

Improvement of light harvesting by inserting an optical spacer (ZnO) in polymer bulk heterojunction solar cells: A theoretical and experimental study

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Abstract—By introducing a thin ZnO layer as an optical spacer, we have demonstrated that inserting this layer between an active layer and a reflective electrode results in a re-distribution of the optical electric field inside bulk heterojunction solar cells. A theoretical analysis by optical modeling showed that the thin ZnO layer could shift the position of the maximum of the electric field into the absorbing layer. Theoretical calculations were compared with experimental results for devices with and without an optical spacer. By using a ZnO optical spacer layer, a significant increase was observed in the short circuit current density of J-V curves. This increase might be due to harvesting more lights and also hole-blocking by the ZnO layer. Both electrical and optical characteristics of the device provided improved results in the power conversion efficiency of the bulk heterojunction solar cell up to 3.49%.

KEYWORDS: Bulk-heterojunction, Optical spacer, Optical modeling, Polymer solar cells, ZnO nanoparticle

I. INTRODUCTION

Polymer bulk heterojunction (BHJ) solar cells offer a great technological potential as a renewable, alternative source of electrical energy. This type of solar cell has been investigated because of its low cost, environmental friendly production, ease of deposition, and flexible applications [1-3]. Most efficient polymer solar cells are based on a mixture of a conjugated polymer and a fullerene derivative (PCBM) as electron acceptor, for which efficiencies of nearly 6% have recently been reported [4-6]. One

limiting parameter in polymer solar cells, as compared to silicon-based devices, is the low short-circuit current density (J_{sc}) which is determined primarily by (1) the fraction of incident photons absorbed in the charge separating layer and (2) the internal quantum efficiency (IQE) defined by the fraction of collected carriers per absorbed photons. Due to the relatively narrow absorption bands and unfavorable overlap of these bands with the solar spectrum, absorption in organic materials is low. To solve this problem, one might simply increase the thickness of the active layer to absorb more light. Because of the low mobility of the charge carriers in the polymer fullerene composites, however, increasing the thickness eventually increases the internal series resistance of the cell and, thereby, reduces the power conversion efficiency. Traditionally, surface texturing is used to increase cell absorption by scattering the light and reducing the reflection in conventional silicon solar cells [7, 8]. However, texturing is practically challenging for organic solar cells because of difficulties in the formation of nanometer scales on topographical surfaces [9]. The use of micro cavities and photonic crystals is another light trapping strategy [10-12]. Nevertheless, there are concerns about processing bottle necks and drawbacks in making periodic patterns, especially in the active layer. An alternative approach is to change the spatial distribution of the optical electric field inside the device by introducing an optical spacer between the active layer and the Al electrode as sketched in Fig. 1[13].

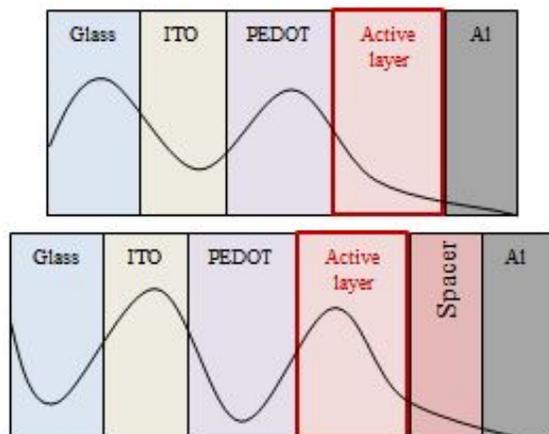


Fig. 1. Schematic representation of the spatial distribution of the squared optical electric field strength inside the devices (up) without and (down) with optical spacer

Reflection from the metallic cathode enables a double pass through the active layer. Moreover, because of the optical interference between the incident and back reflected light, the light intensity at the surface of the metal electrode is zero. Thus, it was demonstrated that insertion of an optical spacer between the active layer and the metal electrode can place the active layer in a more favorable region of the optical electric field [14]. Therefore, the use of an optical spacer between the BHJ film and the metal electrode provides a method for enhancing the photocurrent while keeping the thickness fixed. The optical spacer must be a good acceptor and an electron-transport material with a conduction band edge lower in energy than that of the lowest unoccupied molecular orbital (LUMO) of fullerene. The LUMO must be above (or close to) the Fermi energy level of the collecting metal electrode. It must also be transparent to light with wavelengths within the solar spectrum [5, 14]. Here, we present theoretical and experimental studies, assessing the origin of the increased current in P3HT:PCBM solar cells with a ZnO spacer layer. Theoretical analysis by optical modeling is a powerful mean, increasingly used to understand and predict optical effects in organic solar cells [15-19]. It is based on the transfer matrix formalism [15]. At the early stage of modeling for organic photovoltaic (OPV), optical effects such as reflections and interference were not considering, and the

