Performance of N3 Sensitized Titania Solar Cell under Artificial Light Ambience

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The indoor utilization of emerging photovoltaics is promising but power conversion efficiency (PCE) under indoor lighting is still challenging. Dye sensitized solar cell (DSSC) is one of the most excellent options to harvest and convert the artificial indoor light into electricity. In present study, we investigated the indoor performance of N3 sensitized Titania DSSCs under artificial lighting. We observed a substantial improvement in PEC of solar cell.

Under the indoor illumination of 33 mW/cm², the maximum PCE obtained was 28.21 %. Our findings suggests that the obtained high efficiency N3 sensitized Titania DSSCs under artificial lighting have a good potential application for low power electronic devices for their autonomous operation. Graphical abstract shows the result with glowed LED using three series connected DSSCs.

Keywords: Titania; N3 dye; Dye sensitized solar cell; Artificial light; High efficient; Solar cell

Received 24 November 2018, **Accepted** 2 January 2019

DOI: 10.30919/eseec220

Introduction

Solar cells are the prime candidate for harvesting available indoor light, even if they have been initially developed for outdoor lighting. During the last decade, a large number of photovoltaic technologies such as c-Si, a-Si, polymer PV, CdTe, GaAs, CIGS etc. have been investigated under the full outdoor conditions.¹⁻⁷ DSSC has much attention due to its low cost, easy and inexpensive fabrication, transparency, high photoconversion efficiency and long term stability.⁸ Many researchers examine to improve the PEC by modifying photoanodes, counter electrode, dye and electrolyte. Among them the light harvesting capacity nature of solar cell depends upon photoanode.⁹ The performance of these solar cell technologies depends on the variation in spectral, reflection, temperature, irradiance and power of the modules throughout the day and season.⁴ Therefore it is also important to know how the performance of these solar cell technologies varies with change in operating conditions including indoors than total energy produced outdoor by panels over the course of a time period.^{10,11} Furthermore, these technologies are based on inorganic materials which require high fabrication costs and large amount of energy. Moreover, they often use toxic and scarce materials.¹² The third generation solar cells such as dye sensitized solar cells (DSSCs) have attracted considerable interest during the past two decades due to their potential as a low cost and easily processable alternative to traditional solid state solar cells.¹³ These

solar cells can be used for outdoor and indoor light harvesting and conversion of light into electricity. They showed an outstanding performance under indoor conditions with an artificial light source in comparison with other solar cell technologies.¹⁴ The unique properties of DSSCs make them the best alternative to wired and battery energy sources as they are capable of maintaining a high photovoltage even in diffuse light conditions.¹⁵⁻¹⁷ It has been reported that the DSSCs have larger power conversion efficiency compared to silicon under artificial illumination such as compact fluorescent lamps (CFL) and light emitting diodes (LEDs).¹⁸⁻²¹ Recently, the power conversion efficiency (PCE) with 28.9 % has been reported for DSSC under ambient lighting using dual sensitizer.²²

During functioning of a DSSC, when light is incident on cell, dye molecule absorbs an incident photon and gets excited to an excited state. The excited dye molecule then gets injected to the conduction band of the semiconductor. Then, by reducing redox species of I the oxidized dye molecule is regenerated and returns the dye to its ground state which allows to absorb another photon. Nanostructure semiconductor photoelectrode has large surface area for dye adsorption which enhances the solar cell performance.²³

Here, we show the out performance of N3 sensitized titania DSSC under the indoor lighting. The indoor light harvesting is a nascent technology and hence no standard light illuminations source or testing procedure has yet been established. Our results highlight the PEC value of 28.2 % using N3 dye with 4mA/cm² of anodic spike i.e I_a. Therefore this study is useful for understanding the possible outcomes of solar cell under artificial lightning. Typically, the light intensity under indoor lighting is less and the optical spectrum is completely different compared to outdoor conditions. The optical spectrum for indoor

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lighting is primarily restricted in a narrow spectral band in the visible region. For indoor light, we have compared xenon lamp illumination in solar simulator and commercial white LED lamp illumination having intensities 50 mW/cm^2 and 33 mW/cm^2 respectively. Light intensities affect the photovoltaic performance outcomes from its influence on the cell parameter. Therefore it is important to examine the solar cell under several light illumination condition to understand the possible outcomes. Further the theoretical calculation such as electron lifetime, diffusion length, charge collection efficiency, effective electron diffusion co-efficient and effective electron diffusion length which is useful to study the electron transport properties of solar cell.

