Natural Plant Trifolium Pratense, Mirabilis Jalapa and Bassia Scoparia Extract Used as Photosensitizer in Dye-Sensitized Solar Cell

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ABSTRACT: *The environmental challenges motivated researchers to replace conventional energy sources with clean and green energy. The Dye-sensitized Solar Cell (DSC) is fabricated from environmentally friendly material, however, its efficiency is less than the 1st generation solar cells. In this study, the extract from natural plants such as i) Trifolium pratense ii) Mirabilis jalapa, and iii) Bassia scoparia (Kochia scoparia) are used as a photosensitizer in the DSC. These extracts contain a different class of colorants (dye) specifically betalains, chlorophyll, and anthocyanins. The efficiencies of as-prepared DSC using these dyes were measured and compared with each other. The highest efficiency of 0.18% was recorded in DSC using Bassia scoparia extract. The efficiency of DSC using Trifolium pratense and Mirabilis jalapa extract was 0.15% and 0.05% respectively. The photoanodes of DSCs were characterized using UV Visible spectroscopy, PL emission spectroscopy, and FT-IR. The Bassia scoparia extract was found to give better light absorption, photon emission, and sticking capability with TiO_2 as compare to Trifolium pratense and Mirabilis jalapa. These natural dyes have never been reported for fabrication of DSC.*

KEYWORDS: *Renewable energy; Dye-sensitized solar cell; Natural dyes; Photovoltaic performance.*

INTRODUCTION

The dye-sensitized solar cell (DSC) is a versatile device that is popular for good photovoltaic performance especially under low light conditions, simple and low-cost fabrication, design flexibility, and transparent features [1-2]. The DSC consists of electrolytes between the working electrode and counter electrode. The working electrode is fabricated by depositing a semiconducting layer (TiO_2, ZnO, etc.) on the transparent conducting electrode (like FTO etc.) and loading dye on the surface of the semiconducting layer. The semiconducting layer work

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as electron transport layer, the electrolyte is used as hole transport layer and the dye molecules are used as photosensitizer to absorb the incident light [3].

The first efficient DSC was reported by *O'Regan* and *Gratzel* (1991), in which the TiO_2 mesoporous layer was used as electron transport layer [4]. *Hashmi et al.*, developed inkjet printing method for dye loading, this technique uses less dye solvent for dyeing on the working electrode in DSC and reduced cell assembly cost and time [5]. The light harvesting of the DSC mainly depends on the photosensitizer (dye), which absorbs photon between ultraviolet to infrared region. Several metallic and organic dyes have been synthesized and used as photosensitizers in DSC. Organic dyes have significant absorption coefficients and easy to modify [6-7]. *Freitag et al.,* reported the Power Conversion Efficiency (PCE) of 28.9% under the 1000 lux indoor illumination in which they used organic dyes in DSC [8]. The synthetic dyes provide better efficiency and durability, however, they have some limitations such as higher cost and toxicity as compared to natural dyes [9].

Researchers are focusing to use natural dyes in DSC which are environment friendly and low cost. The natural dyes are mainly classified into four categories, chlorophyll, flavonoids, anthocyanin and carotenoids. In DSC the dye molecules are excited by the light, this interaction is dependent on the type of dye and wavelength of light. Researchers are working to explore natural dye which gives better performance in visible region of light [10]. *Gu et al.*, extracted dye from purple cabbage achieved PCE of 0.162% [11]. *Dhafina et al*., reported PCE of 0.01% and 0.02 % using Ardisia elliptica fruits in DSC [12]. *Atli et al.*, obtained PCE 0.19% using St. Lucie cherry [13]. *Arifin et al.*, investigated natural dye papaya leaves and achieved a PCE of 0.094% [14]. *Kabir et al*., used yellow dye extracted from turmeric obtained PCE 0.378% and red dye extracted from red spinach was 0.134%. [15]. *Liu et al.*, investigated six kinds of plant leaves coded as D1-D6. The dye D1 shows the highest PCE of 1.08% among other dyes [16]. *Torchani et al*., extracted dye from mallow cell obtained PCE of 0.22% [17]. Chlorophyll is the most abundant colour pigment in plants which absorbs photons in the visible range 550-700 nm. Researchers are also working to combine two or more dyes in a single DSC to achieve better efficiency by covering a wide range of light spectrum. This spectrum results in higher absorbance of sunlight and ultimately increases the photovoltaic performance of DSC [18]. *Noor et al.*, reported cocktail of dyes extracted from leaves of black rice and pandanus amaryllifolius, which showed absorption peaks at 536 nm and 665 nm with PCE of 0.72% [19]. *Park et al*., also mixed extracts of gardenia jasminoideellis, gardenia yellow and gardenia blue, they obtained PCE of 0.59% [20]. Till date hundreds of dyes have been extracted from plants and their performances have been checked in DSCs, however there are thousands of plants which need to be explored for dye extraction and their possible use in DSC.

