Simultaneous effect of activated carbon and chlorophyll pigment from leaves of acacia nilotica on the enhancement of electron transfer in DSSC applications

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Abstract

A well-defined attempt has been made to confirm the impact of green dye with activated carbon on the performance of Dye-sensitized solar cell (DSSC) compared to the natural red dye as sensitizer. Green dye from the leaf of Acacia Nilotica combined with the activated carbon derived from coconut Shell and red dye from beet root with activated carbon extracted using ethanol have been used to fabricate dye-sensitized solar cells. The pH of the extracts confirms the acidic nature of the dyes. The chlorophyll pigments in the green dye and activated carbon particles have pronounced effect for the efficiency of the solar cell. It is found that the solar cell with green dye and activated carbon has given an efficiency of 0.69% with fill factor, open circuit voltage and short circuit current density of 0.5361, 0.65 V and 2.0 mA cm$^{-2}$. The red dye with activated carbon as sensitizer has given the efficiency of 0.504%. Analysis of the green dye with activated carbon in DSSC has proven the enhanced electron injection with low charge transfer resistance of 58 Ω and higher rate of reduction of redox couple $I^-/I_3^-$. 

Introduction

To reduce carbon footprints and suppress electricity demand in the coming decades, researchers around the world are focusing on improving electrical efficiency and continuous power generation from renewable energy sources. Solar energy is one of the most important renewable energy sources for power generation. Because solar energy is non-polluting and abundant in nature, and the Sun sends solar radiation to the entire globe without discrimination.

The only challenge was determining how to harvest energy and convert total radiation to electrical energy. The Si-Solar cell is a highly efficient semiconductor material that is manufactured throughout the world. Thin-film photovoltaic cells, such as Perovskite solar cells, dye-sensitized solar cells, quantum dots, organic and polymer solar cells, are another type of solar cell. From above, dye-sensitized solar cells are non-toxic, eco-friendly, and inexpensive and natural based dyes are widely available in nature. According to the findings and literature, green colour pigments from leaves, fruits, and flowers are superior in light harvesting, and dye molecules emphasise the simplest way of electron transfer to the nanoparticle layer. The overall survey concludes that the green extracts of various plants studied by the researcher produce the highest photoconversion efficiency.

A thorough literature review has been reported to consider the references given by the various researchers regarding the green dyes extracted from the leaf of various plants.

Kumara et al [1] extracted the pigments of shisonin and chlorophyll from leaf of shiso to be utilized in DSSC. Cul has also deposited along with the pigments and tested for the performance. It has been found that the cell has given an efficiency of 1.3%. Ho Chang and Yu-jen Lo [2] extracted chlorophyll pigment from pomegranate leaf and anthocyanin pigment from mulberry fruit and used as sensitizers in DSSC separately and in mixed state. The
efficiency has been compared for the cell with dyes separately and in mixture and it is found that the conversion efficiency is about 0.722% for dye mixture. For chlorophyll and anthocyanin, it is found to be 0.597% and 0.548%. Jung-Kun Lee and Menglin Yang [3] reported a thorough review regarding the components of DSSC which includes semiconducting oxide film, dyes, electrolytes, counter electrodes used by various researchers and references are presented more precisely. Red cabbage has been used to extract anthocyanin pigment by Chiang-Yu Chien and Bar-Dar Hsu [4]. The solar cell is fabricated in the presence of coadsorbent, deoxycholic acid with a molar ratio of 1:40 and tested for the efficiency which was obtained about 1.4%. Giuseppe Celogero et al [5] investigated on many dyes from different fruits which contains betalain and anthocyanin which sensitize DSSC. Among various dyes, sensitizers from sicilion prickly pear fruit are reported producing best efficiency of 2.06%. For the fabrication of DSSC, Ananth et al [6] extracted lawsonia inermis seeds with pure TiO2 nanoparticles in anatase phase and pre dye treated TiO2 nanoparticles. The efficiency of the pre dye treated TiO2 nanoparticles with lawsonia inermis seeds was found to be 1.47%, which is 47% higher than the conventional DSSC. Torchani et al [7] investigated the performance of various natural dyes as sensitizers in DSSC by characterising the solar cells. Among the dyes, dyes extracted from Mallows and Henna dyes showed less degradation over time. Syafiner et al [8] created dyes from spinach leaves using ethanol and distilled water, which act as a photosensitizer for DSSC. It was discovered that the cell containing chlorophyll pigment extracted with distilled water had a fill factor of 0.49 and an open circuit voltage of 440 mV. Jitendra Kumar Sharma et al [9] investigated the use of extract from the leaves of the Calotropis gigantea tea plant for the synthesis of CuO Nanoparticles coated on FTO substrate used as a photoanode in DSSC. It was discovered that the proposed solar cell produced more efficient electrical energy of 3.4% with a fill factor of 0.62. Sengupta et al [10] investigated the efficiency of chlorophyll and betalain pigments from spinach and beetroot leaves as sensitizers in DSSC. The combination of spinach and beetroot dye demonstrated a higher efficiency of 0.294% when compared to other dyes reported separately.

