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Synthesis and Characterization of Hybrid Nanostructures Based Natural Dye Sensitized Solar Cell



Abstract: - Dye-sensitized solar cells (DSSCs) are fabricated with perovskite-type Lithium barium doped cerium oxide (LiBaCeO₄) as the photoelectrode materials. The nano particles are prepared by solution combustion technique using glycine as a fuel. To optimize the photoelectric activities of electrode materials hybrid triplex nanoparticles were synthesized. The obtained nanoparticles are characterized using scanning electron microscopy (SEM) for morphological study, UV-visible spectroscopy for UV absorption range, X-ray diffraction (XRD) study to know the formation of a crystalline nature, The FTIR spectroscopy study identifies the functional groups. The triplex LiBaCeO₄ nanoparticles as electrode material for DSSCs performances and photo electric properties are investigated by using surface photo voltage spectroscopy and The SEM and XRD results shows that nanoparticles are uniformly distributed and the obtained particles size lies in the range 37- 59 nm. The UV-visible spectrum exhibited a well-defined absorption steep peak at 220 nm and the absorption peak also canbe observed in the visible region at 515 nm after the particles are adsorbed with dye molecules. The band gap energy is found to be 3.29 eV near short wave UV-visible region. The I-V results of LiBaCeO₄ nano particles with pomegranate dye sensitized solar cell gives short circuit current density of $J_{sc} = 2\text{mA cm}^{-2}$ and open circuit voltage of $V_{oc} = 0.67\text{ V}$ and corresponding solar to electrical energy conversion efficiency of 5.4% is obtained.

Keywords: Barium nitrate, fill factor, Dye sensitized solar cell, Lithium nitrate, Efficiency.

I. INTRODUCTION

Dye-sensitized solar cells (DSSCs) have paying attention due to their prospective in low cost and simple preparation procedure compare to the conventional silicon cells [1]. Dye-sensitized solar cells (DSSCs) are fabricated by nanoparticles of ZnO, TiO₂, CdSe and PbSe as electrode materials because of a large internal surface area for dye adsorption [2]. Currently, researchers are fabricating ZnO-based DSSCs by manipulating the constructions of photo anodes to allow quick electron transport, wonderful light-harvesting and excessive dye-loading [3]. Thereare few challenges to upgrade materials photovoltaic properties. First, the recombination of photo-generated electrons seems to occur more frequently in electrodes material, which deteriorates the cell performance [5]. Second, conventionally dye-loaded photo anode of the material is functional under visible light. It is important to have dyes can absorb not only a visible portion of light it should also absorb greater part of the electromagnetic spectrum consists of ultraviolet (UV) and near infrared (NIR) light [6]. It means 50% of solar irradiation in the UV and NIR regions is not utilized by single nano material based DSSCs. So the effective way to improve these drawbacks is by using hybrid doping of materials to alter their electrical and optical properties [7]. In current years, rare-earth (RE) doped nanoparticles have attracted researchers because of their intra 4f transition optical characteristics. Doping of RE ions increases the conversion of NIR and UV radiation to visible emission also increases electron transport [10]. Gratzel et.al has reported a remarkable energy conversion efficiency of 7.9% for mesoporous TiO₂, DSSCs are being investigated. The progress of the promising photovoltaic device architectures has been well made by considering several factors, for example, (i) the synthesis of light capturing apertures (ii) the use redox couples in an appropriate medium, and (iii) the preparation methods used for synthesis of nano materials and fabrication of thin film electrodes [8]. For the most up to date one, some semiconductors have been applied to built new structures and avoid the oxidation of dye, such as WO₃, TiO₃, ZnO, In₂O₃, Nb₂O₅, SnO₂, SrTiO₃ and Zn₂SnO₄, and so forth [9]. The energy band levels of the semiconductor must match with those of dye molecules to improve the separation of photo generated charges and minimize their recombination in the fabrication of DSSCs, [10]. In addition, the surface morphology, particle sizes and shapes, electro-optical energy band gap of the material, doping concentration, porosity, UV absorption and film thickness of semiconductors must also be well thought-out and optimized. The Perovskite structured LiBaCeO₄ is an n-type semiconducting material with a band gap of 3.1eV [11]. It has been widely investigated on its dielectric, thermal, and photocatalytic properties as an important ceramic

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material. In this work, we employed the solution combustion methods to prepare triplex LiBaCeO_4 nano particles and then assemble them as the working photo electrode in DSSC. The result shows the good photovoltaic performance of the pomegranate dye sensitized LiBaCeO_4 nano particles electrode material. It is probably due to the highest loading amount of dye and best interaction between the semiconductor and the dye used.

