Research Article

Journal of Environmental Nanotechnology

# Optical Study on Co-Sensitization of Betanin Dye with Cadmium Sulfide to Fabricate Dye Sensitized Solar Cell

D. S. Suresh, Sapna Sharanappa, S. P. Vijaykumar, Abdullah Ba Shbil, H. Ganesha and H. Devendrappa*

Department of Physics, Mangalore University, Mangalagangothri, KA, India

Received: 01.07.2024 Accepted: 02.09.2024 Published: 30.09.2024

*dehu2010@gmail.com

## ABSTRACT

Natural dyes are low-cost, eco-friendly, readily available and have optical characteristics which render them useful in energy research. The titanium tetra isopropoxide precursor can be employed using sol-gel synthesis to get titanium dioxide nanoparticles ($TiO_2$ NPs). The FESEM analysis confirmed the deposition of $TiO_2$ NPs on Indium Tin Oxide (ITO) substrate, which is necessary for the photo-conversion mechanism to produce electricity. One of the natural dyes that is extracted from red beets is known as betanin dye (Bd). Due to their aligned energy levels, Bd and cadmium sulfide (CdS) are used together to provide effective optical characteristics, making them suitable as dye sensitizers. The presence of Cd-S, C-N and hydroxyl groups assigned to 500, 1633, and 3300 $cm^{-1}$ respectively, was demonstrated by the FTIR analysis. The energy gap of 2.16, 2.13 and 2.2 eV for Bd, CdS and Bd-CdS composite was estimated using Tauc's plot and UV-visible spectra demonstrated maximum absorbance at 485, 510 and 528 nm, respectively. The Bd absorbs broad light in visible region due to the addition of CdS. The major charge-transport phenomenon in dye-sensitized solar cells (DSSC) involves an increase in photo-current density, due to its luminous nature. However, the recombination of charge carriers prevents the overall performance of the DSSCs. The power conversion efficiency of the DSSC constructed from Bd dye and Bd-CdS composite was 0.234% and 0.367%, respectively. This optical investigation suggests co-sensitization as a sure-fire method to improve the efficiency of DSSCs extracted from natural dyes, making them reliable for future indoor applications.

**Keywords:** Co-sensitization; Dye sensitized solar cells; Energy conversion; Betanin; Cadmium sulfide.

## 1. INTRODUCTION

The renewable resources need to be used to solve energy crisis and natural world is adversely affected by the use of hydrocarbon fuels and their waste. The most promising and simple fabrication in solar cell research is dye sensitized solar cells (DSSC) since this has been evolved from first Grätzel DSSC to achieve power conversion efficiency (PCE) more than 13% recently (O'regan and Grätzel, 1991; Singh and Shougaijam, 2022; Mathew *et al*. 2014). The literature has been reported on inorganic sensitizer and organic dyes used in DSSCs. These dyes are hazardous in addition to other factors like cost, toxicity and availability, which have altered its way of employment. When compared to the expensive rate and limited number of synthetic dyes, natural dyes are more affordable option because of their low cost, convenient use, abundant resource availability, and do not have any environmental hazard (Mathew *et al*. 2014; Piwowar-Sulej *et al*. 2023). It is important to meet the necessary requirements for fabricating more efficient DSSCs incorporating natural dyes as sensitizers. Richhariya *et al*. (2022) reported significance of choosing a strategy for constructing the DSSC. The dye sensitizer has an obvious impact on the overall performance of the device along with choice of semiconductor, electrolyte and counter electrode (Richhariya *et al*. 2022; Teja *et al*. 2023). The natural dyes over the decade show the progress and development in wide range of applications (Yadav *et al*. 2023). Various natural dyes have been extensively studied and tested as affordable substitutes against expensive synthetic organic dye complexes. As compared to them, natural dyes often demonstrate poor photovoltaic response in DSSC because of low charge transfer absorption throughout the entire visible region and weak binding energies with $TiO_2$ thin films, but these dyes are incredibly easy to manufacture and economical in synthesis point of view (Chang *et al*. 2010).

