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Natural pigment based dye sensitized solar cells: photovoltaic studies: a sustainable energy solution

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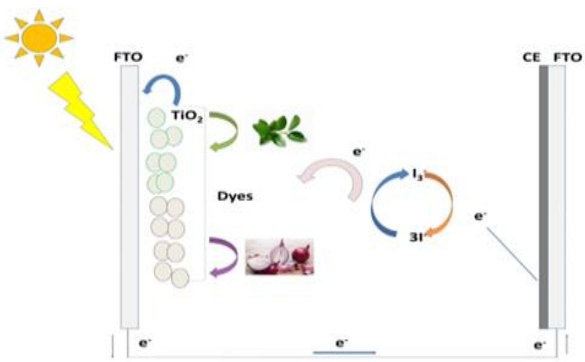
Graphical abstract



Abstract

The present research work is carried out on fabrication of natural pigment-based dye-sensitized solar cells (DSSCs) utilizing Lemon leaves and Onion peels extracted dyes as natural photosensitizers. The absorption spectra analysis shows the major peaks for onion peel at 370 nm and 475 nm and 664 nm for lemon leaf that provides valuable insights regarding the narrowing of band gap of TiO_2 from standard value (3.25eV) to 2.87 and 2.44 eV by incorporation of Onion peel and Lemon leaf pigments respectively and showing the potential of natural pigments for renewable energy applications. The photovoltaic parameters revealed that the Lemon leaf extract exhibited higher power conversion efficiency (0.92%) than the Onion peel extract with an efficiency of 0.054%. Additionally, impedance analysis was conducted to understand the charge transport and recombination processes within the DSSCs which revealed a better lifetime of 0.0349 sec for lemon leaf and 0.0143 sec for onion peel extracted DSSCs. The preliminary photovoltaic results indicated the potential of Lemon leaf and Onion peel extracts as eco-friendly and cost-effective natural photosensitizers for DSSCs using Platinum free counter electrode.

Keywords: TiO_2, DSSCs, Onion peel, Lemon leaves, Natural Pigments, Narrowing band-gap

1. Introduction

Every day since the turn of the 20th century, there has been an increase in the demand for energy. Coal, oil, and natural gas are regarded to be the primary energy sources used in today's society (Zou et *al*. 2016; Dang et *al*. 2018). Fossil fuels are used to create around 90% of all electricity. Without electricity, society as a whole would fall apart in a day. The majority of the thermal energy used for industrial and home purposes, in addition to electricity, is produced by energy sources based on fossil fuels. The issue with producing energy from fossil fuels is that they are not sustainable and are quickly running out (Sorrell et *al.* 2015; Stuckelberger et *al.* 2017). Additionally, using fossil fuels contributes to environmental problems such ozone layer loss, acid rain, acid rain, water pollution, air pollution, land pollution and global climate change. A potential replacement for conventional fossil fuels is the use of sustainable energy sources, such as solar energy, wind power, hydropower, wave energy, tidal energy, biomass, and geothermal energy. Unlike traditional fossil fuels, which are concentrated in a small number of countries, renewable energy resources span a large geographic area (Solangi et *al.* 2011; Zhang et *al*. 2018). The development of low-cost photovoltaic technologies has caught the interest of both academic and industrial research communities due to the ever-increasing trend of demand for sustainable energy supply. Solar energy is the easiest to extract from all other renewable energy sources using a photovoltaic (PV) system. There are three distinct generations of PV cells: Silicon wafer-based solar cells of the first generation, Copper Indium Gallium selenide (CIGS), Cadmium Telluride (CdTe) and Gallium Arsenide (GaAs) of the second generation, and Quantum dot solar cells, Organic solar cells, multi-junction solar cells, and Dye-sensitized solar cells of the third generation (Khatibi et *al.* 2019; Ole et *al.* 2012). Third-generation solar cells include the DSSC, often referred to as the Gratzel solar cell. Since O'Regan and Gratzel first developed a novel type of solar system, DSSC has received a lot of attention due to its straightforward manufacturing method, environmental friendliness, low cost of assembly, and good cell performance in low light conditions (O'regan et *al.* 1991). In this context, dye-sensitized solar cells being researched in particular as effective substitutes for conventional solar cells due to their desired ecological and economic characteristics, simplicity in manufacture, and potential for optimising optical qualities through molecular design (Ahmed et *al*. 2018). The idea of natural photosynthesis served as inspiration for the way DSSCs function, and pigments are crucial to the devices' ability to respond to a wider range of wavelengths. Several approaches, including enhanced oxidation processes, dye quantity reduction, and replacement with natural colours, have been put forward by various research institutions and governmental organisations to address the difficulties of establishing water treatment technology (Taya et *al.* 2015). The sensitizer, which absorbs sunlight and triggers the charge separation process, is one of the crucial parts of DSSCs. It is essential to improve the dyes' capacity for light absorption in order to increase the power conversion efficiency of DSSCs. Investigation and improvement of the dye-substrate interaction, which plays a role in the electron transfer process, are also essential. Ruthenium-based dyes, have a high conversion efficiency of more than 10%, but they are also very expensive and unsustainable (Rajendhiran et *al.* 2023; Gratzel et *al.* 2009). Natural dye is an alternative to costly, harmful dye that must has comparable conversion efficiency and is nontoxic, biodegradable, less expensive, and simple to extract. Natural dyes, which are produced using sustainable bio resources, are biodegradable, affordable, and ecologically friendly options that can effectively replace synthetic colours. In DSSCs, natural dyes derived from plants have been proven to be effective photosensitizers for instance, Abhishek Attri presented a report based on various dyes and observed a photo conversion efficiency of 0.13% with the use of radish leaves as a sensitizer (Attri 2019). In present work, we have investigated the performance of DSSC with natural dyes based on Onion peels and Lemon tree leaves. The study aims to explore the potential of these natural dyes as photosensitizers for DSSCs. By understanding the electronic properties and light-harvesting capabilities of the natural dye solution, the improved light absorption and charge transport within the cell can be optimized. This research aims to identify the potential of natural pigments with motto to improve the efficiency and sustainability of DSSCs with an approach to utilise renewable resources to drive clean energy innovation. With the potential to pave the way for more eco-friendly and accessible solar energy solutions, the natural dyes in DSSCs represents a step forward in our journey towards a greener and more sustainable future.