Experimental Section

Fabrication of Photoelectrode

For fabrication of TiO_2 paste, commercial 0.5 g TiO_2 nano-particles (P25 degussa, Nanoshel LLC, US) were mixed with 2 ml ethanol (Changshu, China) using homogenizer in small glass vessel with subsequent addition of 0.5 g ethyl cellulose (SDFCL, India), 0.25 ml terpineol anhydrous (KPS Ltd., India) and 0.25 ml acetyl acetone (HPCL, India). Gummy slurry form after 30 min and this slurry sonicated using probe sonicator (QSonica-Q700, USA) for 2 hr till no particulates are seen in the paste. Further, in preparation photoelectrode, the TiO_2 films were deposited onto conducting fluorine doped tin oxide (FTO- Sigma Aldrich, USA) having sheet resistance of $15\text{--}18 \Omega/\text{cm}$ glass substrate by modified doctor blade. Before fabricating TiO_2 film, FTO were cleaned in ultrasonic bath by using double distilled water, acetone and ethanol subsequently for 10 min . The FTO substrates were air dried at room temperature. After drying, the edges of the substrate were covered with $100 \mu\text{m}$ transparent plastic paper to prepare the film. Then to form TiO_2 film, one drop of the prepared paste was placed at the center of substrate and the edge of a microscopic slide was used to spread the paste over the substrate. The films then sintered at 450°C for 1 hr . After sintering, thickness of TiO_2 film was observed to be $15 \mu\text{m}$ from SEM cross section. The films were further dipped in 0.1 M thioglycolic acid (Thomas Baker (Chemicals) Pvt. Ltd, India) using dip coating method for 60 sec to etch the TiO_2 film. Before solar cell testing the films were immersed in standard ruthenium based red 'cis-bis(isothiocyanato)bis(2,2'-bipyridyl-4,4'-dicarboxylato)ruthenium(II)' N3 dye (Sigma Aldrich, USA) with the concentration of 0.03 mM solution in ethanol for 90 min for sensitization. X-ray diffractometry (XRD) measurements were performed to further evaluate the structural properties of the TiO_2 film. XRD pattern showed that prepared film were polycrystalline in nature. Supporting information, Fig. S1. Scanning electron microscopy analysis was performed to study morphology of TiO_2 film structure Supporting information, Fig. S2. The sintered films were found porous signifying its suitability for DSSC applications.

Solar cell Assembly

Five layer cello-tape of thickness $\sim 45 \mu\text{m}$ was used as spacers placed at the edge of TiO_2 working electrode with the active area of $0.6 \times 0.3 \text{ mm}$. Platinum coated FTO as a counter electrode facing the TiO_2 photoelectrode was held together using binder clips on opposite end. Polyiodide solution was used as liquid electrolyte consisting of 0.5 M of Lithium iodide (SRL, India), 0.05 M of Iodine (Fisher Scientific, USA) and 0.15 M tertiary butyl pyridine (ACROS Organics, Belgium) in 100 ml Acetonitrile (SDFCL, India). Just before analysis, drops of prepared polyiodide electrolyte were induced in prepared sandwiched cell till no air bubble is present. The light was introduced such that light gets penetrate through the FTO back contact to the dye adsorbed onto the TiO_2 photoelectrode.