Over the years, researchers have turned their attention to natural dyes to identify the alternative to synthetic dyes in DSSCs. Natural dyes are derived from various sources such as *Luffa cylindrica L* (Maurya *et al*. 2016), spinach (Syafinar *et al*. 2015), Aloe vera and Cladode of Cactus (Ganta *et al*. 2017)], red amaranth leaves (Ramanarayanan *et al*. 2017), red cabbage (Pratiwi *et al*. 2014), turmeric plants root (Hossain *et al*. 2017), *Mussaenda erythrophylla* (Rajaramanan *et al*. 2023) and *Alpinia purpurata* (Da-Conceição *et al*. 2023). Moreover, utilizing natural dyes in DSSCs can create additional value for agricultural by-products and promote sustainable practices (Roslan *et al*. 2018; Zhou *et al.* 2011). The DSSCs based on natural dyes is an exciting topic of research due to their optical, eco-friendly and flexible properties. The electrolyte becomes photo-stable when detached dye molecules dissolve into the liquid electrolyte (Kabir *et al*. 2019). Susha *et al*. (2018) reported that the photo-conductivity of Betanin-Cadmium Sulfide (Bd-CdS) nanostructures increased by the addition of CdS. The Bd is a natural red pigment found in red beets suggested to utilize as a dye sensitizer in DSSCs. The CdS is a semiconductor material with excellent light-absorbing properties. It can absorb photons from sunlight that generates electron-hole pairs, which are separated at the interface of electrode-electrolyte (Alkuam *et al.* 2017; Mahapatra *et al*. 2020).

Theoretical optical study reveals bandgap and appropriate band edge location in connection with solar light conversion and water decomposition potential. The CdS represents one of the chalcogenides that are most commonly employed to sensitize $TiO_2$ metal oxide semiconductor. In addition, because of the potential difference that develops at the interface between the CdS and $TiO_2$ semiconductors, electrons may migrate from the conduction band (CB) of CdS to the CB of $TiO_2$ easily due to the band edges alignment of CdS and $TiO_2$ (Yavuz, C. and Ela, 2023). The $I^-/I^{3-}$ redox electrolyte, dye, counter electrode, and dye-sensitized $TiO_2$ photo electrode constitute usual components of a DSSC. When the dye molecule is subjected to light, one electron from its valance band moves towards the conduction band, where electron loss is compensated by an electron in the $I^-/I^{3-}$ redox electrolyte. The counter electrode supplies this electrolyte with one electron. Through the external circuit, an electron across the working electrode's conduction band flows to the counter electrode. The electricity originates from this electron transition. The solar cell is an instance of this redox process. Efforts to reduce production costs and enhance PCE have focused on researching every DSSC component and exploring possible modifications (Ravichandran *et al*. 2022). Innovative techniques such as co-sensitization have the potential to improve the sustainability and efficiency of solar energy conversion technologies in the dynamic field of renewable energy. Creative methods, such as utilizing plant leaf dye as photosensitizers for DSSCs are cost-effective and environmentally beneficial. In addition to promoting sustainable behaviours, using natural dyes lowers the cost and enhances their availability for renewable energy production (Onyemowo *et al.* 2024). Eventually, an extensive examination of the effects of two co-sensitized conformations on DSSCs reveals that the H-H conformation is the most beneficial because of its more intense structure, obvious absorption spectrum red shift and better intermolecular charge transfer (Liu *et al.* 2023).

The present work aims to extract Bd dye through ultrasound assisted extraction method and also to prepare composite of Bd with CdS (Bd-CdS) via probe sonication for 30 minutes which exhibits co-sensitization process. The novelty of work was to fabricate DSSC based on Bd and Bd-CdS dye composites along with $TiO_2$, iodide and graphite as semiconductor, redox electrolyte and counter electrode, respectively.

