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Effect of thickness and temperature on flexible organic P3HT:PCBM solar cell performance

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Abstract

A blend of poly 3-hexylthiophene (P3HT) and [6, 6]-phenyl-C61-butyric acid methyl ester (PCBM) is used as a photoactive layer for simulating a bulk heterojunction organic solar using general-purpose photovoltaic device model (GPVDM) software. The optical and electrical performance of the cell had been analyzed by changing the thickness of each layer and substrate material over a range of operating temperatures from −10 °C to −40 °C. The flexible device exhibits higher PCE compared to a rigid device. The performance of the device was studied using transient simulation at different operating temperatures showing degradation in overall performance while enhancing the de-trapping of charge carrier and the recombination rate. Despite the increase in the number of absorbed photons while increasing the active layer thickness to 300 nm, a successful conversion of photon energy to electrical energy is not possible at higher thickness. The electrical simulation showed that a maximum power conversion efficiency (PCE) can be achieved with 220 nm thick active layer while decreasing the thickness of ITO and PEDOT: PSS layers due to minimization of optical losses.

1. Introduction

Organic photovoltaics is expected to be a significant source for energy production due to its advantages over silicon-based solar cells, such as flexibility, low production cost, and large-area production methods using traditional industrial roll-to-roll techniques like coating and printing [1–3]. Intensive research has been done to improve the efficiency and stability of OPV to compete in the solar cells market. Different cell structures and material compositions have been studied, demonstrating a potential for achieving high power conversion efficiency (PCE) [4, 5]. Over the last few years, a significant increase in PCE has been achieved, recording more than 13% for single-layer polymer solar cells with ITO/PEDOT: PSS as a flexible composite electrode [6] and above 18% for D18:Y6 active material on the rigid substrate [7]. Flexible Organic Solar Cells (FOSCs) are a favorable area of research as they can be adapted as a power source in different applications like wearable electronics [8] and indoor energy harvesting systems [9].

Performance of bulk heterojunction cells is highly related to the proper selection of active layer thickness, the ratio of donor to acceptor, the molecular weight of the organic material, electrode material, and even the amount and the type of organic solvent [10–12]. A proper collective selection of cell parameters is crucial for achieving optimal performance as an enhancement of a certain factor could result in declination in the overall performance. In organic solar cells, the process of photocurrent generation is more complex than inorganic cells, and it happens through multiple steps. Each is associated with a loss mechanism. The process begins with light absorption at the active layer, which results in exciton generation (bound electron-hole pair). The exciton diffuses to the interface between donor and acceptor, where it separates to free electrons and holes. After separation, the electrons move toward the electrode with higher electron affinity (EA) while holes transport to the electrode with higher ionization potential (IP) [13, 14].

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Minimization of loss mechanism is the way to increase cell efficiency. None absorbed photons in the active layer reflected light due to non-ideal transparent electrode and exciton decay in major losses. They are related to the electrode and active layer thickness. Donor-acceptor interface and its related parameters such as interface area, trap density, energetic structure, the concentration of impurity doping, and band bending determine the performance of the cell [15–18]. In addition, the distance from the interface determines the diffusion length of the semiconductor. For example, the diffusion length is about 10 nm in organic materials, which means that an exciton generated at a distance more than 10 nm from the D-A interface will not separate from charging carriers [19–21]. A thin electrode and active layer with maximum interface area between donor and acceptor material are required to match the diffusion length and minimize device resistance. The performance of the cell can be enhanced by proper selection of different layer thicknesses, use of compatibilizers [22], and annealing [23, 24]. Thermal annealing can improve the morphology of different layers, resulting in enhanced electrode transparency, conductivity [25], active layer absorption, and a higher interface area between donor and acceptor. Several simulation studies have been done to assess the performance of organic solar cells with different structures and combinations of donor and acceptor materials by incorporating different models and simulation tools [26–29]. A high efficiency exceeding 10% had been achieved by tuning electrical parameters in a simulation study of the solar cell [30]. The results look much higher than the values that could be achieved by experiments for the materials used in the study. It does not consider how far the parameters are from the experimental data.

Thermal stability is an essential point to address for practical applications of organic solar cells as these devices exhibit low environmental stability, and their performance significantly deteriorates in open environments. Different techniques have been tried to improve the device stability using interface engineering [31], polymer doping [32], tuning donor-acceptor ratio [33], and polymer side chains engineering [34]. However, most of the work focused on measuring the device’s lifetime before and after modification without relating the outcomes to the physical quantities that represent the cell model. As most of the literature links the improvements in the thermal stability to the active layer morphology, studying the effect of temperature combined with changing different layer thicknesses on device performance will be beneficial for understanding the relationship between device structure and the operating temperature.

