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Fabrication of Dye-Sensitized Solar cell using mixtures of Natural dye Extraction from *Elaeocarpus Serratus* Red and Green leaves

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1. ABSTRACT

In the future, global energy demand is deeply uncertain, and it is one of the biggest socio-economic problems. Renewable energy comes from non-depletable sources. Solar energy is one of the renewable energy sources and the most readily available of all energy sources, so it can potentially use effectively. Solar energy can be turned to an accessible form, such as electrical energy, by using solar technologies such as solar power panels. Researchers are interested in producing Dye-Sensitized Solar Cells (DSSC)s utilizing natural pigments that are obtained from plant leaves, fruits, flowers, seeds, algae, and cyanobacteria because it mirrors the natural photosynthesis process. As a result, the pigments in this solar cell absorbs photons from solar radiation and transfers them into energetic electrons. Among them most natural dyes in general, are sensitive to a wide range of visible and UV wavelengths and convert the absorbed energy into electricity. The primary objective of this research is develop a dye-sensitized solar cell using natural dye extract from *Elaeocarpus serratus* red, green and mixture of red and green leaves (red: green 1:1,1:2, 2:1) in ethanol and to investigate the DSSC performances.

DSSC was finally developed using the dye coated TiO_2 film on FTO glass as the anode and the Platinum (Pt) sputtered glass plate as the counter electrode with a liquid form electrolyte. The photovoltaic measurements of the DSSC have been obtained using the computerized PK-I-V 100 I-V analyzer under the illumination of a LED light source with an intensity of 100 mW/cm^2 and direct sunlight. 1:1 mixture of Weralu red & green leaf dye coated DSSC performed well compared to other DSSCs. The photovoltaic measurements of that cell are, short circuit photocurrent density($J_{(SC)}$) 3.842 mA/cm^2, open circuit photovoltage ($V_{(OC)}$) 406.2 mV and the fill factor (ff) around 0.474 with 0.739 % efficiency (η). The UV - visible absorption spectra of natural dyes extracted from Elaeocarpus serratus leaves in ethanol were assessed across the range 200 - 750 nm using CT-2600 Spectrophotometer. The resulting spectra revealed peaks at 664.2 nm, 605 nm, and 533.4 nm for 1:1 combination of red and green dyes. The active pigment in the extraction seems to be chlorophylls, Carotenoids and Anthocyanins.

Incident Photon to Current Efficiency (IPCE) of the DSSCs is measured using VK-IPCE 10 analyzer. 1:1 mixture of red & green leaf dye coated DSSC contributed to obtain highest Quantum efficiency compared to other DSSCs in the wavelength range of 400 nm to 750 nm. The DSSCs were tested over a period of one hour and the efficiency & current of the DSSCs increased prolonged illumination in sunlight.

Key words: Elaeocarpus serratus, Dye-Sensitized solar cell, red, green leaf, and Sri Lankan olive

2. INTRDUCTION

In the future, global energy demand is deeply uncertain, and it is one of the biggest socio-economic problems. According to US energy data, worldwide energy consumption would rise by 28% from 19.2 to 24.6 TW between 2015 and 2040 [1]. Global energy usage is predicted to be 580 million tera joules per year. The rise in global population, increased regional manufacturing and higher living standards are some of the key factors for the increase in energy consumption. [2]

Two primary sources of energy used to generate power are non- renewable and renewable energy sources. Non-renewable energy sources that are frequently used include fossil fuels (coal, natural gas, and petroleum) and nuclear power. Energy is produced by the combustible process of non-renewable fossil fuels [3]. This has an impact not only on the global resources of fossil fuels, but also on ecological systems. As we all know, burning fossil fuels emits greenhouse gasses such as carbon dioxide. These gases form a blanket on the Earth's surface, blocking the discharge of the sun's short rays at night. As a result, we can confidently say that the energy crisis contributes to make the earth a warmer place by causing global warming. [4]

