

Research Article

Fabrication of Natural Dye Sensitized Solar Cell using Tridax Procumbens Leaf and Beetroot Extract Mixer as a Sensitizer

Arjun kumar B¹, Ramalingam G^{1*}, and Vetrivelan V²¹Department of Nanoscience and Technology, Alagappa University, India²Department of Physics, ThanthaiPeriyar Government Institute of Technology, India

*Corresponding author

Ramalingam G, Quantum Materials Research Lab, Department of Nanoscience and Technology, Alagappa University, Karaikudi 630003, Tamil Nadu, India, Tel: 9445295572; Email: ramanloyola@gmail.com

Submitted: 18 December 2020

Accepted: 21 January 2021

Published: 23 January 2021

ISSN: 2334-1815

Copyright

© 2021 Arjun Kumar B, et al.

OPEN ACCESS

Keywords

• Tridax procumbens; Beetroot; NDSSC; Natural pigment; Chlorophyll; Betalain; Lugol's iodine

Abstract

Titanium dioxide (TiO₂) powder was synthesized by co-precipitation method with cetyltrimethylammonium bromide (CTAB) act as a surfactant. The prepared TiO₂ sample was characterized by X-ray diffraction (XRD), Scanning Electron Microscopy (SEM) and UV-Vis absorption spectra to analysis the structural, morphological and optical properties studies were prepared TiO₂ nanopowder was pasted on conducting surface of FTO glass slide by using doctor blade method with triton X-100 as a binder. Chlorophyll and betalain dye pigment extracted from tridax procumbens leaf and beetroot, which was used as a sensitizer for natural dye sensitized solar cell (NDSSC). The optical properties of the natural dyes were also investigated to aqueous lugol's iodine was used as an electrolyte. The 1:1 ratio of Chlorophyll and betalain dye mixer was used as a sensitizer in NDSSC have cell efficiency of 0.11% was obtained under one sun solar simulator.

ABBREVIATIONS

NDSSC: Natural Dye Sensitized Solar Cell; TiO₂: Titanium Dioxide; CTAB: Cetyltrimethylammonium Bromide; FTO: Fluorine doped Tin Oxide;

INTRODUCTION

In our world, continuously increasing population happened in every year and the peoples in most of the countries to face electricity problem. The conventional silicon based solar cell has high efficient with some default problems, such as high expensive and accomplished manufacturing techniques [1]. So, harvesting solar energy is the difficult and at a same time necessary to find another device in low cost. O'Regan and Grätzel are starting persons to build low-cost dye sensitized solar cell (DSSC), and it is also called modern photovoltaic device [2,3]. DSSC is the non-conventional technology with lost cost, simple fabrication technics and user-friendly [4]. The Dye sensitized solar cell have component of wide bandgap semiconductor with photo sensitizing dye molecules, electrolyte (iodide & triiodide ions) and counter electrode. Up to many wide bandgap semiconductors are studied, but TiO₂ and ZnO materials are experimentally suitable for photoanode. TiO₂ has a good material for photoanode with simple preparation, long term thermal and photo stability [1,5-7]. The efficiency in DSSC is depends on the structural and physical properties of the sensitizer. In dye sensitized solar cells (DSSCs), ruthenium-based dyes are used as a sensitizer.

It has high stable excitation state to absorbed entire visible region but, it has high expansive, difficult in preparation of Ru metal complexes and causes environmental pollution [1,8]. To solve this problem, artificial sensitizer in DSSC shifted to natural sensitizer. The natural dye pigments have been extracted from varies plant, leaves, fruits and fungi. It has lot of advantages such as non-toxicity, easy availability, functionalizes in natural dye like organic dyes, simple preparations and temperature compatibility [19]. The antholyanins [10], chlorophyll [11], and betalains [10], are some of examples of natural pigments.

Chlorophyll is responsible of green colored leaf and consists of tetraphyrrole ring surrounded by magnesium ion. It is divided into two types namely chlorophyll a and chlorophyll b. Chlorophyll absorbs red color from white light in 650 - 700 nm range and similarly to absorbs blue-violet region (400 - 500 nm). It usually presented in varies plants such as Tridax procumbens, mimosapudiaetc [8,11]. Betalain is another interesting pigment consisting of red-violet betacyanins and yellow-orange betaxanthins, these has a most probably maximum absorptivity in 535 and 480 nm. The beetroot and Bougainvillea glabra are the examples of betalain plant sources [10,12]. The lugol's iodine is a solution contains potassium iodide with iodine water and this is also called strong iodine solution and iodine-potassium iodide (I₂KI). The lugol's iodine solution in water has redox potential to produce I⁻/I₃⁻ redox couple from the literature [13,14]. In this work, low cost chlorophyll and betalain natural

dye pigment are prepared from tridax procumbens leaf and beetroot extract. To the best of our knowledge, this is the first time to use tridax procumbens leaf extract as a sensitizer. The as-prepared TiO_2 coated FTO glass slides are dipping into natural dyes, so by this way to activate the photoanode in visible region. The photocurrent-photovoltage (J-V), performance of fabricated NDSSC was measured under one sun solar simulator and analyzed.