In this research we have reported the extraction of dye from three plants and their performances have been evaluated in DSCs. We investigated the natural dyes of Trifolium pratense, Mirabilis jalapa and Bassia scoparia (Kochia scoparia) as a photosensitizer in DSC and studied their photovoltaic properties. The performance of these extracts has never been reported earlier for DSC.

EXPERIMENTAL SECTION

Preparation of sensitizers

All fresh flowers were washed several times with tap water to remove mud particles, followed by deionized water to remove the impurities and later dried through the dryer at room temperature. The 10g of dried flowers were crushed into powder and mixed with 100 mL ethanol (96-98% purity) by BDH laboratory supplies-England. The mixture was stirred for 20 minutes at 30ºC and filtered [21]. The extracted dyes were stored in air-tight glass bottles at room temperature. Fig. 1 is illustrating the three extracted dyes of Trifolium prtense, Mirabilis jalapa and Bassia scoparia.

Fabrication of DSC

Fluorine-Doped Tin oxide (FTO) conductive glass sheets (resistance 15 Ω/sq) were cleaned using ultrasonic bath applying detergent, ethanol and acetone for 30 min each. The nanocrystalline TiO_2 paste 18NR-OA (from greatcell Australia) was used. A thin layer (8-10 µm) was printed on FTO substrate of area 0.5cm x 0.8cm (0.4 cm^2) and sintered at 450ºC for 30 minutes, then cool down at room temperature. The photoelectrodes were sensitized with extracted natural dyes and commercial dye N719 (Di-tetrabutylammonium *cis*-bis (isothiocyanate) bis (2, 2′-bipyridyl - 4, 4′-dicarboxylate) ruthenium (II) overnight.

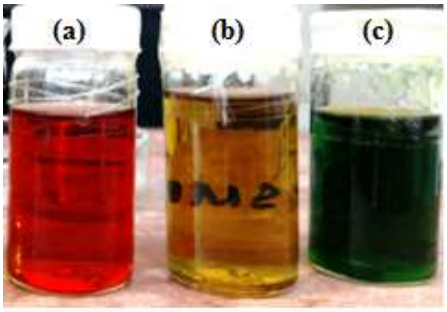

Fig. 1: Dye extracts of plants (a)Trifolium pratense (b) Mirabilis jalapa and (c) Bassia scoparia.

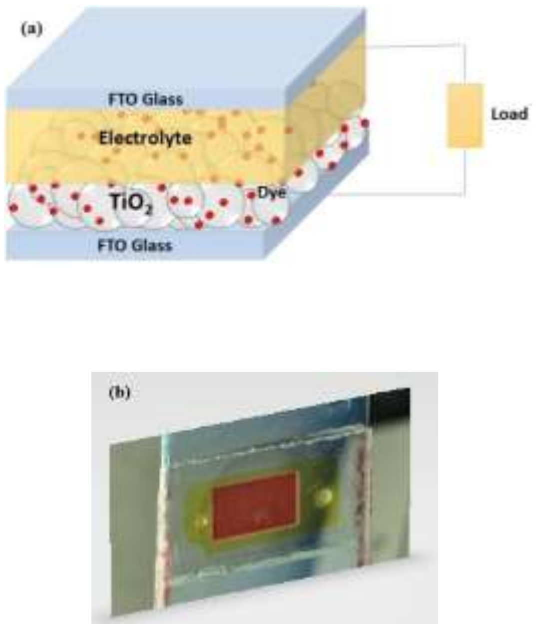

Fig. 2: Dye-sensitized solar cell (a) schematic diagram and (b) fabricated DSC.