Dyes have been extracted from leaf of Henna and beetroot and found the effect of dye on the performance of DSSC. XRD, UV-Visible, SEM and TG/DTA characterization study has been earned out by Sathyajothi et al [11] for further analysis. It has been found that the efficiency of the proposed solar cells for beetroot and henna dye are 1.3% and 1.08%. Hosseinnezhad et al [12] have studied the performance of DSSC incorporated with green dye extracted from the leaf of Sambucus ebulus. Efficiency of the proposed cell has been examined by varying the pH of the solution with dye and found that the acidic solution are less colorimetric and saturated. Ganta et al [13] have segregated chlorophyll and anthocyanin from cladode and aloe vera and combined together in order to use as sensitizer in DSSC. It has been found that the chlorophyll incorporated DSSC has given higher efficiency of 0.740%. Rajita Ramanarayanan et al [14] have extracted two pigments Viz., Chlorophyll and Betalain from red amaranth and used as sensitizer in DSSC. It is found that, the proposed solar cell with chlorophyll pigment has given efficiency of 0.53% with photocurrent density of 1.3 mA cm⁻². Natural dyes are extracted from flower, leaf, root, berk which contains various pigments Viz., anthocyanin, carotenoid, flavonoid and chlorophyll and used by the researchers all over the world. Results of the study are presented precisely by Geetham Richharia et al [15] and the effect on the performance of DSSC has been discussed in detail. Mahmoud A.M.Al-Alwani et al [16] have fabricated solar cell with the dye extracted from the leaf of Alternanthera dentata and bracts of Musa acuminate for its usage as sensitizer in DSSC. It is observed that the cell with chlorophyll has given conversion efficiency of 0.13% which is lower than the anthocyanin (bracts) pigment of conversion efficiency 0.31%. Hosseinnezhad et al [17] have used dyes extracted from Celosia cristata, Saffron, cynoglossum, eggplant peel to fabricate four DSSCs’ and tested for the performance. The study revealed that, sensitizer from celosia cristata has given higher efficiency of 1.38% compared to other sensitizers. Muhammed Zahir Iqbal et al [18] have made a comprehensive on the natural dyes extracted from various parts of the plants and highlighted the advantages over the synthetic dyes in various aspects. References of the researchers regarding the dyes rendering greater efficiency and stability.