Researchers have been working on to find exciting ways around all these challenges. The demand for reliable, financially feasible alternatives to fossil fuels, as well as the development of low-cost renewable energy sources, have inspired research into novel materials for unconventional power generation. Renewable energy is coming from non-depletable sources. They are self-sustainable and natural, with a low or zero carbon footprint. Wind power, solar power, bioenergy (organic matter used as a fuel), and hydroelectric, including tidal energy, are examples of renewable energy sources. [5]

Solar energy, abundant and accessible, surpasses human energy consumption by a factor of 10,000. Conversion to usable forms, like electricity, is achieved through solar technologies such as photovoltaic panels. Photovoltaic cells are classified into three generations: first, second, and third. First-generation cells, known as conventional or wafer-based cells, utilize crystalline silicon, predominantly polysilicon and monocrystalline silicon. Second-generation cells consist of thin film solar cells employing materials like amorphous silicon, CdTe, and Copper indium gallium selenide (CIGS), crucial for utility-scale photovoltaic systems and integrated or standalone power solution. The third generation of solar cells encompasses various thin-film technologies known as emerging photovoltaics, most of which are still under research and development and have not yet reached commercialization. These technologies employ a range of organic and inorganic compounds, often including organometallic substances. Despite historically low efficiencies and limited stability of absorber materials, research continues due to the potential for achieving low-cost, high-efficiency solar cells in the future. [6] [7] [8] [9] [10].

In 1991, Gratzal and colleagues were the first to create an economically viable dye-sensitized solar cell (DSSC) [6]. Many researchers have found DSSCs to be a fascinating subject of study since then. The key components of dye-sensitized solar cells are a photoanode made from dye-coated semiconductor film over transparent conducting Tin oxide film-coated glass (FTO or ITO Glass), an electrolyte in a liquid form such as I_2/I^-_3, and a counter electrode catalyzed with platinum. These photovoltaic cells are non-toxic, economical, simple to build, and function well in both indoor and outdoor conditions. For these reasons, DSSCs are unique to other types of solar cells.

Researchers are interested in producing DSSCs utilizing natural pigments that are obtained from plant leaves, fruits, flowers, seeds, algae, and cyanobacteria because it mirrors the natural photosynthesis process. As a result, the pigment in the solar cell absorbs photon from solar radiation and transfers them into energetic electrons. Among them most natural dyes in general, are sensitive to a wide range of visible and UV wavelengths and convert the absorbed energy into electricity.

The primary objective of this research study is to develop a DSSC using natural dye extract from *Elaeocarpus serratus* leaves in ethanol and for investigating its performances.

3. **METHODOLOGY**

3.1.Extraction of Natural dye

Fresh green and red leaves of Elaeocarpus serratus, commonly known as Olive or Werallu in Sri Lanka, were individually collected, and cut into small pieces. Subsequently, one gram of SL Olive green and red leaves was separately boiled in a beaker over a hot plate at temperatures ranging from 65°C to 70°C for 30 minutes until the leaves lost their color. After the extraction process, the natural dyes obtained from SL Olive green and red leaves were filtered and transferred into separate sample bottles. The dyes were then wrapped in aluminum foil and stored in a refrigerator at 4°C until further use.

3.2. Development of Dye Coated Flim

The subsequent technique was used to clean 2 cm x 1cm pieces of Fluorine-doped Tin Oxide (FTO) plates made of glass. FTO glass plates were cleaned in the beginning in an ultrasonic bath for a five-minute period with distilled water and soap in liquid form. After that, the FTO glass plates were sonicated for 5 minutes with distilled water and a few drops of con.H_2SO_4., Then FTO glass plates were boiled in propyl alcohol in a beaker at 80° C. The FTO glasses were then air-dried using a low-heat hair dryer, and their conducting surface was checked with a conductivity meter.