MATERIALS AND METHODS

All the chemicals used in this synthesis process were received without further purification and deionized water was used as a solvent. TiO_2 powder was synthesized by co-precipitation method at room temperature. Cetyltrimethylammonium bromide (CTAB), was used as a surfactant and to maintain 1:1 ratio of CTAB and Titanium isopropoxide [$\text{Ti}[\text{OCH}(\text{CH}_3)_2]_4$] in entire process. In this typical synthesis, 12.5 ml absolute ethanol ($\text{C}_2\text{H}_5\text{OH}$), in 50 ml water was taken into the beaker, adds 1.82 g of CTAB and continuously stirred the solution well for 1 h to reach clear solution. After that, 1.43 ml titanium isopropoxide was added drop wise in above solution under continuously stirred for 24 h. The resultant solution is milky, and it was centrifuged, washed several times in water and then in ethanol. The final product was dried at 80 °C for 12 h, and then calcined at 450 °C for 2 h to remove unwanted organic templates [15].

Preparation of Nature dye sensitizer

Fresh leaves were collected from tridax procumbens plant in surrounding area of Alagappa University, Karaikudi, India. It was washed with deionized water several times to remove the dust, water soluble solvent on leaves and then dried at room temperature. 4 g of leaves were crushed by using mortar and pestle, dissolved in ethanol and then heated at 40 °C for 45 min. After that, filtered it and covered the sample with aluminum foil and kept it in dark place for 2 days at RT, without exposure to sunlight and sample name called as TPLE. Fresh beetroot was purchased from local market and cleaned with deionized water and the skin was peeled out. Then it was scrapped into many small pieces and 5 g of cut Beet root was crushed it using mortar to prepare the betalain pigment and it was dissolved in ethanol. The remaining process is same as sketch in tridax procumbens leaves extract and finally sample named as BRE. To mix the equal 1:1 volumetric ratio of TPLE and BRE to form mixer dye and it was named as TBM sample. (Figure 1) shows the photograph of as-prepared TPLE, BRE and TBM natural dye samples.

Fabrication of NDSSC

The synthesized TiO_2 powder was pasted on conducting surface of FTO glass slide ($\text{F}:\text{SnO}_2$, Surface resistivity) by doctor blade technique. 0.5 g of as-synthesized TiO_2 powder was ground it well with 0.1 ml acetylacetone ($\text{CH}_3\text{COCH}_2\text{COCH}_3$), for few min using mortar. To add 1:1 ratio of absolute ethanol in water solution and 0.4 ml titronX-100 with continuously ground to make paste. The FTO glass slide was washed with deionized water and then using methanol-acetone (1:1 ratio) solution using sonication. The prepared paste was spread on the FTO plate using glass rod and it was calcined at 400 °C (rate 5 °C per min) for 10 min. To drop the TiO_2 coated FTO glass slide into TPLE and TBM nature dye as prepared earlier and kept it for 2 days. Finally, it was rinsed with deionized water, dried by dryer and named as ND1 and ND₂.



Figure 1 Photograph of extracted dye solutions.

To scrap the graphite carbon pencil on the conducting surface of FTO glass slide, it acts as a counter electrode [20]. The Adhesive tape is a spacer layer to sandwich the photoanode and counter electrode with few drops of lugol's iodine electrolyte in between them [16]. In electrolyte preparation, 2 ml of lugol's iodine in 20 ml water act as an electrolyte. A black mask with an open window (0.25 cm^2) on front surface of TiO_2 coated FTO glass slide to choose the working area of the cell [17].