Solar cell characterization

XRD (D/B max-2400, Rigaku, USA) was used to determine crystalline nature, phase and crystallite size of TiO_2 film. The sample was placed into the diffractometer for analysis. Sample was scanned from 20° to 80° 2θ diffraction angle. Scanning electron microscopy (SEM) (JEOL-JSM 6360-A, USA) was used to study the morphology and cross section of TiO_2 film. The images were obtained at various magnification of $\times 3000$, $\times 10,000$ and $\times 30,000$. UV-Vis spectrophotometer (JASCO V-670, Germany) was used to study optical absorption properties and calculation of band gap of TiO_2 photoelectrodes, spectra of N3 dye and TiO_2 sensitized N3 Dye in diffused reflectance and transmittance mode in the range $200\text{--}800 \text{ nm}$ at room temperature. Photovoltaic parameters of each cell were measured using solar stimulator (Oriel Sol-2A, Newport, USA). AM1.5 sunlight filter with illumination intensities 50 mW/cm^2 and 33 mW/cm^2 LED (Nvis Clean Tech, India). Keithley source meter 2420 (Keithley instruments Ltd., USA) was used to measure J-V characteristics. The incident intensity of illuminating light sources was measured using sun meter (Solaron DSM 01, India) and digital lux meter (Lutron LX-101A, Lutron Electronic Enterprise CO., LTD., Taiwan). Optical luminescence of illuminated light source was measured using optical spectrometer (Ocean Optics HR 4000, USA). All the J-V curves of solar cell were obtained in the dark and under illumination. The J-V characteristics used to obtain open circuit voltage (V_{oc}), short circuit current density (J_{sc}), the maximum voltage point (V_{max}), the maximum current density (J_{max}), maximum power (P_{max}), fill factor (FF), efficiency (Eff). Photocurrent and photovoltage responses with respect to time and electrochemical impedance spectra were measured using potentiostat/galvanostat (Vertex IVIUM Technologies, Netherlands).

Result And Discussion

Optical study

It is clear from Fig. 1 (a) that the spectral distribution of light energy changes with type of illumination source. Unlike xenon lamp, which has broad emission spectra similar to AM1.5G, the LED has narrow spectra mainly in the visible range. The area under curve is observed to be lower for LED lamp from 400 to 700 nm whereas xenon lamp absorbs the maximum wavelength from 300 to 800 nm . As these cells will be ever useful in concentrated solar cell applications which will cause reexamination of the operating limitations which will improve efficiency at 1 sun . The low charge carrier mobility results in high charge accumulation under illumination, which will in turn cause increased recombination which affects overall efficiency of cell.²⁴ The output of any solar cell depends upon the spectral composition of incident light, its intensity and the angle at which the light is incident. The artificial light we are using has less magnitude than solar simulator and we utilized maximum light having narrow emission spectra. Thus the sources of heat affecting the photovoltaic device are minimized using indoor measurements. If we measure photoconversion efficiencies of device under different light intensities it becomes easier to study for which the device is best suited. Here in our study the drastic change in efficiency from 28% to 4% shows that the device is suitable for indoor lightning. The reason for this decrease can be due the spectral distribution and power density of the incident light. If we compare the spectral distribution in the IR region whereas LED shows spectral distribution in the visible region²⁵ as shown in Fig. 1 (a). One more reason to have greater efficiencies is its lower power incident densities. DSC technology delivers better performance under interior lightning because it is able to sustain performance even at low incident power densities and because of the good match between emission spectra of the dye, as both of them mainly span the range of visible wavelength.²⁶

Fig. 1 (b) shows the absorption spectra of TiO_2 photoanode, N3 dye, N3 sensitized TiO_2 photoanode. It is clearly observed spectra of N3 dye and sensitized TiO_2 photoelectrode is extended in visible region.

Fig. 3 (a) Plot of current density as a function of voltage, (b) Power density versus voltage under dark and different artificial illumination sources, (c) comparative plot of Efficiency and fill factor of fabricated device Vs two different light intensities, (d) schematic diagram of layer by layer fabricated DSSC and (e) circuit diagram of solar cell testing with glowing LED using serially connected three dye sensitised solar cell.

addition to the above mechanism there are some condition which reduces the performance DSSC such as before entering the external circuit, the injected electron can get reversely transferred to the oxidized dye via back electron transfer and electrolyte via recombination.²⁹