In this work, we simulate an organic solar cell based on one of the most common and commercially available active layer materials [35] poly(3-hexylthiophene): phenyl-C61-butyric acid methyl ester (P3HT: PCBM) on flexible and rigid substrates, ITO, PEDOT: PSS as a front electrode and Al as back electrode has been assessed in terms of electrical and optical performance using General Purpose Photovoltaic Device Model (GPVDM) simulation software. Effect of each layer thickness on PCE, open-circuit voltage (Voc), short circuit current density (Jsc), fill factor (FF) was studied and analyzed. Transient photocurrent (TPC) and transient photovoltage (TPV) simulations were used to study traps and recombination in the device. The best match of different parameters is combined in a single cell structure. The cell performance was assessed in different operating temperatures considering the consistency of the device parameters with experimentally reported data.

### 2. Proposed design and simulation

An organic solar cell consists of ITO as a transparent electrode on rigid glass substrate and polyethylene terephthalate (PET) flexible substrate, PEDOT: PSS, blend of P3HT: PCBM and Al as a back electrode is simulated using GPVDM software for electrical and optical performance assessment. The device is simulated under 1 sun (AM 1.5G) illumination condition and 25 °C. P3HT: PCBM thickness had been changed from 10 nm to 300 nm with the 10 nm step. The rest of the changed simulation parameters are listed in table 1. The effect of operating temperature (T) on the cell open-circuit voltage (Voc), short circuit current (Isc), fill factor (FF) and power conversion efficiency (PCE) had been investigated. The operating temperature had been increased from −10 °C to 40 °C with an incremental step of 5 °C to investigate the trend of the cell performance in different temperatures assuming the cell will be functional for the whole temperature range.

### Table 1. Simulation parameters.

| Parameter                  | Values               |
|----------------------------|----------------------|
| P3HT: PCBM thickness       | 10 nm–300 nm         |
| PEDOT: PSS thickness       | 10 nm–160 nm         |
| ITO thickness              | 10 nm–160 nm         |
| Substrate material         | Glass/PET            |
| Substrate thickness        | 10 nm–160 nm         |
| Temperature                | −10 °C–40 °C         |
The transfer matrix method is used to obtain the exciton generation rate $G(x)$ inside the active layer by calculating the electrical field and integrating the absorbed photon distribution over the solar spectrum. The optical properties of each layer are represented by the refractive index $n(\lambda)$ and the extinction coefficient $k(\lambda)$. The refractive index is important for the estimation of reflected and transmitted light at each interface between two adjacent layers while the extinction coefficient represents the ability of a layer to absorb photons. The exciton generation rate is given by the following equations [36–38]:

$$Q(x, \lambda) = \frac{1}{2} \varepsilon_0 \varepsilon_r n |E(x)|^2$$  \hspace{1cm} (1)

$$G(x, \lambda) = \frac{\lambda Q(x, \lambda)}{hc}$$  \hspace{1cm} (2)

$$G(\lambda) = \int_{\text{Solar Spectrum}} G(x, \lambda) d\lambda$$  \hspace{1cm} (3)

where $c$ is the speed of light, $\alpha = \frac{4\pi k}{\lambda}$ is the absorption coefficient, $Q$ is the energy flow dissipation, $E$ is the electrical field at position $x$ inside material and $h$ is plank’s constant.

The software uses Poisson’s equation, continuity equation and drift-diffusion equations for electron and hole currents given by (2) to (6) respectively [39]:

$$\frac{d}{dx} \varepsilon_0 \varepsilon_r \frac{d\phi}{dx} = q(n - p)$$  \hspace{1cm} (4)

$$\frac{\partial J_n}{\partial x} = q \left( R_n - G - \frac{\partial n}{\partial t} \right)$$  \hspace{1cm} (5)

$$\frac{\partial J_p}{\partial x} = -q \left( R_p - G - \frac{\partial p}{\partial t} \right)$$  \hspace{1cm} (6)

$$J_n = q \mu_n \frac{\partial E_n}{\partial x} + qD_n \frac{\partial n}{\partial x}$$  \hspace{1cm} (7)

$$J_p = q \mu_p \frac{\partial E_p}{\partial x} - qD_p \frac{\partial p}{\partial x}$$  \hspace{1cm} (8)

where $\varepsilon_0$, $\varepsilon_r$ are permittivity of the free space and the relative permittivity of the organic blend, $\phi$ is the voltage profile, $x$ is the dimension perpendicular to the cell surface, $J_n$ and $J_p$ are electron and hole current densities, $n$ and $p$ are electron and hole densities, $R_{np}$ is the recombination rate of electrons and holes, $G$ is the generation rate and $D_{np}$ is the diffusion coefficient.

