



Study on dye-sensitized solar cell efficiency improvement using methyl orange dye

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Abstract

In this work, different varieties of dye sensitized solar cells are fabricated by simple fabrication process. In this fabrication extract of butea monosperma flower, methylene blue and methyl orange dyes are used as sensitizers. The photovoltaic performance of dye sensitized solar cells (DSSCs) has been studied. The performances of two different types of photo-electrodes are also tested in this work. The morphology and bandgap of TiO₂ (titanium dioxide) and ZnO (Zinc oxide) was observed from XRD, FTIR spectroscopy and UV-vis Spectrum. It is found that TiO₂ based DSSCs have better performance. It also observed that the current density and efficiency was increased from 7.46 to 12.9 mA/cm² and from 1.34 to 6.8% respectively when using methyl orange as a dye. Hence it can be said that methyl orange dye enhanced the photovoltaic performance of DSSC.

Keywords DSSCs · TiO₂ · ZnO · Methyl orange · Methylene blue · Dye

Introduction

Use of renewable energy sources is the most effective way to fulfill the ever-increasing demand for electrical energy. Among all renewable energy solar energy is considered as a suitable source of energy because energy from sunlight in one hour is more than all the energy consumed in an entire year [1].

Dye sensitized solar cells are the low cost, simple fabricated solar cell was first reported by Gratzel [2]. DSSC is multi-layer sandwich like structure having photo electrode, dye, electrolyte and counter electrode [3]. Basically, DSSCs follow the natural process photosynthesis in which the plants convert light from sun into energy by sensitizing a nanocrystalline semiconductor layer using natural or synthetic dyes. DSSC offers many advantages as compare to Si based solar cell like DSSCs are low cost and moderate efficiency solar cell. DSSC work at low intensities and wide angle of incident light beam, great mechanical robustness. It have transparent electrodes [4]. It also found in various colour, so it is used in indoor application also. The dye molecules absorb the sunlight which plays vital role through its capability to absorb the visible photons and an electrolyte is used for dye regeneration. The efficiency of DSSC is based on different dyes and different metal oxides used as the one of the electrode like TiO₂, ZnO, WO₃, FeO₃, SnO₂ etc [5, 6]. Therefore, research focused on different types of dyes and enhance the solar cell conversion

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efficiency [7, 8]. Initially, ruthenium-based complexes were used as dye because of their good photo electrochemical properties [9, 10]. There are many others organic dyes reported in literature. From these dyes, some synthetic dyes like Aniline blue and N719 are very low, and this is because the material that was used as ETL was ZnO nanorods. Besides these dyes there are many natural dyes like red turnip, pomegranate, Crocetin, Bonganvillea, Cherries, Raspberries, Sumac/Rhusfruits, Curcuma longa roots etc. having conversion efficiencies of 1.7%, 1.5%, 0.56%, 0.38%, 0.18%, 0.309%, 1.5%, 0.36% respectively [11–15]. Nur Najiha et al. [16] uses three natural dyes i.e., Avacoda peel, Blue berry and Pomegranate in fabrication of DSSC and found their efficiency as $48.25 \times 10^{-12}\%$, $24.62 \times 10^{-12}\%$, and $15.75 \times 10^{-12}\%$ respectively. Similarly, D. Sinha et al. used natural dyes (i.e., Purple cabbage, Beetroot and their mixture) [17] and found the efficiency of 0.1015%, 0.17% and 0.3827% respectively. H. Setyawadi uses the methyl orange as a dye and found the photovoltaic efficiency of 0.24% [2] as given in Table 1. In this present work three different types of dyes i.e., methyl orange, methylene blue and extract of butea-monosperma flower are reported.

From above literature, it can be seen that the maximum efficiency of DSSC using methyl orange [2] and methylene blue [22] dye were 1.137% and 2.20% respectively. In the present work, fabrication of DSSC using methyl orange and methylene blue dye have been done and the calculated efficiency is 6.8% and 3.35%, which shows an enhancement as 83.27% and 34.32% respectively.