Fig. 3(a) shows the plot of current density as a function of voltage for N3 sensitized titania DSSC under different illumination conditions viz; Xenon lamp (50 mW/cm^2), LED lamp (33 mW/cm^2) and under the dark condition. As seen, our N3 sensitized titania DSSCs has higher current density under the illumination of LED lamp in comparison with illumination under the xenon lamps. The increase in current density suggests increase in light harvesting capacity of N3 sensitized titania DSSCs under the illumination of LED lamp due to good match

between emission spectra of LED lamp with absorption spectra of dye, both of them mainly span of range of visible wavelengths only. Photoelectrodes tested under artificial light exhibited I_d values around $4\text{mA}/\text{cm}^2$. I_d is the anodic spike shown in Fig. 3(a), observed enhancement in current due to the recombination of the photo generated charge carriers at the electrode/electrolyte interface when it was illuminated to light.³⁰ The highest efficiency (28.21 %) has been obtained for N3 sensitized titania DSSC illuminated with LED lamp and the corresponding fill factor was 47 % as shown in Table 1. The enhancement in the efficiency under artificial lighting indicates improvement in performance of DSSC under the artificial lighting compare to conventional solar simulator source. This is because the

spectral distribution of artificial lighting spectrum in the region of 400 to 500nm shown in Fig. 1, overlaps more favorably with absorption spectrum of N3 dye than solar simulator source. Fill factor observed for LED and xenon illumination is 47 % and 67 % respectively. Fill factor of LED is lower than xenon lamp illumination. In LED light we found current density 25.35 mA/cm^2 with $4 \text{ mA/cm}^2 I_d$ value. This fast current decrease affects the fill factor of the cell under LED illumination. Under xenon illumination, no I_d value is observed. The stable current density is the reason for higher fill factor in xenon than LED illumination. Photovoltaic parameters such as efficiency, fill factor, current density and voltage of a cell measured under the irradiance of Xenon lamp and LED lamp supporting information Fig. S3. These results indicate reduction in loss mechanism at reduced intensities of light source. Photovoltaic parameters such as efficiency, fill factor, current density and voltage of 11 DSSCs having $\text{PEC} > 25\%$ under the irradiance of Xenon lamp and LED lamp. Results are quoted after 11 experiments supporting information Fig. S4. It has been observed that when reducing the illumination intensity, the power conversion efficiency increases from 4.13 to 28.21 %. It is observed that current density increases from 3.93 to 25.35 mA/cm^2 . The fill factor decreases with increase in light illumination from 67 to 47 %. Gratzel group³¹ reported DSSC up to 13.1 % at 0.5sun. Wang *et al.*³² reported 20 % efficiency under indoor measurement. Teran³³ reported power conversion efficiency for AlGaAs of 21.1 % which was highest for indoor lightning. Feldt *et al.*³⁴ reported at 25 mW/m^2 max power of $10.7 \mu\text{W/cm}^2$. Barber *et al.*³⁵ reported 19 % efficiency for TiO_2 sensitized N3 dye at 38 mW/cm^2 . According to Green *et al.*³⁶ light emitted by an ideal cell to that by a blackbody Shockley and Quesser showed that the performance of a standard cell was limited to 31 % efficiency. According to Hagfeldt *et al.*³⁷ and Zhang *et al.*³⁸, the increase of I_{sc} was explained by positive shift of the conduction band under UV illumination. Fig. 3(b) is the variation of power density as a function of voltage for N3 sensitized titania DSSC under different illumination conditions. The maximum power density has been observed for LED lamp (9.30 mW/cm^2) followed by xenon lamp (2.13 mW/cm^2). Fig. 3 (c) shows the comparative representation of photovoltaic parameters such as efficiency and fill factor with respect to incident artificial LED and xenon lamp light sources. Fig. 3(d) shows schematic diagram of layer by layer fabricated N3 sensitized Titania photoanode with platinum as counter and polyiodide as liquid electrolyte. Fig. 3(e) shows

circuit diagram of solar cell testing with glowing LED using serially connected three dye sensitized solar cells. The three serially connected devices were illuminated under xenon lamp to glow red colored LED tested for long time duration. The maximum required voltage to glow LED is 2 V and generated voltage by single cell is 0.7 V. These three single cell were connected serially to glow LED. Overall good performance of Titania based DSSCs is observed. This may be due to porous morphology of TiO_2 film sensitized with dye molecules, more electron lifetime, smooth charge transfer process of excited electron from HOMO to LUMO of dye molecule, LUMO to conduction band of TiO_2 then towards external circuit through FTO, platinum coated FTO counter electrode and electrolyte respectively. The suitability of device for dye sensitized solar cell application is observed.

