



Utilization of Naturally Occurring Pigment Lycopene as a Photosensitizer for ZnO based Dye-Sensitized Solar Cells

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Abstract

In this study, we present lycopene-sensitized ZnO (Zinc Oxide) as a photoanode for dye-sensitized solar cells (DSSCs). To obtain ZnO nanopowder, initially zinc peroxide is synthesized and then thermally decomposed. The bandgap of 3.25 eV was estimated by Tauc's formula using the diffused reflectance spectra. Column chromatography was used to isolate lycopene (dye) molecules from a mixture of carotenoids and lycopene. The ZnO films were deposited on the conducting side of Indium-doped Tin Oxide (ITO) glass by the doctor blade method. The dye adsorption time on ZnO film was optimized. These DSSCs show better performance for dye loading times of 24 hours. A photoconversion efficiency of about 0.39% with 281 mV open-circuit voltage (Voc), 176 Acm⁻² photo-current density (Jsc), and a 47% fill factor (FF) has been achieved for a novel lycopene-sensitized ZnO photoanode-based DSSC.

Keywords: Zinc oxide; Lycopene; DSSCs; Carotenoids; Sensitizer.

Received: 24 May 2021; Accepted: 2 October 2021.

Article type: Research article.

1. Introduction

In 1991, a breakthrough in organic photovoltaics was achieved by Graetzel *et al.*^[1] by implementing mesoporous TiO₂ (Titanium oxide) for dye-sensitized solar cells (DSSCs). In DSSCs, dye absorbs light photons and with the help of wide band gap semi-conducting material converts photons to electricity. A review of the literature indicates that various synthetic, organic, and natural dyes were used by researchers^[2-15] as sensitizers in DSSCs. Zhou *et al.*^[16] studied 20 natural dyes for sensitization of TiO₂ based solar cells. And it was observed that mangosteen pericarp extract showed the best conversion efficiency amongst all dyes used. These are the pigments that can be found in different plant parts such as roots, leaves, fruits, and flowers. The natural dyes are biodegradable, non-toxic, and environmentally suitable as compared with the synthetic metal complex-based dyes. Wang *et al.*^[17] studied natural dyes such as chlorophyll and carotenoids as sensitizers. Natural dyes such as betalains,^[18,19] anthocyanins,^[20,21] and carotenes^[22] have been used as sensitizers. Similar to TiO₂, ZnO can be used in the

construction of photoanode in DSSCs. ZnO have certain advantages over TiO₂ such as; (i) wide direct band gap of 3.37 eV, (ii) excitation binding energy of ZnO (60 meV)^[23-25] is higher than TiO₂ (4 meV) and (iii) electron mobility of ZnO (200 cm³V⁻¹s⁻¹) also higher compared with TiO₂ (30 cm³V⁻¹s⁻¹).^[26-27]

The DSSC consists of a thick film of ZnO nanoparticles that accepts electrons from the excited dye since it provides a large surface area for the adsorption of light-harvesting molecules.^[28] Due to the same reason, ZnO finds potential application in photocatalysis,^[29] sensors,^[30] light-emitting diodes,^[31] solar cells,^[32] etc. Keis *et al.*^[33,34] reported photoconversion efficiency of 5% for porous ZnO sensitized with metal complex-based dye. Law *et al.*^[35] used ZnO nanowires for DSSCs and achieved a conversion efficiency of 0.5%. Yafeng *et al.*^[36] showed that the dye-sensitized ZnO nanowire can be used in DSSCs to improve light to energy conversion efficiency. Win *et al.*^[37] had reported the characterization of nano-sized ZnO electrodes with curcumin-derived natural dye extract for DSSCs application. Thambidurai *et al.*^[34-40] reported flowers like ZnO nanorods for DSSCs. Hosni *et al.*^[41] reported a power conversion efficiency of 4.66% for nanorods and 4.21% for nano-spheres of ZnO sensitized D149 dye. Wong *et al.*^[42] have studied the effect of nanoparticle properties on DSSC's performance. The photocurrent density (Jsc) and open-circuit voltage (Voc) recorded by Shinde *et al.*,

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were $179 \mu\text{Acm}^{-2}$ and 326 mV respectively for DSSCs based on lycopene sensitized zirconium dioxide (ZrO_2) photoanode.^[43] To date, lycopene dye was not reported as a sensitizer with ZnO photoanode in DSSCs.