II. IMPLEMENTATION AND EXPERIMENTATION

2.1. Materials and Instruments used

The chemicals used in this study are cerium nitrate, lithium nitrate, barium nitrate and double distilled water was used in the experiment. The synthesized nanoparticles were characterized using the scanning electron microscope SEM using SEM Model S-3400, Fourier transform Infrared (FT-IR) using the Model FTIR-4100. UV-visible spectroscopy by using Beckman DU640. The X-ray diffraction was done using TD-3600 X-ray diffractometer. The light source the Xenon lamp (CHF XQ500W), Trusttech, China) was used and the radiation Meter (FZ-A).

2.2. Synthesis and Characterization of LiBaCeO_4 Nanoparticles

Solution combustion technique mainly uses raw materials such as salts like nitrates, carbonates and metal sulphates, oxidizing agents and carbonates reducing re-agents such as urea, glycine and sucrose. This process uses small aggregation of salts and synthesis nano particles can be carried over in small scale and evaluate the results. The experimental procedure starts with individual sample preparation, cerium nitrate of 2.17 g, lithium nitrate of 0.172 g and barium nitrate of 0.65 g. Combustion reluctance property have a great makeover while selecting fuel and an important role in the synthesis of nano particles [12]. The glycine is selected as reducing re-agent of 0.7 g. The cerium nitrate, lithium nitrate and barium nitrate is independently stirred, heated and combined together in a separate glass beaker and again stirred for 45-60 min. After a few interval of time glycine is added. The solution is added with 4-5 drops of NaOH solution to increase the pH level and tetracycline is added to prevent agglomeration of nanoparticle in constant phase [13]. The sample solution is continuously heated till gel form then immediate combustion takes place then all liquid is evaporated and only ash form is obtained. The residue of powder is collected and then calcinated at 450°C for 3 hours is as shown in fig 1.

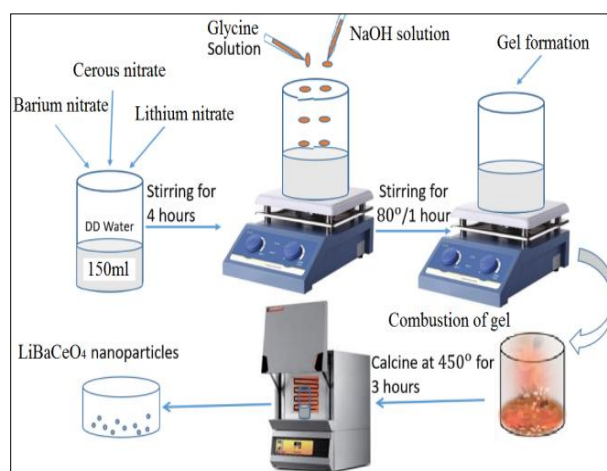


Fig. 1: Schematic preparation of Lithium barium doped cerium oxide (LiBaCeO_4) nanoparticles.

2.3 Construction of Dye-Sensitized Solar Cell

The Dye-sensitized solar cell is a thin film solar cell which acts as semiconductor between anode and electrolyte. The construction of the cell starts with fabrication method with a pair of Indium tin oxide glass which is shown in Fig. 2.1. a). The ITO glass was rinsed it with alcohol and sterilizing it with double distilled water. The conducting side of the glass was checked with the help of multimeter. Then the conducting side of the sterilized ITO glass was coated with LiBaCeO_4 nano paste. The paste is made up of adding LiBaCeO_4 nano with detergent (Triton X-100, Aldrich) in de ionized water of 2 ml and 2-3 drops of vinegar and stirred well in glass plate to create a thick paste so that it anneals together. The Indium tin oxide glass coating on conducting side is as shown in Fig 2.1b). The dye is used to absorb photon molecule and get excited into higher energy state, any dye having anthocyanin component is used as dye [14]. In this work pomegranate juice extract is used as dye and it contains anthocyanin. After dipping ITO glass in dyes as shown in fig.2 c). The dye injects the electrons from valence band to conduction band of LiBaCeO_4 electrode. The Carbon coating is applied on another Indium tin oxide glass conducting side which acts as counter electrode as shown in Fig. 2.1d).

