NATURAL DYE EXTRACTED FROM *Elaeocarpus serratus* LEAVES TO FABRICATE NEAR-INFRARED DYE-SENSITIZED SOLAR CELL

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ABSTRACT

In this investigation, a natural dye was extracted from ripe leaves of *Elaeocarpus serratus* (D1), known as Sri Lankan olive or Weralu to fabricate a near-infrared (IR) dye-sensitized solar cell. For comparison, Dye extracted from *Clitoria ternatea* flower (D2) known as Nilkatarolu was used. Dye-sensitized solar cells were fabricated by using dye-coated TiO_2 films deposited on a fluorine-doped tin oxide glass plate as the anode, platinum-coated FTO glass as the cathode, and iodine/tri-iodide as the redox electrolyte. Optical properties of the dyes were characterized by UV-Vis spectroscopy and the results showed characteristic absorption peaks at 665 nm for D1 and 574 nm and 623 nm for D2. Photovoltaic parameters of the devices were obtained by using a J-V measuring unit coupled to a computer with an IR LED Light source (850nm) of 100mW/cm^2 intensity. DSSC fabricated with D1 dye showed a short circuit current density (J_{SC}) of 45 μA/cm^2, open circuit voltage (V_{OC}) of 173.4 mV and Fill Factor *(FF)* around 0.438 with an Efficiency(η) of 0.003%, whereas the device sensitized with D2 dye showed only a J_{SC} of 7 μA/cm^2 and V_{OC} of 21.4mV. Incident Photon to Current Efficiency (IPCE) measurements demonstrated that the *Elaeocarpus serratus* leaf natural dye exhibits better photovoltaic performance in the near-IR region compared to the *Clitoria ternatea* flower dye.

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1. INTRODUCTION

The demand for reliable and economically feasible alternatives to fossil fuels, coupled with the exploration of low-cost renewable energy sources, has accelerated research on novel materials for unconventional photovoltaics. Solar energy is one of the renewable energy sources which is eco-friendly. The electromagnetic radiation spectrum of the sun has a broad range of energy that includes visible light, Ultraviolet (UV), Infrared (IR), radio waves, x-rays, and gamma rays. However, hazardous radiations are naturally filtered when they reach the earth's surface. Various methods exist for converting solar radiation into practical and usable forms. Among them, solar cells lead to the conversion of solar energy into electrical energy. There are several different generations of solar cells. Solar cells made of crystalline silicon are classified as the first generation, thin-film solar cells as the second generation, and newly developed solar cells with emerging technologies as the third generation, consisting of Dye-sensitized (Gratzel, 1991), organic (Wöhrle D.and Meissner D, 1991), CZTS (Xin X, He M, Han W, Jung J and lin Z, 2011), Perovskite (N.G, Park, 2015), and Quantum dot (Aroutiounian V, Petrosyan S, Khachatryan A and Touryan K, 2001) solar cells. A practically viable dye-sensitized solar cell (DSSC) was first developed by Gratzal and co-workers in 1991 (Gratzel, 1991). Since then, DSSCs have been an interesting field of research among many scientists. The primary components of dye-sensitized solar cells are photoanodes made of dye-coated semiconductor film on transparent conducting oxide film-coated glass (FTO or ITO Glass), an electrolyte (I_2/I^-) and a counter electrode catalyzed with platinum. These types of solar cells are non-toxic, low cost, have high performance at indoor and outdoor conditions and follow a simple fabrication method. Hence, they are different from other types of solar cells.

Researchers are interested in fabricating DSSCs sensitized with natural pigments extracted from plant leaves, fruits, flowers, seeds, algae, and even with cyanobacteria because it mimics the natural photosynthesis process (Eddie Nahúm, 2023). The dye in this solar cell is responsible for absorbing the photons from solar radiation, and then converting those photons into energetic electrons. Generally, most natural dyes are sensitive to solar radiation in the visible and UV range and convert the absorbed energy into electricity. However, very few natural dyes are found to be sensitive to the IR region of the solar radiation which can be utilized in DSSCs. This kind of dye-sensitized solar cells has the possibility of being used in applications such as IR sensors in night vision cameras and thermal detectors.

The objective of this research study is to investigate the characteristics of IR-absorbing dye extracted from *Elaeocarpus serratus* leaves (Sri Lanka olive or Werallu) and to develop a near-IR Dye-sensitized solar cell which can be useful as an IR sensor. This research study also aims to compare this dye and its performance in DSSCs with the natural dye *Clitoria ternatea* which absorbs solar radiation in the UV-visible region.