Baskar et al [19] have extracted red dye from beetroot and green dye from spinach are mixed at different volumes and used as sensitizer in DSSC. It is found that the solar cell with 80% red and 20% green dye has given an efficiency of 0.99% than the cell with individual red and green dye. Kabir et al [20] have used red and green dye in the ratio of 80% and 20% extracted from Malabar spinach and red spinach and used as sensitizer in DSSC. Results of the study revealed that, the cell has given an efficiency of 0.847% compared to the dyes alone which are 0.466% and 0.531%. War Almaz Dhafina et al [21] have extracted anthocyanin from Rubra flower and chlorophyll from androgyous leaves and characterized for the effectiveness of the dye molecules with ZnO surface. It has been found that, the anthocyanin dye has better adsorption properly and good bonding with ZnO surface resulting in better power conversion efficiency of 0.038%. Anees A.Ansari et al [22] have synthesized Lanthanum based up conversion nanoparticles which are higher photochemical and thermally stable for the utilization in Dye sensitized solar cell. The surface functionality of Lanthanum based up conversion nanoparticle has enhanced the photocurrent and its application for upto electronic system construction. Sowmya et al [23] have extracted dyes from the leaves of Prosopis juliflora using the solvents of ethanol and acetone and used as sensitizer in DSSC. From the results, it is observed that an efficiency of 0.3% is obtained which is higher than
DSSC with other leaves from various plants. Mozhgan Hosseinezhad et al [24] have carried out a thorough review on the natural dyes extracted from various plants grown in Iran and presented the performance of DSSC. The study revealed that, dyes from agricultural water such as Walnut and Saffron petal shells can also be used as sensitizers in DSSC. Sabastine C Ezike et al [25] have extracted chlorophyll pigments and used as sensitizer separately. Also combination of the pigments are tried for the performance of DSSC at different volume ratio. Results of the study revealed that cosensitization of dyes has shown higher efficiency than the dyes alone.

Ona Navar et al [26] tested the pigments from algae, cyanobacteria, bacteria, archaea and fungi as sensitizer in DSSC instead of dye from various parts of the plants. It is observed that the cell with pigments from microorganisms cultivated from bioreactors has shown efficiency of DSSC varying between 0.001 and 4.6% which proved the significance of organism as sensitizer. Mozhgan Hosseinehahad et al [27] conducted DFT and CV studies for mix metal free dyes to predict the stability in Dye sensitized solar cell. Results of the DFT and CV study revealed that dye containing thiouindigo and cyanoacrylic acid in TiO2 layer has produced highest photocurrent compared to other five dyes. Ashish Kumar Keshari et al [28] have synthesized TiO2 nanoparticles from various precursors Viz., TiCl4, Titanium isopropoxide and Micro sized TiO2 nanopowder and tested for treatment of water and DSSC. It has been found that conservation of micro sized and TiCl4 precursors with natural dyes is effective on the performance of DSSC. Biswajit Nath and Sujit Roy et al [29] used phase change material in DSSC and tested the photoelectric conversion efficiency between the temperature ranging from 34°C and 48.6°C. Octadecane is used as PCM and it is found that, DSSC has intended temperature equal to 48.6°C.

Figure 1(a) shows the block diagram of proposed DSSC system with Acacia Nilotica as a sensitizer dye. In the present study, co-sensitizer of activated carbon derived from coconut shell with green dye from leaf of Acacia Nilotica and co-sensitizer of activated carbon derived from coconut shell with red dye from the beetroot has utilized as sensitizer for light harvesting in the photoanode of DSSC. The performance has tested and comparison has done for both DSSCs in terms of fill factor, short circuit current density, open circuit voltage and efficiency. The constituents in the green extract have been confirmed with the basic chemical tests and explained. Figure 1(b) shows a schematic sectional view of proposed DSSC system.

Experimental methods

**Extraction of dye from acacia nilotica, beetroot and activated carbon from coconut shell**

*Acacia Nilotica* is an herbal plant taken from the VIT farms, Vellore whose leaf is rich in flavonoids, saponins, tannins and triterpene, steroids and carbohydrates. Leaves of *Acacia Nilotica* have been collected and washed thoroughly with distilled water for removing dust particles. Leaves are allowed to dry in a shadow region for more than 48 h and are crushed into fine powder using pestle and mortar. The crushed fine powder is kept in hot air oven at 50°C for 2 h to remove the volatile impurities. Then it is stirred with ethanol for 2 h and filtered using filter paper to get the dye extract.

