

ZnO nanoparticles with different concentrations inside organic solar cell active layer

Shanmugam Saravanan¹, Yasser A.M. Ismail^{*2,3}, Murugesan Silambarasan¹,
Naoki Kishi⁴ and Tetsuo Soga⁴

¹Centre for Photonics and Nanotechnology, Sona College of Technology, Salem 636 005, Tamil Nadu, India

²Third Generation Solar Cells Laboratory, Department of Physics, Faculty of Science, Al-Azhar University,
Asyut 71524, Egypt

³Al-Azhar Center of Nanoscience and Applications (ACNA), Al-Azhar University, Assiut, 71524, Egypt

⁴Department of Frontier Materials, Nagoya Institute of Technology, Nagoya 466-8555, Japan

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Abstract. In the present work, ZnO nanoparticles (NPs) have been dispersed alone in the same solvent of the active layer for improving performance parameters of the organic solar cells. Different concentrations of the ZnO NPs have been blended inside active layer of the solar cell based on poly(3-hexylthiophene) (P3HT), which forms the hole-transport network, and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM), which forms the electron-transport network. In the present investigations, the ZnO NPs may represent an efficient tool for improving light harvesting through light scattering inside active layer, electron mobility, and electron acceptance strength which tend to improve photocurrent and performance parameters of the investigated solar cell. The fill factor (FF) of the ZnO-doped solar cell increases nearly 14% compared to the non-doped solar cell when the doping is 50%. The present investigations show that ZnO NPs improve power conversion efficiency of the solar cell from 1.23% to 1.64% with increment around 25% that takes place after incorporation of 40% as a volume ratio of the ZnO NPs inside P3HT:PCBM active layer.

Keywords: organic solar cells; ZnO nanoparticles; device performance parameters

1. Introduction

Organic solar cells (OSCs) have become a competitive option for conventional photovoltaics (Aernouts, Aleksandrov *et al.* 2008, Zhou, Zhang *et al.* 2008). Compared to their inorganic counterparts, OSCs offer numerous advantages (low cost fabrication, flexibility, printable on plastic substrates, large-area capability, and so on) and enlarge the application possibilities of solar cells (Li, Shrotriya *et al.* 2007, Mandoc, Koster *et al.* 2007). In addition, the key advantages of OSCs have been identified as: easy integration into other products, new market opportunities (e.g., wearable photovoltaic), manufacturing of OSCs in a continuous process using state of the art printing tools, short energy payback times and low environmental impact during manufacturing

*Corresponding author, Ph.D., E-mail: Yasser_ami@yahoo.com
S. Saravanan and Yasser A.M. Ismail contributed equally as first author

and operations Scharber and Sariciftci (2013).

Bulk-heterojunction (BHJ) OSCs, where a blend of a polymer donor and fullerene acceptor is inserted between a high transparent semiconducting oxide (indium tin oxide (ITO)) anode and a metal (Al, Ag or Au) cathode, have attracted considerable attention worldwide (Scharber, Muhlbacher *et al.* 2006). BHJ OSCs have already shown high power conversion efficiencies (PCEs) of 7-10% (Scharber, Muhlbacher *et al.* 2006, Jorgensen, Norrman *et al.* 2012), demonstrating a bright future for commercialization of such solar cells. However, the conversion efficiency of OSCs still needs necessary improvement to become more efficient economically than inorganic counterparts.

Many research groups (Ismail, Soga *et al.* 2010, Belcher, Wagner *et al.* 2007, Yu, Gao *et al.* 1995, Padinger, Ritterberger *et al.* 2003, Kim, Cook *et al.* 2006) were studying polymer solar cells in order to improve their PCEs. Nanocrystals such as CdSe and TiO₂ can be used as electron acceptors combined with polymer donor in hybrid solar cells. Among these inorganic nanocrystals, ZnO is believed to be one of the most promising electron acceptors due to its environmental friendliness and low crystallization temperature (low-cost production). In addition, ZnO exhibits higher electron mobility with low-cost production. Krebs employed solution processed ZnO in air-stable hybrid organic solar cells which require no vacuum processing steps Krebs (2009). In addition, the solution of ZnO was successfully applied to roll-to-roll processing of inverted OSC devices Krebs (2008). The ZnO nanoparticles (ZnO NPs) are used in the literatures for improving the efficiency of OSCs through two methods; (1) through the deposition separately outside solar cell active layer (You, Chen *et al.* 2012, Mbule, Kim *et al.* 2013) or (2) through the incorporation and blending inside solar cell active layer (Wang and Wang 2008, Beek, Wienk *et al.* 2006, Olson, Pirus *et al.* 2006, Oh, Heo *et al.* 2013). In the first method, the role of ZnO NPs to improve the efficiency of OSCs is limited due to the isolation and separation of ZnO from solar cell active layer. While in the second method, the ZnO NPs are dispersed in a solvent, such as ethanol, which is different than the solvent used for preparing solar cell active layer, such as dichlorobenzene. Therefore, the ethanol can be introduced as an another factor affecting the efficiency of the OSCs. Therefore, it is important to use ZnO NPs, lonely, dispersed in the same solvent of the solar cell active layer for improving performance parameters of the solar cells.