## 2. EXPERIMENTAL METHODS

### 2.1 Materials Used

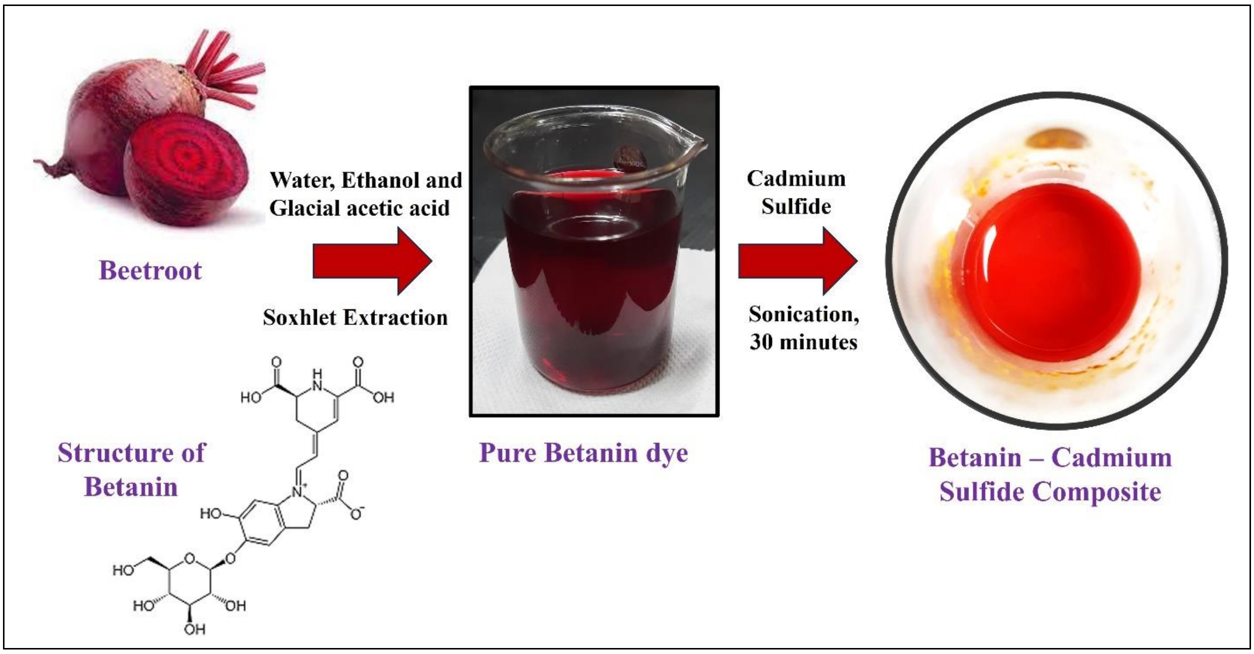
The vegetable red beet purchased from market. The Indium Tin Oxide (ITO) glass substrates, CdS, titanium tetra iso-propoxide, hydrochloric acid, graphite was purchased from Sigma-Aldrich. The ethylene glycol, ethanol, acetone, isopropyl alcohol and distilled water were used as solvents.

### 2.2 Synthesis of Titanium Dioxide Nanoparticles

The $TiO_2$ nanoparticles were synthesized incorporating the sol-gel method, which consisted of dissolving titanium tetra iso-propoxide (TTIP) in 10 ml of ethanol and 35 ml of deionized water to yield a 1M mixture. The resulting mixture was then stirred for ten minutes. After that, 1M of hydrochloric acid (HCl) was added to the mixture drop by drop while it was rapidly stirred for 50 minutes. A transparent yellowish solution was the final product. The resultant solution was baked at 110℃ for six hours for drying. The resulting powder was calcined at 300℃ (Haque *et al*. 2017).

### 2.3 Synthesis of Betanin Dye – Cadmium Sulfide (Bd-CdS) Composite

Sliced red beets were added to (80/20 v/v) ethanol and water mixture. An ultrasound-assisted approach was used to extract the Bd dye (Das *et al.* 2022). For forty-five minutes, the dye solution was continuously agitated in the absence of light. The supernatant was centrifuged for 30 minutes in order to separate the solid components. After that, 20 ml of extracted Bd dye and 20 ml of aqueous CdS were mixed for half an hour using a probe sonicator. The resulting orange color of Bd-CdS composite indicated that it was blended well with the dye mixture (Ahmadi *et al*. 2020).