2. Experimental materials

Onion peels, Lemon leaves used in this study as a sensitizer. FTO conductive glass (sheet resistance: 7Ω/sq), Degussa P25 Titanium dioxide nanopowder (99.5%, Nanoshel India), carbon nanopowder (<100nm, 97%, Sigma Aldrich), Ethyl cellulose, Terpinol, 1-butyle 3-methyl imidazolium iodide (BMII), lithium iodide (LiI), iodine (I_2), 4-tert butyl pyridine, ethanol, acetone, 2-propanol, acetonitrile, HCl, 10% Triton X-100 (Fisher Chemical) and valeronitrile etc. purchased all from Sigma Aldrich.

2.1. Extraction of natural dyes

Onion peels have Anthocyanin and Lemon leaves contain Chlorophyll dyes as natural pigments (Kumara et *al.* 2013), acidified ethanol was used as the extractant for dye extraction from Onion peels and while concentrated ethanol was used for Lemon leaves. For this, 06g of Onion peels were dried and crushed to make a fine powder and mixed in 250 ml of acidified (.01% HCl) ethanol solution. The mixture was left in the dark room for whole day and after that the dye extract was filtered with the use of filter paper. For the preparation of lemon leaf dye, 40 g of air-dried Lemon leaves were crushed into fine powder and disperse into minimum amount of ethanol (95%) to get concentrated solution. The dispersions were heated for 12 hours at 60 °C in closed reflux. Both the dispersions were filtered to get the dye. The subsequent filtering of the solution produced a clean concentrated filtrate that was employed as a natural sensitizer. All dye preparations were kept in the dark and cool place.

2.2. Preparation of TiO_2 paste

TiO_2 paste was prepared by mixing 0.5 g titanium oxide (TiO_2) in 10 mL of ethanol solution. 0.35 g of ethyl cellulose was added to the above mixture, and mixing was done with a magnetic stirrer after adding few drops of Terpinol (Dhungel et *al.* 2010).