In the current investigations, a seed layer of ZnO was deposited using a simple, low-cost Doctor Blade technique. Using XRD (X-ray diffraction), UV-Visible absorption spectroscopy, photoluminescence (PL), and Scanning Electron Microscopy (SEM), the prepared ZnO photoanode was characterized. A novel method was used to synthesize lycopene dye. Further, a systematic study on lycopene sensitized ZnO photoanode was carried out.

2. Experimental

2.1 Materials and methods

Materials: Zinc acetate $\text{Zn}(\text{CH}_3\text{COOH})_2$, hydrogen peroxide (H_2O_2), Ethyl cellulose (SDFCL), α -terpineol (HPCL), acetylacetone (SRL), anhydrous Na_2SO_4 , petroleum spirit, etc. **Methods:** mechano-chemical method and Doctor Blade method.

2.2 Preparation of ZnO powder

In preparation for ZnO , ZnO_2 was used as a precursor. The precursor was synthesized by the prescribed method explained elsewhere.^[39] ZnO_2 was obtained from the reaction between zinc acetate and hydrogen peroxide at room temperature by the mechanochemical method. The dried ZnO_2 was thermally decomposed at around 200°C in a controlled manner and then annealed at 450°C for 90 minutes. Crystal parameters were studied by XRD while surface morphology by SEM. The bandgap was estimated from diffused reflectance spectra (DRS) in absorbance mode. Photoluminescence (PL) was studied by recording the PL spectrum.

2.3 Isolation of lycopene from carrot

The paste of a raw carrot was obtained by grinding carrots in a mixer. The resulted paste was soaked in petroleum spirit for 24 hours. The extract was taken out and dehydrated using anhydrous Na_2SO_4 . The dehydrated extract was concentrated by distillation. The fractionation of carotenoids from the extract was performed by column chromatography using basic alumina as an adsorbent. The lycopene fraction was identified by recording the UV-Visible absorption spectra. The lycopene fraction was concentrated by the distillation method. Fig. 1 shows the structure of the synthesized lycopene dye.

Table 1 shows the detailed description of the extracted lycopene dye used for the sensitization of the ZnO photoanode.

2.4 Fabrication of DSSCs

ZnO films were prepared on indium doped tin oxide (ITO) glass by the doctor blade method. These ZnO films were kept in lycopene for dye adsorption time of 24 hours to obtain lycopene sensitized ZnO photoanodes. Poly-iodide ($0.05 \text{ M I}_2 + 0.5 \text{ M KI}$ in water) is used as an electrolyte. Platinum film coated on ITO glass was used as a counter electrode. Sandwiched type of DSSC was assembled where photoanode and counter electrode were separated by a spacer having a hole of $0.5 \text{ cm} \times 0.5 \text{ cm}$ at the center. The cell was filled with electrolytes and characterized by a JV (Current density vs Voltage) curve. Fig. 2 shows the band diagram & electron transport in lycopene sensitized ZnO photoanode-based DSSCs. The position of the conduction band and valance band of the ZnO is -4.15 and -7.35 eV , respectively (concerning the vacuum level). The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energy levels of the lycopene dye were theoretically calculated using Firefly at B3LYP/6-311G (d, p). The HOMO and LUMO levels energies were -4.65 and -2.321 eV , respectively (concerning the vacuum level).

Table 1. Detail description of extracted lycopene dye.

Nomenclature	IUPAC name	Molecular formula	Molecular Weight (g/Mol)
Lycopene	(6E,8E,10E,12E,14E,16E,18E,20E,22E,24E,26E)-2,6,10,14,19,23,27,31-octamethyltriacont-2,6,8,10,12,14,16,18,20,22,24,26,30-tridecaene.	$\text{C}_{40}\text{H}_{56}$	536