**Scheme 1: The schematic representation of Bd-CdS (Betanin dye - Cadmium sulfide) composite**

### 2.4 Fabrication of Dye Sensitized Solar Cell

The ITO-coated glass substrates undergo cleaning by ultra-sonication for a ten-minute period in distilled water, acetone and isopropyl alcohol as solvents. Implementing a doctor-blade method, $TiO_2$ nanoparticles were deposited on ITO substrates and could function as an anode. The $TiO_2$-coated glass substrates were treated separately with the Bd dye and Bd-CdS composite solutions for a duration of 24 hours in dark, vacuum-sealed atmosphere. Graphite coated ITO can be placed to build a counter electrode. By introducing few drops of iodide electrolyte between the two electrodes and sandwiching the two ITO substrates together to obtain Bd based DSSCs (Saha *et al.* 2016).

## 3. RESULTS AND DISCUSSION

### 3.1 Morphology Analysis

The Fig. 1 shows the morphology of $TiO_2$ nanoparticles to confirm the deposition over ITO substrate which is essential for the photo-conversion into electricity mechanism. After being calcined at 300°C, $TiO_2$ sample showed well defined spherical shapes with an average diameter and length of 25 nm and 100 nm respectively (Abdullah *et al*. 2021). The appearance of aggregated spherical shape linked with each other confirms the rutile phase of $TiO_2$ which is the best choice for the fabrication of photo-anode for DSSCs agreed with reported literature (Li *et al.* 2009).

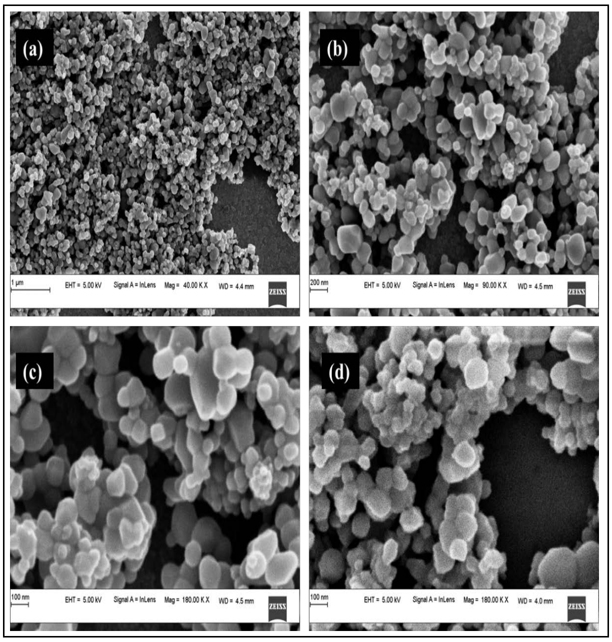



**Fig. 1: FESEM images of deposited $TiO_2$ Nanoparticles on ITO substrate at different magnifications (a-d)**

The aggregation happens throughout the fine spherical formation due to high thermal treatment (*Liu et al*. 2023). The calcined $TiO_2$ nanoparticles with slight variant spherical shapes can create voids between the domains (Kim *et al.* 2021). The void filled with dye allows $ITO/TiO_2$ electrode's larger absorption of light in the visible region while photo-anode treated with dye solution in DSSC mechanism.

## 3.2 FTIR Spectroscopy

Fourier Transform Infrared (FTIR) spectrum analysis provides the chemical bonding, stretching vibrations and functional groups present in the extracted Bd dye, CdS and Bd-CdS composite (Fig. 2).

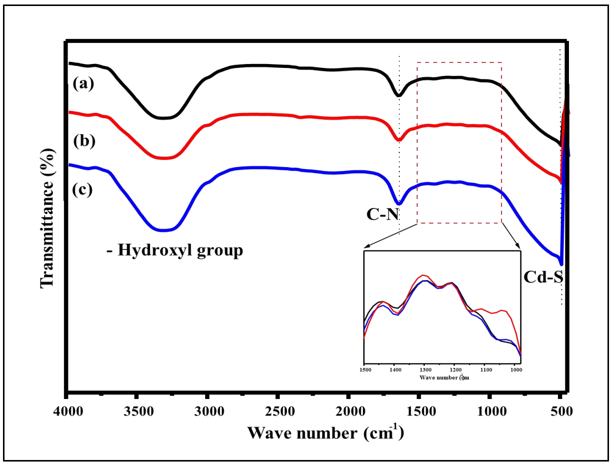



**Fig. 2: FTIR Spectra of (a) Betanin dye, (b) Cadmium sulfide, (c) Bd-CdS composite (inset figure shows magnified FTIR spectrum from 1500 to 975 cm$^{-1}$)**