The counter electrodes prepared with carbon composite-based catalyst layer deposited on FTO glass using the doctor blade method. Carbon composite comprises of 0.4 g carbon soot, 1.2 g graphite powder and 0.4 g of TiO_2 particle, which improves adhesion. The carbon black is found to be suitable component on the counter electrode for electrolyte reduction as well as to improve electrical conductivity [5]. Fig. 2 (a) and Fig. 2 (b) show the schematic diagram and the fabricated DSC respectively. The electrolyte was then injected through a hole in the counter electrode, while the other hole was used for releasing the trapped air. Finally, the drill holes were closed with sealant.

Characterization

The UV-Visible absorption spectra of the extracted dyes measured with a UV-VIS 1700 Spectrophotometer by SHIMADZU (Japan) and fluorescence spectroscopy by RF-1501 Shimadzu Spectrofluorophotometer (Japan). The pH value measured using pH meter (model:3305) by JENWAY, the FTIR spectra of the extracted dyes were measured by FTIR-8400 SHIMADZU (Japan). The XRD characterization was obtained by using D8 Advance X-ray Diffractometer of BRUKER. The surface morphology was examined through field emission scanning electron microscope (FESEM) by TESCAN (Model: MAIA3). The photovoltaic performance of the assembled DSC measured with a source meter (KEITHLEY 2400) under air mass (AM 1.5G) solar simulator of Peccell technology Co. Ltd., with standard solar irradiance (100 mW cm^{-2}). PC software Peccell IV analyzer 2.3 was applied for current and voltage. All the results were plotted in Origin8 Pro SR0 by Origin Lab Corporation.

RESULTS AND DISCUSSION

Absorption spectra

The photosensitizers (dyes) light absorption ability was investigated by UV-vis spectrophotometer. Hence the dyes were extracted in 98% pure ethanol (C_2H_5OH), therefore ethanol was also used as a buffer solution for absorption spectra between 400nm to 800 nm [22]. Absorption spectra of the natural plant's dye and reference dye N719 are illustrated in the Fig. 3. The photosensitizer showed light absorption ability in the visible region. The maximum wavelength λ_{max}, of the reference dye N719 is 514 nm which absorbs green colour and appears purplish red colour [23]. The first λ_{max} of dye Trifolium pratense was 477nm, which absorbs blue colour and second λ_{max} was 537 nm that absorbs green colour as shown in Fig. 3. The absorption peaks of Mirabilis jalapa were observed at 466 nm and 663 nm corresponding to blue and red colours respectively. Similarly, Bassia scoparia dye shows different peaks at 415nm, 437nm, 468 nm and 664 nm corresponding to violet, blue and red colours respectively. Among all the examined dyes Bassia scoparia gave a better result which contains chlorophyll pigments.

Table 1: The plant names, image and pH level of the extracted dyes.

Plant Names	Plant Images	Dyes Classification	pH value of the extracted dyes in ethanol solution
Trifolium pratense		Anthocyanins	6.47
Mirabilis jalapa		Betalains	3.50
Bassia scoparia		Chlorophyll	6.58

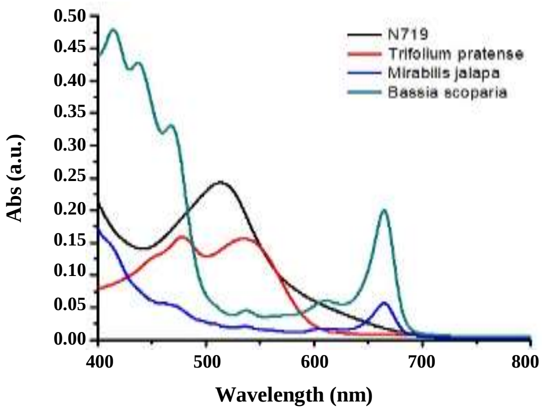

Fig. 3: Absorbance spectra of N719, Trifolium pratense, Mirabilis jalapa and Bassia scoparia