Fresh Vegetable beetroot was taken from the VIT farms, Vellore and it is squeezed to get the juice and dried for more than 48 h. The water content in the juice is evaporated and juice is allowed to dry. The dried powder of the beetroot is crushed using pestle and mortar and kept in hot air oven at 50°C to remove the moisture content from the powder. Then it is mixed with ethanol and stirred well for more than 2 h and filtered using Whatman filter to get red dye extract from beetroot.

Coconut Shell was collected from the VIT farms, Vellore and burnt at 650°C and it is broken down into fine carbon particles by adding NaOH solution drop by drop. The carbonized carbon particles was again converted into activated carbon. Figures 1(c), (d) and (e) represents the photograph of the dyes prepared from *Acacia Nilotica*, beetroot and activated carbon from Coconut Shell.

Results and discussion

Dyes extracted from the leaves of Acacia Nilotica were analysed using FTIR and UV - Visible spectra to identify functional groups and absorption wavelengths for the performance of the proposed solar cell. Shimadzu UV-Visible spectrometer of UV-1700 series has been used to perform the UV-Visible spectroscopic study of the dyes and for the FTIR spectroscopic study, IRAffinity with MIRacle 10 in ATR mode has been utilized. XRD and SEM were recorded for the semiconductor oxide after annealing using the 3rd generation Empyrean, Malvern Panalytical multipurpose diffractometer with MultiCore Optics and Scanning Electron Microscope of EVO 18 model with low vacuum facility and ALTO 1000 cryo attachment respectively.

The FTIR and UV-Visible spectrums of the green dye from the leaves of Acacia Nilotica are shown in figures 2 and 3. The FTIR spectrum shows C–H stretching, confirming the presence of alkyne and alkanes for the dye’s bonding with the TiO2 semiconductor oxide. C–O stretching occurred with the confirmation of secondary alcohol, which is responsible for the photoanode’s good anchoring effect with the semiconductor oxide
Figure 1. (a) Block Diagram of Proposed DSSC. (b) Schematic Sectional view of Proposed DSSC. (c) Green dye extracted from leaves of Acacia Nilotica. (d) Dye extracted from beetroot. (e) Process of preparing Activated carbon from coconut shell.

Figure 2. FTIR spectrum of dye extracted from Acacia Nilotica.
material. The presence of anhydride and alkene functional groups has been confirmed by CO–O–CO stretching and C=\text{C} bonding.

The UV-Visible spectrum of the dye from leaves of Acacia Nilotica demonstrates that the dye has good absorption in the UV region, with high intensity peaks at 420 nm and 430 nm. Following that, the intensity of absorption falls in the visible region and has moderate absorption ranging from 480 nm to 640 nm. It is because flavonoids, saponins, tannins, and carbohydrates absorb well in the ultraviolet and visible regions of the electromagnetic spectrum.

**Carbohydrates confirmation**

15 gm of the dye extracted has been put in 200 ml of distilled water and heated at the temperature of 40°C. Then the water extract is filtered and allowed to dry in a boiling water bath. Gummy residue is obtained and confirmed the presence of carbohydrates.

**Saponin**

The dye is shaken with water and it is observed that foam is produced confirming the presence of saponins in it.

**Flavonoids**

Appearance of red colour when magnesium turnings and 1 to 2 drops of concentrated hydrochloric acid is added confirmed the presence of flavonoids.

**Tannin and Phenolic compounds**

Addition of Ferric Chloride to the dye has changed the colour of Ferric Chloride into Greenish black colour confirming the tannin and phenolic compounds in the dye.

**Steroid acid**

2 ml of chloroform in the dye with 10 drops of acetic anhydride and 2 drops of concentrated sulphuric acid produced a bluish red to cherry red colour, confirming the presence of steroids in the dye.

The above chemical test results for the confirmation of dye constituents are consistent with the findings of Marimuthu Krishnaveni and Ravi Dhanalakshmi [30].