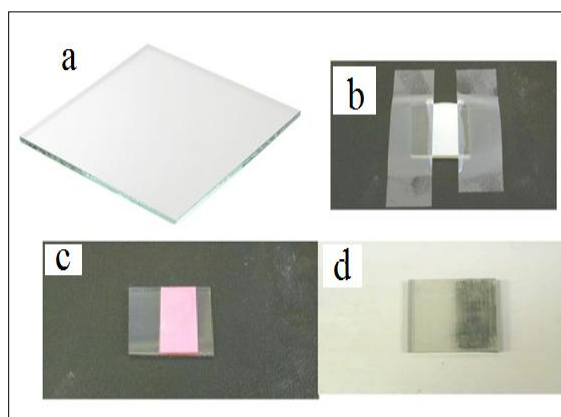


Fig. 2.1: a) Indium tin oxide glass b) The LiBaCeO_4 nano paste coated ITO glass c) After dipping ITO glass in dye d) Carbon coating of counter electrode.

These pair of glass is faced with each other as shown in Fig 2.2.a) The potassium tri-iodine is added in between the glasses as an electrolyte as shown in Fig2.2 b) to gain electron loss by dye molecules during excitation of electron. The electron flows through the outer electrical circuit at the counter electrode the electron is transferred to a hole conducting medium and exited. The charge separation occurs at the surfaces between the dye, semiconductor and electrolyte. These two ITO glasses are coupled together using binding clips and the edges are connected to Linear sweep voltammetry to get Current voltage characteristics, from which efficiency of the solar cell is calculated [20]. A fully mounted dye sensitized solar cell constructed using fabrication method is as shown in Fig. 2.2c).

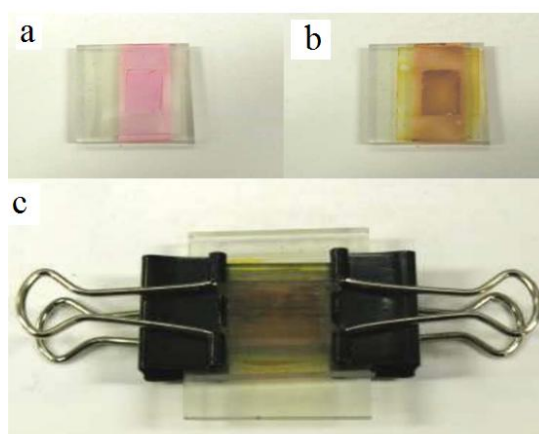


Fig. 2.2 a) ITO glasses facing each other b) After adding electrolyte between ITO glasses c) The fabricated dye sensitized solar cell

III. CHARACTERIZATION AND PHOTOELECTRIC MEASUREMENTS

The X-ray diffraction (XRD) patterns were recorded on a DX-2500 diffractometer (Fangyuan, Dandong) with Cu K radiation (nm) in the 2θ range 10 to 70° . The surface Morphology of the film and thickness were measured by scanning electron microscope (SEM, JSM5600LV, JEOL, Japan). The FTIR spectral studies were recorded within the wave number vary from $4000 - 400 \text{ cm}^{-1}$ employing JASCO 4100 spectrometer, Japan. The optical properties of the synthesized nanoparticles were determined with shimadzu-1800 spectrometer in the spectral range of 200 to 800 nm. The Current-voltage characteristics of fabricated DSSC was measured by an electrochemical workstation (CHI660B, China). For the light source the Xenon lamp (CHF XQ500W), Trusttech, China) was used and the radiation Meter (FZ-A, Beijing Normal University, China) was used to measure the incident light intensity.

IV. RESULTS AND DISCUSSION

4.1. Scanning Electron Microscopy (SEM)

The SEM image shows in fig.4.1. The nano particle has a narrow distribution and homogenous in its structural size it confirms pure nano particle has been examined in the spectroscopy. The particle examined SEM image indicates uniformly distributed nanoparticles are all in spherical shape, as the diameter of the sample particles decreases the number of pores decreases. In fewer areas sample contain only few pores and larger pores are observed in less compared to uniformly distributed sample [15]. The results obtained from Fig.4.1 explains that its nano particle size ranging from size 30-60 nm size range particles surface will behave as good absorption medium by

comparing UV rays at the range of 1.46eV [16]. The sample examination reveals it has regular size and uniformly distribution indicates, it has long holding time of charges when applied to p-n junction applications. Absence of agglomeration in the particle is clear shows the sample is less contaminated.