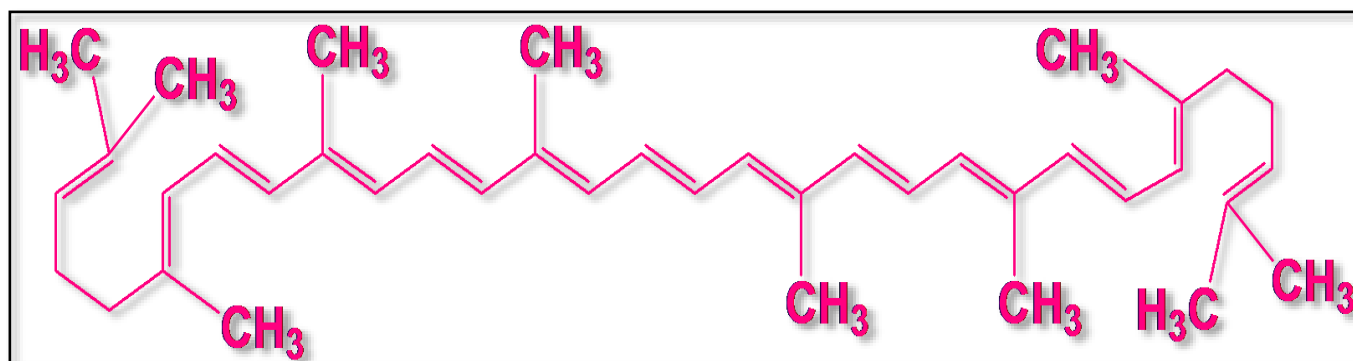


Fig. 1 Structure of the lycopene dye.

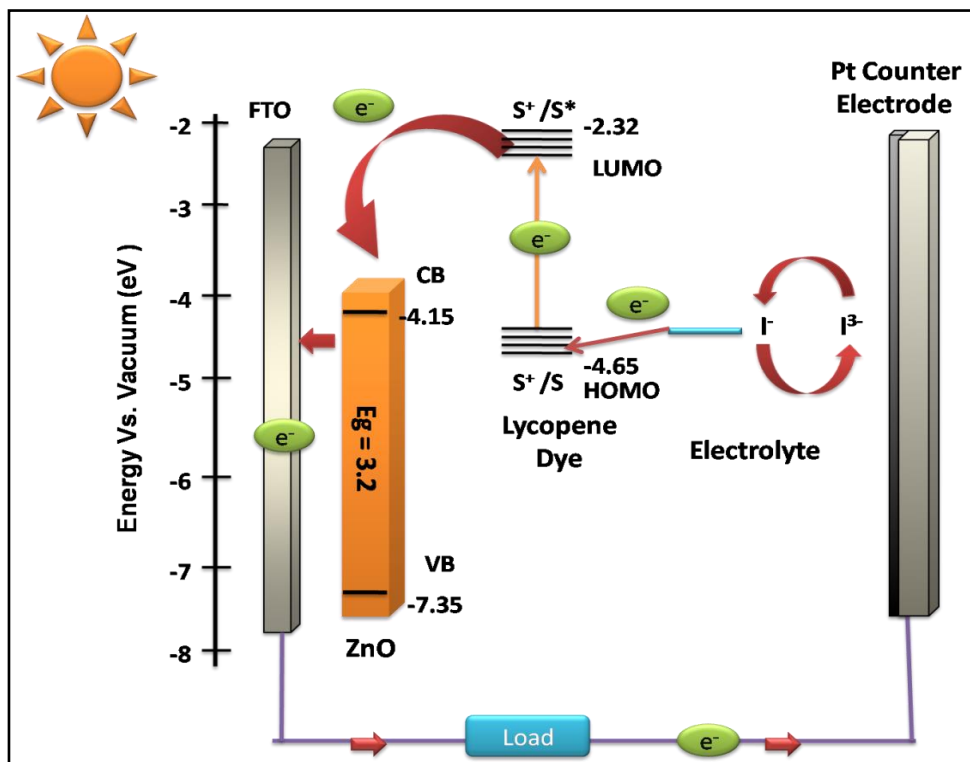


Fig. 2 Band diagram and electron transport in lycopene sensitized ZnO photoanode based DSSC.

2.5 JV Characteristics

The DSSCs (irradiated under 100 mW/cm²) were characterized for different parameters under standard conditions. The overall efficiency of the lycopene sensitized cells was evaluated in terms of open-circuit voltage (V_{oc}), short current (J_{sc}), and fill factor (FF) using the following formulae.

$$\text{Fill Factor (FF)} = \frac{P_{\text{max}}}{J_{\text{sc}} \times V_{\text{oc}}} \tag{1}$$

where P_{max} is maximum power output.

$$\text{Efficiency } (\eta) = \frac{J_{\text{sc}} \times V_{\text{oc}} \times \text{FF}}{P_{\text{in}}} \tag{2}$$

where, J_{sc} – short circuit photocurrent density (Acm⁻²),

V_{oc} – open circuit voltage (volts),

P_{in} – intensity of the incident light (Wcm⁻²),

J_{max} – maximum photocurrent density (A cm⁻²) and

V_{max} – maximum voltage (volts) in the JV curve.