To prepare the TiO2 paste, a mixture of 0.25 g of 20 nm TiO2 powder, 0.1 ml of 0.1M HNO3, a drop of Triton-X 100, and a drop of PEG 400 was combined and processed until a thick paste formed. This paste was then applied onto the conductive surface of the FTO glass plate using the doctor blade method. Subsequently, the cells were sintered at 450°C in a furnace for 30 minutes and left to cool. The TiO2-coated glass plates were then immersed in separate test tubes containing ethanol extracts derived from Elaeocarpus serratus green leaves, red leaves, and solutions combining red and green leaf natural dyes in ratios of 1:1, 1:2, and 2:1 for a duration of 15 hours

3.3. Development of DSSC

One of the most fundamental components of a DSSC is the electrolyte. In this investigation, an I_2/I^-_3 electrolyte in liquid form was utilized, which was prepared in a volumetric flask by dissolving 0.127 g of iodine (I_2) and 0.83 g of potassium iodide (KI) in 10 ml of acetonitrile and ethylene carbonate in an 8:2 ratio. For a duration of 5 hrs, the solution was stirred continuously in order to make sure that all solid particles were completely dissolved [11].

The DSSC was finally developed using the dye coated TiO_2 film on FTO glass as the anode and the Platinum (Pt) sputtered glass plate as the counter electrode, which were connected side by side and fasten together using crocodile clips. The liquid form electrolyte was transferred into the capillary gap between the two plates.

3.4.DSSC and Natural Dye Characterization

A UV-visible spectrometer was used to analyze the absorption spectrum of a natural dye extract of *Elaeocarpus serratus* leaves. The photovoltaic measurements such as open circuit voltage $(V_{(OC)})$, short circuit Current $(I_{(SC)})$, short circuit current density$(J_{(SC)})$, fill factor *(ff)* , efficiency (η) , series Resistance $(R_{(S)})$, and shunt Resistance $(R_{(Sh)})$ of the DSSC have been obtained using the computerized PK-I-V 100 I-V analyzer under the illumination of an LED light source with an intensity of 100 mW/cm^2. The incident photon to current conversion efficiency (IPCE) of the DSSC has been examined using a computerized VK-IPCE-10 analyzer.

4. RESULTS AND DISCUSSION

Due to the presence of the natural pigments, flowers, fruits, and leaves of plants appears in a wide spectrum of colors such as green, red, orange, etc. Natural plant leaves contain ~~s~~ number of active pigments, but the most frequent natural pigments are carotenoids, anthocyanins, and chlorophylls.

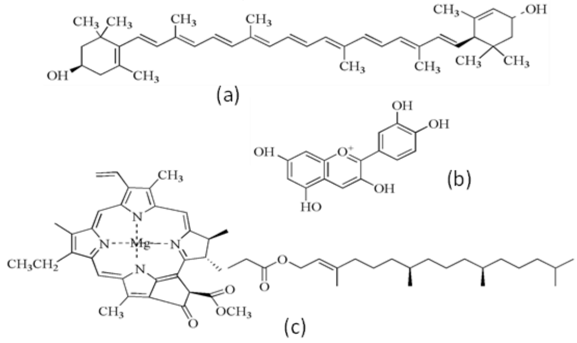

Figure 1*: Structures of pigments in leaves* ***(a)*** *Xanthophyll (Carotenoid)* ***(b)*** *Cyanidin (Anthocyanin)* ***(c)*** *Chlorophyll a* [14]

Plant leaves initially appear green in color due to the presence of chlorophyll pigments present in the leaves and they help to carry out the photosynthesis during the daytime. Then the leaves' color changes to orange yellow and red with age [12] [13]. Most green leaves contain chlorophyll as the major pigment and anthocyanins which give the red color and Carotenoids which give the yellow color to the leaves at minority levels. Chlorophyll (green color) disintegrates from these leaves with age, but other pigments, such as naturally occurring carotenoids, remain, turning the leaves yellow or orange. Anthocyanins, which give leaves their red color, are created in larger numbers as the amount of chlorophyll disappears. Addition to the chlorophyll pigment, Carotenoids is a photosynthetic pigments 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. Carotenoids can be classified into two major groups:

one is carotenes which contains hydrocarbon groups and another one is xanthophylls which [14] contains oxygenated hydrocarbons groups. The chemical structures of these pigments are shown in Figure1.