The broad absorption peak appeared at around 3300 cm$^{-1}$ was assigned to stretching vibration of hydroxyl group (-OH) which is auxochrome attached to chromosphere that has an ability to alter wavelength or intensity absorption (Yameen *et al*. 2022). The aromatic C-H stretching vibration noted at around 1378 cm$^{-1}$ (Devadiga and Ahipa, 2020) and the absorption band at around 3248 cm$^{-1}$ were assigned to hydrogen bonded symmetric and asymmetric N-H stretching vibrations. The strong absorption peak at 1649 cm$^{-1}$ was ascribed to C-N stretching vibration. Absorption bands at 1118 and 1016 cm$^{-1}$ were assigned to C-H in plane bending vibrations (Seoudi *et al*. 2010).

## 3.3 UV-Visible Spectrum Analysis

Fig. 3 represents the UV Visible spectra and Tauc's plot of Bd, Cds and Bd-CdS composite in the wavelength range of 400 to 600 nm. The maximum absorbance at 485, 510 and 528 nm corresponding to Tauc's plot gives calculated energy gap of 2.16, 2.13 and 2.2 eV for Bd, CdS and Bd-CdS composite, respectively (Sengupta *et al.* 2015; Saeednia *et al.* 2019).

The broad absorption of Bd and slight absorption of CdS enables more light absorption when they undergo co-sensitization process and there is an increase in energy gap in Bd-CdS composite due to the presence of auxochrome hydroxyl attached to chromophore group (Rached *et al.* 2023; Shaikh *et al.* 2024). The investigation on light absorption and its associated factors indicates that broad light absorption and precise energy level alignment alteration can render them suitable for use as dye sensitizer materials in DSSCs.

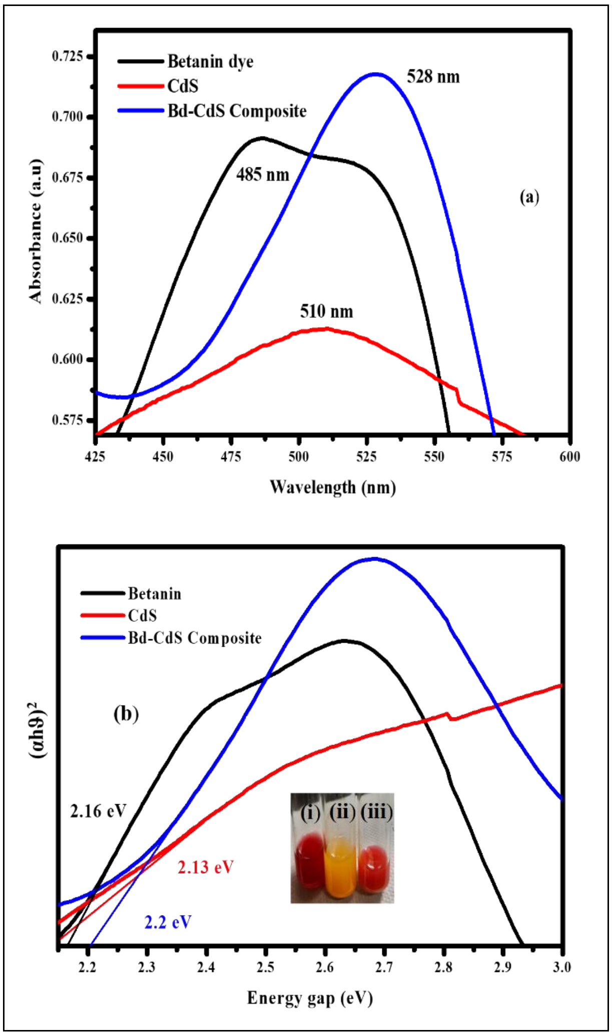



**Fig. 3: (a) UV-Visible spectra and (b) Tauc's plot of Betanin dye (Bd), Cadmium sulfide (CdS) and Bd-CdS composite (inset figure shows colour appearance of samples)**