In the present study, ZnO NPs, dispersed in dichlorobenzene, are incorporated inside the active layer of the famous OSC based on a BHJ blend of regioregular poly(3-hexylthiophene) (P3HT) (which forms the hole-transport network) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) (which forms the electron-transport network). For examination the behavior of the most common OSC under blending with a third material, the effect of different concentrations of the ZnO NPs on device performance parameters is investigated for P3HT: PCBM solar cell. The indium-tin-oxide (ITO)/poly(3,4-ethylene dioxythiophene)-blend-poly(styrene sulfonate) (PEDOT:PSS)/P3HT:PCBM:ZnO/Al bulk heterojunction solar cell is constructed with varying ZnO NPs concentrations incorporated in the solar cell active layer. The optical absorption spectroscopy, photocurrent spectroscopy, and current density/voltage (J/V) characteristics under AM1.5 white light illumination with an intensity of 100 mW/cm² are investigated for the solar cells.

2. Experimental procedures

2.1 Materials and solutions

Regioregular P3HT (from Merck, Germany) and PCBM (from Nano Spectra, Japan) were purchased and used without further purification. Equal weight ratios of P3HT and PCBM were dissolved in 1,2-dichlorobenzene (Tokyo Chemical Industry Co. LTD, Japan) for preparing a 50 mg/mL solution concentration of the solar cell active layer. The P3HT:PCBM blend was vigorously stirred at 150 rpm under 60°C for more than 24 h outside glove box. The ZnO NPs with diameters from 20 to 50 nm were obtained by solution combustion method (Saravanan, Silambarasan *et al.* 2014, Silambarasan, Saravanan *et al.* 2014). The ZnO NPs were dispersed in 1,2-dichlorobenzene (1mg/mL) under harsh sonication for 3 h in air and mixed with P3HT:PCBM blend solution in different volume ratios from 20 to 50% of the total volume with further stirring for 1 h at room temperature under nitrogen atmosphere in a glove box to maximize mixing while avoiding touching the vial cap. The total volume of the 1,2-dichlorobenzene, as a solvent, was kept constant in all the cases.

2.2 Film and device fabrication

For optical absorption measurement, the P3HT:PCBM and P3HT:PCBM:ZnO blend films with different concentrations of the ZnO NPs were prepared by spin-coating (2000 rpm) the blend solutions onto clean ITO-glass substrates with thermal annealing using a digitally controlled hotplate at 110 °C for 10 min under nitrogen atmosphere in a glove box. The film preparation and thermal annealing conditions were kept the same as those of the corresponding complete devices for accurate comparison.

For organic solar cell fabrication, the ITO-glass substrates ($\sim 10 \Omega/\square$) were sequentially cleaned in an ultrasonic bath using acetone (twice) and methanol (once), rinsed with deionized water, and finally dried in flowing nitrogen. To increase the work function of the ITO electrode and to improve the electrical connection between ITO and organic active layer, a layer of PEDOT:PSS (Clavios) was spin-coated (2000 rpm) onto ITO-glass substrate in air and dried using a digitally controlled hotplate at 150°C for 10 min under nitrogen atmosphere in a glove box. On the top of insoluble PEDOT:PSS layer, the P3HT:PCBM and P3HT:PCBM:ZnO blend solutions, with different ZnO NPs concentrations, were spin-coated (2000 rpm) and dried, for removing any residual solvent, at 110°C for 10 min under nitrogen atmosphere in a glove box. An approximately 150-nm-thick Al electrode was thermally deposited onto the active layer using a vacuum deposition system at a pressure of about 3×10^{-4} Pa through a shadow mask to obtain 25 identical cells on one device with an active area of $3 \times 3 \text{ mm}^2$. Then, the devices were annealed in a nitrogen atmosphere at 140°C for 4 min for increasing the electrical contact between solar cell active layer and Al electrode. The complete organic solar cells, with a configuration shown in Fig. 1, were stored in the dark under nitrogen atmosphere until measurements.