Working principle of DSSC

A typical dye sensitized solar cell consists of anode and cathode made up of fluorine doped tin oxide glass (FTO) which

Table 1 Review of DSSCs using different natural and synthetic dyes

Dyes	Type of Dyes	Efficiency (%)	References
Eosin Y	Synthetic	0.399	[18]
Crystal violet	Synthetic	0.249	[18]
Methyl orange	Synthetic	0.115	[18]
Ru complex	Synthetic	2.58	[18]
Harda fruit	Natural	3.52	[19]
Fabric dye	Synthetic	0.80	[19]
Methyl orange	Synthetic	0.24	[2]
Monodye-N719	Synthetic	1.68	[20]
Betanin	Natural	1.69	[20]
Crocin acacetin	Natural	1.85	[20]
Indigo	Natural	1.88	[20]
Papaya leaf	Natural	0.17	[21]
Alizarian red	Synthetic	0.62	[22]
Methyl orange	Synthetic	1.137	[2]
Methyl orange	Synthetic	0.756	[23]
Methyl orange	Synthetic	1.57	[22]
Methylene Blue	Synthetic	2.20	[22]
Methylene blue	Synthetic	0.85	[22]

allows light pass through the cell, semiconductor oxide layer (TiO_2/ZnO), dye sensitizer, first type of dye was Polypyridyle compounds of Ru (II) F. Some researchers reported the “black dye” and different ruthenium complexes. Besides these synthetic dye different researchers work with natural dye like shiso leaf pigments, Calafate fruit, Henna, Bamboo leaves, California blackberries, Strawberries, Pomegranate, Raspberries, and Cherries etc. In this present work both natural and synthetic dye and electrolyte (Iodide, tri-iodide) are used. DSSC works on the principle that, when sun light strikes on the photoanode the photons of wavelength corresponding to the energy difference between highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of the dye, are absorbed by the dye sensitizer (natural or chemical). The electrons are then moved to the conduction band of TiO_2 and move to the photo-anode. The electrons from the photoanode flow through the outer circuit via load and re-enter through the cathode. The tri-iodide electrolyte receives an electron from cathode and is reduces to iodide. The oxidized dye receives one electron from iodide electrolyte and oxidizes the iodide into tri-iodide. The oxidized dye reduces to normal state and process continues [24, 25].

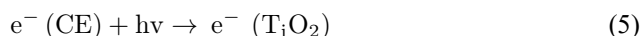
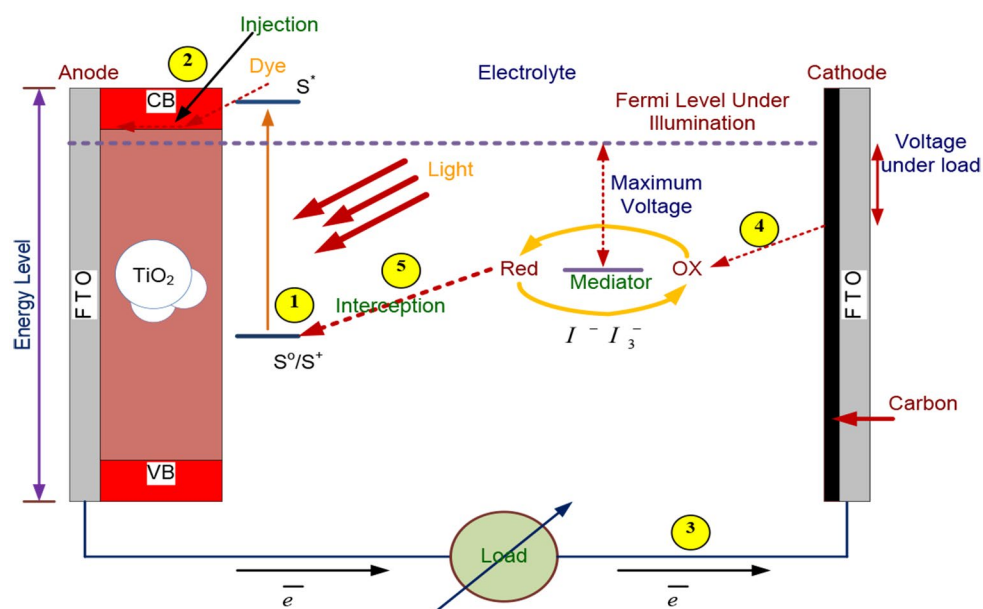
The TiO_2 nanoparticles have high surface areas which are required for dye absorption. The typical DSSC components include two electrodes i.e. the working electrode coated with TiO_2 , dye molecules get attached to the surface of TiO_2 , an iodide electrolyte i.e. I^-/I_3^- was used and the counter electrode (cathode) is generally made of platinum or carbon coated FTO glass. The working of DSSC is shown in Fig. 1. The incoming photon is absorbed by the dye molecule on the surface of the TiO_2 particle and an electron moves from ground state S^0 to excited state S^* shown in Eq. (1). The excited electron is injected to the conduction band of the TiO_2 particle leaving the dye molecule to an oxidized state S^+ shown in Eq. (2). The injected electron moves through the porous TiO_2 to the FTO glass and finally through an external load to the counter-electrode (cathode) shown in Eq. (3). At the cathode the electron is transferred to tri-iodide in the electrolyte shown in Eq. (4) and the cycle completes by reduction oxidation reaction as shown in Eq. (5).