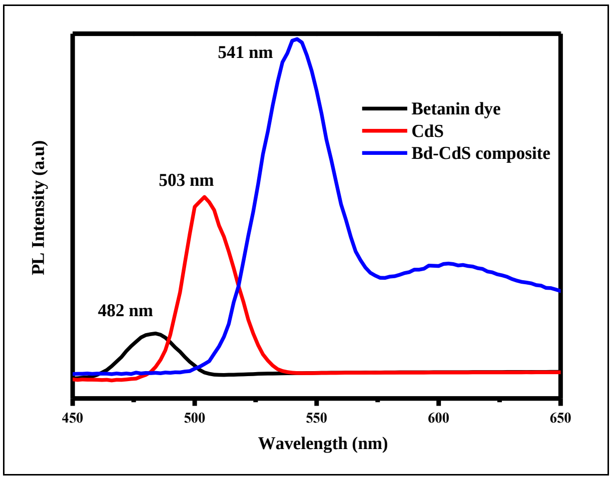



**Fig. 4: a) PL spectra of Betanin dye (Bd), Cadmium Sulfide (CdS) and Bd-CdS composite**

### 3.4 Photoluminescence (PL) Spectrum Analysis

Fig. 4 demonstrates the PL spectrum of Bd, CdS and its Bd-CdS composite which exhibits maximum emission wavelength at 482, 503 and 541nm, respectively and shows their florescent nature (Arbuj *et al*. 2013).

Although the Bd solution has a wider absorption spectrum, it emits light at a lower wavelength. Inclusion of CdS in Bd causes an obvious increase in photon emission in broad PL intensity, which may aid in more significant charge-carrier recombination in the Bd-CdS composite (Lim *et al*. 2015). The energy level alignment of Bd displays good light absorption property whereas CdS has good emissive property (Rempel *et al*. 2012). The CdS material can enhance photo-conductivity when intermixed with Bd dye as reported (Susha *et al*. 2018). The Bd-CdS composite was good in light absorption as well as emission that limits the performance of DSSCs through recombination of charge carriers as evident from PL spectrum analysis.

### 3.5 Photovoltaic Performance of Betanin Dye (Bd) and Bd-CdS Composite

Fig. 5 (a) represents J-V characteristics of Bd and Bd-CdS displaying photovoltaic behavior of dye and its composite. From the optical analysis it is evident that Bd and Bd-CdS composite have sufficient optical properties for use as dye sensitizers in DSSCs. The light absorbed from Bd dye and its composite can excite the electrons into excited state and enable charge transport through iodide electrolyte (Labat *et al*. 2012). The charges are collected at the counter electrodes. The electrons transport through titanium dioxide and graphite coated ITO substrate electrodes as shown in Fig. 5 (c). Due to this device aligned with well blended energy levels, the photon and electron interaction improves photo-current in Bd-CdS composite (Smrithi *et al*. 2022). The Bd-CdS composite based DSSC displayed best performance due to its strong charge transport mechanism, which enhances photo-current density in it. The PCE of DSSC is improved when gold acts as a counter electrode from 1% to 2% (Barichello *et al*. 2022).

The PCE of DSSC calculated by using the following equations.

$$\mathrm{FF} = \frac{\mathrm{Jm\ Vm}}{\mathrm{Jsc\ Voc}} \qquad \text{............. (1)}$$

$$\eta = \frac{\mathrm{Jsc\ Voc\ FF}}{\mathrm{Pin}} \qquad \text{............. (2)}$$

where, $J_{sc}$ - Short circuit current density (mA/cm$^2$), $V_{oc}$ - Open-circuit voltage (V), FF - Fill factor (%), $J_m$ - Current density at maximum power point, $V_m$ - Voltage at maximum power point, $P_{in}$ - Incident Power (mW/cm$^2$), η - Power Conversion Efficiency (%).