optical field was assumed to decrease exponentially along the propagation of direction. In 1999, Pettersson *et al.* [15] pointed out that the thickness of thin film in organic solar cells is normally less than the wavelength of incident light, and therefore optical interference effects becomes important to the optical electric field distribution inside the device. They applied the transfer matrix theory described by Heavens [20], which considered the interference effects for light wave propagating in the thin films, to the organic photovoltaic devices. Absorption, transmission, reflection, and phase shift can all be described by the real and imaginary parts of the complex refractive index ($\tilde{n} = n + ik$, with n as the refractive index and k as the extinction coefficient) of each layer. The optical electric field at each position can be calculated by the resulting transfer matrix system. The enhancement in the photocurrent due to the presence of an optical spacer is determined by the thickness and the optical constants (refractive index and extinction coefficient) of the optical spacer and also by the thickness and optical constants of all the other layers within the BHJ solar cell including the indium/tin oxide (ITO), the poly(ethylenedioxythiophene) (PEDOT: PSS), and the BHJ material. Here, in our device design and optimization, we first tuned the thickness of the active layer to maximize the short-circuit current density and the optical distribution inside the active layer. Then, we inserted a layer of ZnO nanoparticles (cast from solution) as an optical spacer in order to further enhance the light absorption. The theoretical and experimental results indicated that the optical spacer simultaneously improved the electrical and optical properties of the solar cell. Therefore, by using ZnO after an active layer, the need for vacuum-deposition and using air-sensitive calcium as an interfacial layer is eliminated. Fig. 2 shows the molecular structure of P3HT and PCBM, the device structure and the energy levels of the components used in the devices. An increase in the power conversion efficiency of the fabricated devices using ZnO, as an optical spacer, is shown for the solution-processed BHJ solar cells based on P3HT and PCBM as

the donor and the acceptor components respectively.

II. METHODS

A. Theory and simulation (Model)

Polymer solar cells consist of multiple thin film layers with different optical properties. In the optical transfer-matrix model used in this research to calculate the distribution of light intensity in polymer solar cells, the layers are assumed as simple to be parallel, smooth, and of uniform thickness. The transfer-matrix method which is applied to solar cells has been described in detail elsewhere [15, 21-25], and so only the essential features of the model will be described here. The only parameters required by the model are the complex index of refraction $\tilde{n}_j(\lambda) = n_j(\lambda) + ik_j(\lambda)$ and thickness d_j , $j=1,2, \dots, m$, of each layer in the solar cell (the glass substrate is assumed to be optically thick). Light of intensity I_0 is assumed to be incident normal to the substrate, the refractive index of air is taken to be 1, and multiple reflections at the air/substrate and substrate/multilayer interfaces are taken into account [21, 22].

Calculation of the transfer matrix provides the optical electric field $E_j(x)$ in layer j as a function of distance x to the right of the $(j-1)$ interface, from which the total absorbed power in layer j at position x can be obtained as [15]:

$$Q_j(x) = \alpha_j I_j(x) = \frac{2\pi c \epsilon_0}{\lambda} n_j k_j |E_j(x)|^2 \quad (1)$$

where $\alpha_j = 4\pi k_j / \lambda$ is the absorption coefficient of layer j , $I_j(x) = (c \epsilon_0 n_j |E_j(x)|^2) / 2$ is the light intensity (time-averaged energy flux) in layer j at position x , c is the speed of light, and ϵ_0 is the permittivity of free space. It has been shown that integration over $Q_j(x)$ gives a measurement of the total energy dissipation in the active layer [26]. The power dissipated in layer j relative to the incident light intensity is:

$$D_j = D_{tot} \left[\int_0^{d_j} Q_j(x) dx \right] / \left[\sum_{i=1}^m \int_0^{d_i} Q_i(x) dx \right] \quad (2)$$

where $D_{tot} = I_s(1-T-R)/I_0$ [23, 24] is the total power dissipated (absorption efficiency) in the m -layer stack (not including the substrate) relative to the incident intensity I_0 , I_s is the light intensity incident at the substrate/multilayer interface [24] and T and R are the transmissivity and reflectivity of the multilayer stack respectively (not including the substrate).