Characterization Studies

X-ray Diffraction pattern was recorded from PANalytical X'pert Pro using $\text{CuK}\alpha$ radiations in 2θ range from 20° to 80° to analysis the phase and crystal structure of TiO_2 nanopowder. The morphological studies of as-prepared TiO_2 nanopowder were scanned on using Scanning Electron Microscope (SEM, VEGA 3 TESCAN). The optical analysis of power was conducted by using UV-Vis spectrophotometer (Elico, SL-159). The photocurrent density-photovoltage (J-V), performance of NDSSCs using natural dye extract was taken out from precision source/measure meter (Agilent, Model: B2901A), under one solar simulator (Royal Enterprises, Chennai, Model: 100L).

RESULTS AND DISCUSSION

The fabricated natural dye sensitized solar cell with novel lugol's iodine as an electrolyte in (Figure 2). The fabricated cell has components such as, nanostructured semiconductor (TiO_2 & ZnO), on FTO glass slide, Sensitizing layer (natural extract), lugol's iodine electrolyte (I^-/I_3^-) [13,14], and counter electrode (graphite carbon on FTO glass slide). During working process, natural dye extract absorbed the photons and created electron-hole pairs. The electrons are injected into conducting band of wide band gape of Nanostructured TiO_2 semiconductor, FTO glass slide and then passed through load, to produce electricity. After that electron reach the counter electrode, then electrolyte and finally to reach the natural dye sensitizer (regeneration of dye). Lugol's iodine is an iodine solution which contains potassium iodide with iodine water and it has I^-/I_3^- redox ions to use as an electron mediator between photoanode and counter electrode [13,14].

Structural studies

The X-ray diffraction pattern of TiO_2 powder was analyzed in between the 2θ range is from 10 to 80° represented in the (Figure 3). The sharp and intense peaks of the pattern show that

highly crystalline and it matches the standard JCPDS file (Card No: 21-1272) [15]. The diffraction peaks located at 25.2° , 36.9° , 37.8° , 38.5° , 48.0° , 53.8° , 55.0° , 62.1° and 62.6° corresponding to the diffraction plane at (101), (103), (004), (112), (200), (105), (211), (213) and (204). The as-prepared TiO_2 nanopowder has tetragonal structure. The average crystallite size (D) of prepared TiO_2 powder was found out by using the Debye Scherrer's equation [18], $D = 0.9\lambda / \beta \cos\theta$; Where, λ is the wavelength of X-ray ($\lambda = 0.1540$ nm), D is average crystallite size (nm), β is full width half maxima of the peak (in radians), θ is Bragg Diffraction angle ($^\circ$). The calculated average crystallite size was 15 nm. In this XRD pattern, no other peaks are founded, and it confirmed that prepared TiO_2 nanopowder have formation of single phase structure.

SEM micrograph analysis

The scanning electron microscope (SEM) is a scanning device to study the structure of material in microscopic range. The SEM micrographs of synthesized TiO_2 powder with different magnifications are shown in the (Figure 4). In SEM investigation, agglomerates of Nano size regime to form the micro-structured powder.

Optical studies

UV-Vis absorption spectrum of TiO_2 powder is in range between

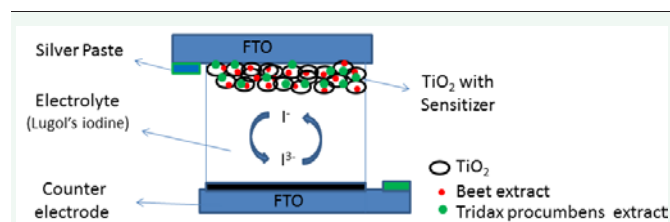


Figure 2 Schematic diagram of fabricated natural dye sensitized solar cell.

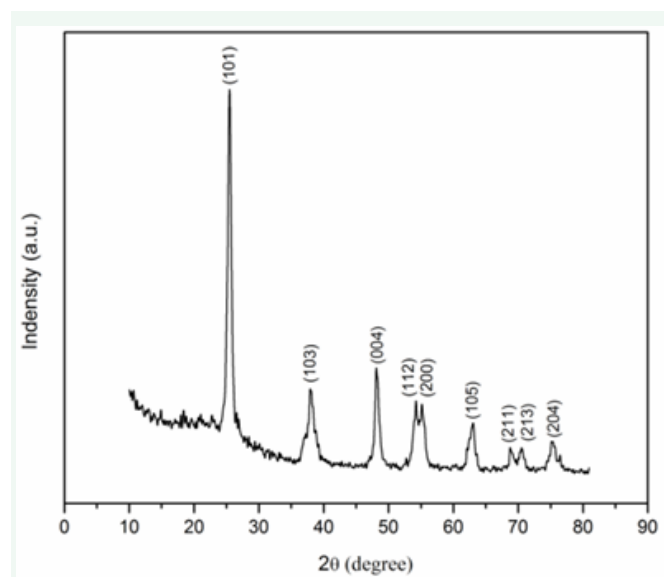


Figure 3 XRD pattern of synthesized TiO_2 nanopowder.

