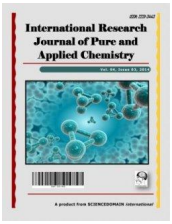

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New Metal Free Organic Dyes Based DSSC with Graphite-PSS: PEDOT Counter Electrode

Sarita Bose[1] and K. R. Genwa[1*]

[1]Department of Chemistry, J. N. V. University, Jodhpur, India.

Authors' contributions

This work was carried out in collaboration between both authors. Author SB preformed the statistical analysis, managed the literature search and wrote the first draft of the manuscript with assistance and supervision from author KRG. Author KRG read and approved the final manuscript.

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ABSTRACT

Dye sensitized solar cells (DSSCs) have emerged as an efficient third generation solar cells. This is a photo-electrochemical cell where the junction consists of a semiconductor–electrolyte interface. The DSSC was fabricated using TiO_2 nanoparticles, wide band gap semiconductor, as anode. Graphite-PSS: PEDOT as effective counter electrode due to high work function which enhances cells performance and iodide-triiodide as redox electrolyte. Though conventional Si-Solar cells have still dominated the market, DSSCs provide a cheap and efficient alternate for solar energy conversion. Till date metallic dyes based DSSCs have showed highest efficiency. But higher cost, relative low abundance of most metals like Ruthenium and environmental hazards limits their extensive use in the research. DSSC needs to be less cheap and more efficient than Si-solar cells to replace later commercially. Organic dyes offer a low cost alternate at the cost of efficiency for DSSC. In this paper new organic dyes Phloxin B, Amido Black and Indigo Carmine were used as photosensitizers. The photoelectrochemical properties were observed for these dyes in different solvents at 1.5 AM condition.

**Corresponding author: E-mail: krg2004@rediffmail.com;*

Keywords: Dye sensitized solar cell; nanoparticles; organic photosensitizers; PEDOT-PSS.

1. INTRODUCTION

Dye sensitized solar cells (DSSCs), Grätzel cells [1], have attracted great attention over the last 15 years owing to their prospectus of high energy conversion efficiency and low production cost. They can be engineered to be a flexible platform, providing a potential rugged energy harvesting platform. A typical DSSC is constructed with dye-adsorbed wide band gap oxide semiconductor (such as TiO_2), electrolyte generally I^-/I^{-3} redox couple and a counter electrode (graphite/ PSS-PEDOT). In these solar cells, dye-sensitized metal oxide electrodes play an important role for achieving high performance since the porous metal oxide films provide large specific surface area for dye loading and the dye molecule possesses broad absorption covering the visible region or even part of the near-infrared (NIR) [2]. As a vital component, the sensitizer has the function of absorbing light and injecting electron into the conduction band of semiconductor. Thus the modification on dye sensitizer is important to gain a promising photo-to-current conversion efficiency (η). Overall peak power conversion efficiency for DSSCs has reached upto 11% [3,4] and still higher efficiencies have been achieved recently worldwide. Current record for prototypes lies at 15% [5]. As far as high efficiency is considered ruthenium based dyes are superior to any other photosensitizers till date. Metallic dyes due to higher stablility, good redox properties and broad absorption spectrum are choice photosensitizers for DSSCs. Inspite of high performance of Ru-based dyes, higher cost and environmental issues limit the large scale application of these dyes. Thus metal free organic dyes are better alternative in the research area for DSSC. There is a large scope for replacing costly metallic dyes with organic dyes. Though organic dyes are less efficient than metallic dyes, they offer a low cost, easy structural modification and environment friendly option for the researchers. These dyes also possess higher extinction coefficient than metallic dyes. In the now popular DSSC, TiO_2 nanoparticles are frequently used with organic dyes to increase the cell's efficiency. Recently several groups have developed metal free organic sensitizers to overcome the prohibitive cost of ruthenium metal complexes [6-10]. Wang et al. [11] reported efficiency upto10.3% for a metal-free organic sensitizer. Indoline [12-14], triphenylamine [15,16] and carbazole [17-19] derivatives have proved promising results with efficiency close to that of Ru-dyes [20]. Some organic dyes like hemicyanine dyes [21], thienothiophene and thiophene based dyes [22], oligothiophene dyes [23,24], and coumarine-based dyes [25,26] have also shown impressive photovoltaic performance. Triphenylene diamine dyes showed efficiencies in the range of 0.64–0.96% with and without ZnO compact layers [27]. Recently a molecularly engineered porphyrin dye, coded SM315, with the cobalt(II/III) redox shuttle resulted in power conversion efficiency of 13% [28]. Research is also going on developing Pt free counter electrodes like carbon nanotubes [29,30], graphite [31-33], PEDOT [34,35], PEDOT: PSS [36], etc as an alternate counter electrode. These are low cost and possess good catalytic activity. The performance of DSSC is now beyond the second generation of solar cells, but the result of the huge research would qualify them as third generation devices, which are expected to deliver electric power in a larger scale at a lower price per Watt [37]. The efficiency increase in the solid state version of DSSCs from about 5% to over15% have been reported within two years of timescale, which comes mainly from the efforts in the perovskites based mesoporous solar cells developments [38]. To further improve the efficiency, many photochemists have made efforts to develop new materials for electrodes and looking for new redox couple. Continuous efforts have been made to design and synthesize high efficiency sensitizers [39-41]. Although satisfactory progress has been made in preparing high performance DSSCs with organic sensitizers, there is still a long way to go for future large-scale commercial production.