The absorption spectra greatly depend on pH and temperature. The pH value of dye influenced by the temperature and pH; that is if temperature is increased the pH value decreases which tend to change the absorption spectra [24, 25]. In order to avoid temperature effects in the absorption all the dyes were analyzed for UV absorption at room temperature (around 25ºC).

The pH values of the extracted dyes remain in the range of 3.50- 6.58 as shown in Table 1, which are relatively stable and give less dark colour [26, 27]. However in non-aqueous solutions, there are generally no pH values defined. These spectra were monitored at room temperature around 25 ºC. The dyes extracted from Trifolium pratense plants contains anthocyanins as red dye, betalains dye from Mirabilis jalapa reveals yellow dye and Bassia scoparia plant dye contain chlorophyll exhibits green dye as given in the Table 1.

Where E is the energy of the photon, h is Plank's constant, *v* is the frequency, c *is* the speed of light and λ is the wavelength. The graph plotted with the help of Origin software as represented in Fig. 4.

The bandgap energy of the dyes N719, Trifolium pratense, Mirabilis jalapa and Bassia scoparia are 2.2 eV, 2.15 eV, 3.0 eV and 2.5 eV respectively. The absorption coefficient of the extracted dyes was determined by Eq. (3),

$$Absorption\ coefficient(\alpha) = 4\pi k/\lambda \quad (3)$$

Fluorescence spectroscopy analysis

The fluorescence spectra correspond to particular dyes which are estimated by the analogies that the maximum absorption wavelength identified. The excitation range 350 nm to 600 nm was set, as shown in Fig. 5. The reference dye N719 showed highest fluorescence with a maximum excitation efficiency peak at 377 nm near ultraviolet; Trifolium pratense peak appeared at 400 nm, while Mirabilis jalapa and Bassia scoparia dyes showed little peaks at 461 nm and at 401 nm respectively [28].

The fluorescence emission spectra of extracted dyes were analyzed between 600nm to 750nm. The two dyes showed highest peaks, such as dye N719 at 597nm and Bassia scoparia at 639nm, while the Trifolium pratense peak appeared at 621nm and Mirabilis jalapa at 622nm. Dye N719 and Bassia scoparia, which emitted more photons over a wide range of wavelengths, so dye N719