2.3. Preparation of photoanode

Firstly, Fluorine-doped tin oxide (FTO) substrates were ultrasonically cleaned in ultrasonicator bath with deionized water, acetone and 2-propanol for each cycle of 15 min and then treated by UV-ozone system for 15-20 min. The photoanode were prepared using the method reported (Lokman et *al*. 2019), with the deposition of three layers of dye coated TiO_2 on top of FTO substrate by doctor's blade using 3M scotch tape to control the thickness upto 16 micrometres, annealing is done in a heating furnace for 30 min at 450˚C. Subsequently, TiO_2-coated fluorine-doped tin oxide (FTO) was stored in a dark area and submerged in the dye solution for a maximum of 24 hours. The excess dye was removed using ethanol and let to air dry.

2.4. Preparation of counter electrode

The counter electrode was prepared using the same FTO cleaning process and the same blade method adopted in depositing the TiO_2 making a Carbon nano paste (Kouhnavard et *al.* 2016) onto the FTO substrate and then film was annealed for 30 minutes at 450˚C in a furnace.

2.5. Fabrication of DSSC

The natural dye-sensitized photoanode and Carbon coated Counter electrode were assembled together to fabricate a DSSC by adding few drops of redox electrolyte solution (I_3^-/I^-) in between them. The electrolyte solution was prepared deploying 0.1 M LiI, 0.05 M I_2, 0.6 M BMII, and 0.5 M 4-tert-butyl pyridine in 17 mL of acetonitrile and 3 mL of Valero nitrile solution. To acquire a uniform solution, the mixture was stirred for 2 hrs as shown in **Figure 2**. 1µm thick counter electrode was prepared by using 10% Triton X-100 as a solvent.

3. Results and discussion

The absorption spectrum of natural dyes was carried out by UV/Vis/NIR spectrometer (PerkinElmer lambda 750). Surface morphological study of counter electrode was investigated by using field emission scanning electron microscope (FESEM- MIRA 3 TESCAN). J-V performance of DSSC was carried out by solar simulator (Verasol ORIEL LSS-7120) at AM 1.5 G under irradiation. The charge transfer, electronic and ionic processes were investigated by using Impedance analyser (Auto lab PGSTAT30) at 0V under dark condition.

3.1. UV-VIS NIR Spectroscopy

The specific absorption spectra for the extracts of Lemon and Onion loaded on TiO_2 layer were studied. Different DSSCs were prepared using different dyes over the TiO_2 layer. **Figure 1(a, b)** show the absorption spectra of these samples dissolved in ethanol. The Onion peel extract was shown a peak around 370 nm, it's correlated to the anthocyanin. These peaks could enhance the charge transfer reaction in the assembled DSSC when placed under solar radiations (Jalali et *al*. 2020).

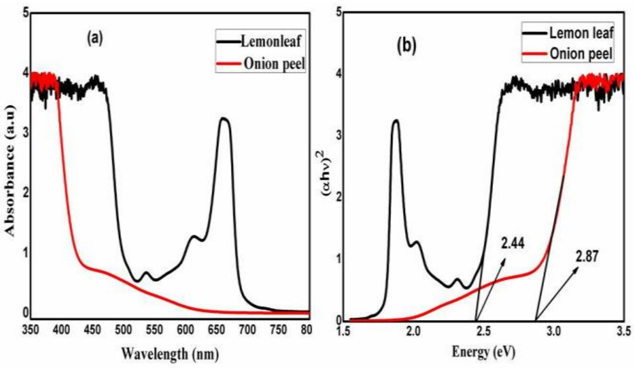


Figure 1. (a) Absorption spectra of TiO_2 coated dyes (b) Tauc plot of different dye coated TiO_2 electrode

Usually, the absorption peaks in visible range are dispersed through the wavelength range of 400–900 nm. The Lemon leaves extract displayed two major absorption peeks at 475 nm and 664 nm which represent the component of the dyes extract is chlorophyll (Rekha et *al.*2019). The chlorophyll dye coated TiO_2 represented the first absorption spectral peaks spreading between wavelength of 400-490 nm and second absorption wavelength 630-700 nm (Strack et *al.* 2003; Azeredo et *al.* 2009). The absorbance peak at 400-490 nm indicates the $\pi \rightarrow \pi^*$ transition, while the absorptions are observed to peak at 664 nm reflecting the $n \rightarrow \pi^*$ transition. Due to conjugated complex structure, transitions in chlorophyll require less energy for any electronic transition than in non-conjugated systems (Bruice 2004). In the fabricated DSSC, these peaks might be responsible to accelerate the charge transfer reaction under sunlight. These results showed that lemon leaf extract coated TiO_2 layer impregnated by chlorophyll pigment, which enables it to be employed as DSSC photosensitizers due to their visible spectrum light absorption. Lemon leaf dyes have the ability to increase absorbance and expand the area where light is absorbed.