1.1 Structure and Working of DSSC

A typical DSSC contains five components: a conductive mechanical support, a semiconductor film, a sensitizer, an electrolyte, and a counter electrode as shown in Fig. 1. Working electrode is composed of a porous layer of titanium dioxide nanoparticles, covered with a molecular dye that absorbs sunlight, like the chlorophyll in green leaves. The titanium dioxide is immersed under an electrolyte solution, above which is a graphite coated with PEDOT-PSS counter electrode is placed to form a sandwich like structure.

The mechanism of DSSC is quite different to the conventional silicon solar cell. Charge separation in a conventional photovoltaic built on *p-n*

junction is fundamentally different from the one in the DSSCs. Normally the silicon acts as both the source of photoelectrons, as well as providing the electric field to separate the charges and create a current. DSSCs in contrast to conventional silicon solar cells have separate sources for these functions. In the dye-sensitized solar cell, the bulk of the semiconductor is used solely for charge transport, the photoelectrons are provided from a separate photosensitive dye. Charge separation occurs at the surfaces between the dye, semiconductor and electrolyte. The schematic diagram representing the reactions involved is shown in Fig. 1. A DSSC is a photoelectrochemical cell mimicking the principle of photosynthesis. When sunlight passes through the transparent electrode coated with dye loaded TiO_2, it excites the electrons of the dye (step a). The excited electrons are injected into the conduction band of the semiconductor, from where they reach the cell anode (step b). Electrons are migrated from the anode to the counter electrode which closes the circuit powering the load (step c). Regeneration of oxidized dye occurs through the donation of electrons from an iodide/triiodide redox electrolyte (step d). Triiodide is reduced in turn at the counter electrode (CE) through electron recapture process (step e). The voltage generated is equal to the difference between the energy level of the electron in the solid TiO_2 and the redox potential of the electrolyte.

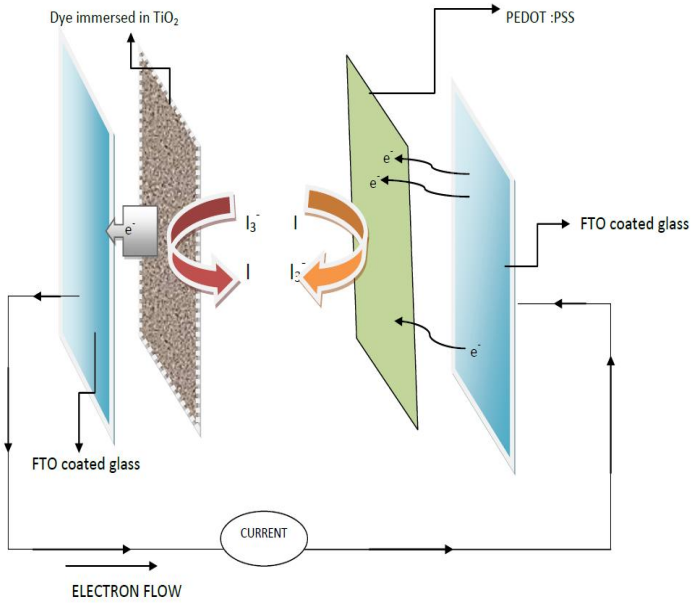


Fig. 1. Structure and working of DSSC

The overall reactions involved are given below:

S + *photon*	⟶	S^*	**(a) Excitation process**
S^* **+ TiO_2**	⟶	$e^-_{(TiO)_2}$ **+ S^+**	**(b) Injection process**
$e^-_{(TiO)_2}$ **+ C.E**	⟶	**TiO_2 +** $e^-_{(C.E)}$ **+ load**	**(c) Energy generation**
$S^+ + \frac{3}{2}I^-$	⟶	$S + \frac{1}{2}I_3^-$	**(d) Regenration of dye**
$\frac{1}{2}I_3^- + e^-_{(C.E)}$	⟶	$\frac{3}{2}I^-$ **+ C.E**	**(e) e^- recapture reaction**

2. MATERIALS AND METHODS

2.1 Materials

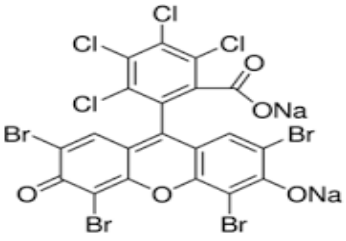
TiO_2 nanoparticles(anatase), acetic acid (pH3-4), Triton-X 100, (0.5M) KI, (0.5M) I_2, ethylene glycol, ethanol, acetone, dimethylsulphoxide (DMSO), PEDOT: PSS and double distilled water and Conducting FTO glass slides.

2.2 Experimental Method

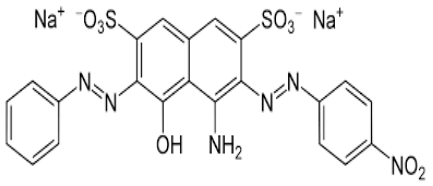
FTO glass slides were cleaned with various solvents in ultrasonicator. Nanocrystalline TiO_2 paste was prepared in acetic acid solution (pH3-4). One two drops of surfactant triton- X was also added to increase binding capacity of the paste. This semiconducting paste is sonicated for one hour with 15 min time gaps to avoid heating through sonication. The slides were masked by scotch tape at three sides and the TiO_2 paste is then applied on the slide through doctor blade method. The slide was allowed to dry at room temperature then sintered at 500°C for one hour. Sintering is done for making TiO_2 layer mesoporous. Mesoporous structure helps in enhancing surface area for dye loading and greater light scattering by the TiO_2 particles. The slide is then digested with photosensitizer for 12 hours in dark for proper adsorption. Excess dye was washed with the same solvent used for dissolving the dye and dried. Counter electrode was prepared by applying thin graphite coating and PSS-PEDOT (mix in DMSO in 3:1 ratio) was spin coated upon graphite layer. It was sintered at 150°C for 15 min. Finally both slides were joined together through binder clips. Electrolyte was inserted from the sides of slide which gets distributed evenly through capillary action. The complete cell is then placed under solar spectrum for I-V charaterization under 1.5 AM sun condition.

2.3 Structure and Description of Dye

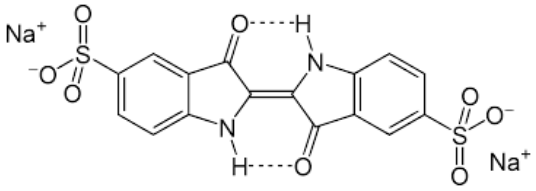
New organic dyes Phloxin B, Indigo Carmine and Amido Black were used as photosensitizers for the fabrication of DSSC. Solubilities of these dyes in various solvents (DW, ethanol, acetone and DMSO) were observed and corresponding absorption spectra were taken. Phloxin B and Amido Black were soluble in all the solvents while Indigo Carmine was insoluble in acetone and ethanol. The structures of the dyes are given below:



Phloxin B



Amido Black



Indigo Carmine

3. RESULTS AND DISCUSSION

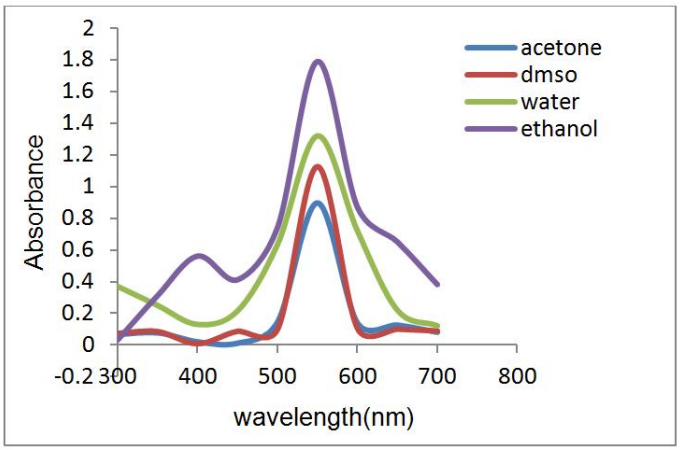
3.1 Spectral Properties of Dyes by UV/Vis Spectrophotometer