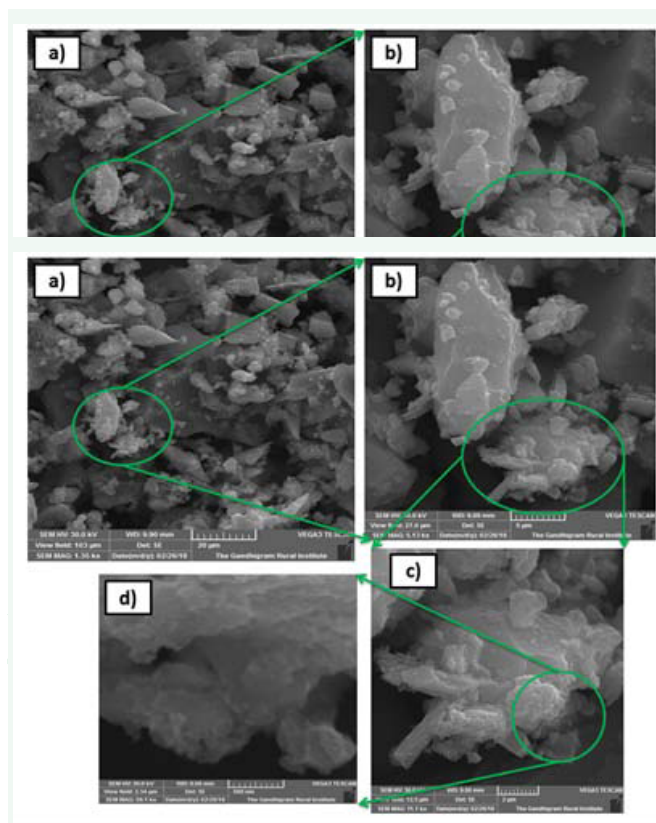


Figure 4 SEM images of TiO_2 nanopowder.

200 and 900 nm as shown in the (Figure 5a). Strong absorption band is found in the Ultraviolet Region. A plot $(\alpha h\nu)^{1/2}$ versus $h\nu$, known as Tauc's plot can be used to determine the optical band gap of the semiconductor material as shown in (Figure 5b). The optical bandgap of the synthesized product was determined to be 2.9 eV and corresponding to the excitation wavelength of TiO_2 is 428 nm. The band gap of bulk TiO_2 material is 3.2 eV [19], higher than synthesized TiO_2 powder. The (Figure 5c), shows the spectra of prepared natural dye extract in range between 200 to 900 nm. From tridax procumbens leaf extract (TPLE) spectrum there are two strong absorption peaks found at 664.5 and 423 nm in the region of green and blue, it's corresponding to chlorophyll [8]. In beetroot extract (BRE), absorption is found at 474 nm corresponding to yellow orange betacyanins [10]. In 1:1 mixer dye pigment (TBM), three absorption peaks at 415, 472 and 664 nm, because of both chlorophyll and betacyanin pigments are presented. The 1:1 mixer (TBM) has absorbed more visible photons compare to tridax procumbens extract; it also has more electron excitation and good sensitizer for NDSSC.

Solar cell analysis

Figure 6 shows the photocurrent to photovoltage (I-V) characteristics curve to study the performances of the fabricated natural dye sensitized solar cell. The chlorophyll dye pigment extracted from tridax procumbens leaf and betalain dye pigment extracted from beetroot is the sensitizer to sense the photons, to produce the excited electrons and these natural dye extracts are low cost dye technology. The overall performance of open circuit