2. METHODOLOGY

2.1. Extraction of Natural dye

Ripe *Elaeocarpus serratus* (known as Sri Lanka olive or Werallu) leaves with red patches were taken and cut into small pieces. One gram (1g) of chopped leaves were first boiled in water to remove any polar pigments such as anthocyanin that are present in the leaves. After filtration, the leaves were boiled again with 2.5 ml of ethanol for 30 minutes at 80°C in a beaker placed in a water bath until the leaves became colourless. After following the extraction, the dye was filtered and collected into a sample bottle. This bottle with dye was then wrapped with aluminium foil and kept in a refrigerator at 4°C until used. *Clitoria ternatea* flower dye (Nilkataluu) was directly extracted in ethanol using a similar procedure.

2.2. Development of Dye Coated Flim

Fluorine-doped Tin Oxide (FTO) glass plates that were cut into 2 cm x 1 cm pieces and cleaned using the following procedure. The FTO glass plates were first cleaned for five minutes in an ultrasonic bath after being placed in a beaker containing water and a few drops of cleaning agents. The plates were then sonicated for 5 minutes in a bath with distilled water and a few drops of con.H_2SO_4. Cleaned plates were then boiled in isopropyl alcohol at 80° C in a beaker and dried using a hair dryer with low heat.

For the preparation of TiO_2 paste, 0.25 grams of TiO_2 nanoparticle (20 nm) powder, 0.1 ml of 0.1M HNO_3, a drop of Triton-X 100, and a drop of PEG 400 were mixed together and ground until forming a thick paste. The doctor blade technique was used to apply the prepared TiO_2 paste on the conducting surface of the FTO glass. After that, the cells were sintered for 30 minutes at 450°C in a furnace and allowed to cool to room temperature. Afterwards, TiO_2 film-coated FTO glass plate was immersed in ethanol extract of both dye solutions separately in test tubes for 15 hours.

2.3. Fabrication of DSSC

The electrolyte for the dye-sensitized solar cells was prepared by using 0.127 g of iodine (I_2) and 0.83 g of potassium iodide (KI) dissolved in 10 ml of acetonitrile and ethylene carbonate in an 8:2 ratio in a volumetric flask. The entire mixture of the solution was then stirred for a further five hours to make sure that all the solid particles were dissolved completely.

Devices were fabricated by sandwiching the dye-coated TiO_2 films on FTO glasses as the photoanode and the Platinum (Pt) sputtered glass as the counter electrode, using crocodile clips. The electrolyte was poured into the empty space built up by the capillaries between the two plates.

2.4. Characterization of Natural dye and Solar cell

The absorption spectrum of the dye extract of *Elaeocarpus serratus* leaves was determined by using a UV-visible spectrometer. The photovoltaic measurements of the near IR dye-sensitive solar cell were obtained under the illumination of an inferred LED light source (850 nm) of intensity 100 mW/cm^2 using the computerized PK-I-V 100 I-V analyzer. Using a computerized VK-IPCE-10, the incident photon to current conversion efficiency (IPCE) of the near- IR DSSC was measured.

3. RESULTS AND DISCUSSION

The enormous variety of pigments that plants synthesize is linked to numerous physiological and/or biological processes, including photosynthesis and reproduction, for example, to entice insects to pollinate flowers. There are many different kinds of pigments in leaves, but the main ones include carotenoids, anthocyanins, and chlorophylls. The chemical structures of these dyes are shown in Fig1.

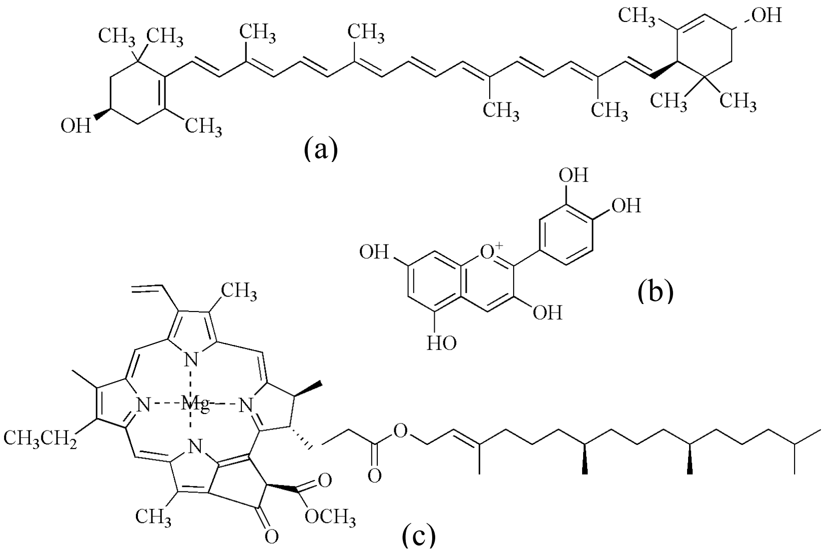

Figure 1: Structures of pigments in leaves **(a)** Xanthophyll (Carotenoid) **(b)** Cyanidin (Anthocyanin) **(c)** Chlorophyll a