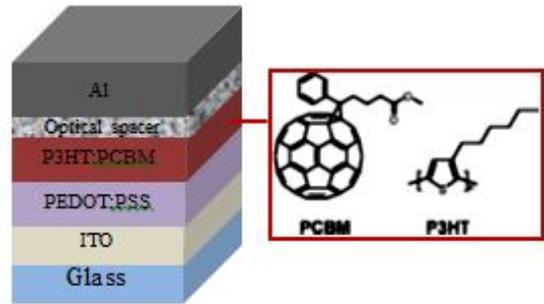
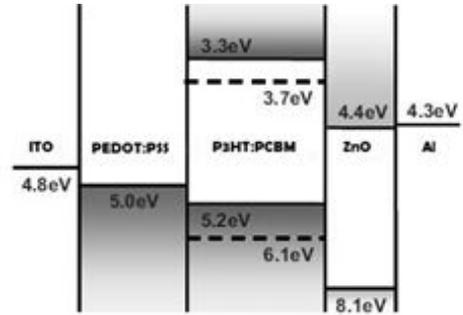


Fig. 2(a). Energy level diagram of the components of the polymer bulk heterojunction solar cell. (b) Device structure of polymer solar cell and molecular structure of P3HT and PCBM

One of the most indicative quantities calculated in the description of solar cells is the external quantum efficiency (EQE). This is a measurement of the ratio of electrons collected at the electrodes to photons incident on the device surface. EQE is typically calculated as

$$EQE = \frac{hcJ_{sc}}{I_0 \lambda q} 100 \quad (3)$$

where q is the electron charge and J_{sc} is the short circuit current density. Using the optical model presented above, another measurement of the quantum efficiency is derived. The maximum possible EQE (EQE max) for a given excitation wavelength is equivalent to the fraction of the energy dissipated in the active layer

$$EQE \text{ max} = D_{Active} 100 \quad (4)$$

EQE max is the EQE assuming that every photon dissipated in the active layer contributes to the measured current. This means that internal quantum efficiency is to be considered equal to one. The following equation results from Eq. (3) and (4):

$$J_{SC, \text{max}} = \frac{I_0 \lambda q D_{Active}}{hc} \quad (5)$$

In this paper, we calculated the variation of optical electric field distribution and the maximum short circuit current density ($J_{Sc, \text{max}}$) by inserting an optical spacer.

III. EXPERIMENTS

A. Preparation of ZnO nanoparticles

ZnO nanoparticles were prepared using an adapted procedure based on the work of Weller *et al.* [27] and Janssen *et al.* [28]. The general procedure used for the preparation of nanoparticles was as follows. At first, zinc actatedihydrate (0.5 g) was dissolved and stirred in methanol (20 mL) at 60° C for 30 minutes. A solution of potassium hydroxide in methanol (0.25 g/10 mL) was slowly added into the zinc actatedihydrate solution (It took about 10 to 15 minutes to completely add the KOH solution). Then, the reaction mixture was stirred at 60 °C for 2 hours. The resulting solution (stock solution) was washed and transferred into a plastic tube and then centrifuged at 3300 rpm for 2 minutes to separate the precipitate and mother liquor. Finally, the ZnO precipitate was dissolved in 2-methoxyethanol with the concentration of 40 mg/mL.

B. Device Fabrication

Polymer solar cells were prepared according to the following procedure. An ITO-coated glass substrate was first cleaned with a detergent, then ultrasonicated in acetone, ethanol and isopropyl alcohol, and subsequently dried in an oven. Then, a hole injection layer (PEDOT:PSS, Aldrich) was spin-cast (5000

rpm) with a thickness of ~50 nm from the

aqueous solution. The substrate was dried for 10 minutes at 140 °C in air. For the active layer, a blend of P3HT (Rieke Metals) and PCBM (SES Research) was used in a 1:1 weight ratio. This solution was then spin-cast at 1500 rpm on the top of the PEDOT layer. The prepared ZnO solution was spin-coated on the active layer with 3000 rpm in an N₂-filled glove box. The ZnO film was annealed at 80 C for 10 minutes in the glove box. Finally, Al (250 nm) was deposited sequentially under 2×10^{-6} Torr by thermal evaporation through a shadow mask to form an active area of ~ 9 mm².