**Table 1. Comparison of solar parameters of natural DSSCs**

| Material | Open Circuit Voltage (V) | Short Circuit Current density (mA/cm$^2$) | Fill Factor (%) | Power Conversion Efficiency (η) (%) | Reference |
|---|---|---|---|---|---|
| Grape | 0.427 | 1.81 | 42.69 | 0.33 | (Zhang *et al*. 2022) |
| *Canna indica L*540. | 0.54 | 0.82 | 58.71 | 0.26 | (Luo *et al*. 2009) |
| Cowberry | 0.556 | 0.40 | 53.95 | 0.12 | (Luo *et al*. 2009) |
| Begonia | 0.537 | 0.63 | 70.94 | 0.24 | (Zhou *et al*. 2011) |
| Rhododendron arboretum zeylanicum | 0.402 | 1.15 | 62.72 | 0.29 | (Hao *et al*. 2006) |
| Black rice | 0.551 | 1.14 | 50.94 | 0.32 | (Fernando *et al*. 2008) |
| $TiO_2$QD with MWCNT/$TiO_2$ | 0.539 | 0.095 | 37 | 0.331 | (Onyemowo *et al*. 2024) |
| Sandoricum koetjape | 0.04905 | 0.04233 | 46 | 0.0181 | (Sabarikirishwaran *et al*. 2023) |
| **Betanin dye** | **0.5641** | **0.595** | **69.8** | **0.234** | **This Work** |
| **Bd-CdS Composite** | **0.565** | **0.8965** | **72.6** | **0.367** | |