Chlorophyll gives leaves their green colour, however, in some plants and leaves, this pigment loses its green appearance and turns wildly colourful. Chlorophyll disappears from these leaves with age, but other pigments, such as the naturally occurring carotenoids, which are present in the leaf remain turning leaves yellow or orange. Chlorophyll and Carotenoids are very long-chain water-repelling pigments that are synthesized in plastids of plant cells. Anthocyanins, which give leaves their red colour, are produced in greater quantities when the quantity of chlorophyll declines. They are water-soluble. Carotenoids and Anthocyanins known as auxiliary or accessory pigments, improve the use of white light for photosynthesis in addition to chlorophyll, which absorb light in the red and blue spectrum. These molecules also transfer their electrical excitations to chlorophyll. The carotenoids, which have triple-banded spectra change when they interact with other substances, shifting to longer wavelengths. Generally, Carotenoids are classified into two classes: Carotenes which are hydrocarbons and xanthophylls which are oxygenated hydrocarbons.

Therefore, the green and red colour pigments in *Elaeocarpus serratus* leaves are probably Chlorophyll and Xanthophyll facilitated with hydroxyl groups that could chelate to the TiO_2 film. Polar pigments such as anthocyanin could be minimized in dye extraction by boiling leaves in water prior to ethanol.

3.1. UV-Visible absorption spectrum of the dye

According to Perera B.K.S (2023), the research utilized *Clitoria ternatea* (Nil Katarolu) flowers' natural dye, which was extracted directly into ethanol. The anthocyanin dyes present in the extract were recorded in Figure 2, which exhibits the UV-Vis absorption spectra of the natural dyes extracted from *Clitoria ternatea* flowers and *Elaeocarpus serratus* leaves.

The absorption measurements were carried out in the wavelength range from 400 nm to 1000 nm. The peaks at 665 nm correspond to the *Elaeocarpus serratus* leaves dye and 574 nm and 623 nm corresponds to the *Clitoria ternatea* flower dye respectively.

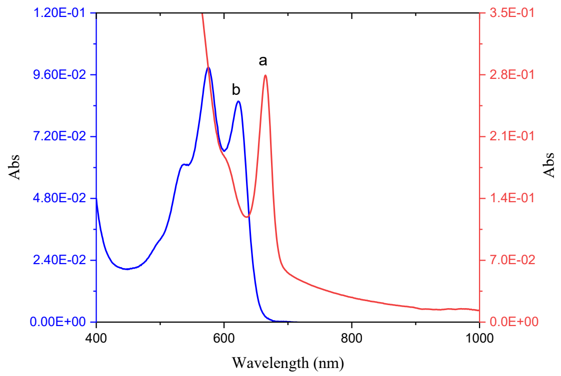

Figure 2: UV-Vis absorption spectra (a) *Elaeocarpus serratus* leaf dye (b) and *Clitoria ternatea* dye

Figure 2 shows the Tauc plot, which was generated from the absorbance spectrum data of the natural dyes.

Following Tauc relationship is used to find the minimum band gap between the Highest Occupied Molecular Orbital (HOMO) and Unoccupied Molecular Orbital (LUMO) of the *Elaeocarpus serratus* and *Clitoria ternatea* natural dyes (Cheng C, Zhang H, Li F, Yu S and Chen Y, 2021).

$$(\alpha h\vartheta)^n = A\left(h\vartheta - E_g\right)$$

$$\alpha = \frac{4\pi k}{\lambda}$$

Where , A= Constant, $h\vartheta$ is the photon energy, α is an absorption coefficient, k is the absorbance,

and E_g is the band gap between LUMO and HUMO levels of the natural dye. Corresponding n values are n = 2 and n =1/2 for direct band gap and indirect band gap. Since dyes have a direct band gap, n was taken to be equal to 2, and the band gap energy was determined using the Tauc plot drawn $(\alpha h\vartheta)^n$ vs $Ah\vartheta$. From the Tauc plot band gap energy was determined as 1.77 eV for

Elaeocarpus serratus dye and 1.92 eV for *Clitoria ternatea* dye where absorption energy for *Elaeocarpus serratus* leaf dye is in near- IR region.