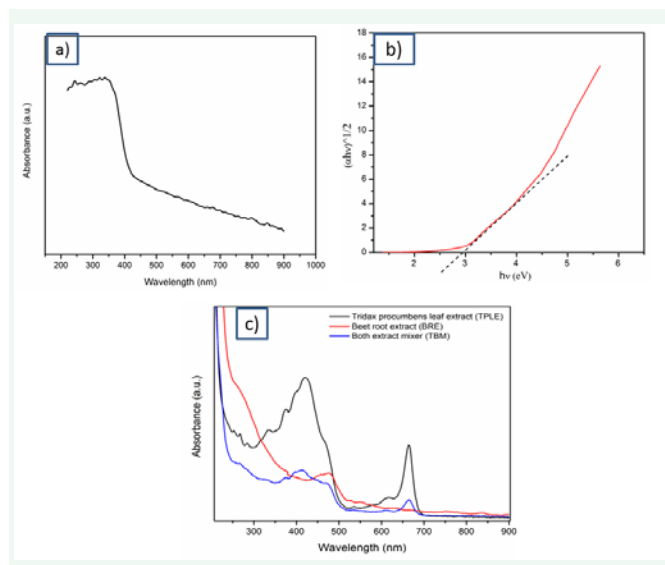


Figure 5 Optical studies of as-prepared TiO₂ powder and natural dye extracts, a) UV-Vis absorption spectra of TiO₂ powder, b) Tauc's plot of TiO₂ powder and c) Absorption spectra of prepared Natural dye in ethanol.

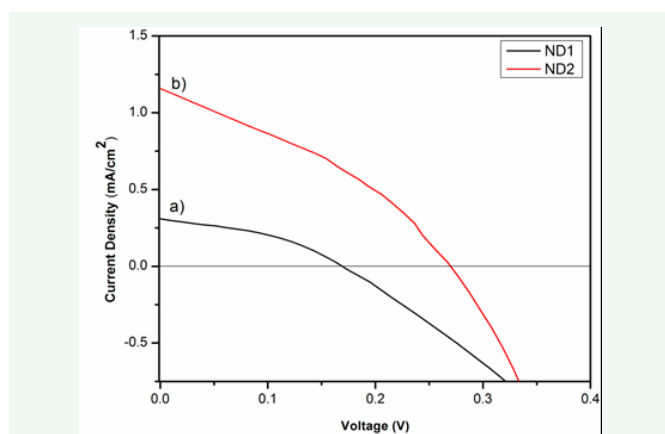


Figure 6 Photocurrent density-photovoltage (J-V) curves based on a) Tridax procumbens leaves extract (TPLE) as a sensitizer and b) mixer (TBM) dye pigment as a sensitizer.

Table 1: Summarized detail of fabricated NDSSCs parameter values.

	J_{sc} (mA/cm ²)	V_{oc} (V)	Fill Factor	Efficiency (%)
ND1	0.31	0.17	0.400	0.02
ND2	1.16	0.27	0.345	0.11

Abbreviations: J_{sc} – Short Circuit current; V_{oc} - Open circuit voltage

voltage (V_{oc}), short circuit current density (I_{sc}), fill factor (FF) and cell efficiency (η) of fabricated natural dye sensitized solar cells are given in the (Table 1).

The output of NDSSC with chlorophyll dye pigment as a sensitizer of open circuit voltage, short circuit current density, fill factor, and efficiency of cell are 0.17 V, 0.31 mA/cm², 0.4 and 0.02%. After mixing betalain dye pigment into chlorophyll

dye pigment in 1:1 ratio, the efficiency is strictly increase from 0.02 to 0.11%. The power conversion efficiency of chlorophyll and betalain mixed sensitizer in NDSSC is 5.5 times, increased approximately. The co-sensitization of betalain in chlorophyll dye (1:1 ratio) gives more electron transpare to semiconducting nanostructured TiO₂, because of this mixer dye pigment has more photon sensitizer in visible region compare to chlorophyll dye pigment and it was confirmed from UV-visible spectrum.

CONCLUSION

Tetragonal TiO₂ powder was successfully synthesized by co-precipitation method and characterized. Novel aqueous lugol's iodine as an electrolyte used in NDSSC to produce iodine redox couple. The chlorophyll and betalain natural dye pigment mixer of 1:1 ratio have higher visible light absorber confirmed from absorption spectrum and used as a sensitizer of NDSSC with higher efficiency of 0.11% compare to chlorophyll dye pigment sensitized solar cell.

ACKNOWLEDGEMENTS

The corresponding author Dr. G. Ramalingam extends his gratitude and acknowledgment for the financial support from RUSA 2.0 Grant No. F.24-51/2014-U.