Accordingly, the green and red pigments in *Elaeocarpus serratus* leaves are most likely Chlorophyll, Anthocyanin and Carotenoids (Xanthophyll) with hydroxyl groups that can chelate on the TiO_2 coating.

4.1. Photovoltaic Measurements of the DSSCs

The photovoltaic measurements such as open circuit voltage $(V_{(OC)})$, short circuit current $(I_{(SC)})$, short circuit current density($J_{(SC)}$),, fill factor *(ff)* , efficiency (η) , series resistance $(R_{(S)})$, and shunt resistance $(R_{(Sh)})$ of the DSSC have been obtained using the computerized PK-I-V 100 I-V analyzer under the illumination of an LED light source with an intensity of 100 mW/cm^2.

Figure*2* illustrates the open circuit voltage vs. short circuit current density of DSSCs fabricated from *Elaeocarpus serratus* red leaf, green leaf, and a combined mixture of red and green leaf ethanol extract. The cell was illuminated with sunlight at outdoor for one hour and gradual increment in photocurrent could observed.

According to the graph, the DSSC which is fabricated by using mixture of 1:1 ratio *Elaeocarpus serratus* red and green leaf Ethanol extract has produced higher current density than other DSSCs. The photovoltaic measurement of that cell is $J_{(SC)}$ 3.842 mA/cm^2, $V_{(OC)}$ 406.2 mV and the fill factor around 0.474 with 0.739 % efficiency. The Series resistance, and shunt resistance of The DSSC are 86.445 Ω and 5577.557 Ω respectively.

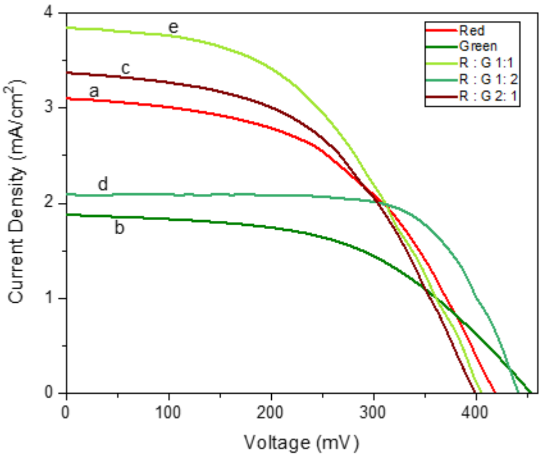

Figure 2: The open circuit voltage vs. short circuit current density (J vs V) characteristics graph of the cells sensitized with dye extracts of ethanol extract from *Elaeocarpus serratus* (a) red leaf (b) green leaf (c) 1:1 ratio mixture of red & Green leaf (d) 1:2 ratio mixture of red & Green leaf (e) 1:1 ratio mixture of red & Green leaf

The Table 01 below shows the photovoltaic characteristic measurements of the *Elaeocarpus serratus* red leaf, green leaf, and a combined mixture of red and green leaf ethanol extracts.