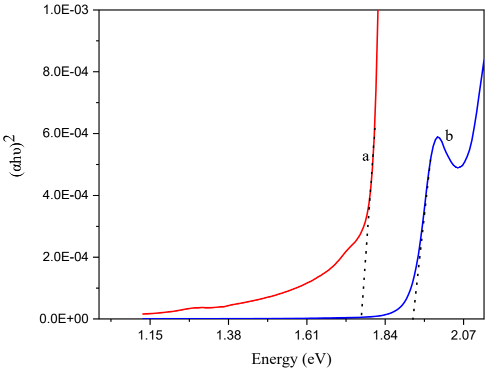

Figure 3: Tauc Plot for Band gap energy of (a) *Elaeocarpus serratus* leaf dye and (b) *Clitoria ternatea* flower dye

3.2. IV Characteristics Measurements

Under the illumination of an Infrared LED Light source (850nm) with the intensity of 100mW/cm^2, photovoltaic characteristics such as open circuit voltage *(V_{OC})*, short circuit current density *(J_{SC})*, fill factor *(FF)*, and efficiency *(η)* of the Dye-sensitized solar cells were measured using the computerized P-K-I-V 100 I-V Measuring unit.

Figure 4 illustrates the open circuit voltage vs. short circuit current density of the near- IR DSSC. According to the plot, with the IR LED light Source, 45 µA short circuit current density (JSC), and a 173.4 mV Open circuit Voltage (VOC) were obtained for DSSC sensitized with *Elaeocarpus serratus* leaf dye extract. The fill factor (FF) of the cell was around 0.438 and its efficiency (ɳ) was 0.003%. For the *Clitoria ternatea* flower dye extract, 7 µA/cm^2 short circuit current density (JSC), and a 21.4 mV Open Circuit Voltage (VOC) and its efficiency (ɳ) was less than 0.001% for the IR LED light Source (850 nm) at 100 mW/cm^2 light intensity. It is evident from the J-V measurements of the cell that the DSSC sensitized with *Elaeocarpus serratus* leaf dye is more sensitive to IR radiation.

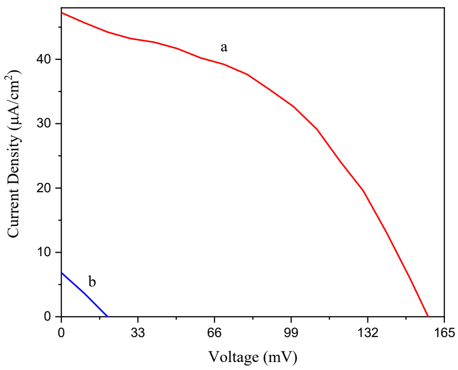

Figure 4 : JV Characteristic curve of (a) *Elaeocarpus serratus* leaf dye and (b) *Clitoria ternatea* flower dye in Dye-sensitized solar cell (DSSC) under an IR Light source (850 nm) with 100 mW/cm^2 intensity.

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3.3. IPCE Characteristic of the Near IR-DSSC

Figure 5 shows the IPCE vs Wavelength graph measured by using computerized VK-IPCE-10 system. The maximum near- Infrared absorbance wavelength of the IPCE is in the range of 650 nm to 900 nm.

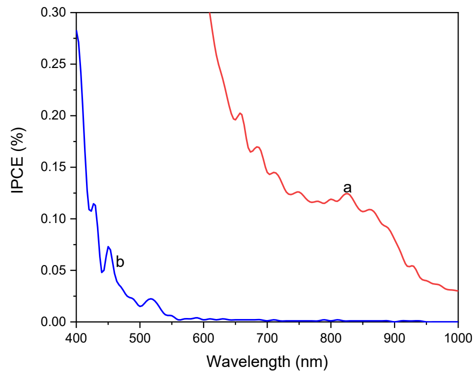

Figure 5: Incident photon-to-current efficiency (IPCE) Characteristic curves of Dye-sensitized solar cells (DSSCs) (a) *Elaeocarpus serratus* (SL Olive) leaf and (b) *Clitoria ternatea* (Nil Katarodu) flower dye extracts

According to Figure 5, *Elaeocarpus serratus* leaf natural dye extracted in ethanol is sensitive and converts its absorbed photons to electrons in the near- Infrared wavelength range compared with *Clitoria ternatea* flower dye extract known as Nilkatarolu which contains anthocyanins.

4. CONCLUSION

A natural dye was extracted from *Elaeocarpus serratu*s (Weralu or Sri Lanka olive) leaf for the sensitization of Near- Infrared DSSC. The solar cell produced a short circuit photocurrent Density *(J_{SC})* of 45µA and an open circuit voltage *(V_{OC})* of 173.4mV with an efficiency *(η)* of 0.003%. The fill factor *(FF)* of the cell was around 0.438 for the Infrared LED Light source (850 nm) with an intensity of 100mw/cm^2. The red colour pigment in *Elaeocarpus serratus* leaves is likely to be xanthophyll and the green colour pigment is Chlorophyll that is present in the dye extraction. Compared to other natural dyes the active pigments in *Elaeocarpus serratus* leaf seem to be more stable and sensitive to the near-infrared region of the electromagnetic radiation. So, we can use this kind of near-IR Dye-sensitized solar cell as Infrared light sensors.

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