The organic dyes exhibited different UV-visible absorption spectra. The spectra of Phloxin B, Amido Black and Indigo Carmine dyes in different solvents were taken. The absorption curves for the dyes in different solvents can be seen in Fig. 2 (a), (b) and (c).

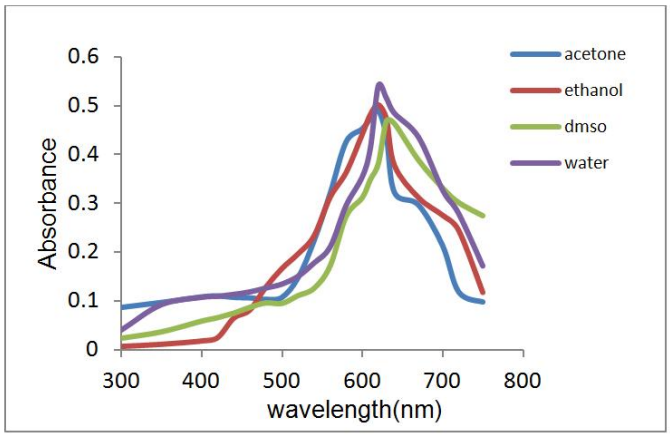
The shift in absorption maxima can be easily seen through the figures. In case of Phloxin B, maximum absorption is seen in ethanol with broad spectrum in the visible region followed by water, DMSO and acetone. Amido black has almost similar absorption but there is slight broadening with water and DMSO. Indigo Carmine was insoluble in ethanol and acetone. Fig. 2(c) clearly shows that indigo carmine has higher absorption peak in DMSO than water.

3.2 Morphological Study of TiO_2 Electrode

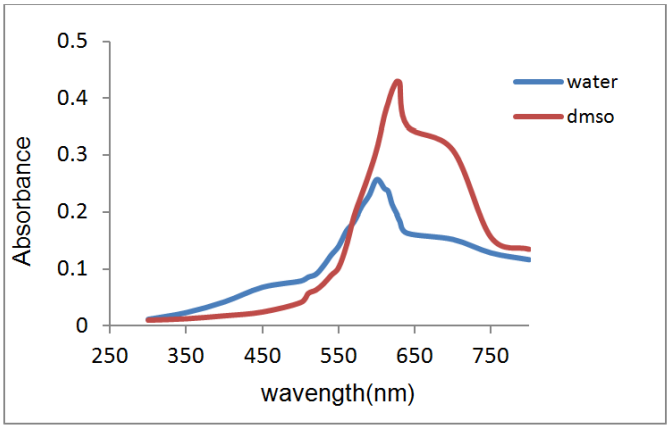
The mesoporous structure of the TiO_2 layer was examined by scanning electron microscopy (SEM). The electron transport rate, which highly depends on the crystallinity, morphology, and the surface area of semiconductors, affects the efficiency of DSSCs [42]. The SEM images were examined at different magnification (2.00 K and 17.6 K) as shown in Fig. 3. Images clearly show that the TiO_2 layer is highly porous and evenly distributed. Granular porous particles of TiO_2 can be seen clearly.



(a)



(b)



(C)

Fig. 2. Absorption spectra of (a) Phloxin B, (b) Amido Black and (c) Indigo Carmine dye in different solvents