The solution is obtained for the active layer while treating other layers as ohmic components. Since the area of the device is small ($A = 2.45 \text{ mm} \times 2.45 \text{ mm}$), the 1-D simulation is valid for the device assuming homogenous composition of donor and acceptor materials [40]. The electrical parameters of the cell were obtained by fitting experimental data to the electrical model by the software developer and it is reported in the literature [41]. The fitted parameters correspond to a device fabricated using a concentration of 20 mg ml$^{-1}$ of P3HT with weight ratio of 1:0.8 of donor to acceptor and an annealing temperature of 165 °C for 10 min. We increased the mesh gradually from 10 elements up to 3 × 10$^4$ to minimize the error of the output. A mesh of 1 × 10$^4$ elements is selected for error minimization at acceptable computational speed.

Table 2 represents the electrical parameters used to simulate the organic solar cell. Although the values of electron and hole mobilities are out of the range that will result in a maximum power conversion efficiency as reported in [42]. The value we used can be trusted as it based on experimental data and values in the same range were reported [43, 44].

### 3. Materials and methods

An inverted structure organic solar cell was prepared from a blend of P3HT:PCBM with a 10 mg mL$^{-1}$ concentration dissolved in dichlorobenzene (DCB). The blend was prepared by dissolving P3HT in DCB while heating at 60 °C for 2 h, then PCBM was added to the solution to mix with P3HT. The mixture was left under the same conditions overnight. A flexible sheet of PET-coated ITO with a sheet resistance of 60 Ω sq$^{-1}$ is used as a substrate. The substrate was cleaned, and the active blend was deposited using the spin coating technique. The thickness was controlled by changing the spinning speed between 1000 rpm and 4000 rpm. A thin layer of PEDOT:PSS was deposited on the active layer at a spinning speed of 1500 rpm after filtration with a 0.45 μm filtering syringe. The top electrode was made from Ag. All materials were purchased from Sigma Aldrich. Scanning electron microscopy (SEM) is used to characterize the active layer surface.
4. Results and discussion

Investigation of electrical and optical performance of the organic solar cells by changing different layer thickness resulted in various effects on the cell $V_{oc}, J_{sc}, \eta, PCE$ and photon absorption. The macroscopic electrical parameters were obtained by the single diode model of the solar cell given by figures 1 and 9 [45] in combination with the drift diffusion model.

$$I(V) = I_s \exp \left( \frac{qV}{nkT} \right) \left( 1 + \frac{R_s}{R_{sh}} \right) = I_d \left( \exp \left( \frac{qV}{nkT} \right) \right) (V - IR_s) - I_L$$

where $V$: voltage bias, $I_d$: saturation current, $q$: elementary charge, $k$: Boltzmann constant and $T$: temperature (K).

Table 2. Electrical parameters of the device.

| Symbol | Quantity | Value |
|--------|----------|-------|
| $\mu_n$ | Electron mobility | $2.48 \times 10^{-7}$ m$^2$V$^{-1}$s$^{-2}$ |
| $\mu_p$ | Hole mobility | $2.48 \times 10^{-7}$ m$^2$V$^{-1}$s$^{-2}$ |
| $\varepsilon_r$ | Relative permittivity | 3.8 |
| $E_g$ | Band gap | 1.1 eV |
| $N_{e}^{np}$ | Effective electron trap density | $3.8 \times 10^{20}$ m$^{-3}$eV$^{-1}$ |
| $N_{h}^{np}$ | Effective hole trap density | $1.45 \times 10^{21}$ m$^{-3}$eV$^{-1}$ |
| $E_{HOMO}$ | P3HT:PCBM HOMO electron affinity | $-4.9$ eV |
| $E_{LUMO}$ | P3HT:PCBM LUMO electron affinity | $-3.8$ eV |
| $R_s$ | Series resistance | 19.5 $\Omega$ |
| $R_{sh}$ | Shunt resistance | 1.14 $\Omega$ m$^2$ |
| $A$ | Device area | 2.45 mm $\times$ 2.45 mm |
Figure 2, represents the efficiency of the cell against active layer thickness. The efficiency sharply increased from 0.16 % at 10 nm to 3.94% at 70 nm. A peak efficiency of 4.34% was obtained at a thickness of 220 nm. Although the cell with 220 nm thick active layer had the highest efficiency, the maximum $V_{oc}$ and $J_{SC}$ were recorded at 70 nm and 300 nm, respectively.