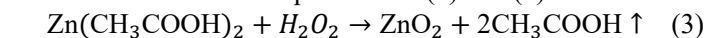
3. Characterizations

ZnO films deposited on ITO glass and lycopene sensitized ZnO photoanodes were characterized by various characterization techniques. XRD (X-ray diffractometer, model Bruker D8 with CuK α radiation of wavelength 1.54Å), UV-Visible Absorption Spectroscopy (JASCO V-670), SEM (Scanning Electron Microscopy, JSM-7600F) and EDS (Energy-dispersive X-ray spectroscopy) to confirm structural, optical, morphological and compositional properties respectively. The DSSCs based on lycopene sensitized ZnO photoanode was investigated using a Keithley 2400 source meter and solar simulator (ENLITECH Model SS-F5-3A) under 100 mW/cm² incident light intensity.

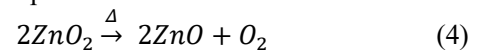
4. Results and discussion

4.1 Reaction mechanism

The ZnO powder was formed by following chemical reaction^[44] as shown in equation No. (3) and (4).



Thermodynamically ZnO₂ is more stable than zinc acetate hence O₂²⁻ of H₂O₂ displaces acetate from zinc acetate.



ZnO₂ is thermally unstable at a higher temperature and hence on heating at 200 °C it decomposes to ZnO with the evolution of O₂ gas.

4.2 Structural properties

Fig. 3 shows the XRD pattern of the ZnO powder. The XRD pattern indicates that highly crystalline ZnO having a wurtzite crystal structure (JCPDS card No. 36-1451).

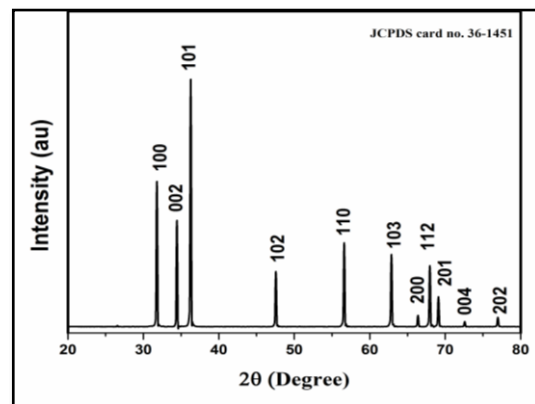


Fig. 3 XRD spectrum of ZnO powder.

A similar type of XRD pattern was observed by different researcher, The lattice dimensions were calculated from the observed XRD pattern which is found with a wurtzite structure ($a = 3.2478\text{\AA}$, $c = 5.1992\text{\AA}$, Crystal density = 5.69 g cm^{-3} , Zn-O bond length = 1.977\AA). The average crystallite size was estimated by Debye-Scherrer's formula and was found to be $\sim 24\text{ nm}$.

4.3 Morphological properties

Surface morphology was studied by SEM images as shown in Fig. 4. The SEM image of ZnO powder shows the nanoparticles with the porous agglomerated crystallites having grains of a somewhat spherical shape. The average grain size of ZnO was obtained from the SEM image and observed to be 70 nm . Fig. 5, shows the SEM image of (top view) of ZnO photoanode having porous and rough structure useful to dye adsorption.

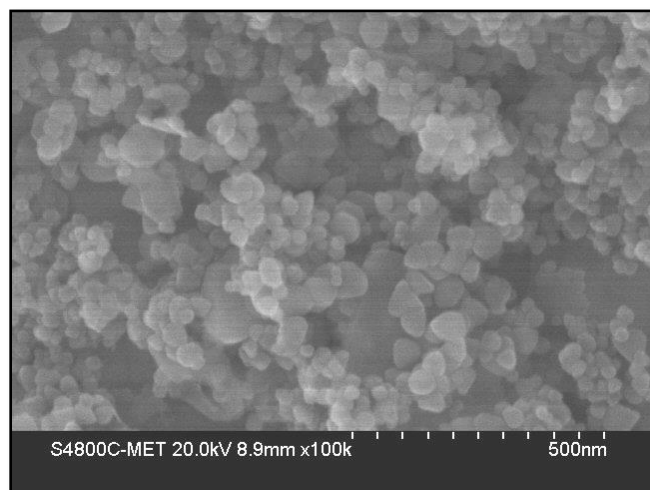


Fig. 4 SEM of ZnO powder.