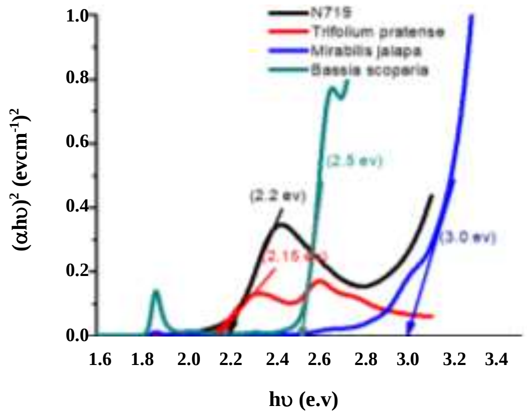

***Fig. 4:** The bandgap of N719, Trifolium pratense, Mirabilis jalapa and Bassia scoparia.*

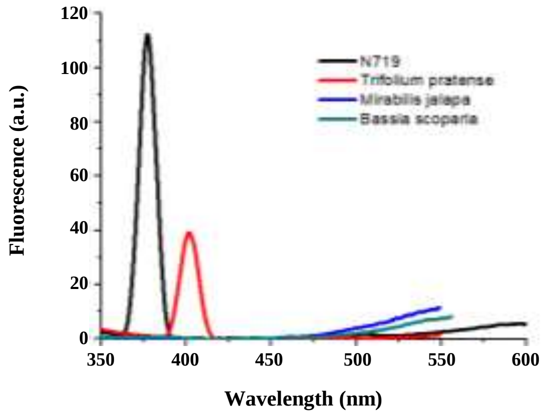

***Fig. 5:** The excitation spectrum of N719, Trifolium pratense, Mirabilis jalapa and Bassia scoparia.*

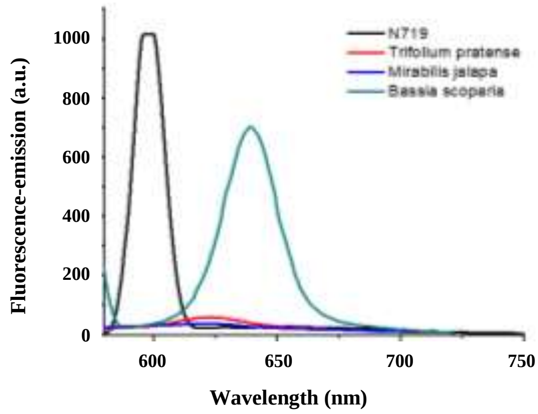

***Fig. 6:** The emission spectrum of N719, Trifolium pratense, Mirabilis jalapa and Bassia scoparia.*

and Bassia scoparia show the higher light harvesting abilities and resulted the greater charge generation. Fig. 6 shows the fluorescence spectra which were observed at room temperature [28].

Fourier transformed infrared (FTIR) analysis:

FTIR is an analytical technique which is used to identify functional groups of the dyes. To examine the extracted dyes, the absorption bands range was selected from 3900 cm^{-1} to 600 cm^{-1}, operated 60 scans/second with a resolution of 2 cm^{-1}. Prior to analysis, stage of the FT-IR was rinsed with ethanol to avoid the possibility of any contamination. Infrared spectra of the dyes are shown in Fig.7. Trifolium pratense dye broad peak shows the first region at 3324 cm^{-1} corresponding to alkynes C-H stretch and the second sharp peak at 1022 cm^{-1} similar to C-O group which exhibits carboxylic acid and hydrogen bonding affiliation. In the Mirabilis jalapa IR spectrum, the first peak appeared at 3317 cm^{-1} while second peak appeared at 1016 cm^{-1} which contain C-O stretching and carboxylic group respectively. The Bassia scoparia first region appeared at 3324cm^{-1} and second peak at 1025 cm^{-1} which signify the same structure of C-O stretching and carboxylic group. Similarly the reference dye 719 first peaks appeared at 3353 cm^{-1} and second peak at 1030 cm^{-1} which are C-O stretching and carboxylic group [29-30].

X-Ray Diffraction (XRD) of TiO_2

The XRD studies were made to investigate the crystalline structure of the TiO_2 screen printed film on the FTO substrate as shown in Fig. 8. The highest peak was observed at 2theta = 25.22° as per source COD1010942 it confirms the TiO_2 anatase, crystalline phase of tetragonal and orientation plane (101).

Scanning Electron Microscopy (SEM) for surface analysis of TiO_2

The surface morphology of TiO_2 paste over FTO was examined by Field Emission Scanning Electron Microscope (FESEM) as shown in Fig. 9. The result indicates that no significant changes observed over the entire surface of the sample, same roughness can be observed at different resolution of the Fig. 9 [31-33].

The elemental composition of the layers was confirmed by the Energy Dispersion Spectroscopy (EDS) analysis as shown in Fig. 10. The result shows the high peak