Anode:



Cathode:



Fig. 1 Working principle of the dye-sensitized solar cell

Due to the energy level positioning in the system the cell is capable of producing voltage between its electrodes and across the external load. The maximum theoretical value for the photo voltage at open circuit condition is determined by the potential difference between the conduction band edge of the TiO_2 and the redox potential of I^-/I_3^- pair in the electrolyte. The operation of the cell is regenerative in nature, since chemical substances are neither consumed nor produced during the working cycle.

Materials and methods

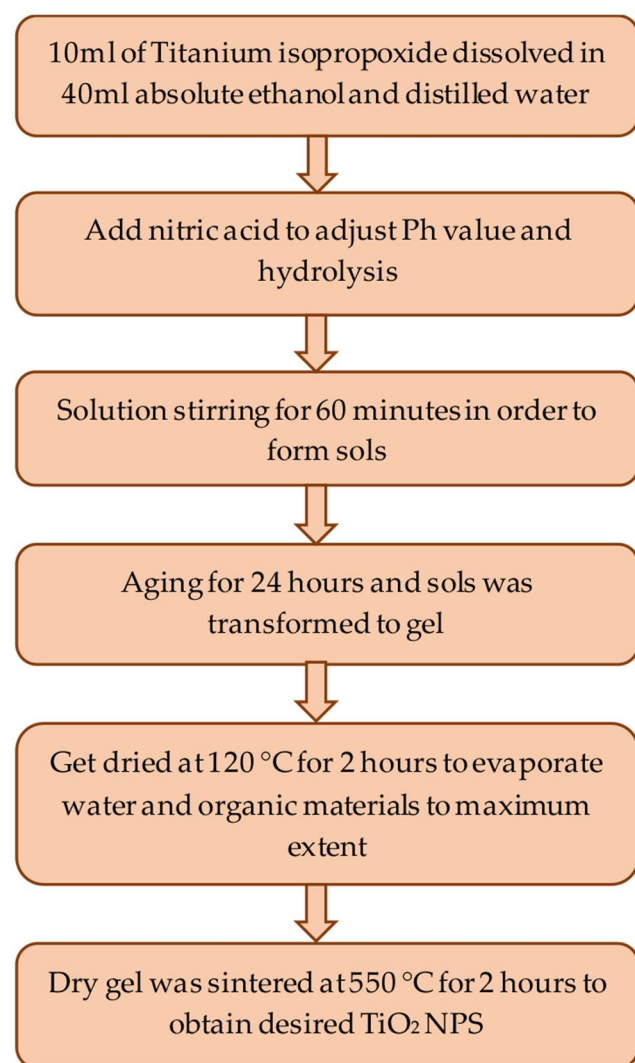
Material used

Titanium isopropoxide, ethanol, nitric acid, Zinc acetate di hydrate, NH_4OH were procured from center of nanotechnology, AMU, Aligarh, India for preparation of TiO_2 and ZnO . All the apparatus (like beaker, furnace, magnetic stirrer, sonicator, furnace) used in the fabrication of DSSC was procured from Centre of Nanotechnology, AMU, Aligarh, India.

Synthesis of TiO_2

Firstly, TiO_2 nanoparticles is synthesized for coating the conducting side of glass electrode of the cell. Take Titanium (IV) isopropoxide (TTIP)(10 ml) and dissolve in ethanol (50 ml) and distilled water is also add in this solution in terms of molar ratio of $\text{Ti}:\text{H}_2\text{O}=1:4$. The steps involving the synthesis of titanium dioxide is shown in Fig. 2.