The energy band gap of the Lemon leaf and Onion peel dyes was calculated using the Tauc-plot equation 1(Khakhal et *al.* 2020)

$$(\alpha h\nu)^2 = \beta(h\nu - E_g) \qquad (1)$$

Where h, E_g, β, α and ν represent Plank constant, band gap, proportionality constant, absorption coefficient and frequency respectively. The Tauc's plot for Lemon leaf and Onion peel dyes are shown in **Figure 3(b)**. The bandgap energy of the Lemon leaf and Onion peel dye coated TiO_2 are found 2.44 eV and 2.87 eV respectively, which is lower than the standard band gap of pristine TiO_2 (3.25eV). As a results, narrowing of the band gap is observed for both dyes, Lemon leaf band gap energy much more lowered as compare to Onion peel can be ascribed to more numbers of conjugated bonds due to which the electron may quickly excited from the VB (valance band) to the CB (conduction band). By providing additional electron states, Natural dyes can enhance the light absorption properties of TiO_2, leading to improved photovoltaic performance (Natrayan et *al.* 2024).

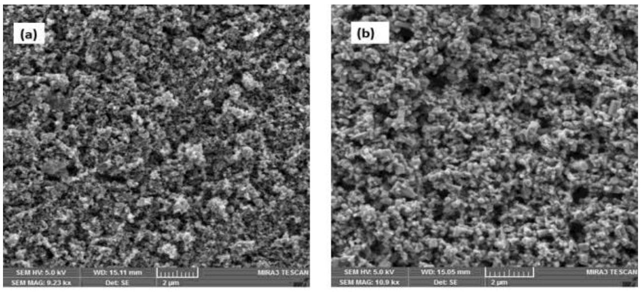


Figure 2. FESEM images of (a) Onion peel extract coated TiO_2 (b) Lemon leaf extract coated TiO_2

3.2. Surface Morphology

The surface morphology of the TiO_2 nanoparticles coated with Lemon leaf and Onion peel extracts directly affects the efficiency of DSSCs. The Lemon leaf or Onion peel dye-coated nanoparticles exhibit a homogeneous particle size distribution. In **Figure 2**, higher degree of agglomerated clusters connected to each other on the surface are visible through which electron transport is established in the resulting DSSCs. Thus, the SEM surface morphology images reveal that natural pigments are suitable for DSSC as electron acceptors. Optimal surface characteristics are responsible for improved light absorption, efficient electron transfer, and to achieve better power conversion efficiency (PCE) in DSSCs (Faisal 2020).

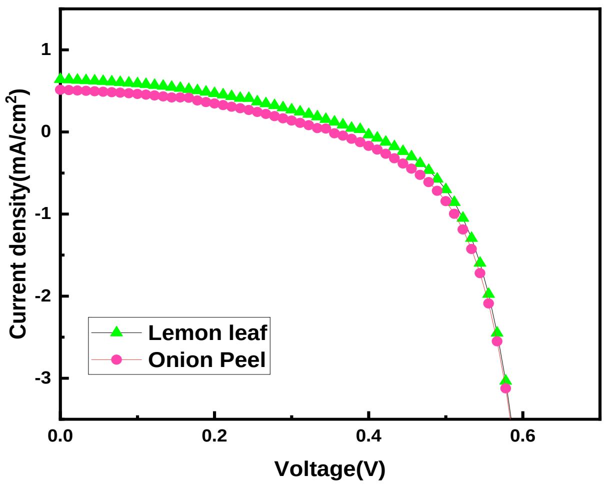


Figure 3. Current-voltage curve for DSSC using Lemon leaf and Onion peel dyes at room temperature under AM 1.5 G illumination condition

3.3. J-V Characteristics

DSSCs were created to test the suitability of natural and direct dye to affordable, environmentally friendly devices. **Figure 3** compares the current density and voltage (J-V characteristics) of the Lemon leaf dye and Onion peel dye, and **Table 1** lists the related performance metrics with co-operating the current work with some earlier reported results. The J-V tests were carried out at room temperature with AM 1.5 G illumination (100 mW/cm^2). Short circuit current (J_{sc}) of 0.49mA/cm^2 and open-circuit voltage (V_{oc}) of 0.35 V, FF of 38.02 and efficiency of 0.054are displayed by DSSCs made using Onion peel dye. In comparison to Onion peel-based dye, Lemon leaf based DSSC offers high Jsc (0.50mA/cm^2), V_{oc} (0.38V), and FF of 39.46%, all of which contribute to a PCE of (0.92%).