The fill factor of the cell gradually increased while increasing the active layer thickness to 40 nm with value of 0.78 and decreased gradually to a value of 0.61 at 300 nm.

At the thickness of maximum efficiency, the fill factor dropped by 15.17% from its maximum value at 40 nm $\Delta FF = FF_{40nm} - FF_{220nm} = 0.118$

The short circuit current density decreased by 3.11% from its maximum value at 300 nm $\Delta J_{sc} = J_{SC_{300nm}} - J_{SC_{220nm}} = 3.5 \text{ mA/m}^2$ and the open circuit voltage is decreased by 19.2% $\Delta V_{OC} = V_{OC_{220nm}} - V_{OC_{70nm}} = 0.12V$

The maximum thickness of 300 nm does not offer a considerable advantage as the increase in short circuit current is less significant compared to the decrease in $PCE$, $V_{oc}$, $FF$ and the cost of the photoactive material. The optical simulation obtained by TMM showed that the number of absorbed photons increases with the increase

![Figure 3. Number of absorbed photons in the active layer.](image)

![Figure 4. Photon distribution absorption for (a) 40 nm, (b) 70 nm (c) 220 nm (d) 330 nm thick active layer.](image)
in active layer thickness in a behavior close to linear as illustrated in figure 3. There are some points at which the absorption deviates from being linear. These points correspond to the range of thicknesses (200 nm–220 nm) around the point of maximum efficiency at 220 nm and the maximum $V_{oc}$ in the range of 60 nm–100 nm.

As the electrical parameters of the cell does not follow the same trend as figure 3, it means that only a portion of the absorbed energy had been transformed into electrical energy.

Figure 4 represents the absorbed photons distribution in four different active layer thicknesses (40 nm, 70 nm, 220 nm, and 300 nm) which corresponds to maximum values of $FF$, $V_{oc}$, PCE and $J_{sc}$. In the organic solar cell with 40 nm thick layer of P3HT:PCBM, the distribution of the absorbed photons is close to the electrodes, specially the PEDOT:PSS layer. If their energy is greater than the bandgap of the active layer, the possibility of charge carrier generation and collection is high which results in a higher fill factor. In cell with active area of 40 nm, 70 nm and 220 nm most of the absorbed photons have wavelength between 300 nm and 650 nm. On the other hand, at 300 nm thickness the absorbed photons have wider range of wavelengths which could explain why the maximum power conversion efficiency does not correspond to the thickness of higher number of absorbed photons as the energy of photons with longer wavelength is not enough to overcome the bandgap.

The absorption of the 70 nm layer is higher than the 40 nm although the later has a slightly higher fill factor. In 220 nm and 300 nm layers two spots of absorption appear but the number of photons close to electrodes in the 220 nm layer is higher. The performance of 220 nm layer cell is better in terms of power conversion efficiency. This indicates that the success of charge carrier’s generation and collection is more possible in the 220 nm cell as illustrated in figures 5(c) and (d).
A peak in generation rate curve corresponds to the spots that appear in the absorbed photon distribution graphs. Each peak of the 220 nm thickness has a maximum value above the HOMO where the 300 nm active layer has a single peak above the HOMO. The area enclosed between the HOMO level and the peaks are bigger than the area enclosed by the single peak of the 300 nm active layer. From the previous information the 220 nm is more efficient in exciton generation than the 300 nm thick active layer. Despite the thicker cell has higher $J_{sc}$ which could imply that it has better performance in terms of charge collection, but it comes in cost of reduction in $V_{oc}$, FF and the PCE.

The optical simulation of small thickness layers combined with the previous results explain why the thin cells have higher fill factor and open circuit voltage and they decrease with higher thicknesses until they reach a minimum value at 300 nm. As per figures 5(a) and (b), the peaks within the active material of 40 nm and 70 nm thickness approach higher energy bands near to the LUMO which directly affects the $V_{oc}$ and the FF of the two cells. Figures 6 and 7 represent the effect of changing the thickness of ITO and PEDOT: PSS on the performance of the solar cell. The decrease in the power conversion efficiency of the device corresponds to the increase in the thickness of both layers. In the ITO layer, the efficiency slightly decreased by 0.13% from a thickness of 10 nm to 160 nm at a thickness of P3HT: PCBM and PEDOT: PSS equal to 220 nm and 100 nm, respectively. This slight decay in PCE is due to the increase in the number of absorbed photons in the ITO layer, which reduces the number of generated excitons in the active layer. In PEDOT: PSS, the effect is more significant, as the efficiency significantly decreases by about 0.79%. Thus, the decrease in $V_{oc}$ can be neglected while the decrease in $J_{sc}$ is much significant. To better understand the system performance, the study of recombination in the active layer is considered. Coulomb’s attraction forces strongly bound generated charge carriers in organic photovoltaics, and they separate to free holes and electrons at the interface between donor and acceptor. The unsuccessful dissociation of the exciton to free carriers is known as geminate recombination (GR), as the generated pair decay to a ground state [46]. A common type of recombination in organic devices is non-geminate recombination. It happens when two free carriers of opposite charges generated from two different photons recombine on their way to their respective electrodes. Langevin model well describes Non-geminate recombination. The recombination rate is given by Langevin theory as the following equation [47]:
Figure 9. SEM images of the active layer surface showing the formation of donor/acceptor clusters distribution.