Add nitric acid to the solution coming from first step and the stirred the resultant solution for 60 min. After this sols

**Fig. 2** Flowchart showing the steps for the synthesis of TiO_2 NPs

transformed into gel and then this gel is converted into dry TiO_2 by evaporating water at 120 degrees. Dry gel is finally sintered at 550 degrees for 2 h and get TiO_2 nano particles.

Synthesis of ZnO

ZnO nanoparticles were synthesized for coating on the conducting side of FTO glass for fabrication of another DSSC. Take 2 ml of ethanol and dissolved in the solution of water and tea and the whole procedure of synthesized ZnO is shown in Fig. 3.

Fabrication process of the DSSCs

Firstly, add glycol in these nano particles and sonicate for 15 min to homogenize TiO_2 and ZnO nanoparticles. By using doctor blade method, nanoparticles are deposited on conducting side of FTO glass with the help of glass rod with the active area of 1 cm^2 . After this put this electrode in the furnace at 250° C and then cool at room temperature and photoanodes were immersed in all dye's solution in dish for 48 h. After two days dyes were absorbed by material and rinsed with distilled water air dried and then put one drop of iodide electrolyte on the dye electrode. Placing a conducting electrode with graphite coating on top of another electrode that had already been manufactured and fastened with binder clips allowed for the assembly of the sandwich type DSSC. The experimental steps involving in fabrication of DSSCs of three different dyes (extract of Buteamonosperma flower, methylene blue and Methyl orange) are shown in Fig. 4.

The conversion efficiency of different DSSCs can be calculated by using Eqs. (6)–(7) as,

$$\eta = \frac{V_{oc} (V) \times J_{sc} (mA/cm^2) \times ff}{\text{light intensity (mW/cm}^2\text{)}} \quad (6)$$

Where,

V_{oc} = Open circuit voltage.

J_{sc} = short circuit current density.

$$\text{Fillfactor} = \frac{V_m \cdot J_m}{V_{oc} \cdot J_{sc}} \times 100 \quad (7)$$

Results and discussion

Characterization of TiO_2 and ZnO

Before using nanomaterials for DSSC fabrication, it is essential to understand their reaction mechanisms and verify their proper synthesis through various characterization techniques. The synthesized TiO_2 were examined by XRD

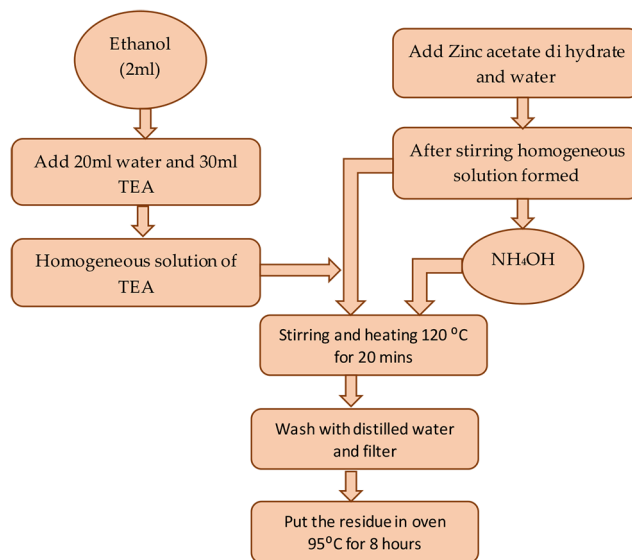


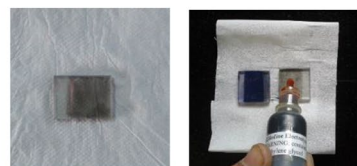
Fig. 3 Flowchart showing the steps for the synthesis of ZnO nanoparticles



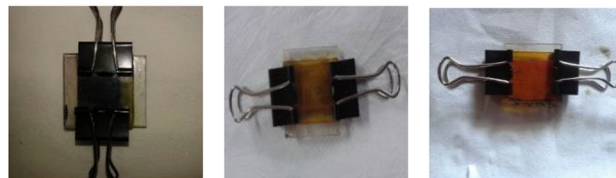
(a) Pasting of TiO_2/ZnO on FTO glass



(b) dipped the coated electrodes in dyes
[(i)methylene blue, (ii)extract of butea mono sperma flower, (iii)methyl orange]