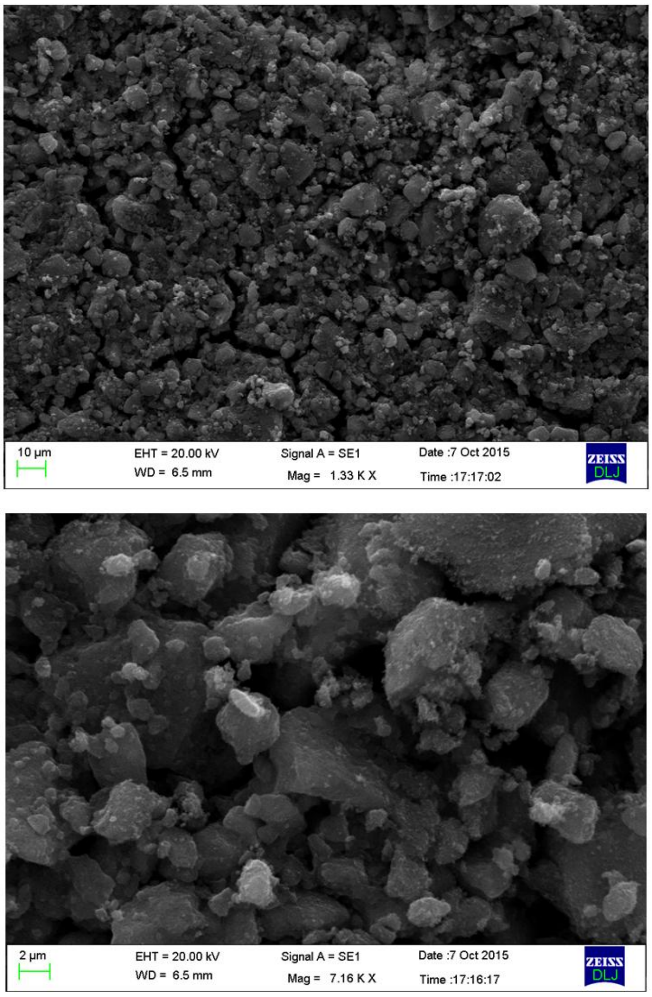


Fig. 3. SEM images of TiO_2 layer coated on FTO glass slides at different magnifications

Table 1. Photoelectrochemical parameters of the DSSCs with a) Phloxin B, b) Amido Black and c) Indigo Carmine dyes in various solvents

a) Phloxin B

Solvents	Voc(mV)	Jsc(µA/cm^2)	Fill factor	η (%)
Water	591	329	0.46	0.093
Acetone	633	373	0.45	0.107
DMSO	586	420	0.40	0.1008
Ethanol	640	368	0.48	0.1142

b) Amido black

Solvents	Voc(mV)	Jsc(µA/ cm^2)	Fill factor	η (%)
Water	525	284	0.48	0.071
Acetone	607	264	0.54	0.086
DMSO	645	421	0.53	0.146
Ethanol	578	278	0.59	0.083

c) Indigo carmine

Solvents	Voc(mV)	Jsc(µA/ cm^2)	Fill factor	η (%)
Water	489	251	0.56	0.0698
DMSO	543	337	0.45	0.0825

3.3 I-V Characterisation

The device performance were analysed for DSSCs sensitized with Phloxin B and Amido Black dyes and Indigo carmine dye. Fabricated cells were characterized using a variable load resistor through the circuit. The photovoltaic parameters (open circuit voltage (V_{oc}), short circuit current density (J_{sc}) and photoelectric conversion efficiencies (η)) of the DSSCs with active area of 1 cm^2 were observed. The comparative results are summarised in Table 1(a), (b) and (c). The open circuit voltage (V_{oc}) and short circuit current (Isc) of the solar cells were observed measured under 100 mW/cm^2 at AM 1.5G condition. The typical J-V curves for DSSCs using Phloxin B and Amido Black dyes in water, acetone, DMSO and ethanol is shown in Fig. 4 (a) and (b). J-V curve for the cell sensitized with indigo carmine dye in water and DMSO is shown in Fig. 4 (c). The J-V curve shows maximum power point (PP), which correspond to the highest value obtained by the product of voltage and current. These maximum voltage and current are represented as, V_{MP} and I_{MP} respectively. Corresponding fill factor (FF) of the device is calculated by the following equation:

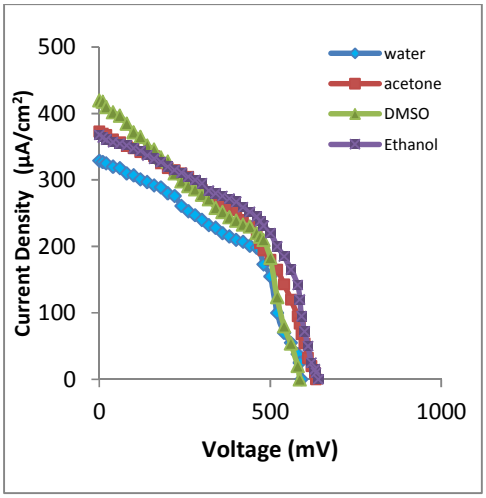
$$FF = \frac{V_{MP} I_{MP}}{V_{OC} I_{SC}}$$