Table 1: Photovoltaic Measurements of the DSSCs

	Open circuit voltage ($V_{(OC)}$) mV	**Short circuit current density ($J_{(SC)}$) mA/cm^2**	**Fill factor**	**Efficiency (η)**	**Series resistance ($R_{(S)}$) Ω**	**shunt resistance ($R_{(Sh)}$) Ω**
Red Leaf	420.0	3.101	0.497	0.647	85.166	3277.111
Green Leaf	453.8	1.88	0.509	0.482	84.975	2087.276
R : G 1 : 1	406.2	3.842	0.474	0.739	86.445	5577.558
R : G 1 : 2	440.0	2.485	0.580	0.634	51.811	6243.712
R : G 2 : 1	400.0	3.369	0.499	0.672	77.727	3745.185

4.2.UV-Visible absorption spectrum of the dye

The absorption spectrum, a unique pattern of wavelengths absorbed by different pigments, reveals their identities. Chlorophyll absorbs blue-violet light, while chlorophyll b absorbs red-blue light. Carotenoids absorb blue-green and violet light, reflecting yellow, red, and orange light. Organisms often have pigment combinations, broadening their absorption spectrum. To assess pigment types in photosynthetic plants, spectrophotometry is employed. This technique measures transmitted light to determine absorption, unveiling which wavelengths are absorbed. By extracting leaf pigments and analyzing them in a spectrophotometer, researchers identify the absorbed light wavelengths. [15].

In this study diluted solution of red dye extract, green dye extract and the dye mixtures (red: green 1:1, 1:2, 2:1) of the *Elaeocarpus serratus* were prepared in two different concentrations to observe the absorption of these solutions under UV spectrum and visible spectrum separately by using CT-2600 Spectrophotometer**.** For UV spectrum and visible spectrum, all 5 dye samples were prepared to have the same concentration

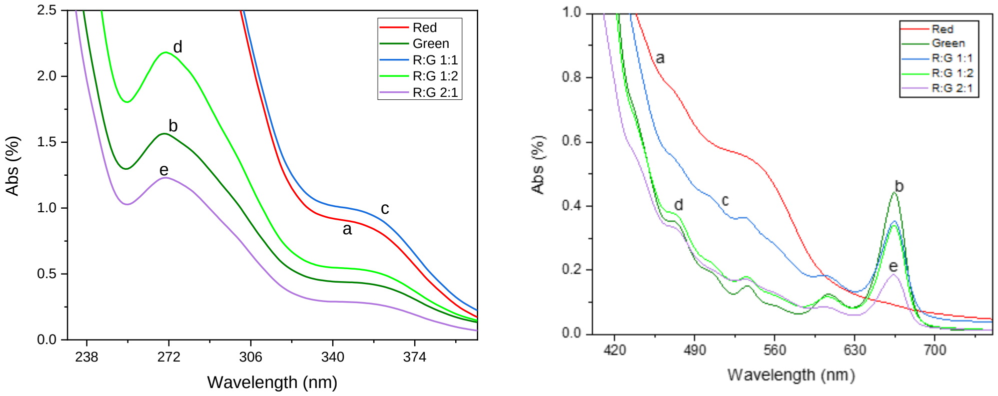

Figure 3 Figure 4

Figure 3 & 4: Absorption spectrum in UV range and visible rang of the ethanol extract from Elaeocarpus serratus (a) red leaf (b) green leaf (c) 1:1 ratio mixture of red & Green leaf (d) 1:2 ratio mixture of red & Green leaf (e) 1:1 ratio mixture of red & Green leaf.

Figure 3 and figure 04 separately shows the UV &Visible absorption spectrum of the *Elaeocarpus serratus* red leaf, green leaf, and a combined mixtures of red and green leaf ethanol extracted natural dyes in the UV (200 nm to 400 nm) and visible (400 nm to 750 nm) ranges.

Comparing the visible light absorption spectra of dye solutions with the same concentration following can be observed. According to the spectrum analysis graph, the ethanol extracts from Elaeocarpus serratus red leaves exhibited a single peak at 535.2 nm. Green leaves, on the other hand, showed four major peaks at 644.80 nm, 607.70 nm, 536.20 nm, and 405.41 nm. A 1:1 mixture of red and green dye exhibited three peaks at 664.2 nm, 605.00 nm, and 533.4 nm. A 1:2 mixture of red and green dye displayed four peaks at 664.2 nm, 606.00 nm, 535.5 nm, and 472.3 nm. Lastly, a 2:1 mixture of red and green dye exhibited three peaks at 664.3 nm, 604.4 nm, 536.2 nm, and 472.2 nm.