(c) Graphite coated on Cathode (d) put a drop of Iodide electrolyte



(e) Fabricated DSSCs of different dyes

Fig. 4 Steps involving in the fabrication of different DSSCs

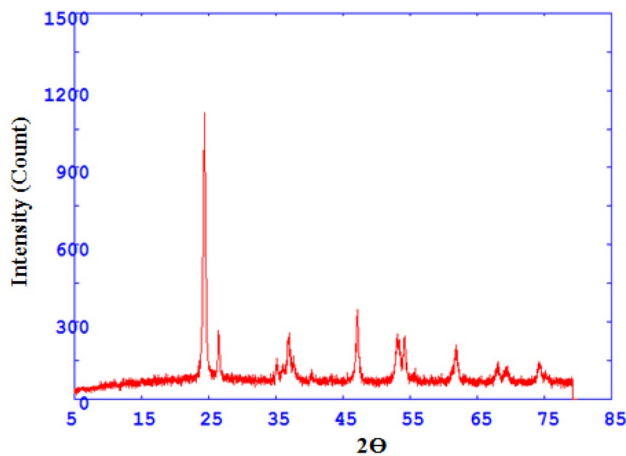


Fig. 5 XRD pattern of TiO₂ NPs

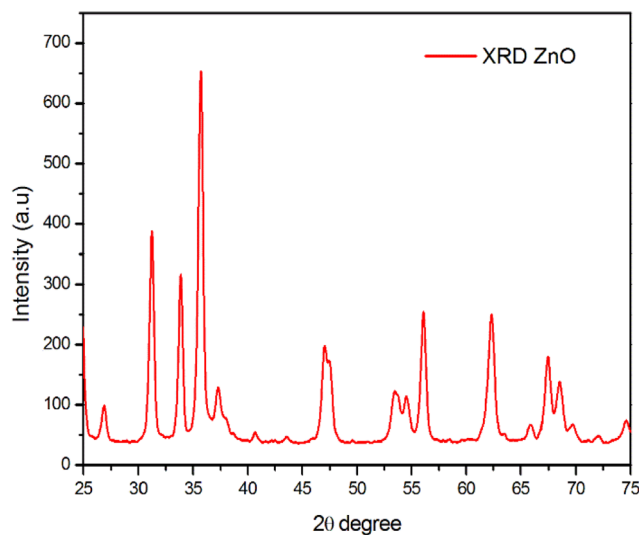


Fig. 6 XRD pattern for ZnO

Cu radiation at 2θ angle ranging from 0° to 80° as shown in Fig. 5.

The XRD characterization was performed by Rigaku Miniflex II, XRD in 2θ range of 20° – 80° with CuK α radiations $\lambda = 1.5418 \text{ \AA}$, 15 mA, 30KV. The data was analysed by Powder X software package, the XRD peaks were analysed and observed according to (JCPDS 21-1272), from full width half maximum, the crystallite size was calculated by Scherer formula, the average crystallite size (D) is determined using Eq. (8) as,

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (8)$$

Where 'K' - the shape factor (0.9), ' λ ' - wavelength and ' β ' - full width at half maxima, ' θ ' - diffraction angle. The average crystallites size of TiO₂ calculated by XRD peak at 25.3° was 9.6 nm.