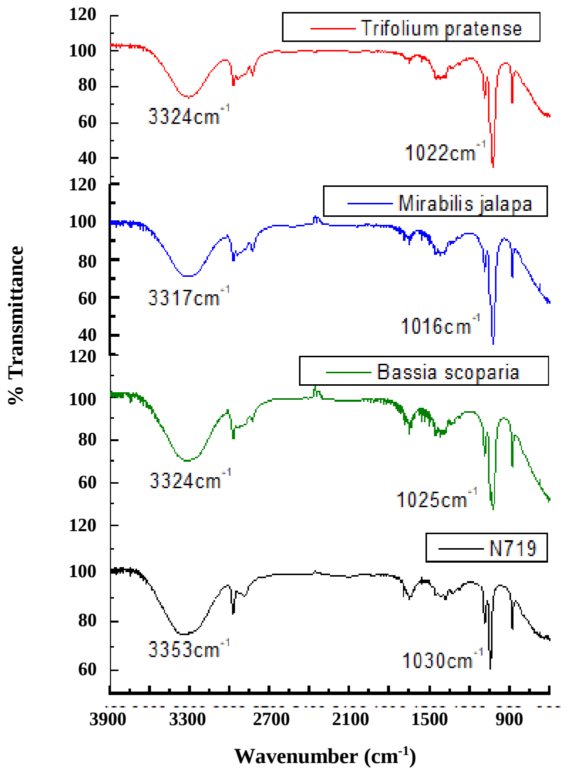

Fig. 7: FT-IR spectra of Trifolium pratense, Mirabilis jalapa, Bassia scoparia and dye N719.

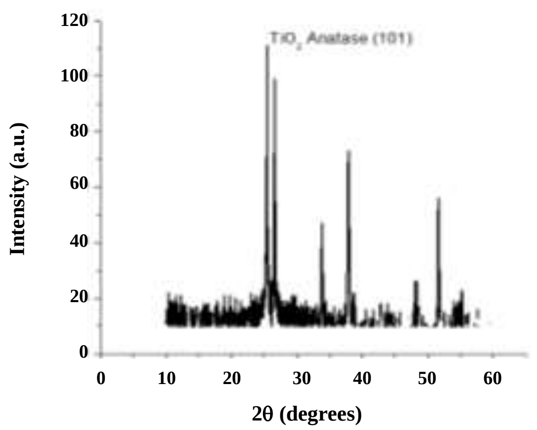

Fig. 8: The result of XRD X-ray diffraction.

of titanium (Ti) at 4.5 keV followed by oxygen (O). Other elements such as Si and Na may be the impurities or contamination over the substrates.

Photovoltaic performance

The photovoltaic performance of the DSCs were determined with the help of short-circuit current (I_{SC}), current density (Jsc), open circuit voltage (V_{OC}), fill factor and power efficiency (η) comparing with reference dye N719 as shown in Table 2. For the photovoltaic performance standard solar irradiance (100 mW/cm^2) was used for an active area 0.16 cm^2 using black tape mask over the screen printed area 0.4 cm^2 (0.8 cm x 0.5 cm) of TiO_2. The DSC efficiency and fill factor were calculated according to the following Eq. (4) and Eq. (5) [33].

$$\text{Efficiency} = \frac{\text{Jsc} \times \text{Voc} \times \text{FF}}{\text{Pin}} \quad (4)$$

Where FF is fill factor and P_{in} is an incident source of light.

$$\text{Fill factor} = \frac{\text{J max} \times \text{V max}}{\text{Jsc} \times \text{Voc}} \quad (5)$$

The power conversion efficiency of the extracted dyes is shown in Fig.11. Among the three extracted dyes, Bassia scoparia shows high PCE.

CONCLUSIONS

This study conducted in two parts; natural plants dye extraction and fabrication of DSC. Three different types of plants were selected for dye extractions including Trifolium pratense, Mirabilis jalapa and Bassia scoparia. The extraction of mentioned dyes was made at 30ºC temperature. Prior to the fabrication of DSC, the properties of extracted dyes were investigated through FT-IR, fluorescence spectroscopy and UV-Vis spectroscopy. The characterization results revealed that these extracts are suitable candidates for DSC. These extracts contain several pigments such as; anthocyanin in Trifolium pratense, Betalains in Mirabilis jalapa and Chlorophyll in Bassia scoparia. The extracted dyes are environment friendly and have better efficiency in absorbing photon. The extracted dyes were utilized in the fabrication of DSC. The photo anode of DSC was fabricated using screen printing of TiO_2 paste on FTO substrate, and dip coating of extracted dyes. The efficiencies of as-prepared DSCs were measured and compared with each other. The highest efficiency of 0.18% was recorded in DSC using Bassia scoparia extract. The efficiency of DSC using Trifolium pratense and Mirabilis jalapa extract was 0.15% and 0.05% respectively. These natural dyes have never been reported for fabrication of DSC. Nature is so vast and yet millions of natural plants are to be discovered as a photosensitizer.