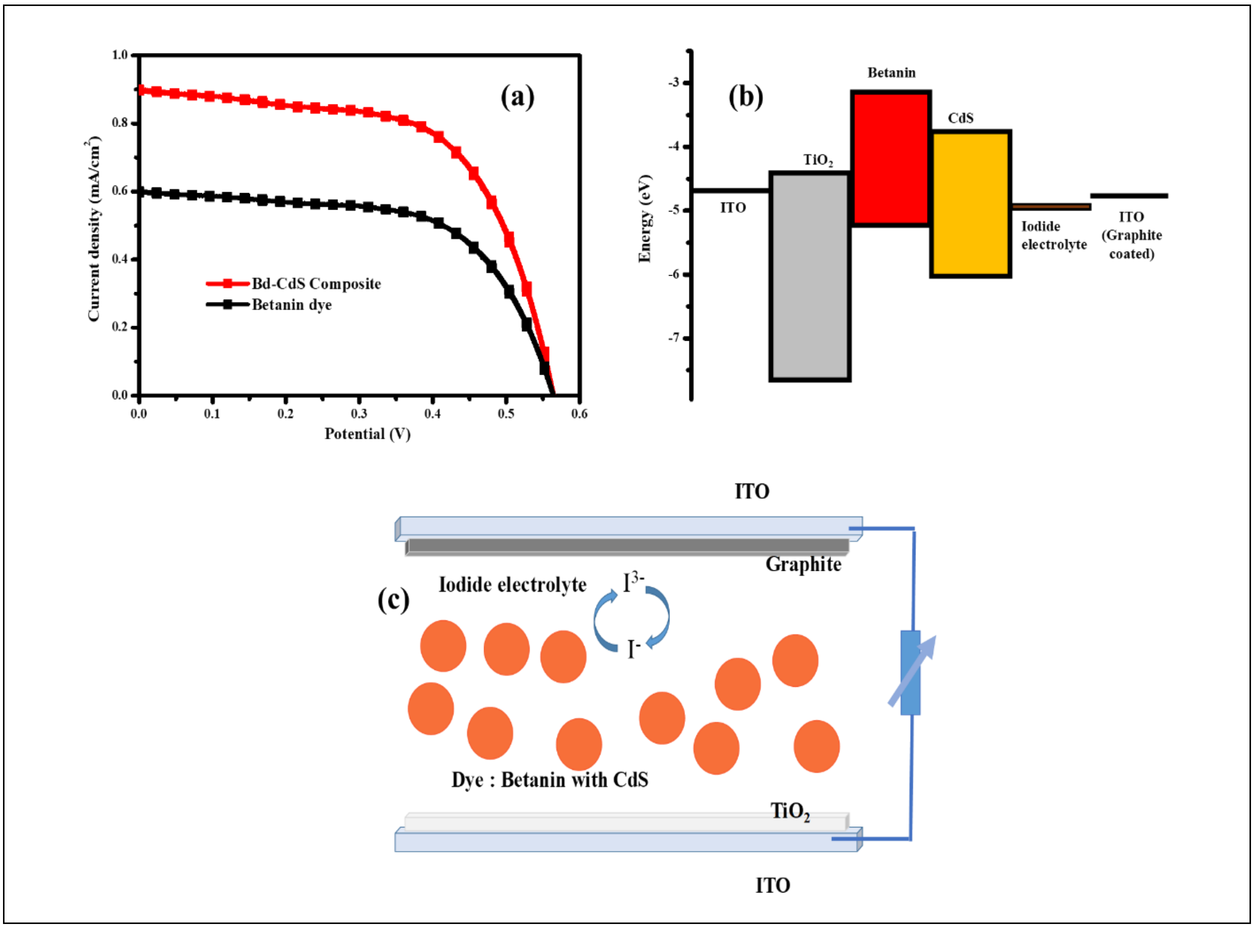



**Fig. 5: (a) J-V characteristics, (b) energy level alignment, (c) Mechanism of fabricated DSSCs based on Bd and Bd-CdS composite**

The performance of DSSC based on Bd and its composite and comparison with previously reported results that grape (Zhang *et al*. 2022), Canna indica L540, Cowberry (Luo *et al.* 2009), Begonia (Zhou *et al*. 2011), Rhododendron arboretum zeylanicum (Hao *et al.* 2006), Black rice (Fernando *et al.* 2008) and also in recent studies of $TiO_2$ QDs through co-sensitization (Onyemowo *et al*. 2024) and Sandoricum koetjape (Sabarikirishwaran *et al.* 2023) are shown in Table 1.

In our work, $TiO_2$, iodide, Bd and its composite and graphite can act as semiconductor, electrolyte, dye sensitizer and counter electrode, respectively. As per optical analysis and above-mentioned results, Bd with its composite enhances PCE from 0.234% to 0.367% by the process of co-sensitization of CdS with Bd dye. Given its economic and environmental advantages over other materials, Bd-CdS composite emerges as a promising candidate for future energy conversion applications. By employing it along with suitable photoactive materials and using a mixture of dyes through the co-sensitization process, its effectiveness can be enhanced (Srivastava *et al*. 2022).

## 4. CONCLUSION

Along with $TiO_2$ nanoparticles synthesized from sol-gel method, Bd dye and Bd-CdS composite were prepared from ultrasound assisted approach and probe sonication method, respectively. The FTIR spectrum revealed the presence of Cd-S, C-N, and -OH interactions, attributed at near 500 cm$^{-1}$, 1633 cm$^{-1}$ and around 3300 cm$^{-1}$, respectively. Maximum absorbance evident from UV-visible spectrum at wavelengths of 485, 510, and 528 nm and Tauc's plot shows energy gap values of 2.16, 2.13, and 2.2 eV for Bd, CdS and Bd-CdS composite, respectively. The PL spectrum intensity confirmed energy level alignment and charge carrier recombination that limits the PCE is 0.234% and 0.367%, respectively. According to this optical study, co-sensitization with mixture of dyes is a promising way to improve natural dyes with compatible materials, which can further enhance the PCE of DSSCs in the future renewable energy technology.

## AUTHOR CONTRIBUTION STATEMENT

Research Plan, Methodology and Conceptualization: D. S. Suresh, H. Devendrappa; Data

collection and analysis: D. S. Suresh, Sapna Sharanappa, S. P. Vijaykumar, Abdullah Ba Shbil; Experiment performed and preparation of original draft: D. S. Suresh; Paper review and editing: Ganesha H.; Paper Review, Editing and Supervision: H. Devendrappa;

## CONFLICTS OF INTEREST

This is certified that there is no conflict of interest with results present in the manuscript.

## ACKNOWLEDGMENT

The authors would acknowledge Professor Dr. Balachandra G. Hegde, Rani Channamma University, Belagavi for the fabrication of DSSC facility, The PURSE Laboratory, Mangalore University for their FESEM facility and also, I would like to thank Ministry of Tribal Affairs, Government of India for availing fellowship through National Fellowship for ST students (NFST) scheme.

## FUNDING

This research did receive grant from National Fellowship for ST students (NFST) scheme.

## COPYRIGHT



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