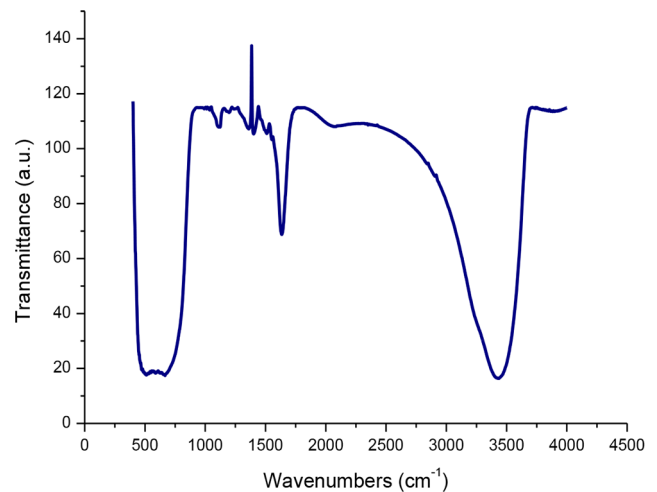


Fig. 7 FTIR pattern of TiO₂ NPs

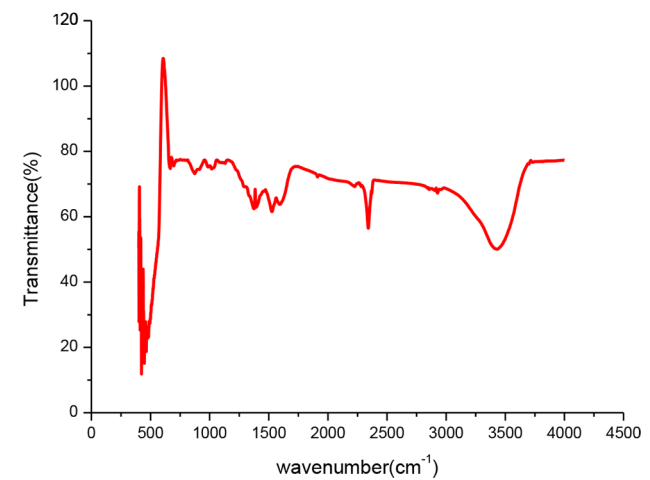


Fig. 8 FTIR pattern of ZnO NPs

The largest peak of TiO₂ NPs at $2\theta = 25^\circ$, which is same as the peak of XRD pattern of TiO₂ present by Safia A. Kazmi and Ahd Zyoud et al. [26, 27].

Figure 6 shows XRD of ZnO nanoparticles with wavelength $\lambda = 1.54056 \text{ \AA}$ Cu α . The interplanar distance ' d ' is calculated by using formula:

$$2d \sin\theta = n\lambda \quad (9)$$

The 2θ peaks at 31.2 , 33.8 , 35.48 , 37.36 , 46.9 , 55.9 , 62.47 and 67.7° confirm its anatase structure. Broad diffraction peaks suggest extremely tiny size of the crystallite, and the sample's intense XRD peaks demonstrate that the produced nanoparticles were crystalline. The crystallite size is calculated as 15.4 nm.

Figure 7 and 8 shows the FTIR patterns of TiO₂ and ZnO. From Fig. 7, it is shown that Ti-O-Ti and Ti-O is present at 400 – 700 cm^{-1} which is similar as in [28]. In Fig. 8 the peak

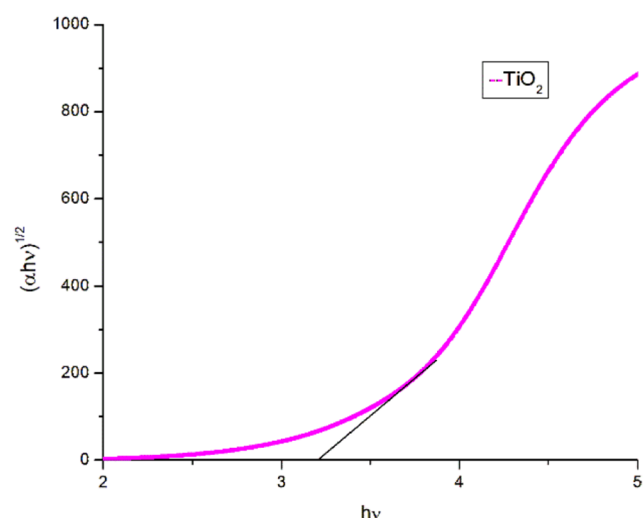


Fig. 9 Bandgap of TiO₂ NPs

at 500 cm⁻¹ which shows the Zn-O stretching band. The peaks are near 1600 cm⁻¹ which is due to OH bending of water. It exhibits a band at 590 cm⁻¹. This observed vibration band may be assigned as M-O-M (metal-oxygen-metal) stretching vibration. The bandgap of TiO₂ and ZnO NPs are 3.2 eV and 3.35 eV are shown in Figs. 9 and 10 respectively.