REFERENCES

- Kushwaha R, Srivastava P, Bahadur L. Natural Pigments from Plants Used as Sensitizers for TiO₂ Based Dye-Sensitized Solar Cells. *J Energy*. 2013; 654953: 1-8.
- Chandra Maurya I, Neetu, Arun Kuma rG, Srivastava P, Bahadur L. Natural Dye Extracted From Saracaosoca Flowers as Sensitizer for TiO₂-Based Dye-Sensitized Solar Cell. *J Sol Energy Eng*. 2016; 138: 1-6.
- Brian O'Regan Michael Grtazel. A low cost high efficiency solar cell based on dye-sensitized colloidal TiO₂ films. *Nature*. 1991; 353: 737-740.
- Michael Grtaze I. Photoelectrochemical cells. *Nature*. 2001; 414: 338-344.
- JintingJ, Seiji I, Motonari A, FuminW. Preparation of TiO₂ nanocrystalline with 3-5 nm and application for dye-sensitized solar cell. *J. Photochem. PhotobiolA Chem*. 2007; 189: 314-321.
- Qifen Z, Christopher SD, XiaoyuanZ, Guozhong. ZnO Nanostructures for Dye-Sensitized Solar Cells. *Adv Mater*. 2009; 21: 4087-4108.
- Zhong-sheng W, Hiroshi K, Takeo K, Hironori A. Significant influence of TiO₂ photoelectrode morphology on the energy conversion efficiency of N719 dye-sensitized solar cell. *Coord Chem Rev*. 2004; 248: 1381-1289.
- Rajalakshmi K, Banu N. Extraction and Estimation of Chlorophyll from Medicinal Plants. *Int J Sci Res*. 2015; 4; 209-212.
- Abdel-Latif MS, Mahmoud BA, Taher ME. Dye-Sensitized Solar Cells Using Dyes Extracted From Flowers, Leaves, Barks, and Roots of Three Trees. *Int J Renewable Energy Res*. 2015; 5; 294-298.
- Ramamoorthy R, Radha N, Maheswari G, Anandan S, et al. Betalain and anthocyanin dye-sensitized solar cells. *J Appl Electrochem*. 2016; 46; 929-941.
- Ridwan MA, Noor E, MS Rusli, Akhiruddin. "Fabrication of dye-sensitized solar cell using chlorophylls pigment from sargassum," *IOP Conf. Series: Earth and Environmental Science*. 2018; 144: 012039.
- Isah.KU, AhmaduU, IdrisA, KimpaMI, EssangU, et al. Betalain pigments

- as natural photosensitizers for dye-sensitized solar cells: the effect of dye pH on the photoelectric parameters. *Mater Renew Sustain Energy*. 2015; 4: 39.
13. Gottardi W. Redox-potentiometric/titrimetric analysis of aqueous iodine solutions. *Fresenius J Anal Chem*. 1998; 362: 263-269.
 14. Hopkins TM, Alexander MH, James AL, Kathleen L. Combining Micro-Computed Tomography with Histology to Analyze Biomedical Implants for Peripheral Nerve Repair. *Neurosci Methods*. 2015: 255; 122-130.
 15. Satyanarayana RG, Krishnamoorthy A, Christopher Y, Grtazel M. Synthesis of mesoporous titanium dioxide by soft template-based approach: characterization and application in dye-sensitized solar cells. *Energy Environ. Sci*. 2010; 3: 835-845.
 16. Benekphal NP, lez-pedro VG, Boix PP, Chavhan S. Colloidal PbS and PbSeS Quantum Dot Sensitized Solar Cells Prepared by Electrophoretic Deposition. *J Phys Chem C*. 2012; 116: 6391-16397.
 17. Zhonglin D, Zhenxia oP, Santiago FF, Zhao K, Donghui L. Carbon Counter-Electrode-Based Quantum-Dot-Sensitized Solar Cells with Certified Efficiency Exceeding 11%. *J Phys Chem Lett*. 2016; 7: 3103-3111.
 18. Bindu P, Sabu Thomas. Estimation of lattice strain in ZnO nanoparticles: X-ray peak profile analysis. *J Theor Appl Phys*. 2014; 8: 123-134.
 19. Madhusudan ReddyK, ManoramaSV, Ramachandra ReddyA. Bandgap studies on anatase titanium dioxide nanoparticles. *Materials Chemistry and Physics*. 2003;78: 239-245.
 20. Binti Zulkifili AN, KentoT, Daiki M, Fujiki A. The Basic Research on the Dye-Sensitized Solar Cells (DSSC). *J Clean Energy Technol*. 2015; 3: 382-387.

Cite this article

Arjun Kumar B, Ramalingam G, Vetrivelan V (2021) Fabrication of Natural Dye Sensitized Solar Cell using *Tridax Procumbens* Leaf and Beetroot Extract Mixer as a Sensitizer. *JSM Nanotechnol Nanomed* 8(1): 1076.