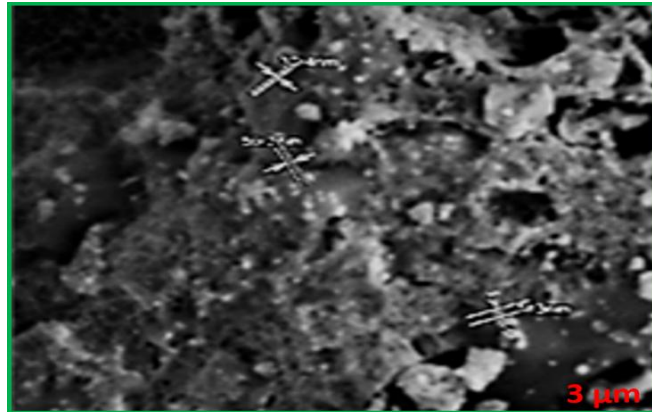


Fig. 4.1: SEM image of LiBaCeO₄ nanoparticles.

4.2. X-Ray Diffraction (XRD)

XRD analysis is mainly applied in identification of the following geometry Phase Identification, Lattice parameter, Phase fraction analysis, Diffraction pattern, translational symmetry and Peak intensities [23]. The average particle size, interplanar spacing between atom is calculated using Debye-Scherrer formula and Brag's law. The LiBaCeO₄ diffraction peaks indices corresponds to 2θ [Figure 2] with the most highest positioning indicates the prominent peak, these results correlated to hkl indices show signs of pure with structure of nano particles samples. The highest peak in the X-ray diffraction graph shows that nanoparticles are amorphous in nature and fermi level is low. The nano particles in the highest prominent peak indicate its high performance in wide range of temperatures. The deformation per unit length in the graph having the dominant peak at 2θ , nano particles residual stress can be oriented from the samples.

The size of the crystal is D (nm) which is calculated using Debye- Scherrer Formula [17].

$$D = KY/\beta \cos\theta \quad (1)$$

D is the size of the crystal in nm, K is the Debye constant that is 0.94, γ is the wavelength of the x-ray, β is full width at half maximum(FWHM) and θ is the diffraction angle. The average crystalline size was calculated from X-ray line broadening using Debye scherrer formula and the value found is between 5.3 nm to 26.66 nm. The peaks at $15^\circ(111)$, $29^\circ(110)$, $38^\circ(200)$, $45^\circ(211)$, $50^\circ(220)$, $58^\circ(222)$, indicates well crystalline nature of synthesized particles and The lattice parameter values founded from the XRD data are almost similar compare to the standard JCPDS card data with the JCPDS card numbers, Li₂O-JCPDS#00-36-1451, BaO JCPDS card no. 26-0178 , CeO₂(JCPDS Card # 34-0394) of CeO₂ as shown in the Fig. 4.2.

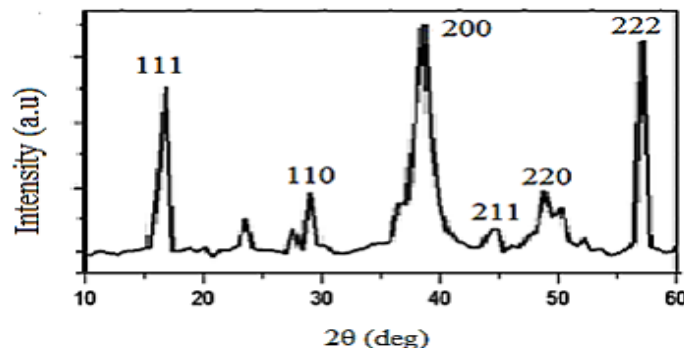


Fig.4.2: XRD pattern of LiBaCeO₄nanoparticles.

4.3. UV-Visible Spectroscopy

The UV-vis absorption spectrum for the LiBaCeO₄ particle and dye-sensitized nano particles at room temperature is as shown in fig.4.3. The pure LiBaCeO₄exhibit intense absorption band with a steep edge at ~ 200 nm (corresponding to 3.2eV in energy [19]. The optical absorption in the wavelength region shorter than 400 nm is mainly attributed to the electron transition from the top of the valence band to the bottom of the conduction band