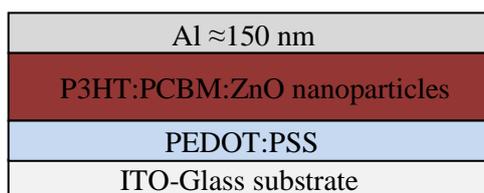


Fig. 1 The schematic illustration of organic solar cell with ZnO nanoparticles incorporated inside P3HT:PCBM active layer

2.3 Measurements

The optical absorption spectroscopy of the P3HT:PCBM and P3HT:PCBM:ZnO blend films with different concentration of ZnO NPs was measured using a JASCO V-570 ultraviolet/visible/near-infrared (UV/vis/NIR) spectrophotometer with incident optical light towards the coated side of the films and using an ITO-glass substrate in the reference position. The external quantum efficiency measurement was performed for the devices using a halogen lamp and a monochromator. The J/V characteristics of the devices under white light illumination were determined using standard solar irradiation of 100 mW/cm^2 (AM1.5) with a JASCO CEP-25BX spectrophotometer J/V measurement setup with a xenon lamp as the light source and a computer-controlled voltage-current source meter (Keithley 238) at 25°C under nitrogen atmosphere. The J/V characteristics were determined for several cells in one device, and we considered the solar cell having the best performance parameters in that device. All measurements were performed soon after preparation of the devices to avoid any change in the photoelectric properties caused by aging.

3. Results and discussion

3.1 Effect of ZnO NPs concentration on light harvesting of the solar cell

It is important to know that, the spectral range of the optical absorption compared to solar spectrum, absorption value, and absorption wavelength are the factors affecting the total number of absorbed photons by solar cell active layer, where the limitation in the light absorption across the solar spectrum limits the photocurrent of the solar cells. The number of absorbed photons and, therefore, the generated photocurrent in a solar cell increases when the absorption range and absorption peak value increase for solar cell active layer. The number of absorbed photons in a solar cell active layer, also, increases when the active layer absorbs in the red or near infrared region of the solar spectrum, where there is the dense number of the light photons.

Fig. 2 shows the effect of concentration of ZnO NPs on the optical absorption of the P3HT:PCBM solar cell active layer. As shown in this figure, the absorption of P3HT in the films occurs around the wavelength of 506 nm during the range 388-648 nm, while the PCBM absorbs around the wavelength of 332 nm during the range 288-388 nm. As shown in Fig. 2, the increase in ZnO NPs concentration from 20% up to 40% increases the absorption peak value of the solar cell active layer, while at 50% as a higher ZnO NPs concentration the absorption peak value is decreased. We can observe from Fig. 2 that, in the time at which the solid contents of both P3HT and PCBM do not change (25 mg of both) in the active layer of all solar cells, the absorption peaks of both P3HT and PCBM are increased with increasing ZnO NPs solution from 20% up to 40%, which are blended in the same solar cell active layer.

The observed increase in the absorption peaks of both P3HT and PCBM is attributed to optical scattering caused by ZnO NPs which can reflect the light to be absorbed inside active layer. The blended ZnO NPs induces a roughness of the active layer surface that increases light scattering and, therefore, absorption peak value of the solar cell active layer (Oh, Heo *et al.* 2013). The reduced optical absorption at 50% of ZnO NPs concentration may be attributed to the decrease in the active layer thickness, where the amounts of ZnO aggregates may remove small amounts of the P3HT:PCBM solution during the spin coating process. This action can mostly take place due to the

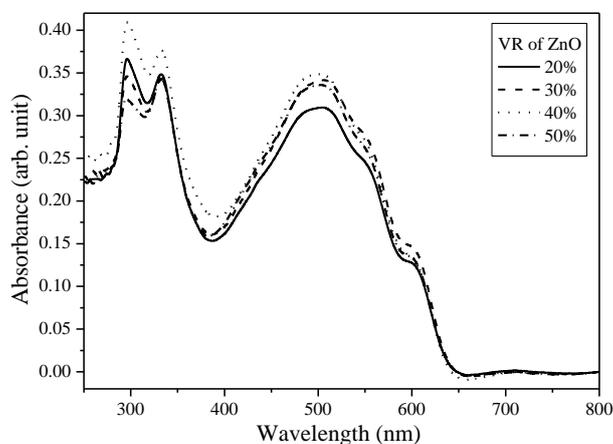


Fig. 2 UV-visible absorption spectra of P3HT:PCBM active layer containing various volume ratios of ZnO nanoparticles

higher viscosity (high concentration) of the P3HT:PCBM solution as 50 mg/mL.