C. Device and Film Characterization

The J - V characteristics of the solar cells were measured by a Keithley 2400 source meter. The J - V curves and PCE parameters of V_{OC} , J_{SC} , FF were obtained under exposure to an AM 1.5 Global solar simulator with an intensity of 100 mW/cm². Absorption of the layer was measured by a UV-vis-NIR spectrophotometer system (ocean optics HR 4000). The thicknesses of the individual layers in the cells were determined by the cross-sectional SEM images (TESCAN SEM system).

IV. RESULTS AND DISCUSSION

Figure 2(b) shows the polymer bulk heterojunction solar cell structure used in this study, which consists of a glass/indium tin oxide (ITO)/ (PEDOT: PSS)/ (P3HT:PCBM)/ an optical spacer/ Al (aluminum). The optical constants of the layers were obtained from the literature [29, 30]. Figure 3 presents the

calculated distribution of the squared incident optical electric field for two device structures with and without an optical spacer for a wavelength of 400 nm. As it is seen, $|E|^2$ is strongly influenced by insertion of an optical spacer after the active layer in the device.

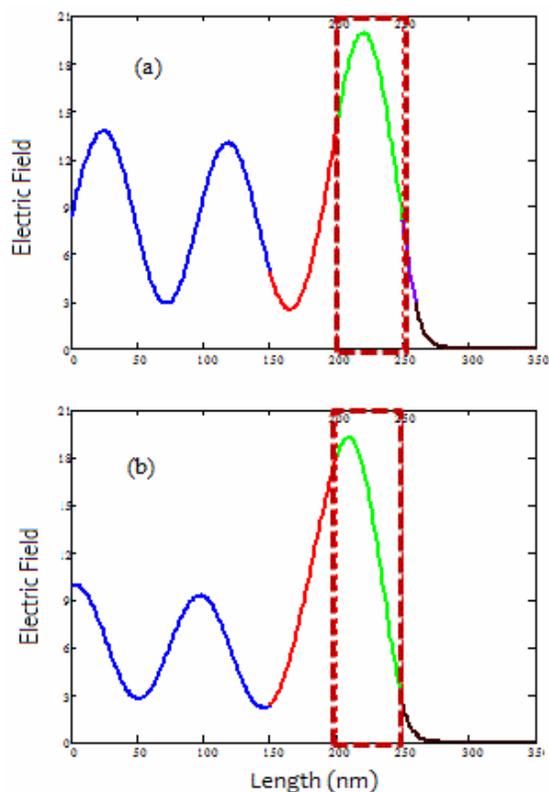


Fig. 3. Calculated optical electric field in devices with a 50 nm thin active layer, (a) with and (b) without 10 nm ZnO optical spacer.

Without an optical spacer, the position of the maximum optical electrical fields of the relevant wavelengths of the incident light is situated outside the thick photoactive layer. However, by insertion of an optical spacer, the maximum shifts into the absorbing layer. From these calculations, one would predict a significantly higher photocurrent for such a thin photoactive layer as a result of improvement in coupling. To quantify this effect, the maximum short-circuit current (JSC, max) generated in the active layer was calculated, assuming an internal quantum efficiency (i.e. the conversion efficiency of the photons absorbed to the collected charges) of

unity and under illumination of AM1.5 at 100 mW/cm².

The absorption spectra of P3HT:PCBM (10 mg:10 mg)/mL in chlorobenzene is shown in Fig. 4. The peak absorption is around 500 nm. So, the calculations are performed at this wavelength. For bulk heterojunction solar cells based on P3HT:PCBM, the calculated JSC, max oscillates with the active layer thickness due to interference effects (Fig. 5, solid line). A maximum is observed between 70 and 90 nm. By inserting a ZnO layer, this maximum is varied. The extent of this shift corresponds to the thickness of the ZnO layer. As it can be seen in Fig. 5, with an active layer thickness less than 50 nm, cells containing 10, 50 and 140 nm of ZnO show a higher current density; however, with an active layer thickness between 50 and 100 nm the devices with a 10 and 140 nm spacer generate higher currents.