The FTIR and UV-Visible spectrums of the dye extracted from beetroot are shown in figures 4 and 5. The presence of O–H stretching in the FTIR spectrum confirms the carboxylic and alcoholic group. The functional groups listed below have a strong anchoring and bonding effect with TiO\textsubscript{2} semiconductor oxide. The absorption wavelength ranges from 225.30 nm to 434.64 nm based on the UV-Visible spectrum. It concludes that the constituents in the dye extracted from beetroot absorb in the ultraviolet region of the electromagnetic spectrum, but have no effect on solar radiation absorption in the visible region.

**XRD, SEM and EDAX of activated carbon from coconut shell**

Figure 6 represents the XRD spectrum of the activated carbon obtained from coconut shell. The amorphous nature of the activated carbon prepared from coconut shell has been confirmed from the broad peaks obtained at 24° and 43° corresponding to (002) and (100) plane respectively. The activated carbon has shown good absorption due to larger surface area with micropores and SEM image is represented in figure 7. To calculate the...
size of the activated carbon, Debye-Scherer formula has been used with the shape factor of 0.9 due to amorphous nature of the sample. Knowing the values of \( \lambda \), \( \beta \) and \( \theta \) which are wavelength of anode material, full width half maximum and diffraction angle, the size of the activated carbon is found to be 89.7 nm. Figure 7 represents the SEM image of the activated carbon and it is found that the surface of the carbon is smooth and homogeneous. It seems to be flakes with micropores which has surface area for large electron injection capability in the semiconductor oxide material.

The elemental composition has been analyzed with the help of Energy Dispersive x-ray analysis. EDAX spectrum of the activated carbon which also confirms the presence of carbon in the material is represented in
figure 8. Absence of any other element supports the XRD pattern as well as purity of the prepared sample. Due to the atmospheric oxygen, a small amount of O peak has also been observed.

**Electrolyte and counter electrode**

0.67 g of lithium iodide, 0.13 g of iodine, and 10 ml of acetonitrile were thoroughly mixed in a beaker using a mechanical stirrer for 15 min. The mixture serves as the electrolyte in the proposed solar cell. As the counter electrode, a graphene-coated FTO substrate was used.

**Fabrication of solar cell**

Two cells have been fabricated with two different photoanodes viz., one with green dye extracted from the leaves of *Acacia Nilotica* incorporated with activated carbon prepared from coconut shell and other one with red dye extracted from beetroot. The two cells have prepared with $I^-/I_3^-$ electrolyte and graphene based counter electrode. The photograph of the two cells prepared from the dye Extracted from leaves of *Acacia Nilotica* with activated carbon prepared from coconut shell and from dye from beetroot with activated carbon prepared from coconut shell is represented in figures 9(a) and (b). Five solar cells for each dye had fabricated and tested. Among
the five cells, two cells with each dye had used and in terms of reproducibility, the cells are perfectly sandwiched with photoanode, electrolyte and counter cathode to show JV characteristics.

The fabricated cells are subjected to JV characterization to find the significant parameters of fill factor, open circuit voltage, short circuit current density, maximum current and voltage to predict the performance. Solar simulator with Xenon lamp of power of 100 mW cm$^{-2}$ incorporated with Keithley 2400 Graphical Series SMU has been used to record the JV characteristics study for the Dye sensitized solar cell. JV characterization graphs of the solar cells have been represented in figure 10. The solar cells are fabricated in the laboratory condition with humidity and ambient temperature of 72% and 32 °C.

From the JV characteristics of the solar cell utilizing the green dye (Acacia Nilotica and activated carbon) has shown significant impact on the electron injection, charge transfer and reduction of redox couple. It is found that, the oxidation of the dye after absorption of photon energy and regeneration of dye molecules by reduction happened within picosecond. The green dye extracted from the leaves of Acacia Nilotica has the pigment of flavonoid which made a good bond with the semiconductor oxide material and the activated carbon from coconut shell in the dye prevents the recombination of electrons. This is in turn increases the electron injection to the TiO$_2$ material and the electrons reaches the FTO transparent substrate. The injected electrons move towards the counter electrode via the external circuit constituting the photocurrent. It is possible to find the

Figure 9. (a) Solar Cell with Dyes from Acacia Nilotica and activated carbon from coconut shell. (b) Solar cell with dyes from Beetroot and activated carbon from coconut shell.