Photovoltaic characteristics of DSSC using different dyes

Photovoltaic performance of different material based DSSCs using three types of dyes are shown in Figs. 12 and

Fig. 11 Experimental setup for measuring the performance of DSSCs

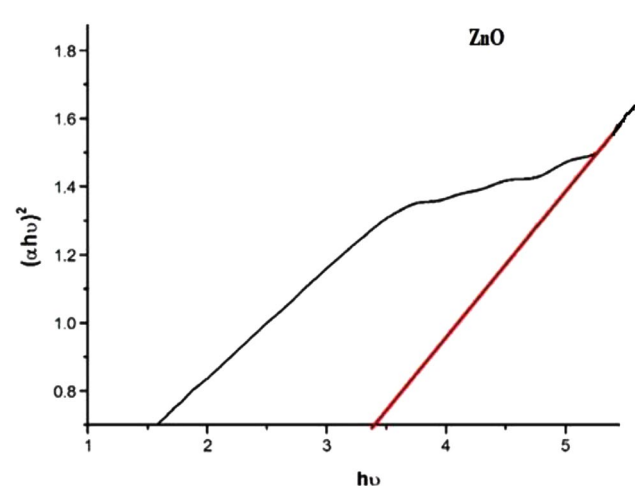
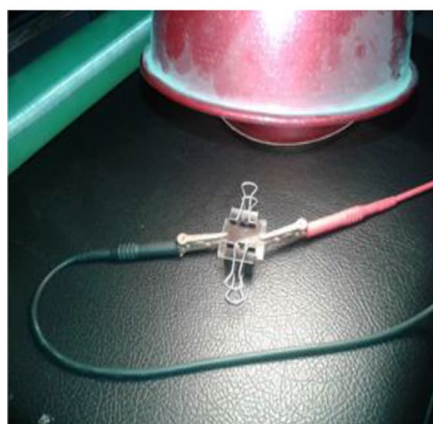


Fig. 10 Bandgap of ZnO NPs

13. The experimental set up for measuring the performance of DSSCs is shown in Fig. 11. The photovoltaic performance of all fabricated DSSC were measured under AM 1.5 (100mW/cm²) and were summarize in Table 2.

Figure 12 shows the photovoltaic performance TiO₂ based DSSC with different dyes. It is shown from the characteristics that the efficiency of TiO₂ based DSSC using extract of buteamonosperma flower, methylene blue and methyl orange are 1.34%, 3.35%, and 6.8% respectively. From this it can be said that the DSSC using methyl orange as a dye has better performance having current density of 12.9 mA/cm², voltage 0.82 V and efficiency of 6.8%.

Table 2 Comparison between PV performances of fabricated DSSCs

Materials	Dyes	V _{oc} (V)	J _{sc} (mA/cm ²)	Fill factor	Efficiency,η(%)
TiO ₂	Methyl orange	0.82	12.9	0.64	6.8
	Methylene blue	0.62	8.4	0.62	3.35
	Extract of butea mono sperma flower	0.33	7.46	0.54	1.34
ZnO	Methyl orange	0.70	12.2	0.49	5.5
	Methylene blue	0.62	7.3	0.54	2.77
	Extract of butea mono sperma flower	0.31	6.2	0.59	0.87