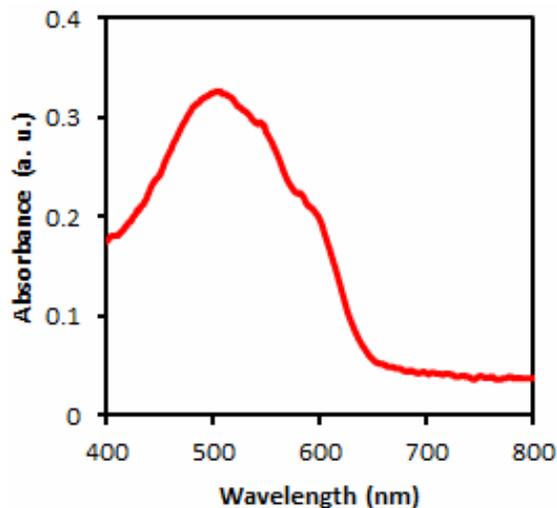


Fig. 4. The absorption spectra of P3HT:PCBM (10 mg:10 mg)/mL in chlorobenzene.

Since at a low thickness, the risk of a short circuit is high, a thickness of 90 nm was chosen for the active layer. Figure 6 represents the variation of optical electric field inside the P3HT:PCBM (90 nm) at different ZnO thicknesses. As it can be seen, the distributions of the squared optical electric field strength are more appropriate inside the devices with 10 and 140 nm optical spacers. Since these distributions were calculated for a 500 nm wave length, one expects them to result in

more light absorption inside the active layer and, thus, to result in an increase of short-circuit current density as shown in Fig 5. For devices with photoactive layer thickness of 90 nm and without an optical spacer, short-circuit current density is 11.27 mA/cm^2 . This value increases to 12 mA/cm^2 by insertion of a 10 nm layer of ZnO. The increase in the current density is due to the optical effect of ZnO. As can be seen when the thickness of active layer is optimized, increase in the current density due to ZnO layer is not significant. This prediction is consistent with report of Anderson and co-workers [31]. An optical electric field distribution inside the cell is plotted in Fig. 7. As it can be seen, by insertion of a 10 nm ZnO layer in addition to a change in the distribution of intensity, a separation occurs between the maximum intensity and the metal cathode. This separation can lead to a reduction of electron-hole pair recombination near the cathode. Hence, it is expected that a ZnO layer can also electrically improve the cell performance.

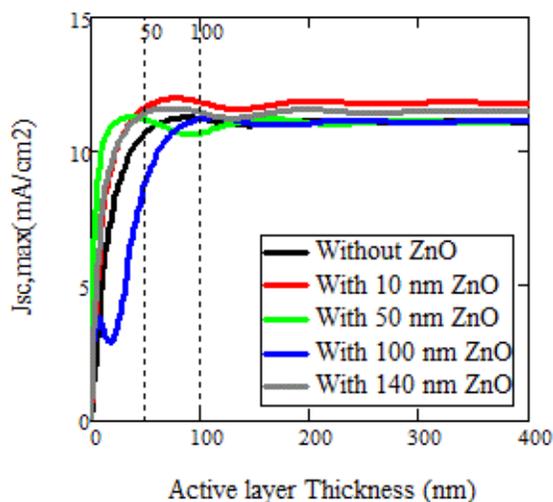


Fig. 5. Calculated short-circuit current as a function of active layer thickness assuming a 100% IQE and illumination with AM1.5 (100 mW/cm^2) for different thicknesses of the ZnO layer.

To verify the theoretical results, polymer bulk heterojunction solar cells were fabricated based on P3HT:PCBM. On the basis of these theoretical results, the optimum thicknesses of the active layer and the optical spacer were considered to be 90 and 10 nm respectively.

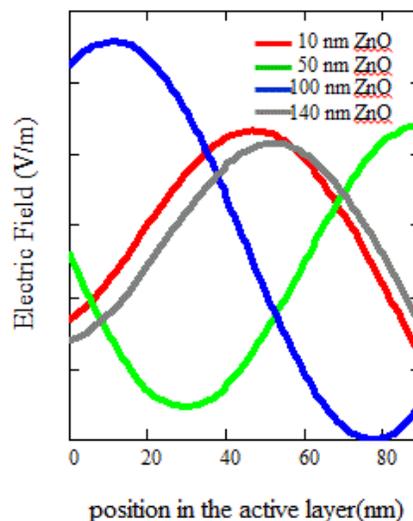


Fig. 6. Calculated optical electric field for the wavelength of 500 nm in devices with a 90 nm thick active layer for different thicknesses of the ZnO layer.