(band-to-band transition). After the LiBaCeO_4 particles are adsorbed with dye molecules, a new absorption band at the peak of 515 nm emerges in the visible region, which is caused by the intralig and transitions ($\pi-\pi^*$) of dye molecules. The amount of adsorbed dye determines the number of photo excited electrons by visible light in the dye-sensitized LiBaCeO_4 particle, thus we examined the amount of adsorbed dye by using UV-vis spectroscopy, where the pomegranate dye was desorbed using a 10–4 M aqueous NaOH solution from the surfaces of the three LiBaCeO_4 films. Figure 5(a) illustrates the UV-vis absorption spectra of LiBaCeO_4 NPs and pomegranate dye-sensitized NPs. Figure 5(b) shows the optical absorption spectra of LiBaCeO_4 NPs and NPs containing the desorbed dye samples in the wavelength range from 200 to 800 nm. The absorption band at ~ 500 nm results from the light absorption of dye. Obviously, the order of the amount of the adsorbed dye of LiBaCeO_4 NPs is greater than the pristine NPs. This is in consistent with that of the photocurrent density. In general, the more the amount of dye adsorption, the more the light harvest, thus giving rise to the larger photocurrent density [18]. Therefore, the difference in the photocurrent densities for the three DSSCs is associated with the different amount of adsorbed dye on the surface of the NPs.

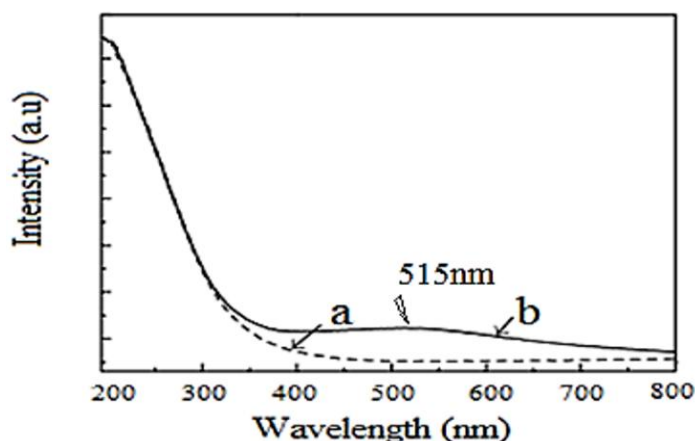


Fig.4.3: UV-Vis absorption spectrum of a) LiBaCeO_4 NPs and b) LiBaCeO_4 NPs with dye.

4.4. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR is used to identify the possible functional groups are obtained after the synthesis of nano particles is as shown in Fig. 4.4. The bands centered at 1521 to 2359 cm^{-1} and the shoulder at 1500 cm^{-1} have the stretch vibration, their functional group is C=C and C-H and intensity is medium. The variation in spectrum in all wavelengths of sample LiBaCeO_4 indicates sample is less contaminated around main peak spectrum at 2992 cm^{-1} . The peaks at 3400 and 3000 cm^{-1} have very strong and broad intensity which indicates the presence of Sp^2 carbon-hydrogen breakdown [20].

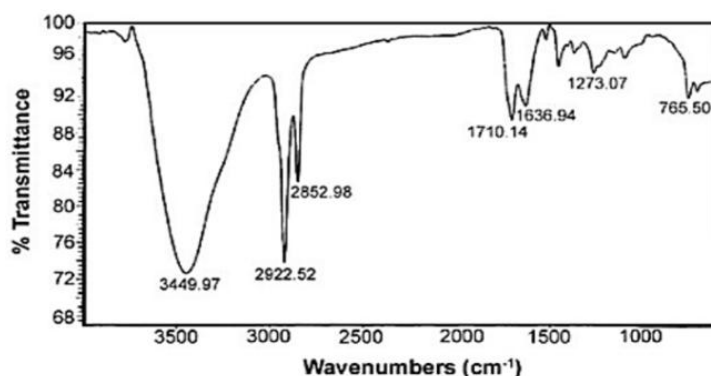


Fig. 4.4: FTIR spectra of LiBaCeO_4 NPs

4.5. Photovoltaic Performance of Cells

The performance evaluation of LiBaCeO_4 dye-sensitized electrodes were used as the working electrodes in sandwich solar cell set up is as shown in Fig. 4.5.1. The I-V characteristics for LiBaCeO_4 was obtained by 0.6 cm^2 NPs sample was kept at open cell under the light irradiation of 100 (measured by Radiation Meter). The sample exhibits a typical behavior of the I-V curve for LiBaCeO_4 NPs sample. The obtained characteristics curve of photo current versus voltage characteristics of LiBaCeO_4 nanoparticles is as shown in Fig 4.5.2.