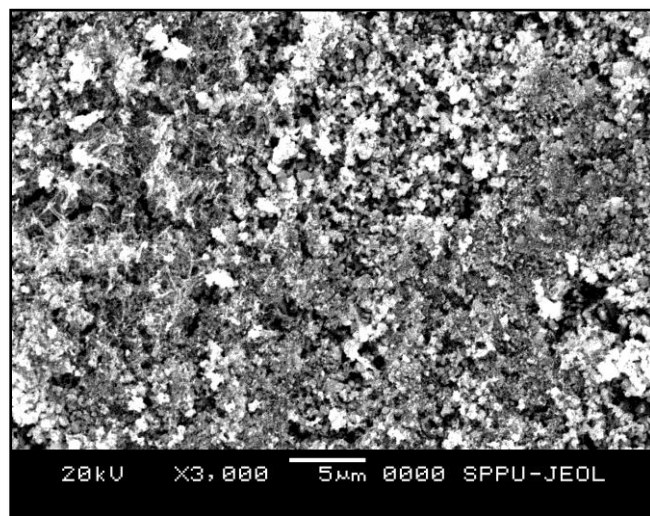


Fig. 5 SEM image of the ZnO photoanode (Top view).

4.4 Optical properties

Fig. 6 shows the UV-Visible absorption spectra of the ZnO photoanode. The band gap was calculated from DRS spectra

in the absorbance mode. Fig. 7, shows the graph of $(\text{absorbance})^2$ against energy (eV) of ZnO powder. The observed band gap is 3.25 eV .

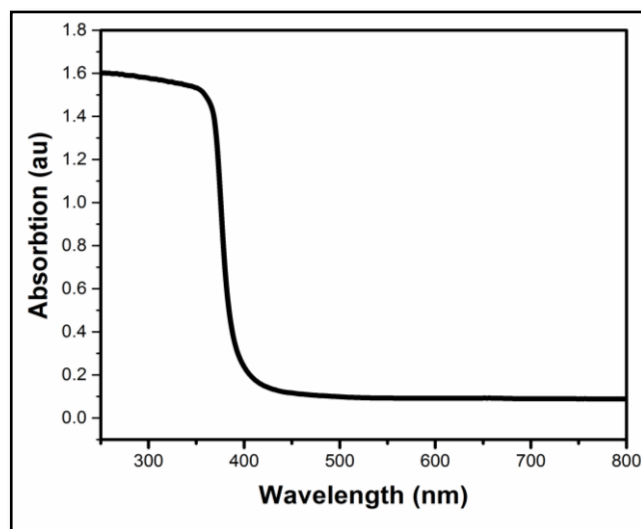


Fig. 6 Absorbance spectra of the ZnO photoanode.

Fig. 8, shows the photoluminescence (PL) spectra of the ZnO. It indicates the presence of sharp emission peaks at 384, 395, 451, 468, 567, 628, and 668 nm while the broad peak at 426 nm. The peaks at 384 and 395 nm are related to the band-to-band transitions (valence band to conduction band). These emission bands at these wavelengths arise in PL spectra due to electron-hole recombination and it is a measure of the band gap of semiconductor material. Vanheusden *et al.*^[50] attributed the origin of such emissions in the ZnO to the transitions of electrons between photo-excited holes and singly ionized oxygen vacancies. The peaks at 426, 451, and 468 nm arise mainly due to defects in the ZnO crystal. They are termed as deep level emissions and they originate from point defects such as oxygen vacancies and interstitial Zn^{2+} sites.^[51,52]