Figure 8 shows the J–V characteristics of the Bulk heterojunction solar cells fabricated with and without an optical spacer. The photovoltaic characteristics of these devices, including the short-circuit current (J_{SC}), the open-circuit voltage (V_{OC}), the fill factor (FF), and the conversion efficiency (Eff), are summarized in Table 1. As it can be clearly seen, insertion of a spacer layer has a significant impact on the cell performance. The short-circuit current density in the cell without an optical spacer is 5.7 mA/cm^2 . However, an increase up to 10.57 mA/cm^2 can be seen in the short-circuit current density due to insertion of an optical spacer. The difference between the photocurrents magnitude obtained by the model (Fig. 5) and the experimental data might be due to the fact that the actual internal quantum efficiency is less than unit (which is assumed 1 in the model calculation). However, the increase in the current density is more than theoretical prediction. This considerable increase in the current density could be due to the electrical effect of the ZnO layer.

As mentioned before, separation between the maximum intensity and the metal cathode can lead to a reduction of electron-hole pair recombination near the cathode and increases

the current density of the devices. In the device with the optical spacer experimental result gives the current density of 10.57mA/cm^2 . This value is close to the simulation result (12mA/cm^2). This is demonstrated that the light harvesting occurs by insertion of spacer layer. As a result, the ZnO layer increases absorption by shifting the optical electric field and decreases recombination by increase in probability of exciton dissociation. In fact, the increase can be explained by two aspects, optical aspect and electrical aspect. According to the optical aspect, the ZnO layer increases absorption by shifting of the optical electric field and, therefore, the current density is increased. According to electrical aspect, the ZnO layer acts as a hole-blocking layer and prevents recombination of the electron-hole pair near the metal electrode by creating a distance between the cathode and the active area.

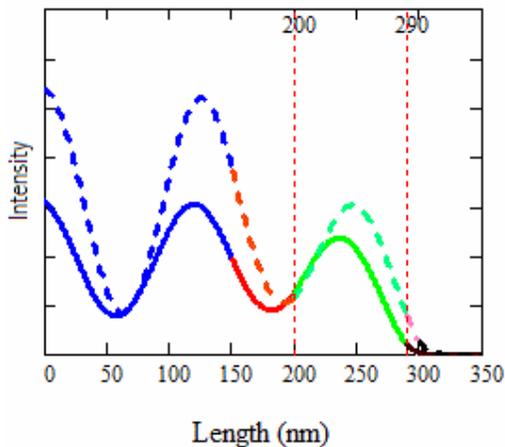


Fig. 7. Calculated distribution of optical intensity inside the device at 80 nm active layer thickness with a 10 nm ZnO layer (dash line) and without a ZnO layer (solid line).

Table 1. Characteristics of bulk heterojunction solar cells fabricated with and without an optical spacer

Device	V_{OC} (V)	J_{SC} (mA/cm ²)	FF	Eff. (%)
Without ZnO	0.58	5.7	0.55	1.87
With 10 nm ZnO	0.6	10.57	0.55	3.49

In a conventional device, a thin layer of LiF or Calcium is deposited by thermal evaporation

before a metal electrode for blocking of a hole. But in the present research, the deposition of the ZnO layer carried out by a solution process, and it eliminated the need for a vacuum deposition technique. Therefore, the device fabrication process became easier.

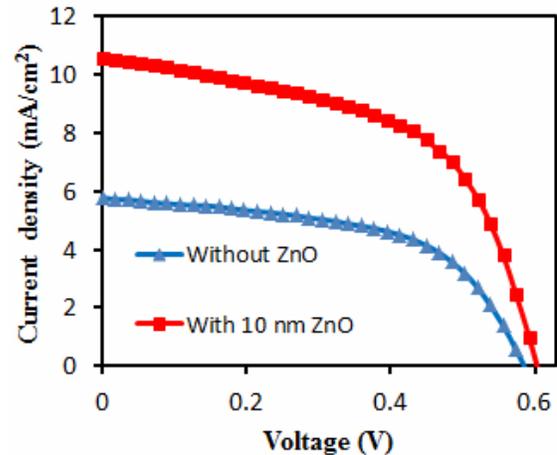


Fig. 8. J-V curves of polymer solar cells with and without an optical spacer

V. CONCLUSION

Absorption of light in polymer bulk heterojunction solar cells could be improved by optimizing the thickness of the active layer followed by inserting a ZnO optical spacer between the active layer and the metal contact. It was shown that an optical spacer can improve light absorption and cause hole-blocking at the cathode while simultaneously reducing the charge recombination. A combination of electrical and optical improvements causes the power conversion efficiency of polymer bulk heterojunction solar cells to increase up to 3.49%.

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