This PCE value of Lemon leaf-based dye is one of the best for DSSC made with natural dyes that has been recorded (Kumar et *al.* 2021; Kharkwal et *al.* 2021; ADU et *al.* 2023). The improvement in short circuit current density can be ascribed to increase in dye attachment to the surface of TiO_2. As the TiO_2 surface absorbs more dye molecules it can generate a greater number of photons from sunlight, which leads to a rapid electron injection (Inbarajan et *al.* 2022). With Lemon leaf electrolyte, the short circuit current density is greater than Onion peel electrolyte for DSSC. This is because the electrolytes may have lowered TiO_2's Fermi level. The DSSC with Lemon leaf electrolyte showed an increase in current density as a result of the rate of facile electron injection into the conduction band. Additionally, this may indicate a decrease in the rate of electron recombination to the dye's hole and the electrolyte's triiodide ion. Nonetheless, there is a 10% difference in the open circuit voltage between the DSSC with Lemon leaf electrolyte and the DSSC with Onion peel electrolyte. The Fermi level has been decreased in the direction of the redox mediator potential, as this data verifies. This data also suggests that iodide ion conductivity has a significant role in the J_{sc} of solar cells containing I^-/I_3^- mediator. Lemon leaf electrolyte has more I^-ions than Onion peel electrolyte in terms of the number density of charge carriers. This demonstrates that the Lemon leaf electrolyte may accelerate the exchange of I^- to I_3^- and I_3^- to I^- ion more quickly, which will cause the dye molecules to regenerate fast, while Onion peel electrolyte has shown a lower number of I^- and a slower pace of redox process than Lemon leaf electrolyte (Hassan et *al.* 2016).

Table 1. Photovoltaic Parameters of DSSCs

S.No.	Dyes	V_{oc} (V)	J_{sc} (mA/cm^2)	FF	Efficiency	Ref.
1.	Onion Peel	0.33	0.96	25.64	0.05	(Adul et *al.* 2023)
2.	Chlorophyll and Betalain Pigment	0.43	0.09	55.00	0.04	(Philibus et *al.* 2016)
3.	Onion Peel	0.48	0.24	46.63	0.06	(Ammar et *al.* 2019)
4.	Red Cabbage	0.51	0.21	46.61	0.06	
5.	Papaya Leaves	0.32	0.36	56.00	0.07	(Suyitno et *al.* 2015)
6.	Lemon Leaves	0.59	1.08	10.00	0.03	(Maabong et *al.* 2015)
7.	Onion Peel (A)	0.35	0.49	38.02	0.05	Present Work
8.	Lemon Leaves (B)	0.38	0.50	39.46	0.92	

4. Impedance analysis

The impedance spectra, recorded at 0.1 V, spanned a frequency range from 1 Hz to 100 kHz. **Figure 4** illustrates the Nyquist plot of both dye-based devices in dark conditions. Generally, the Nyquist plot of DSSC shows three frequency regions (Kharkwal et *al.* 2024). The high-frequency region can be recognised to the charge transfer resistance at Carbon/electrolyte interface. The middle frequency region represents to the charge transfer recombination resistance at TiO_2/dye/electrolyte interface. The low frequency region is represented to the diffusion properties of the redox couple (I^-/I_3^-) in the electrolyte (Ammar et *al.* 2019). In **Table 3**, R_s and R_{rec} represent the series and recombination resistance of the cell. The high value of recombination resistances (1704.1 Ω**)** implies a slower rate of electron recombination between injected electrons and I_3^- in the electrolyte, particularly in Lemon leaf dye (Amogne et *al.* 2020). The observed increase in short-circuit current density and conversion efficiency aligns with the results of Lemon leaf dye-based device. In Onion peel dye-based devices, recombination resistance decreases, as shown in **Table 2**. The high internal resistance within the cell could be the reason of high recombination rate. The DSSC's high internal resistance, attributed to significant recombination, leads to poor performance (Ghann et *al.* 2017). The carrier lifetime values precisely correspond to the recombination resistance (R_{rec}) of devices with different dyes (Maiaugree et *al.* 2015).