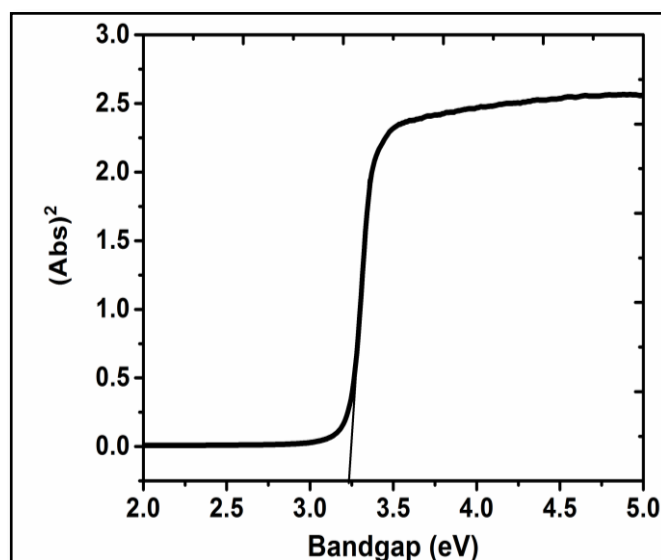


Fig. 7 Plot of $(\text{Absorbance})^2$ Vs energy for ZnO.

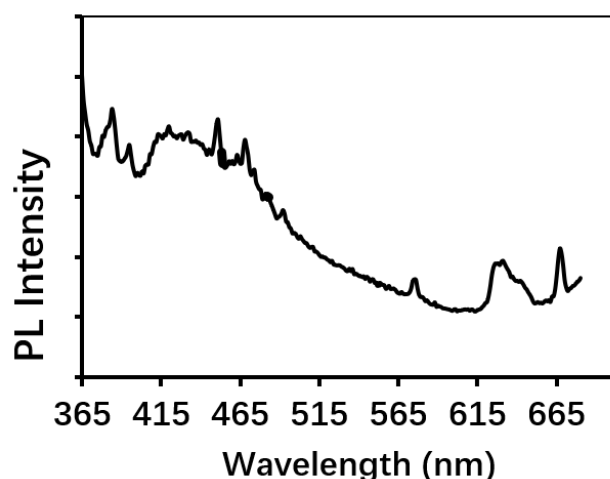


Fig. 8 Photoluminescence (PL) spectra of the ZnO.

The lycopene is orange-red colored highly and conjugated hydrocarbon. The absorbance spectra of the lycopene (shown in Fig. 9), which show peaks at 446, 472, and 505 nm. The observed spectrum is a characteristic spectrum of lycopene.^[53]

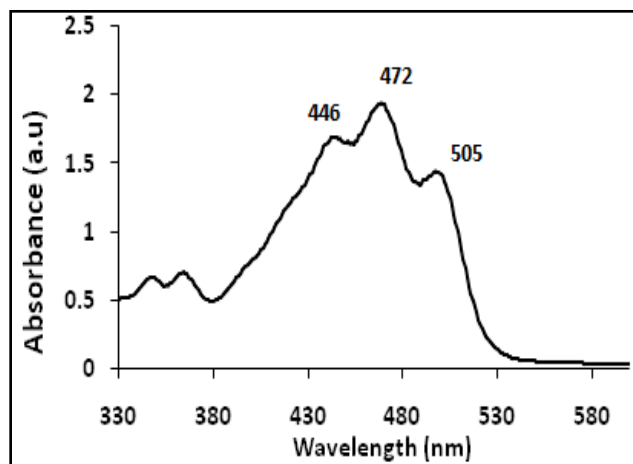


Fig. 9 Absorption spectra of lycopene dye.

4.5 JV characteristics of the cell

Fig. 10, shows the JV curve for lycopene sensitized ZnO photoanode based DSSCs.

The power conversion efficiency of ~0.39% with V_{oc} ~281 mV, J_{sc} ~176 $\mu A/cm^2$, and FF ~47%. The photovoltaic parameters of lycopene sensitized ZnO photoanode-based DSSCs are summarized in Table 2.

Table 2. Photovoltaic parameters of lycopene sensitized ZnO photoanode based DSSCs.