The optical absorption spectroscopy is an efficient tool for detecting the organization of the molecules in crystalline domains as stated by (Vanlaeke, Swinnen *et al.* 2006). As shown in Fig. 2, the absorption peak of regioregular P3HT does not observe neither red nor blue shifts with increasing ZnO NPs concentration. This means that, the incorporation of ZnO NPs inside P3HT:PCBM active layer does not alter the conjugation and molecular organization of the P3HT polymer.

3.2 Effect of ZnO NPs concentration on external photocurrent quantum efficiency of the solar cell

The external quantum efficiency (EQE) spectra of the ITO/PEDOT:PSS/P3HT:PCBM/Al and ITO/PEDOT:PSS/P3HT:PCBM:ZnO/Al solar cells with varying ZnO NPs concentration in the active layer blends are shown in Fig. 3 in the wavelength range from 300 to 800 nm. The contribution of P3HT to photocurrent of the solar cells is observed around the wavelength of 550 nm as shown in Fig. 3, where the P3HT has an absorption peak at 506 nm, as shown in Fig. 2. Fig. 3 shows a small contribution of PCBM to photocurrent of the solar cells around the wavelength range of 335-450 nm, while the PCBM absorbs around the wavelength range of 288-388 nm, as shown in Fig. 2. As it can be observed from Fig. 3 that, the incorporation of ZnO NPs inside P3HT:PCBM active layer tends to improve the photocurrent of the investigated solar cells. The observed improvement in photocurrent increases with increasing ZnO NPs concentration in the solar cell active layer from 20% to 40%, while at 50% of ZnO NPs concentration the photocurrent decreases compared to 40%. The improvement of EQE is attributed to an improvement in electron mobility in the solar cell active layer due to the use of ZnO (Li, Shrotriya *et al.* 2007) which helps to transport photo-generated electrons from both P3HT and PCBM to the cathode. Also, in the time at which the ZnO acts as electron transporter, it enhances electron acceptance strength among with PCBM molecules. Then the number of photo-generated charge carriers is increased in the solar cell active layer, and, consequently, the EQE is improved. In addition, the active layer

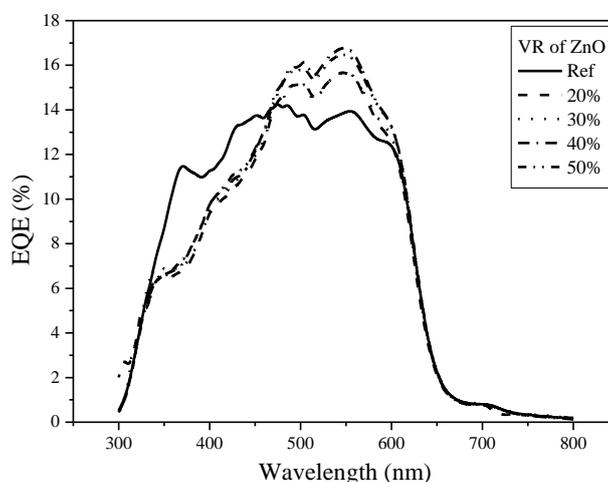


Fig. 3 External quantum efficiency spectra of the BJJ solar cells without and with different volume ratios of ZnO nanoparticles inside the active layer

roughness, induced by ZnO NPs, is the reason for improving optical absorption which in turn improves the photocurrent of the solar cells. However the EQE is reduced for 50% of ZnO NPs concentration due to the decrease in the optical absorption, as shown in Fig. 2.

In the present paragraph we prefer to illustrate the contribution of ZnO to electron acceptance in solar cell active layer. The ZnO NPs may be able to dissociate the generated excitons in the solar cell active layer at the interface between P3HT and ZnO NPs as a result of energy difference between the lowest unoccupied molecular orbital (LUMO) of donor P3HT at 3.53 eV Wang and Wang (2008) and the conduction band of ZnO NPs at 4.19 eV (Beek, Wienk *et al.* 2006). Therefore, the excitons at P3HT/ZnO interface can be easily dissociated and the charge carriers can be generated in the solar cell active layer.