DSC shows better performance in interior lightning because it is able to sustain performance even at low incident power densities as it is good match between the emission spectra of LEDs with the absorption spectrum of the dye, as both of them mainly span the range of visible wavelengths only.¹⁸

Electrochemical Impedance Spectroscopy

Electro Impedance spectroscopy carried out in 1MHz to 0.1Hz frequency range with -0.8V constant applied potential. Fig. 4 (a) shows the bode plot with characteristic frequency peak in the middle frequency range. LED lamp shows shift towards higher frequency peak compared to xenon lamp and dark. The observed shift in frequency is mentioned in Table 2. Any shift of peak from higher frequency to lower frequency reveals a more rapid electron transport process. The mean electron life time calculated from the relation $\tau_{\text{eff}} = \frac{1}{2\pi f_{\text{max}}}$, where, f_{max} is the frequency at the top of the intermediate frequency arc.³⁹ The electron lifetime observed under dark condition, Xenon lamp and LED lamp is observed to be 7.39, 4.66 and 2.52 mili second respectively. When light is incident on cell, the electron are generated and current flows through the external circuit, which reduces the charge recombination resistance and this reduces the electron lifetime. Similar trends in EIS result in dark and light was observed by Wu-Qiang *et al.*⁴⁰.

The EIS study is carried out to determine electron transport and recombination resistance. The experimental impedance data, determined using nyquist plot is shown in Fig. 4(b). the experimental impedance data given by nyquist plots in Figure were fitted to an equivalent circuit based on the diffusion-recombination model.⁴¹ The series resistance of

Fig. 4 Electrochemical Impedance spectra (a) Bode plot and (b) Nyquist plot with equivalent circuit of N3 sensitized titania DSSCs under dark and different artificial illumination sources.

Table 1 Typical solar cell performance parameters, Cell Area 0.18 cm².

Lamp	Illumination Light Intensity		V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	η (%)
	(mW/cm ²)	Lux ($\times 10^3$)				
LED	33	225	0.76	25.35	47	28.21
Xenon	50	342	0.77	03.93	67	4.12

Table 2 Electron transport properties of fabricated cell.

Sample Under Light Illumination Condition	R_{tr} (Ω)	R_{ct} (Ω)	f (Hz)	$\frac{R_{tr}}{R_{ct}}$	L_n (μm)	η_{cc} (%)	τ_{eff} (msec)	t_d (msec)	D_{eff} (cm ² sec ⁻¹)	L_{eff} (μm)
	Electron Transfer Resistance of Counter	Charge Transfer Resistance of Photoanode	Frequency		Diffusion Length	Charge Collection Efficiency	Mean Electron Lifetime	Effective Electron Diffusion Time	Effective Electron Diffusion Coefficient ($\times 10^{-7}$)	Effective Electron Diffusion Length
Dark	6.8	62	21.54	9.1	18.12	90.12	7.39	0.81	4.4	18.12
Xenon Lamp	5.4	35	34.15	6.4	15.28	86.63	4.66	0.72	5.0	15.28
LED Lamp	3.7	11	63.1	2.9	10.35	74.83	2.52	0.85	4.2	10.35

solar cell calculated from nyquist plot is $R_s = \sim 18$. The impedance spectra consist of three semicircles. The elements related to interface of counter electrode and electrolyte include the electron transport resistance R_{tr} , the charge transfer resistance R_{ct} which is related to the recombination of electrons at TiO_2 /dye/electrolyte interface. This EIS equivalent circuit fitted parameters R_{tr} and R_{ct} can be used to calculate the parameters such as diffusion length (L_n), charge collection efficiency (η_{cc}), effective electron diffusion coefficient (D_{eff}), effective electron diffusion length (L_{eff}), which is useful to study the electron transport properties of solar cell. We have obtained the experimental impedance data at various light intensities. Usually, the semicircle in lowest frequency range assigns the electrolyte interface by Warburg diffusion process of I and I_3 .⁴²