Photovoltaic parameter	
V_{oc} (V)	0.279
J_{sc} (mA/cm^2)	0.176
FF (%)	47
Efficiency (%)	0.39

The conversion efficiency reported for ZnO-based natural DSSCs was 0.0277% with anthocyanin dye extracted from

strawberry,^[54] Hatem *et al.*^[45] reported not more than 0.1% conversion efficiency for seven different natural dyes. The ZnO sensitized by natural dye lawsone showed a conversion efficiency of 0.68% as reported by Khadare *et al.*^[55]

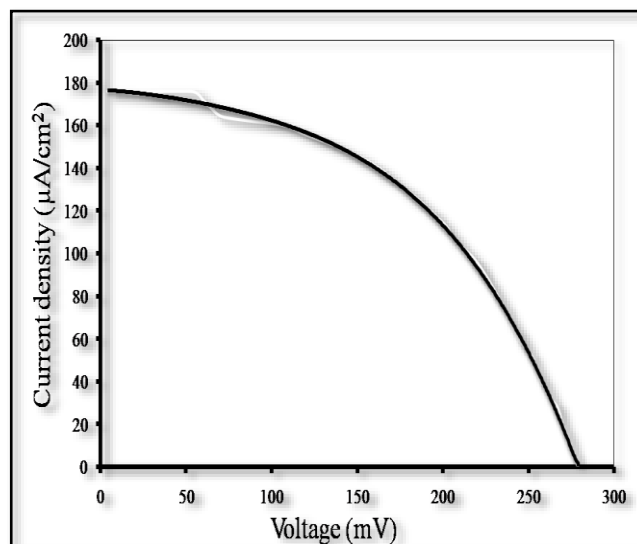


Fig. 10 JV curve of the lycopene sensitized ZnO photoanode-based DSSCs.

The performance of ZnO-based DSSCs for four different natural dyes such as sappan wood, noni leaves saw flowers, and black rice was studied by Magaraphan *et al.*^[56] and they observed conversion efficiency of less than 0.06%. Wang *et al.*^[57] reported a charge-separation process at the donor/acceptor interface for bulk hetero-junction organic solar cells for carotenoids such as fucoxanthin, β -carotene, and lycopene and it was observed that the hole mobility of lycopene was $2.1 \times 10^{-2} \text{ cm}^2/(\text{V s})$, which was three times greater than those of fucoxanthin and β -carotene. The lycopene shows better performance for photovoltaic organic solar cells because of its superior charge-carrier transport. The carrot-derived natural dye for DSSCs was reported by Win *et al.*^[58] for the $\text{TiO}_2\text{-ZrO}_2$ photoanode with photo-conversion efficiency of 0.03%. Wang *et al.*^[17] reported conversion efficiency increases with the increase of that the conjugated double bonds in carotenoids in DSSCs. Yamazaki *et al.*^[22] used natural carotenoids such as crocetin and crocin as a photo-sensitizer and the best performance was observed for which containing carboxylic as a functional group. Senthil *et al.*^[59] studied ZnO nanorod-based solar cell sensitized with natural dyes extracted from the beetroot, strawberry, and rose, it gives better performance power conversion efficiency for strawberry with photoconversion efficiency of 0.22%.

The present study on lycopene as a sensitizer showed low efficiency but it is higher than most of the natural dyes. The lower efficiencies are due to the absence of functional groups such as carboxylic and hydroxyl groups in the lycopene. Due to these reasons, the lycopene does not strongly bind to ZnO and shows lower conversion efficiency. Another point to note is that the electrolyte in this study is Poly-iodide (0.05 M I_2 +

0.5 M KI in water). Water is employed as a solvent in this process. For DSSC, this is not a favorable alternative in terms of charges. This could be a factor in the low Jsc. Efficiency suffers as a result. One of the most effective techniques to address this issue is to use polymer-based electrolytes such as (PEG-based Gel Polymer Electrolytes).^[60]

5. Conclusions

In the present work, we have successfully synthesized ZnO nanopowder by the mechanochemical method. The ZnO thin films were deposited by the Doctor Blade method. The X-ray diffraction studies confirm the wurtzite structure of ZnO. The adsorption of natural lycopene dye of the ZnO films was to a very less extent (from UV-Visible absorption). The photovoltaic parameters such as Voc, Jsc, FF, and efficiency were calculated to be 281 mV, 176 μAcm^{-2} , 47%, and 0.39% respectively. These DSSCs show better performance for dye loading time of 24 hours. The lower conversion efficiency of these cells may be due to the weak bonding of the lycopene to the ZnO because of the absence of the functional groups in the dye.

Acknowledgment

KMG is greatly thankful to Prof. Ramakrishna More Arts, Commerce and Science College, Akurdi, Pune, India, and Advanced Physics Laboratory, Department of Physics, Savitribai Phule Pune University, Pune, India for experimental and characterization facilities and constant motivation.

Conflict of interest

There are no conflicts to declare.

Supporting information

Not applicable.

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