One can observe from Fig. 3 that, the resulted improvement in photocurrent is only related to the contribution of P3HT segment to photocurrent, while the contribution of PCBM to photocurrent, in the reference cell, is deteriorated after adding ZnO NPs in the solar cell active layer, although the PCBM molecules exhibit obvious optical absorption, as shown in Fig. 2. The quenching in the contribution of PCBM to photocurrent is mostly attributed to recombination of the electrons found in the LUMO of the PCBM molecules. In this situation, it is difficult to understand the relationship between this recombination owing to presence of ZnO in the solar cell active layer.

3.3 Effect of ZnO NPs concentration on the performance parameters of the solar cell

In the present work, we have measured J/V characteristics of the ITO/PEDOT:PSS/P3HT:PCBM/Al and ITO/PEDOT:PSS/P3HT:PCBM:ZnO/Al devices as a function of ZnO NPs concentration in the solar cell active layer, as shown in Fig. 4. From the analysis of the J/V characteristics, we have obtained the photovoltaic performance parameters, i.e., open circuit voltage (V_{OC}), short circuit current density (J_{SC}), fill factor (FF), and power conversion efficiency (PCE), of the investigated devices. Figs. 5 and 6 show the variations of these parameters with increasing the concentration of ZnO NPs in the solar cell active layer. Fig. 5 shows an increase in

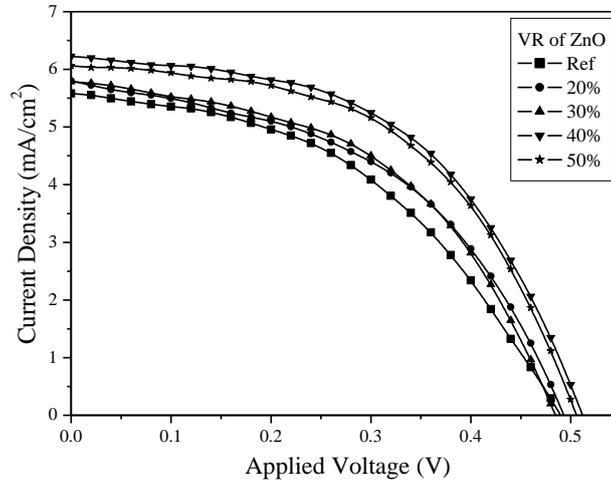


Fig. 4 Current density-voltage characteristics of the BHJ solar cells without and with ZnO nanoparticles incorporated inside solar cell active layer

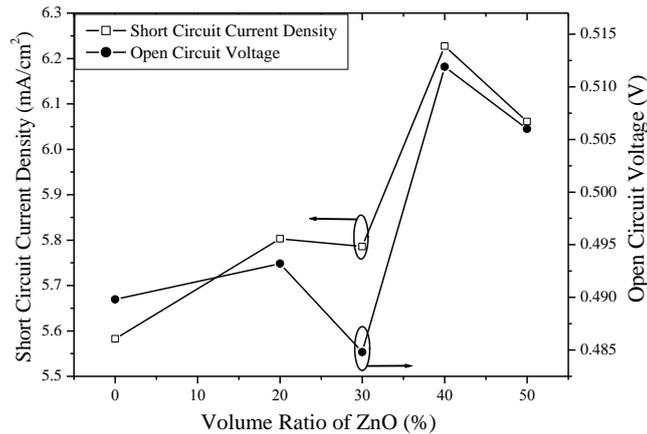


Fig. 5 Short circuit current density and open circuit voltage of the BHJ solar cells under the variation of volume ratios of ZnO nanoparticles incorporated inside solar cell active layer

both J_{SC} and V_{OC} with increasing ZnO NPs concentration up to 40%, while at the 50% of the ZnO NPs concentration the J_{SC} and V_{OC} are reduced. The FF, as shown in Fig. 6, increases continuously with increasing ZnO NPs concentration up to 50%. At this concentration the FF increases nearly 14% compared to the non-doped sample.