Figure 10. The external quantum efficiency of the solar cell.
where $g$ is the recombination factor.

The charge carrier mobility determines the recombination rate. Our system studied the effect of active layer thickness and temperature using transient photovoltage simulation (TPV). In TPV, a laser pulse is used to illuminate the solar cell with background illumination at open-circuit conditions. The rise in the open-circuit voltage corresponds to the generated charges induced by the laser pulse. The decay of the voltage is a measure of the recombination rate of charge carriers [48, 49]. Figure 8 represents the decay in $\Delta V_{oc}$ on a log scale with active layer thickness. It follows an exponential decay. As the recombination rate is proportional to the decay in the voltage, the performance of the 220 nm active layer can be explained by the low recombination rate.

Scanning electron microscopy images of the active layer surface show the formation of the bulk-heterojunction regions in relatively large clusters in areas near to the center of the cell while close to edge regions have a relatively small area donor-acceptor cluster as figure 9(a) shows. The large area clusters affect the photogeneration due to recombination of carriers while in small clusters probability of carriers generation and collection is high. The area of a large cluster is much higher than the diffusion length of the charge carriers, that’s why high recombination is expected in this sample as represented by figure 9(b). SEM images 9(c)–(i) show that the cell is highly sensitive to the ambient unwanted molecules that interact with the active layer causing degradation.

The external quantum efficiency of the fabricated device is measured which represents the ability of the cell to convert light to electric current. The external quantum efficiency is about 60% as shown in figure 10 which is in the range reported in the literature. The results support the qualitative results from the SEM data that the performance is limited by recombination.

Solar cells are supposed to operate in an open environment with a high level of disturbances in weather conditions. Thus, over a single day, a working solar cell could be exposed to a different range of temperatures.
The TPC simulation results in figure 11 show that the area under the current curve of the device increases by increasing temperature, meaning that the heat enhances the process of charge excitation and collection.

The results can be divided into two sections. The first one ends before the vertical dashed line in figure 11, represents the deep traps in the active layer. The current is decaying with a fast slope, and all curves are almost coincident with each other, meaning that the system has a small number of deep traps. In contrast, the curves get far from each other after the vertical line with a slower decay rate, indicating that the system has a considerable of shallow traps that are more dominant in limiting the device performance than the deep traps. Although transient results reveal that the device performance could be improved by heat, the PCE decreases with increasing the operating temperature from −10 °C–40 °C as in figure 12. A possible explanation could be that the improvement in the number of excited charge carriers above the HOMO due to the additional heat is not significant as the number of shallow traps in the system.

5. Conclusion

Combined optical and electrical stimulation of organic solar cell based on P3HT: PCBM photo-active blend had been carried out to study the effect of different layer thickness on the cell PCE, Voc, Jsc, and FF. The highest efficiency (PCE = 4.34%) was achieved at 220 nm thick P3HT: PCBM layer due to the absorption of high energy photons close to the electrodes, generating free charge carriers as illustrated by the outcomes of the transfer matrix simulation. The device exhibits low optical losses on PET substrate compared to glass substrate resulting in better performance. That is why we suggest the use of flexible substrates for device fabrication. The thickness of different device layers is important to the device’s efficiency. It could drop by 1% at a higher thickness of ITO and PEDOT: PSS layers due to the wide absorption spectrum of both layers combined. Although the operating temperature reduces the PCE of the device, it has positive effects on the recombination rate and excitation of trapped charges as given by transient results. Future studies will be experimental to verify the outcomes of the simulation to relate the preparation conditions of the device to the physical parameters. Parameter extraction of the fabricated solar cell will be done to compare the real device parameters extracted from the experiment to parameters used in the simulation.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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