Tabe 3: Peak absorption wavelengths of plant photosensitive pigments [15] [16]

Plant Pigment	**Abs Wavelength (nm)**
Chlorophyll	425, 470, 606, 640, 660
Carotenoids	450 – 455
Anthocyanin	510 to 520
Xanthophylls	435, 494

According to the UV-Visible spectrum data, *Elaeocarpus serratus* leaf ethanol extract containing pigment chlorophyll, carotenoids, and anthocyanin

Figure 5 shows the Tauc plot of the *Elaeocarpus serratus* ethanol extract, plot was generated from the visible spectrum data of the *Elaeocarpus serratus* ethanol extract of red leaf, green leaf, and mixture of red & green leaf

The Tauc relationship is used to determine the minimum band gap between the Highest Occupied Molecular Orbital (HOMO) and Unoccupied Molecular Orbital (LUMO) of the *Elaeocarpus serratus* Ethanol extraction of natural pigments.

$$(\alpha h\vartheta)^n = A\left(h\vartheta - E_g\right) \longrightarrow \text{Tauc Relationship}$$

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

Here, 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. n = 2 for the direct band gap and n = 1/2 for the indirect band gap [17] [18] [19]. Since dyes have a direct band gap, n was taken to be equal 2, and the band gap energy was determined using the plot drawn $(\alpha h\vartheta)^n$ vs $Ah\vartheta$.

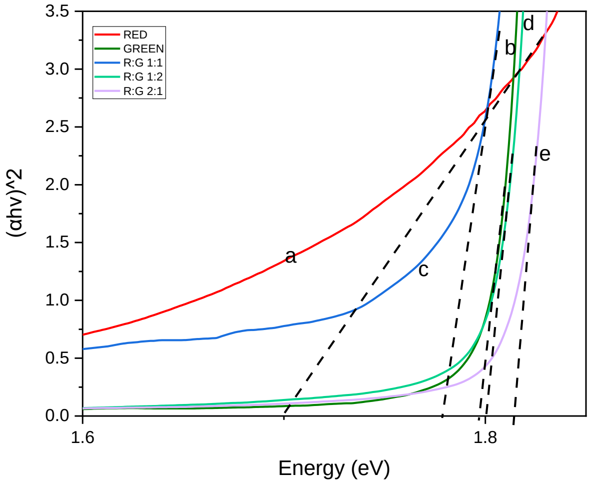

Figure 5: Tauc Plot for energy gab in visible range of the ethanol extract from *Elaeocarpus serratus* (a) red leaf (b) green leaf (c) 1:1 ratio mixture red & green leaf (d) 1:2 ratio mixture of red & green leaf (e) 1:1 ratio mixture of red & green leaf

The energy gab between the Highest Occupied Molecular Orbital (HOMO) and Lowest Unoccupied Molecular Orbital (LUMO) in the Elaeocarpus serratus ethanol extract varies depending on the mixture of red and green leaf dyes used. According to the graph analysis, the energy gaps are measured to be 1.70 eV for red dye, 1.79 eV for green dye, 1.77 eV for a 1:1 mixture, 1.80 eV for a 1:2 mixture, and 1.81 eV for a 2:1 mixture of red and green leaf dyes.

4.3. IPCE Characteristic of the DSSCs

Figure 6 shows that Incident Photon to Current Efficiency (IPCE) measured by using computerized VK-IPCE-10.