Table 2: Photovoltaic performance of DSC under simulated (100 mW/cm^2)

Dyes	I_{sc} (mA)	J_{sc} (mA/cm^2)	V_{oc} (mV)	Fill factor (%)	Efficiency (%)
Trifolium pratense	0.05	0.34	0.58	0.75	0.15
Mirabilis jalapa	0.02	0.16	0.46	0.72	0.05
Bassia scoparia	0.06	0.42	0.58	0.76	0.18
N719	1.42	8.86	0.79	0.71	5.02

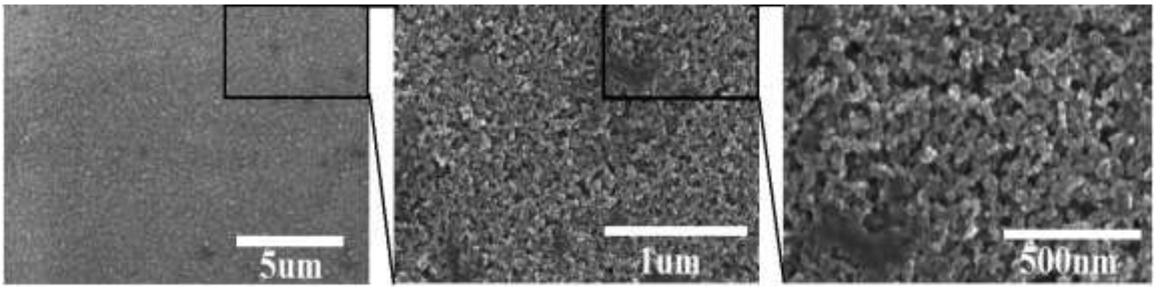

Fig.9: The FESEM result of the TiO_2 paste over FTO with different magnifications.

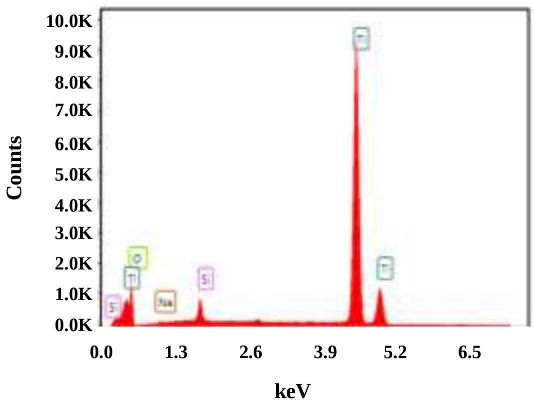

Fig. 10: EDS of TiO_2 coated FTO.

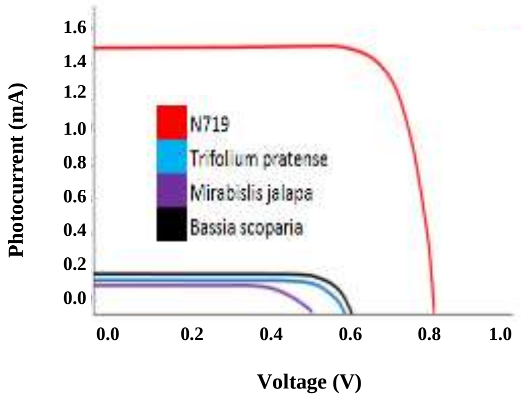

Fig. 11: Current and voltage curves of the fabricated DSCs.

Nomenclature

DSC	Dye sensitized solar cell
Voc	Open circuit voltage
Jsc	Short circuit current density
Isc	Short circuit current
FF	Fill factor
η%	Efficiency in %
FTO	Fluorine dop tin oxide
TiO_2	Titanium dioxide
PCE	Power conversion efficiency

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