Where, V_{MP} = voltage at power point
I_{MP} = current at power point
V_{oc}= open circuit voltage (highest voltage when current is zero)
I_{sc} = short circuit current (highest current when voltage is zero)

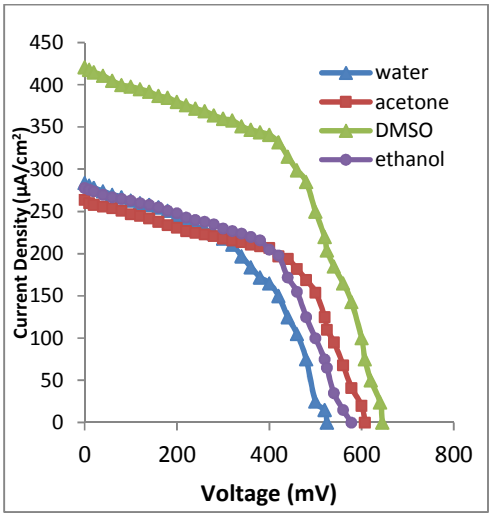
The cell performance was observed for all the cells in different solvents. The comparative performance and photoelectrochemical parameters are summarized in Table 1. Conversion efficiency of the device was determined by the following equation:

$$\eta = \frac{V_{OC} I_{SC} FF}{P_{in}}$$

Where, Ƞ = conversion efficiency of the cell
V_{oc} = open circuit voltage (highest voltage when current is zero)
I_{sc} = short circuit current (highest current when voltage is zero)
FF = Fill Factor
P_{in} = power input (here, 100 mW/cm^2)



(a)



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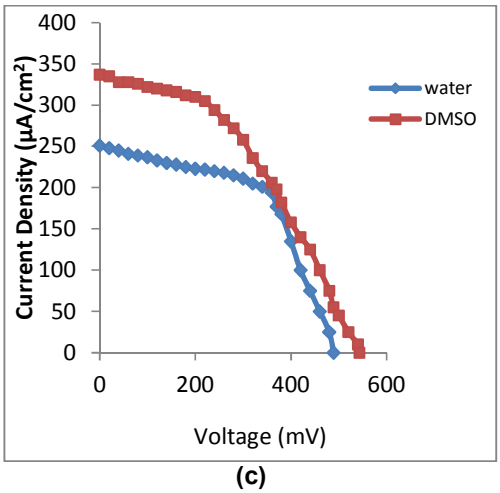

(c)

Fig. 4. J-V curves for the DSSC sensitized with a) Phloxin B, b) Amido Black and c) Indigo Carmine dyes in different solvents

The result of the Table 1 shows that the values of conversion efficiencies are affected by the solvent used for the dyes. Though the values of voltage observed were good, still the dyes showed the low conversion efficiency. This is due to low current generation by the dyes. Phloxin B showed highest conversion efficiency of 0.1142 with the open circuit voltage (Voc) 640 mV, short circuit current (Jsc) 368 μA/ cm^2 and fill factor (ff) 0.48 in ethanol. Amido Black showed highest efficiency of 0.146, open circuit voltage (Voc) 645 mV, short circuit current (Jsc) 421 μA/ cm^2 and fill factor (ff) 0.53 in DMSO. Indigo carmine was least efficient with 0.0825 conversion efficiency, Voc 543 mV, Jsc 337 μA/ cm^2 and ff 0.45 in DMSO. Lowest efficiencies were observed in water for all the dyes.

4. CONCLUSION

In summary, the paper reveals that metal free organic dyes are successful candidates in point of view of the lower cost. The results show that Amido Black dye in DMSO had highest conversion efficiency i.e. 0.1440% and the corresponding Jsc was 421 μA/ cm^2, Voc was 645 mV and fill factor (FF) was 0.53. Phloxin B showed highest of 0.1142 % efficiency in ethanol with Voc equals to 640 mV, Jsc 368 μA/ cm^2 and fill factor (FF) was 0.53. Indigo Carmine though showed poor result; still in DMSO its efficiency was better. Although the efficiencies of these dyes are not so high but still there is scope for better performance with these dyes with better counter electrode and electrolyte combination. Further improvements are possible with modification of organic dyes and additional encouraging research is required to boost performance of DSSCs in future.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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