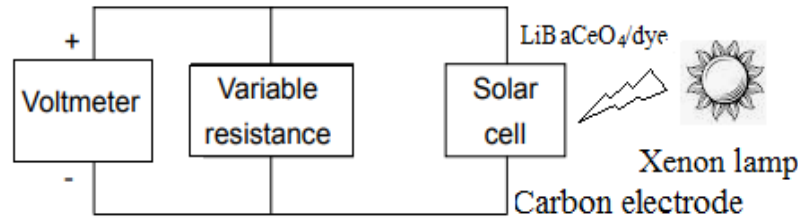


Fig 4.5.1: Performance evaluation set up of dye sensitized solar cell.

The open-circuit voltage (V_{oc}) and the short-circuit current density (J_{sc}) for LiBaCeO₄ are 0.67V and 2mA (cm⁻²) respectively. The fill factor ff , which is defined as $P_{max} / (J_{sc} \times V_{oc})$, where P_{max} is maximum power output it is the product of ($J_{max} \times V_{max}$) for the LiBaCeO₄ sample and it is calculated to be 49%.

$$FF = (J_{max} \times V_{max}) / (J_{sc} \times V_{oc}) \quad (2)$$

The energy conversion efficiency under white-light irradiation can be obtained from the following equation [21]:

$$\eta(\%) = \frac{J_{sc} [mA cm^{-2}] \times V_{oc} (V) \times ff}{I_0 [mW cm^{-2}]} \times 100\% \quad (3)$$

where, I_0 is incident light intensity. The energy conversion efficiency for the LiBaCeO₄ sample is 5.4%. The LiBaCeO₄ cell properties can also be varied by different amounts of adsorbed dye on the electrode. The amounts of absorbed dye can be evaluated using optical absorption measurements. Measured Current -Voltage characteristics of a dye sensitized solar cell is $V_{oc} = 0.67V$ and short circuit current density $J_{sc} = 2 \text{ mA (cm}^{-2}\text{)}$ and efficiency is 5.4%.

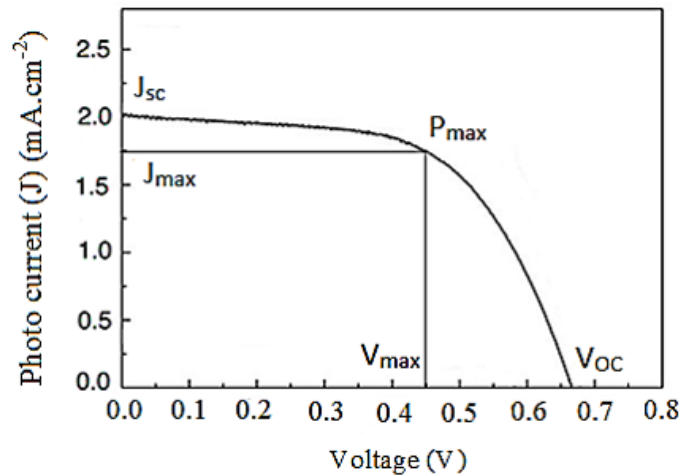


Fig.4.5.2: Characteristics curve of LiBaCeO₄ nanoparticles

V. CONCLUSION

The hybrid nano particles were synthesis using the solution combustion technique. The XRD and SEM results of hybrid NPs is having regular dispersion and grain size ranging from 30-60 nm. UV-Vis spectrum shows absorption of light in near UV regions (200nm) and after injecting the dye extends up to both UV and visible range at 515nm. The optical absorption in the wavelength region shorter than 400 nm is mainly attributed to the electron transition from the top of the valence band to the bottom of the conduction band (band-to-band transition). In the IV characteristics the efficiency of DSSC was 5.4% for the obtained V_{oc} and J_{sc} of 0.67V and 2mA (cm⁻²) respectively. The nano particle prepared from LiBaCeO₄ shows quite good fefficiency and can be applied for solar cell application.

About the Corresponding Author:

Dr. Gayitri H M, Ph.D. Associate Professor, Dept. of Electronics and Communication Engineering, JSS science and Technology University. SJCE, Mysuru, Dr. Gayitri's work focuses on nanomaterials synthesis, fabrication of polymer nanocomposites for various electronic application. She received her Ph.D from University of Mysore and has published more than 20 papers in top tier journals. Currently she is exploring new approaches to energy storage applications.

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