Table 2. Different Impedance parameters for DSSCs

S. No.	Dyes	$R_s(\Omega)$	$R_P(\Omega)$	CPE parameters		Capacitance (C_P) (μ_F)	τ_n (Sec.)
				Q_o	N		
1	Lemon leaf dye	97.51	1704.1	3.843×10^{-5}	0.81	2.045×10^{-5}	0.0349
2	Onion peel dye	118.74	1020.2	2.923×10^{-5}	0.82	1.403×10^{-5}	0.0143

Lemon leaf-based DSSC exhibits minimal recombination due to efficient charge transfer between the dye and TiO_2. A longer carrier lifetime is associated with higher recombination resistance, and a high diffusion length, restricting from a prolonged carrier lifetime, improves photocurrent and charge collection rates (Faraz et *al.* 2021).

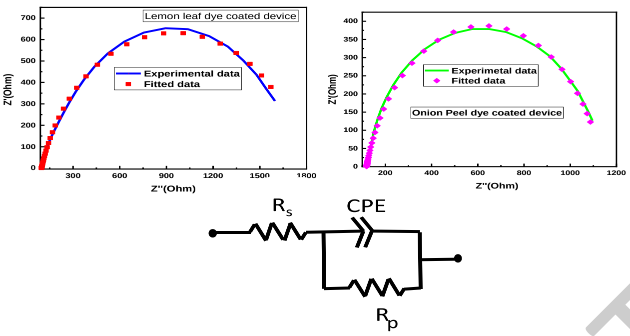


Figure 4. Impedance spectra for DSSC made with Lemon leaf and Onion peel dyes. The electrical equivalent circuit model (EECM) of the DSSCs understudy

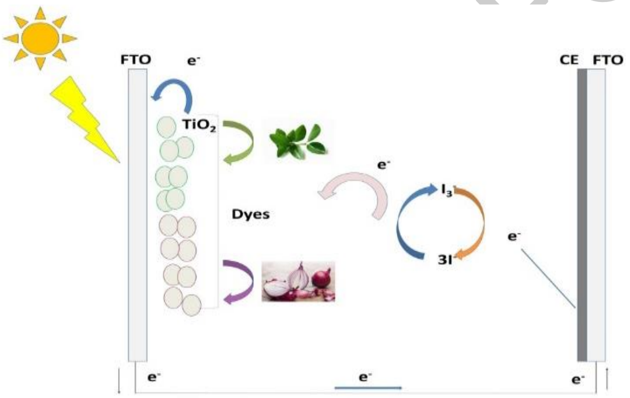


Figure 5. Schematic of a DSSC sensitized with the extracted dyes

5. Conclusion

The outcomes of this research propose that natural dye-based sensitizers can provide fair photovoltaic performance for DSSCs. The narrowing of band-gap of TiO_2 by the utilization of Lemon leaf and Onion peel extracts in DSSCs opens up avenues for greener and more sustainable energy solutions, promoting the development of eco-friendly technologies for a cleaner and more sustainable future. We optimized factors such as the dye concentration, annealing temperature, the thickness of the photo-anodes, surface morphology and absorption of dyes. These parameters were carefully adjusted to maximize the light absorption and charge transport efficiency of the DSSCs. However, it has been observed that the conversion efficiency for natural dyes is not sufficient enough for fabricated DSSC, but by considering other factors like type of electrode materials, electrolytes, mixing of dye solutions etc. may provide better performance in near future works. It is further observed that in case of Lemon leaf dye-based device PCE optimized in AM 1.5 G under irradiation is slightly more than Onion peel dye. Based on our findings, it can be concluded that the natural pigment based DSSCs must be further investigated as greener, cost-effective, good alternative to the costly Ruthenium complexes and propose a metal free, climate adaptable solutions in renewable energy sectors. As the efficiency of DSSC depends on several factors, further investigations on optimization of some important parameters like materials of electrodes and electrolytes used will be considered in our future experiments to enhance the efficiency of DSSC using combinational approach to fabricate DSSC.

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