Figure 10. JV Characteristics of Solar cell with dyes extracted from leaves of Acacia Nilotica and Beetroot with activated carbon from coconut shell.
transient photocurrent which is based on the varying biasing voltage which will clearly explain the conjoint trend between the theoretical and experimental values.

It is found that, the proposed solar cell with green dye and activated carbon has produced maximum current density of 1.7 mA cm$^{-2}$ with maximum voltage of 0.41 V. The cell produced open circuit voltage of 0.65 V with short circuit density of 2.0 mA cm$^{-2}$. Fill factor of 0.5361 is obtained giving the efficiency of 0.697%. In case of red dye from beetroot and activated carbon contains betalain consists of betacyanin and betaxanthins has not responded to the light photons as that of the green dye. In this context, the time taken to inject the electrons to the TiO$_2$ material is less than that of the recombination with the electrolyte. Therefore the oxidation of dye molecule with the absorption of photon energy is not done significantly compared to the process taking place in the solar cell with green dye and activated carbon. It is found that, the solar cell with dye from beetroot and activated carbon has given an efficiency of 0.504% with fill factor of 0.427. From the JV characteristics of solar cells have been drawn with trendline to good interpolation and the equations for current density in terms of voltage has been derived.

Also the EIS model has been developed and MATLAB program has been written and the following characteristics parameters of the fabricated dye sensitized solar cell are evaluated [31]. The parameters are Cell characteristic resistance, $R_{cell} = 325.0000$ (ohms) Normalized $V_{oc}$, $V_{oc} = 24.0774$ (units) Normalized series resistance, $r_s = 0.0031$ (units) Approximate fill factor, FF, with $R_s$, $FF_{approx} = 0.8295$ More accurate FF valid for $r_s < 0.4$ and $V_{oc} > 10$ FF, 0.8293. The Series resistance in fabricated solar cell has three consequences viz., the flow of electrons that is the current through the solar cell, the resistance in the junction between the various interfacial layers and the resistance on the front and back of the solar cell. The series resistance will influence the fill factor with the decrement of the value to a certain extent [32].

**Conclusion**

The solar cell with green pigments and activated carbon has higher efficiency compared to the cell with dye from beetroot and activated carbon. The trend line equations for the cells are found as for solar cell with dye from leaves of *Acacia Nilotica* and activated carbon from coconut shell is

$$Y = -5.7205x^2 + 1.4717x + 2.0493$$

$$R^2 = 0.9917$$

For solar cell with dye from beetroot and activated carbon from coconut shell is

$$Y = -4.3921x^2 + 0.403x + 1.9557$$

$$R^2 = 0.9943$$

The electron injection to the TiO$_2$ is taken place in the solar cell with green dye and activated carbon is found to be 68 ps whereas for the solar cell with red dye is found to be 54 μs. Minimum time taken for electron injection for the green dye and activated carbon is due to the presence of flavonoids, saponins, tannins and triterpen, steroid and carbohydrate in the leaf and surfactant effect of activated carbon. The surfactant effect of the activated carbon protects the dye molecules from degradation and used for the repetition of the photocurrent production without interruption. The electron transfer resistance for the electrons for the reduction of redox couple for the solar cell with green dye and activated carbon is found to be 58 Ω which is very small.

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**Figure 11.** Series resistance diagram for the proposed DSSC.
Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Authorship contribution statement

All the authors contributed to the manuscript and approved for submission. Dhana Sekaran P worked on validation, formal analysis, investigation, methodology, writing original draft.

Marimuthu. R performed writing, review and editing

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