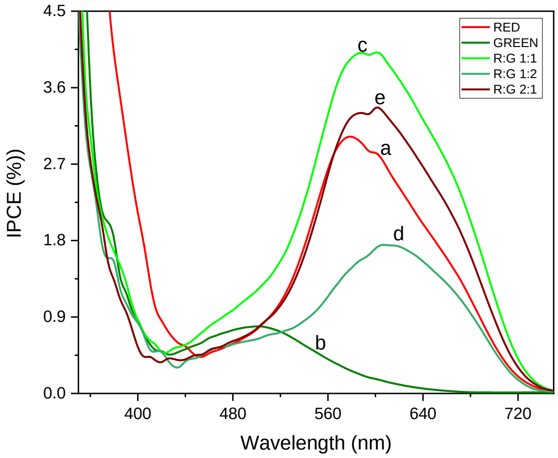

Figure 6: Incident photon to current efficiency graph of the ethanol extract from *Elaeocarpus serratus* a) red leaf (b)green leaf (c) 1:1 ratio mixture of red & Green leaf (d) 1:2 ratio mixture of red & Green leaf e) 1:1 ratio mixture of red & Green leaf DSSCs

Based on the graph, it’s confirmed that the DSSCS is functioning efficiently in the wavelength range of 400nm to 750nm. According to the IPCE Graph the maximum wavelength of DSSCs generated utilizing a mixture of red and green *Elaeocarpus* *serratus* leaf ethanol extract is 600nm to 605nm, maximum wavelength of the red and green leaf ethanol extract DSSCs is respectively 580nm and 500nm.

1:1 mixture of *Elaeocarpus serratus* red and green leaf ethanol extract DSSC is contributed to obtain highest current Comparing with other DSSCs

5. CONCLUSION

The ground-breaking aspect of this study lies in the remarkable performance of dye-sensitized solar cells (DSSCs) employing Elaeocarpus serratus ethanol-extracted natural dyes. Notably, the 1:1 mixture of Weralu red and green DSSC exhibited superior photovoltaic characteristics, boasting a short-circuit current density ($J_{(SC)}$) of 3.842 mA/cm2, an open-circuit voltage ($V_{(OC)}$) of 406.2 mV, a fill factor of approximately 0.474, and an efficiency (η) of 0.739%. Comparative analysis revealed that this DSSC outperformed others, showcasing higher short-circuit current density and efficiency. The short circuit current and efficiency of the red, green, mixture of 1:2 and mixture of 2:1 dye sensitized soler cells are respectively, 3.101mA/cm^2 & 0.647%, 1.88mA/cm^2 & 0.482%, 2.485mA/cm^2 & 0.634% and 3.369mA/cm^2 & 0.672%.

The UV &Visible absorption spectrum of the Elaeocarpus serratus leaves ethanol extracted natural dyes were measured in the UV (200 nm to 400 nm) and Visible (400 nm to 750 nm) ranges and peaks obtained at 664.2nm, 605nm and 533.4nm for 1:1 mixture of red & green dye. The active pigment in the extraction seems to be Chlorophylls, Carotenoids and Anthocyanins. The presence of these supporting pigments may boot the sensitization process in a wider band that results high photovoltaic performance in the Elaeocarpus serratus green leaves sensitized DSSC.

The determination of the energy gap between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) using the Tauc relationship revealed distinctive values for various pigment sources. Notably, the 1:1 mixture of red and green dye exhibited a band gap of 1.77 eV, indicating the potential for efficient photon absorption and utilization in solar cell applications.

Additionally, the incident photon-to-current efficiency (IPCE) analysis demonstrated the efficient functioning of DSSCs in the wavelength range of 400 nm to 750 nm. The utilization of a mixture of red and green Elaeocarpus serratus leaf ethanol extract in DSSCs displayed a maximum wavelength in the range of 600 nm to 605 nm, with the 1:1 mixture contributing to the highest current. This finding underscores the significance of the natural pigment blend in achieving enhanced DSSC performance.

In conclusion, the study's key discoveries include the outstanding performance of the 1:1 mixture of Weralu red and green DSSC, the identification of active pigments, and the realization of efficient DSSC functionality within specific wavelength ranges. These findings hold promise for advancing the field of dye-sensitized solar cells and their application in sustainable energy solutions.

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