The observed increase in the J_{SC} , as shown in Fig. 5, may be attributed to the increase in optical absorption through light scattering, which is able to generate excess of charge carriers in the solar cell active layer. Actually, optical absorption is not the solely responsible for improving J_{SC} that can be affected further by charge carrier transfer and acceptance strength, which may be improved by the aid of the ZnO NPs blended in the solar cell active layer. For the same reasons, the J_{SC} in Fig. 5 behaves in the same agreement with the behavior of EQE under the influence of ZnO NPs upon the solar cell, as shown in Fig. 3. Therefore, the decrease in the J_{SC} at 50% of ZnO NPs

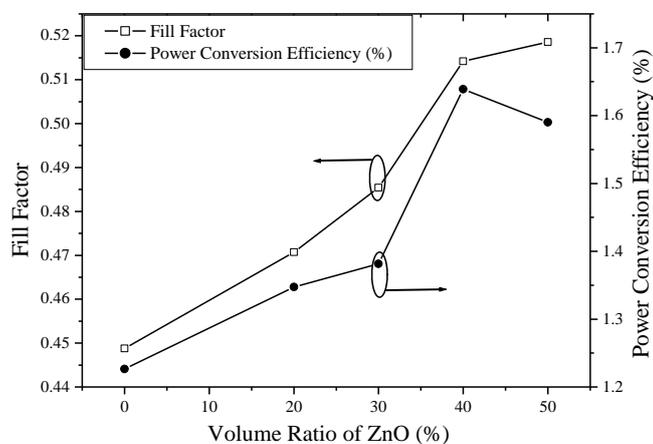


Fig. 6 Fill factor and power conversion efficiency of the BHJ solar cells under the variation of volume ratios of ZnO nanoparticles incorporated inside solar cell active layer

concentration is attributed to the decrease in optical absorption (as shown in Fig. 2) of the solar cell active layer.

Fig. 5 shows a slight increase in V_{OC} around 0.02 V with increasing ZnO NPs concentration up to 40%. The general perception in organic solar cells is that, the V_{OC} is determined by the energy difference between the highest occupied molecular orbital (HOMO) of the donor and the LUMO of the acceptor (Yamanari, Taima *et al.* 2009). The LUMO and HOMO levels of P3HT are 3.53 and 5.2 eV, respectively Wang and Wang (2008). The LUMO and HOMO energy levels of PCBM are 3.7 and 6.1 eV, respectively Wang and Wang (2008). The conduction energy level of ZnO nanoparticle is 4.19 eV (Beek, Wienk *et al.* 2006). Therefore, the slight increase in V_{OC} with increasing ZnO NPs concentration can be attributed to the change in band alignment caused by the introduction of ZnO NPs into solar cell active layer (Kruefu, Peterson *et al.* 2010).

The continuous increase in FF with increasing ZnO NPs concentration up to 50%, as shown in Fig. 6, can prove that, the ZnO NPs are the efficient tool for enhancing charge carrier transfer in the P3HT:PCBM active layer. The improvement in the charge carrier transfer is due to the decrease in series resistance of the solar cell active layer by the aid of ZnO NPs, which are blended inside active layer blend of the solar cell. The behavior of FF at higher ZnO NPs concentration (50%) in comparison with the behavior of optical absorption, EQE, and J_{SC} proves our previous suggestion assumed that the ZnO aggregations at higher concentration reduce optical absorption, EQE, and J_{SC} as a result of reduction of active layer thickness through the removing little of P3HT:PCBM solution during active layer preparation. Depending on the behavior of all previous performance parameters, the PCE of the solar cells behaves as appeared in Fig. 6 under the variation of ZnO NPs concentration. Fig. 6 shows that, the best ZnO NPs concentration is the 40% at which the PCE of the solar cell reaches 1.64%. This means that the incorporation of ZnO NPs in the active layer improves the PCE of the solar cell from 1.23% to 1.64% with increment around 25%.

4. Conclusions

The dispersion of ZnO NPs alone in the same solvent of the active layer shows an improvement

in optical absorption and performance parameters of the organic solar cell composed of bulk heterojunction blend of P3HT and PCBM. From the present investigations, it can be concluded that, the ZnO NPs are efficient tool for: (1) improving light scattering inside active layer that tends to improve optical absorption of the solar cell; (2) improving electron mobility in the solar cell active layer that tends to improve EQE and performance parameters of the solar cell; (3) improving electron acceptance strength which tends to improve the number of photo-generated charge carriers, and consequently the EQE and performance parameters are improved for the solar cell. Our investigations show that, the incorporation of ZnO NPs in the active layer improves the PCE of the solar cell from 1.23% to 1.64% with increment around 25%. This improvement takes place after the incorporation of 40% of the ZnO NPs inside P3HT:PCBM solar cell active layer. The FF increases nearly 14% compared to the non doped sample when the doping is 50%.

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