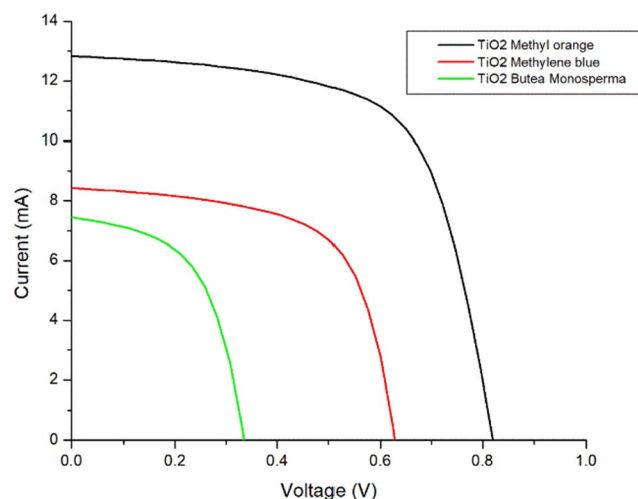


Fig. 12 I-V characteristics of DSSCs using TiO_2 NPs

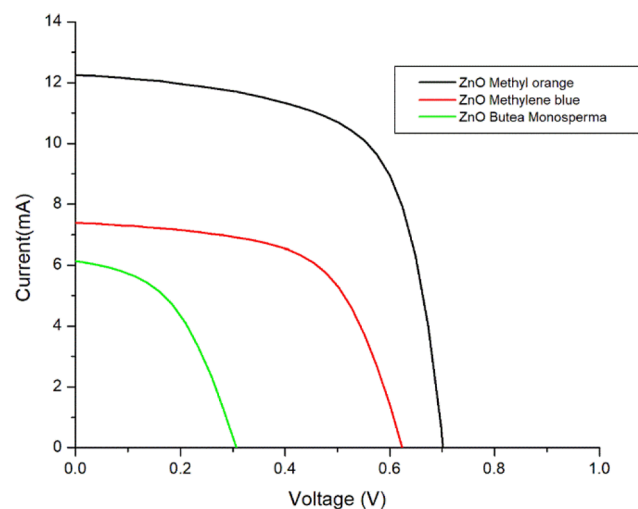


Fig. 13 I-V characteristics of DSSCs using ZnO NPs

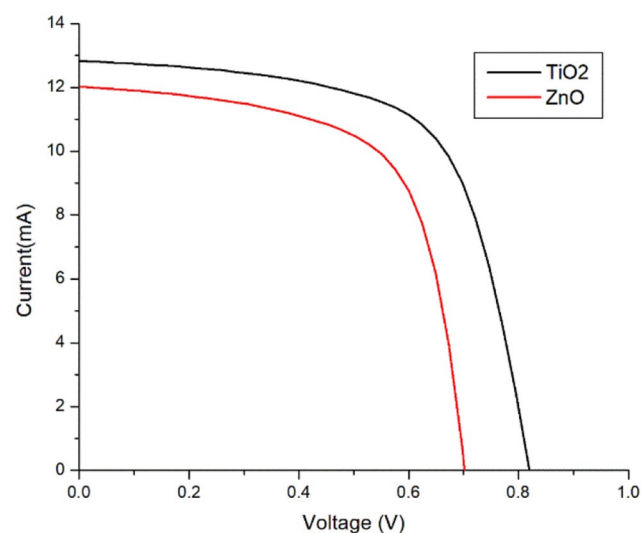


Fig. 14 Comparison of DSSCs using different NPs

Similarly, from Fig. 13, ZnO based DSSC have an efficiency of 0.87%, 2.77%, and 5.5% when extract of butea mono sperma flower, methylene blue and methyl orange respectively used as dye. It is also clear from these results that methyl orange gives the better performance having current density of 12.2 mA/cm^2 , voltage is 0.70 V and efficiency is 5.5%.

The comparison of different nanomaterial based DSSC are also done in this present work, which is shown in Fig. 14; Table 2. From these, it can be observed that the TiO_2 based DSSCs have better performance as compared to ZnO based DSSCs. Figure 14 also shows the performance of DSSC using three different dyes. It is seen from the graphs that the methyl orange has better performance than the other dyes.

Conclusions

In the present work, DSSC using different nanomaterials as well as using different dyes have been fabricated and their performance was investigated. Initially both ZnO and TiO_2 are used as anode material and different natural dyes are used as sensitizers the overall efficiency was very low, so to increase the efficiency various experiments were conducted.

- When ZnO is used as anode material and methyl orange as dye the DSSC achieves current density of 12.2 mA/cm^2 , open circuit voltage is 0.70 V and efficiency is 5.5%.
- If the anode material is replaced by TiO_2 , for the same methyl orange dye an enhancement in values of cell parameters is observed. The V_{oc} increases to 0.82 V, I_{sc} to 12.9 mA and efficiency of DSSC to 6.8%.
- When calculated efficiency of DSSC using methyl orange dye and methylene blue dye in the present work are compared with the maximum efficiency of DSSC using methyl orange and methylene blue dye reported in literature [2, 22]. The calculated efficiency is 6.8% and 3.35%, which shows an enhancement in % efficiency as 83.27% and 34.32% respectively.

So, it can be concluded that the methyl orange dye with TiO_2 photo-electrode enhanced the photovoltaic performance of dye sensitized solar cells.

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