With the help of EIS nyquist plot, the diffusion length is calculated using formula $l_n = \sqrt{\frac{R_{ct}}{R_{tr}}}$.⁴³ Further effective electron diffusion co-efficient in the photoanode is given by $D_{eff} = \frac{R_{ct}}{R_{tr}} * \frac{l^2}{\tau_{eff}}$,⁴² where l is the thickness of TiO_2 film. Charge collection efficiency is calculated using $\eta_{cc} = 1 - \frac{R_{tr}}{R_{ct}}$.⁴⁴ Effective electron diffusion length calculated using $L_{eff} = \sqrt{D_{eff} * \tau_{eff}}$.⁴⁵ Effective electron diffusion length calculated using $\tau_D = \tau_{eff} * \frac{R_{tr}}{R_{ct}}$.⁴⁶ The calculated values are given in table no 2.

Stability Test

The open circuit voltage decay and photocurrent stability of N3 sensitized titania DSSCs analyzed under the different light sources for the short time period (1000 s). The measurements were carried out for

un-sealed DSSC device. Fig. 5 shows the stability test for N3 sensitized titania DSSC with variation of current density and voltage of N3 sensitized titania DSSC as a function of time. As seen when light source is ON, both voltage and current density of DSSC suddenly enhances. Photoanodes exhibited I_d values around 4mA and the observed anodic enhancement in current is due to the recombination of the photo generated charge carriers at the electrode/electrolyte interface when it was illuminated to light.²⁸ The rise in voltage and current density is maximum for DSSC exposed to LED lamp followed by exposure to xenon lamp. It is interesting to note that the current density and voltage remain constant when the light sources are ON suggesting excellent stability of N3 sensitized titania DSSC for indoor lighting. We expect high rise in current density and voltage for sealed N3 sensitized titania DSSCs because sealing enhances the lifetime of dye and the electrolyte.

Conclusion

We have studied the indoor performance of N3 sensitized titania DSSCs under artificial lighting. It is observed that there is substantial improvement in the performance of solar cell efficiency. In standard AM1.5G 50mW/cm² sunlight, the PCE reaches above 3 %. Remarkably, under indoor illumination of 33 mW/cm² the maximum PCE of this new DSSC design is 28.21 %, which results in a power output of 9.31 mW/cm². This power is sufficient to establish the independent operation of a wide range of electronic devices in the indoor environment with reasonable solar cell size as low power electronic devices typically consume 100 W in sleep mode. The electron transport properties were studied using electrochemical impedance spectroscopy, which supports JV characteristics. The present

Fig. 5 Stability test for N3 sensitized titania DSSC (a) Change in open circuit voltage and (b) Change in current density under the illumination of LED Lamp and xenon lamp.

study suggests that the N3 sensitized titania DSSCs are ideally suited for artificial light applications and proved that an LED can glow with the help of this DSSC. We expect our results to have a major practical impact on low power electronic devices as harvesting the artificial light energy for power electronic devices require low electric power for their autonomous operation. Overall, our findings suggests that the obtained high efficiency N3 sensitized titania DSSCs under artificial lighting have a good potential application for low power electronic devices for their autonomous operation.

Author contributions

HMP conceived the work and CVJ, VSK and HMP designed the experiments and wrote the manuscript. CVJ and VSK fabricated and characterized the solar cells: assembled the devices. HMP and SRJ coordinated the work. CVJ and VSK equally contributed the work.

Notes

The authors declare no competing financial interest.

Acknowledgements

We acknowledge the Board of College and University Development, Savitribai Phule Pune University Pune, India for financial support. VSK and CVJ are thankful to Dr. Rajendra Prasad MB, Dr. Prashant K. Baviskar, Dr. Vikram P. Bhalekar, Ms. Parvin S. Tamboli, Mr. Ravi V. Ingle, Mr. Niyamat I. Beedri for fruitful discussion.

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