract with various concentrations

Concentration	Voc (V)	Jsc (mA/cm2)	FF (%)	Eff. (10^{-3}%)
2.5%	0.44	0.079	65.0	22.8
5.0%	0.32	0.066	63.9	11.9
7.5%	0.46	0.104	65.0	31.1
10.0%	0.45	0.105	69.0	32.2

The absorption area and intensity change when the concentration of the dye solution causes an increase in the percentage of LHE, and will then affect the Jsc value indirectly (Conradie, 2024; Faqih et al., 2019). The LHE value has the same trend as the Jsc value, as well as the trend in the DSSC efficiency value. The 10% sample showed the best efficiency value of 0.032%. The values of Voc, Jsc, FF, and efficiency at varying dye solution concentrations are summarized in Table 9.

Research by Muryani et al. (2019) reported the best DSSC efficiency results with variations in the concentration of Binahong leaf dye at a concentration of 5 g/100mL, which is 0.0662%, and decreases until, at a concentration of 1 g/100mL, the efficiency value becomes 0.0362%. Chien & Hsu (2013) conducted research by varying the concentration of anthocyanins from Brassica oleracea extract with a range of 3 µM - 6 mM. The resulting efficiency increases with increasing concentration until it reaches the optimal concentration at 3 mM, then decreases again as the concentration increases.

Table 10 shows the statistical analysis of the effect of concentration on DSSC performance, revealing several interesting relationships between the dye concentration and the optical characteristics and performance parameters. The Pearson correlation coefficient for the concentration and dye efficiency (η) was 1.000, which is highly significant (p = 0.000), indicating a perfect positive correlation between the concentration of the dye solution and DSSC efficiency. This suggests that as the dye concentration increases, the DSSC's efficiency also improves, highlighting the importance of concentration in optimizing DSSC performance.

The correlation between concentration and dye absorbance was moderate (0.800). Still, it was not statistically significant (p = 0.200), indicating that while concentration appears to influence dye absorbance, this effect is not strong enough to be considered statistically significant in this analysis. Similarly, the correlation between concentration and band gap energy (0.400) was insignificant (p = 0.600), implying that the concentration of the dye solution did not have a noticeable impact on the band gap energy.

Table 10. Statistical analysis for the Concentration's Dye effect on DSSC performance

Pearson Correlation		Concentration	Dye absorbance	BandGap	LHE	Voc	Jsc	FF	HOMO	LUMO	η
η	Correlation Coefficient	0.800	0.400	0.400	1.000*	0.80	1.000*	0.94	0.200	0.200	1.00
	Sig. (2-tailed)	0.200	0.600	0.600	0.000	0.20	0.000	0.05	0.800	0.800	0.00
	N	4	4	4	4	4	4	4	4	4	4

**. Correlation is significant at the 0.01 level (2-tailed).

In contrast, the correlation between concentration and light harvesting efficiency (LHE) was perfectly significant (1.000, p = 0.000), indicating a strong positive relationship. This suggests that higher concentrations of the dye significantly improve the light-harvesting efficiency of the DSSC, which is crucial for maximizing the energy capture of the device. The correlation between concentration and both Voc (0.800, p = 0.200) and Jsc (1.000, p = 0.000) was also positive and significant, highlighting that concentration considerably impacts these performance parameters, particularly Jsc, which showed a perfect correlation with concentration.

The correlation between concentration and fill factor (FF) was 0.949, with a p-value of 0.051, which is marginally statistically significant. This indicates that while concentration strongly influences the fill factor, the relationship is just shy of being definitively substantial at the 0.05 level. The correlation between concentration and HOMO and LUMO was both weak (0.200) and not statistically significant (p = 0.800), suggesting that concentration has little to no impact on these parameters in the context of DSSC performance.

In summary, the statistical analysis indicates that dye concentration significantly affects DSSC efficiency, light harvesting efficiency, Jsc, and fill factor, with the most substantial impacts observed on efficiency and Jsc. However, concentration appears to have less effect on dye absorbance, band gap energy, and the HOMO/LUMO values. This suggests that optimizing the concentration of Dewandaru fruit extract in DSSC dye solutions is crucial for enhancing performance, particularly regarding energy efficiency and light absorption.

Comparing these findings with those from Chien and Hsu (2013), which report that anthocyanin from red cabbage shows a high affinity for TiO2 nanoparticles and achieves a light-to-electricity conversion efficiency of over 1.4%, significantly higher than the previously reported efficiencies of 0.5–0.6%, highlights the potential of natural dyes in DSSCs. In their study, optimizing factors such as pH, concentration, and immersion time was key to improving DSSC performance. Similarly, our study shows the importance of pH and concentration in optimizing the performance of DSSCs. However, the maximum efficiency we observed (0.0322%) is lower than that reported by Chien & Hsu, suggesting that further optimization, including exploring additional natural dyes or coadsorbents, could improve the performance of Dewandaru fruit extract-based DSSCs. Furthermore, the presence of natural copigments in the crude extract, as observed by Chien & Hsu, may also enhance the efficiency of our DSSC, and future studies could explore this aspect in more detail.

4. Conclusion

In conclusion, the study on the effect of solution pH and concentration of Dewandaru fruit extract as a natural dye on the optical characteristics and performance of DSSC demonstrated that both factors significantly influence the device's performance. Variation in pH affects the absorption peak intensity, band gap energy, and the HOMO and LUMO values, with optimal performance observed at pH 3.00, yielding a Voc of 0.425 V, Jsc of 0.053 mA/cm², fill factor of 68.7%, and efficiency of 0.01548%. As the solution's acidity increased, the DSSC's performance decreased. The concentration of the extract did not notably affect the band gap energy but influenced the HOMO and LUMO values, broadened the absorption region, and slightly increased the LHE percentage. The best DSSC performance was achieved at a 10% concentration, producing a Voc of 0.45 V, Jsc of 0.105 mA/cm², fill factor of 69.0%, and efficiency of 0.0322%. This study shows that both solution pH and extract concentration play crucial roles in optimizing the optical properties and enhancing the performance of DSSC. Further optimization of the solution pH and extract concentration should be explored to improve the efficiency of Dewandaru fruit extract-based DSSCs. Additional factors, such as immersion time and coadsorbents, could enhance performance. Future research could also examine the role of natural copigments in the crude extract, which may contribute to improved light-to-electricity conversion. Additionally, exploring different extraction methods and using other natural dyes could improve the DSSC's optical properties and overall efficiency.

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Suburb	Energy source (number of respondents; multiple responses allowed		
	Electricity	Wood	Paraffin
Par 1	2	13	8
Par 2	3	17	13
Par 3	1	5	9

Source: Reference(Year)

List of Figure

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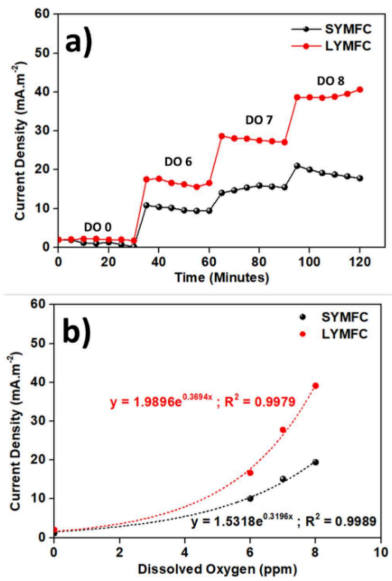

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Effect of Solution pH and Concentration of Dewandaru Fruit Extract as a Natural Dye on Optical Characteristics and Performance of Dye-Sensitized Solar Cells

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